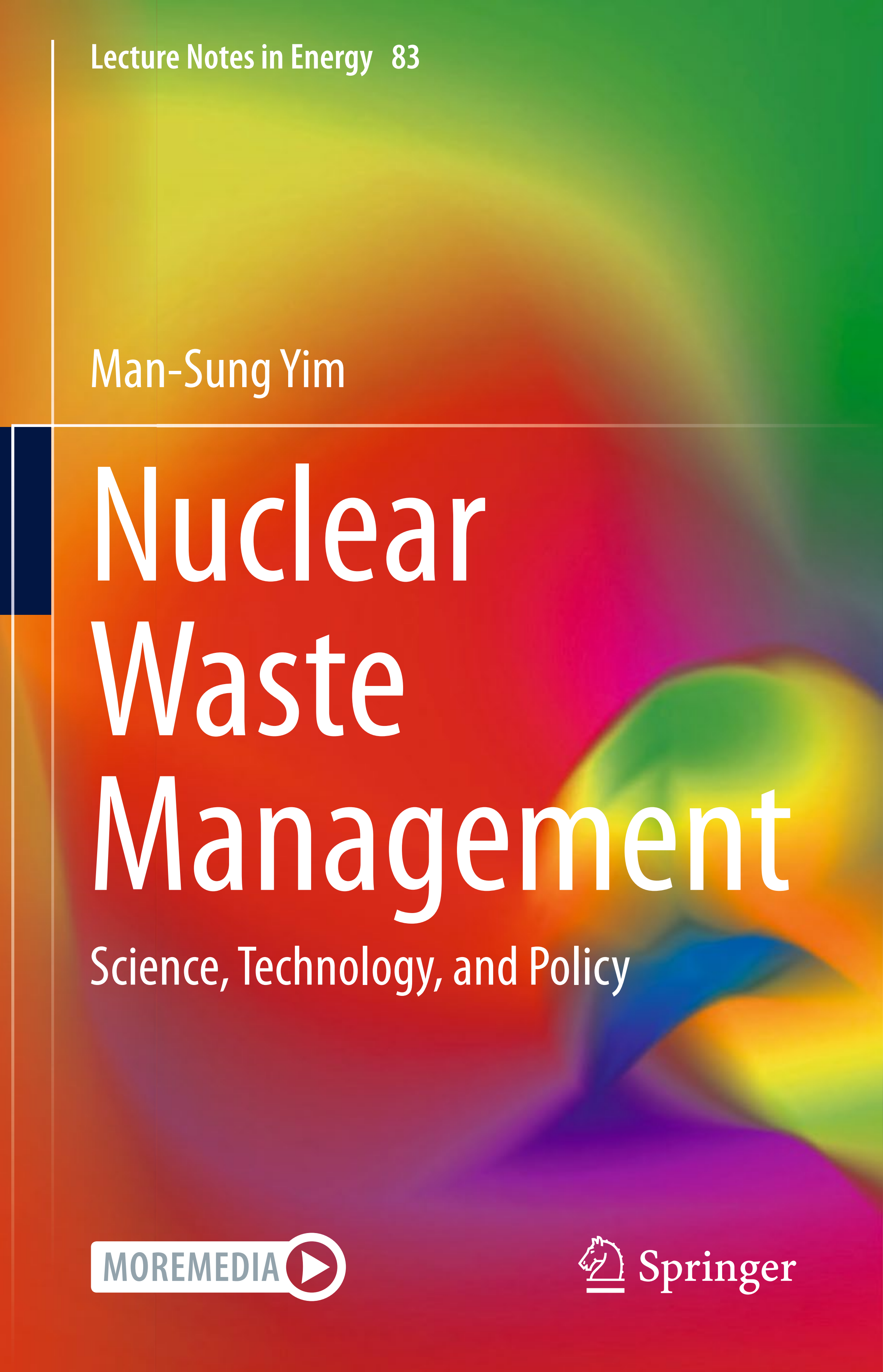

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Man-Sung Yim

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Science, Technology, and Policy

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To Soyoung, Eunji, and Minjie, and my Lord.

Preface

Technological and environmental risks are ubiquitous in today's life. Conscientiously or unconscientiously, we make decisions about risk. At the same time, dealing with risk is an unpleasant or uncomfortable matter as the word "risk" involves a situation with the potential for undesirable consequences. Risk of nuclear waste is an important example. Every country with a nuclear power program or using radioactive materials must deal with risk of nuclear waste.

Under the concern of global climate change, nuclear power is gaining renewed interests. Growth in the global use of nuclear power technology is expected. At the same time, any increase in activities in nuclear power generation implies increased nuclear waste burden on the society. If nuclear power were to make sustainable contributions to the society, the problem of nuclear waste must be solved.

Achieving success in nuclear waste management has been challenging although countries consider the problem of nuclear waste a topic of grave national importance. The challenge is in both technology and social arena. Technical challenges in nuclear waste management originate from the fact that nuclear waste can remain radioactive for very long periods of time. Time periods to be covered in nuclear waste management go beyond the typical time scales of technological enterprises. The social issues present bigger challenges. There is a large gap between the scientific community's achievements and the public's perception toward nuclear waste, although technologies for nuclear waste management have significantly advanced.

This book is a very small effort to address the challenge. When I first took a course in nuclear waste management as a student, the professor said there is no good textbook for the class. I think that is still the case today. While addressing the challenge at hand demands people with good understating of the various issues involved, studying the subject of nuclear waste management has been difficult due to the required coverage of many different disciplines. This book, as an educational tool to study nuclear waste management, covers diverse topics of relevance including basic sciences, available technologies, and the use of policy to support the overall scheme of nuclear waste management. Attempts were also made to address the connection between technical and social issues in nuclear waste management.

Although this book is mainly for graduate students in a nuclear engineering curriculum, the book may serve a reference for anyone interested in the subject matter.

This book still has limitations. The coverage is mainly based on US experiences although attempts are made to include the experiences from other countries. Specific details in some areas may not be enough to certain readers. While the use of SI units is necessary, some commonly used units (such as curie) are sometimes used in the book. Hopefully, the book can be improved through the feedbacks received.

I don't claim expertise in all areas covered in this book as this book is based on the efforts and contributions made by many. I learned the materials from my former professors and mentors, even from the students. Some are based on self-study. My goal was to provide a useful tool for the next generation of people with enough interest in the subject matter.

Using this opportunity, I would like to acknowledge the special support and mentorship of late Prof. Dade Moeller of Harvard University and Dr. Scott Simonson, the former professor of MIT. They have made very special impacts on my personal and professional growth. The generous support from Prof. Richard Lester of MIT is also very much appreciated, who developed some of the earlier education resources for the study of nuclear waste management. This book is an outgrowth of a course I have taught at MIT, North Carolina State University, and KAIST over the years. I am grateful for the emotional support and interest in this book from my former colleagues in the Department of Nuclear Engineering at North Carolina State University and my current colleagues in the Department of Nuclear and Quantum Engineering at KAIST. In particular, I am very grateful for professional support from Prof. Soon Heung Chang of KAIST (currently, president of Handong Global University). Also, this book could not have been completed without the help of many individuals. In particular, I want to mention Ms. Carol Hornibrook of KAIST for the professional friendship over the years and her contributions to Chaps. 13 and 14. The willing help from her colleagues from the US nuclear industry on those two chapters is also much appreciated. The help and support from Prof. David McNelis of the University of North Carolina, Dr. Jun Li of AREA, and Prof. Petr Vaganov of the University of Saint Petersburg are also specially noted. I also want to recognize special help from Mr. Seongwoo Kang and Mr. Junho Kwon of KAIST. Mr. Kang made significant contributions to the book by working out the solutions to many of the example problems and preparing figures and tables in the book. Mr. Kwon also made significant efforts by drawing figures, obtaining copyrights for the cited materials, and making necessary edits. The students (Ms. Youngeun Jung. Dr. Sana Ullah, Mr. Youngjun Kim, to name a few) in the Nuclear Energy Environment and Nuclear Security (NENS) group at KAIST (the students of my research group) also provided helping hands in the preparation of the manuscripts. The encouragements from my admin support team from NENS (Ms. Miryoung Kim), Nuclear Nonproliferation Education and Research Center (Ms. Jieun Choi and Yunhwa Choi), and the International Office (Mr. Eunjaee Lee, Ms. Suyun Ahn, Ms. Yeseon Kim) of KAIST are much appreciated. I would also like to recognize a number of students in my NQe541 class at

KAIST who provided feedback on the draft manuscripts of the chapters. These include Da In Choi, Wooseup Kim, Harkjoon Lee, Il Kwon Jung, Wonsuk Yang, Yunha Lee, Hiba Al-Khodire, Suood Al Mazourie, Shakirah Shukor, Guembou Shouop, Ismail Samya, Julius Federico, Rysken, Selato Peter Selato, Min Seok Kim, Jaejoon Kim, Dokyu Kang, Adi Wijayanto, Do Cong Binh, Edmund Mvula, Sayed Husain M. Abdulla, Umma Tamim, Zephania Mege, Jisu Kim, Risha Diah Rhamadhani, Andhika Prawira, Jaehyung Sim, Mustapha Faru Abdulazeez, Min Hwan Jung, Yonadan Choi, Hye-Seung Kim, Chang-Jong Kim, Hyungsan Lim, Haewan Cho, Bitna Lee, Giang Vu, Woojin Kim, Junyong Kim, Daekwon Eun, Youngho Chae, Inkyo Kim, Jangsu Oh, Jungnam Chang, Woongjae Wang, Sungwoo Yang, Munkyung Choi, Rami Ahmed, Ali Abushqair, Hoomsuk Hoorkwap, Jolly Kaitheth, Anitta Mouaxia, Machibya Matulanya, Lonah Ongayo, Mungkol Touch, Dunong Tran, Eyoel Tulu, Olof Schargberg, Jieun Joo, Hanlim Cha, Jongwook Ko, Loius Durandy, Chulmin Kim, Tiv Sothea, Seungkyu Choi, Solchan Han, Youngtaek Ji, Junghwan Kim, Yongheum Cho, Namseok Kim, Sukki Cho, Shuchang Liu, Viet Phuong Nguyen, Kyo Nam Kim, Haneol Lee, Chan Kim, Jee Min Ha, Young Ho Shin, Aznan Ismail, Chimedtseren, Kabiru Muhammed, Kristyo Rumboko, Seung Park, and Jae Hwan Yang.

Special thanks also go to my parents and parents-in-law who made my journey in nuclear waste management possible through their tireless support over the years. Lastly, but not least, I am most grateful to my wife and my two daughters, for their understanding, support, patience, encouragements, and prayers in bringing this effort to fruition. They are part of the journey and efforts embedded in this book.

The future of nuclear power technology, to a large extent, depends on the progress to be made in nuclear waste management. I hope this book contributes to the progress.

Daejeon, Soul-tʹukpyolsi, Republic of Korea Man-Sung Yim

Contents

Chapter 1
Introduction

Abstract While nuclear energy provides the benefit of abundant energy supply as a low carbon energy source, it also produces nuclear waste. This chapter describes the nature of the problem of nuclear waste and introduces overall activities conducted to manage nuclear waste. An overview of nuclear waste generation from various sectors in the world is also provided as background for the discussions in the following chapters.

Keywords Nuclear energy · Nuclear waste · Safety · Risk · Nuclear waste generation

Nuclear waste is radioactive materials as by-products or wastes from the operation of a nuclear facility or any facility using radioactive materials. Thus, it is radioactive materials produced from the use of radiation or radioactive materials, or from nuclear processes for which no further use is foreseen. The term, *radioactive waste*, is also widely used for nuclear waste. In this book, *nuclear waste* refers to radioactive waste as a more general term as it includes broader class of materials such as mixed waste that is both radioactive and chemically toxic.

Nuclear energy provides the benefit of abundant energy supply as a low carbon energy source. At the same time, nuclear energy comes with the problem of nuclear waste. The global use of nuclear power and the operation of related fuel cycle activities have produced and will continue to produce a large amount of nuclear waste. In addition, modern uses of radioactive materials in various industries and R&D institutions have led to generation of nuclear waste. Nuclear weapons production programs also produce nuclear waste. While production of nuclear waste continued over the years, progress in managing nuclear waste has been slow. Among the countries where nuclear power program started very early in their nuclear technology development, such as the U.S., the U.K., France, German, and Russia, the slow progress in nuclear waste management is particularly noticeable. None of these countries has an operating facility for permanent disposal of highly radioactive

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type of nuclear waste to date. Especially, the case of the U.S. highlights the challenges associated with nuclear waste management, where the relevant effort began in the 1950s and continued over the years with finalizing the site selection process in 2002 by announcing Yucca Mountain as the nation's high level radioactive waste disposal site. This plan was subsequently cancelled in 2010. The expectation is that the demand for the disposal of nuclear waste will grow significantly in the next several decades as nuclear power remains an importance source of low carbon energy. There are also cases of success stories such as Finland and Sweden. In these countries, significant progresses have been made after encountering major initial difficulties. Finland is expected to operate world's first geological repository for permanent disposal of highly radioactive nuclear waste within a few years. Sweden is very likely to follow the same path with a few years' time lag. These examples illustrate the challenges and at the same time possibilities in nuclear waste management. This book address these challenges and possibilities.

1.1 The Nuclear Waste Problem

Nuclear waste can be of almost no concern or extremely harmful depending on the types of nuclides included. The so-called NORM (naturally occurring radioactive materials) is an example of nuclear waste with almost no concern and the so-called spent nuclear fuel represents the opposite, the extremely harmful type of nuclear waste. Also, the problem could be short-term or very long-term depending on the half-lives of the nuclides included. For instance, most of the nuclear waste generated from hospitals are short-lived in radioactivity while the nuclear waste from nuclear power plants contain very long-lived radionuclides. Therefore, appropriately managing nuclear waste begins with understanding the hazard and time scales involved. Based on such understanding, technologies are called upon to control the hazard of nuclear waste. In this process, answers to the following questions are sought.

"What are the technologies available to control the hazard of nuclear waste?"
"How is safety ensured through the use of technologies?"

The most important goal of nuclear waste management is to provide safety. Technologies are applied to protect humans and sustain the desired level of safety in nuclear waste management. Once risk prevention or reduction is made using technologies, the methods of physical and biological sciences are used to assess the remaining risk as probabilities and consequences of events that may cause harm from nuclear waste. The remaining risk must be acceptable from the point of the people potentially affected by the presence of nuclear waste. Unfortunately, the estimated risk does not determine whether the result is acceptable by the public. The result does not dictate how people should feel about being safe.

Safety is a basic human need. It is the condition of being protected from harm. The condition of safety is verified when a person feels safe or secure. Feeling of safety depends on personal conditions and value judgments. In fact, it is possible for people to feel safe while safety is not provided or vice versa. This happens when people do not fully comprehend the level of harm they are exposed to. For example, some people use coal to heat their homes during winter while feeling safe, and die while sleeping due to carbon monoxide poisoning. Because of the subjective value judgments involved, achieving safety or agreeing on what is safe becomes a very difficult task. This is even truer when achieving safety is required for a group of people or at the societal level. Discussions of these issues are provided in Chaps. 16 and 17.

Concept: Distinction Between Hazard and Risk and Between Risk and Safety
A hazard is any agent that can cause harm or damage to humans or the environment. Risk is the likelihood for occurrence of an event that is harmful and destructive. Risk exists when exposure to a hazard occurs. At the same time, a hazard poses no risk if there is no exposure to that hazard.

Safety is the condition of being safe from harm. Therefore, safety depends on how a person feels about the risk. If the person feels free from harm of risk or secure from threat of danger, harm, or loss, then there is safety. Thus, safety depends on personal value judgment. While risk estimation is part of addressing safety, the process of risk interpretation and judgment is also important in safety. The process is driven by personal experiences and social values beyond the realm of technical science. To feel safe, the person also needs to trust the parties involved with the risk source or risk assessment.

Another important goal in nuclear waste management is to secure nuclear materials in nuclear waste away from the concern of potential misuse. As the materials such as plutonium in nuclear waste can be used for nuclear weapons purposes, the risk associated with misusing or mishandling nuclear waste must be carefully managed. The related activities are part of nuclear security and nuclear nonproliferation which are discussed in Chaps. 7 and 15.

While the majority of the radionuclides contained in nuclear waste are short-lived, some remain radioactive for very long periods of time. The half-lives of these very long-lived ones are beyond the time scales of human institutions (~1000 years). This leads to a question “how long should the effort of nuclear waste management be maintained?” Or, a more realistic question is “how long can the effort be maintained?” Science or technology may not be able to fully answer these questions.

Typically a material becomes waste when it becomes no longer useful. Nevertheless, deciding whether a material is still useful (or not) is not straightforward. The distinction between waste and resource depends not only on the reuse potential of the material but also on personal values of the decision maker. This is also true with nuclear waste. In particular, whether the spent nuclear fuels from nuclear reactors are waste or not is related to a government’s energy policy or strategy.

Development and implementation of technologies are an essential part of nuclear waste management. The technologies enable the efforts in all phases of nuclear waste management including production, handling, treatment, storage, shipment, possible reuse, and final disposal of nuclear waste. These aspects are described throughout the book (Chaps. 2, 6, 7, 8, 10). Technologies are also available to separate long-lived radionuclides in nuclear waste (e.g., reprocessing) or to transmute them into short-lived or stable ones (as discussed in Chap. 8). Use of specific technology requires the examination of the issue of performance as well as cost. As the cost can be significantly high, the resulting cost-benefit tradeoff analysis demands the discussions on how to utilize the limited resources from the perspective of achieving social goals. In that case, the process of technology selection and implementation requires social consensus.

With many actors and stakeholders involved with the management of nuclear waste, the process of achieving social consensus becomes complicated. As the problem deals with health and safety of the public in both the present and the future generations, the federal (central) government is naturally the key actor. At the same time, the waste producers, e.g., nuclear utilities, local governments, professional societies, the expert community, various interest groups, and the general public are all important part of the stakeholders. The neighboring countries may even be a stakeholder if the planned activity is near the border of a country. Having many stakeholders makes the dialogues among them very challenging. At the same time, the problem to be solved requires deliberations of diverse issues such as scientific, technical, financial, ethical, and philosophical issues. With personal or social value choices involved, the debates surrounding nuclear waste are naturally very contentious. Ultimately the problem requires government decision as public policy choice through a political process (see Chap. 2).

Once the choice is made as public policy, policy will control the process of technology development and implementation to provide safety in nuclear waste management. To specifically guide such process, regulations and standards are developed with the goals of ensuring safety of the citizens and workers and protection of the environment from nuclear waste. Therefore, demonstration of safety and environmental protection through regulatory compliance becomes an essential element of nuclear waste management (see Chap. 2).

1.2 Brief Overview of Nuclear Waste Generation

There is a total of 34.8 million m^3 of nuclear waste (as a cumulative figure) in the world as of 2018 according to the International Atomic Energy Agency (IAEA 2018). Among them, 0.022 million m^3 (less than ~0.7 million ton) is so-called high level waste (HLW), the most hazardous type of nuclear waste. HLW is generated in the form of spent nuclear fuel or in association with its recycling. Spent nuclear fuel or used nuclear fuel refers to nuclear fuels released from nuclear reactors after their service irradiation periods. These wastes contain some of the very long-lived radionuclides, thus present a very long-tern problem. In addition, about 10.3 million m^3 is so-called very low level waste and 23.9 million m^3 and 0.567 million m^3, belong to low level waste and intermediate level waste, respectively. As the names imply, very low level waste is the category of nuclear waste with very low content of radioactive materials. The next level in terms of radioactivity content is low level waste. Intermediate level waste is the one in between the high level waste and low level waste. Depending upon the contents, these wastes can present a short-

Table 1.1 Total amount of stored/disposed nuclear waste in the world (IAEA 2018)

	Solid radioactive waste in storage (m^3)	Solid radioactive waste in disposal (m^3)	Storage and disposal combined (m^3)
Very low level waste	2,356,000	7,906,000	10,262,000
Low level waste	3,479,000	20,451,000	23,930,000
Intermediate level waste	460,000	107,000	567,000
High level waste	22,000	0	22,000
Subtotal	6,317,000	28,464,000	
Total	**34,781,000**		

term or very-long-term problem. The exact definition of these wastes is described in Chap. 6 (Sect. 6.4.5). The summary of the breakdown of different waste class materials is shown in Table 1.1. Nuclear waste is also produced from uranium mining and purification of its products in support of fuel preparation for nuclear power plant. This type of waste is not included in the 34.8 million m^3 figure as it is a very low level, bulk material waste. While the exact figure of the total cumulative volume of this very low level bulk material is not available, its rough estimation is about 2 billion m^3 (IAEA 2008).

To put these numbers into perspective, a large (1150 MWe) coal power plant produces about 0.73 million m^3 of waste as ash every year. Also the world produces about 6.7 billion m^3 (~2 billion tons) of solid waste in every year (Kaza et al. 2018) among which over 1.3 billion m^3 (400 million ton) is hazardous waste (The World Counts 2020).

Besides the operations for uranium mining and processing, nuclear power plants are the largest producer of civilian nuclear wastes. The wastes include spent fuels (Chap. 7) and low and intermediate level wastes (Chap. 13). Low and intermediate level wastes refer to various byproducts of plant operations such as radioactive resin, chemical sludge, and also daily refuse such as contaminated paper, gloves, and plastics. They are generated mostly from the processing of radioactively contaminated liquids, gases, or solids. The contaminated parts, hardware, and equipment from the nuclear reactor system are also part of the waste.

The so-called nuclear fuel cycle facilities also produce nuclear waste (see Chap. 6). Nuclear fuel cycle facilities are for the acquisition and processing of uranium for nuclear reactor utilization (e.g., uranium mining facility, uranium enrichment plant, or fuel fabrication plants). They also include the facilities to handle spent nuclear fuel for packaging, storage, and disposal. The liquid nuclear wastes generated from recycling of spent nuclear fuel are of particular importance as they contain very high levels of long-term radioactivity. In general, nuclear wastes from nuclear fuel cycle facilities are solid, liquid and gaseous materials with low levels of radioactivity contamination.

There are also institutional sources of nuclear waste in both the governmental and private sector. These include scientific research organizations, medical operations using radioisotopes for diagnosis, therapy, and research, and industries producing and using radiochemical or radioisotope sources. Most of these wastes carry low levels of radioactivity and often contain radionuclides with short half-lives.

In some countries, the military sector also produces nuclear wastes through activities related to nuclear weapons program, naval nuclear propulsion system utilization, research and development, or the related fuel cycle operations.

Table 1.2 shows the breakdown of nuclear waste generation per the sources of waste up to early 2000s based on the cumulative global inventory of nuclear waste (IAEA 2008). The largest source of nuclear waste is uranium mining and related processing covering more than 99.8% of the total. Among the waste from uranium mining and processing, 86% is from the civilian nuclear fuel cycle with the rest from defense and nuclear weapons related uranium production. Excluding the waste from uranium mining and processing, low and intermediate level waste takes up about 89% of the total volume while spent fuel and HLW cover the rest (11%). Among the low and intermediate level waste, about 30% is generated in association with nuclear power plant operation, about 55% in association with nuclear weapons production, and the remaining 15% from institutional activities.

1.3 Conclusion

Nuclear waste management covers a wide variety of issues and activities, including defining what is nuclear waste, assessing its risk, agreeing on the level of safety to be provided, determining who is responsible or who are to be protected, developing and applying necessary technologies, demonstrating safety performance of technologies, and engaging in activities related to social communications.

The technical community believe that the necessary technologies required for the management of nuclear waste have been developed and are available. In contrast, popular opinion among the public is that there are no solutions to the problem of nuclear waste. This gap between technological progresses and social perception divides the discussions surrounding nuclear waste. It is necessary to examine each side’s perspectives and to discuss if such gap can be narrowed. Such effort is needed to ensure that today’s nuclear waste does not put undue burden on the present as well as the future generations.

Table 1.2 Breakdown of global inventory of radioactive waste by IAEA, cumulative up to early 2000s (IAEA 2008)

Waste source	Low and intermediate level waste		Spent fuel		High level waste		Waste from uranium mining and milling	
	Volume (m^3)	Activity (TBq)	Mass (Metric ton of heavy metal)	Activity (TBq)	Volume (m^3)	Activity (TBq)	Volume (m^3)	Activity (TBq)
Nuclear fuel cycle	2.2×10^6	1.2×10^6	1.8×10^5	2.8×10^{10}	3.4×10^4	4.2×10^7	1.6×10^9	2.8×10^4
Institutional activities	1.1×10^6	7.0×10^5						
Defense and weapons related	4.0×10^6	7.0×10^5				3.1×10^7	2.5×10^8	4.6×10^3
Total	$\mathbf{7.3 \times 10^6}$	$\mathbf{2.6 \times 10^6}$	$\mathbf{1.8 \times 10^5}$		$\mathbf{8.3 \times 10^5}$	$\mathbf{7.3 \times 10^7}$	$\mathbf{1.8 \times 10^9}$	$\mathbf{3.3 \times 10^4}$

Homework

Problem 1.1 List key activities involved in nuclear waste management. Briefly explain the importance of each activity for overall management of nuclear waste.

Problem 1.2 People argue that there are no solutions to the problem of nuclear waste. The technical community claims that the necessary technologies required for the management of nuclear waste have been developed. Discuss why such differences might exist.

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Chapter 2
Policy and Regulations for Nuclear Waste Management

Abstract Management of nuclear waste is largely a public policy problem requiring decisions by government. Policy for nuclear waste management defines goals, responsibilities, procedures, appropriation of resources in a national program while addressing various ethical issues involved. Policy also guides and supports technical activities including technology developments necessary to effectively support nuclear waste management. This chapter describes the process of policy making and how to conduct policy analysis. Development of laws, standards, and regulations as part of national policy for nuclear waste management is also discussed.

Keywords Goals · Responsibilities · Procedures · Policy making process · Policy analysis

As mentioned in Chap. 1, integration of various disciplines and activities are essential for nuclear waste management. Such integration is required for the formation of relevant policy toward the specified goals. Various aspects considered for the integration are described in this chapter.

2.1 Role of Policy

As stated in Chap. 1, management of nuclear waste demands decisions. The decisions cover a wide spectrum of issues. Examples of such decisions are reflected in the following questions.

"*What constitutes nuclear waste?*"
"*How should nuclear waste be disposed of?*"
"*What is the level of safety to be maintained in managing nuclear waste?*"
"*What is the role of local government vs. federal government in nuclear waste management?*"
"*What is the role of the public in managing nuclear waste?*"

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Answering these questions require not only scientific/technical expertise but also consideration of social, ethical and political aspects of the issues. Also, as various stakeholders are involved, answers to these questions will depend upon who is asked. Eventually government makes decisions to resolve the disagreements among the parties involved.

Public policy (hereafter, simply called policy) is a decision made by government. It is a decision made by government to achieve social goals within the national political and institutional context. The decision includes choices over allocation and mobilization of resources. Government of different ideological orientations or worldviews would make different choices with respect to setting goals and how its resources are to be utilized toward the goals.

Policy settles many of the disputes including the debates on the goals of nuclear waste management. As the resources are limited, securing necessary resources is also within the scope of policy. Policy also guides and supports necessary technological developments. Therefore, policymaking precedes any technical activities in nuclear waste management.

History tells that past mistakes in nuclear waste management were largely due to lack of appropriate national policy (OTA 1985). Note that generation of nuclear waste started in the 1940s and 1950s in several countries with the development of nuclear technology. However, national efforts for developing policy for nuclear waste in these countries didn't start until 1960s. Good national policy is a prerequisite for success in nuclear waste management. Policymaking enables organization and execution of a range of necessary institutional activities, thus controls the overall structure of and progress in nuclear waste management.

2.2 How Policy Is Made

Policy is a decision made under uncertainty for social problem solving. This is the very fact of policy for nuclear waste. Significant areas of scientific and societal uncertainty exist in dealings with nuclear waste mainly due to the very long-term nature of the problem and involvement of many stakeholders.

The process of policymaking attempts to address the issue of uncertainty or conflicts through participation of various actors in different stages. Thus policymaking represent complex interactions of individuals, institutions, ideas and interests. The stages begin with selection and screening of agenda as part of so-called policy cycle. The policy cycle is the process through which public policy is designed and implemented for problem solving. It includes agenda setting, policy-formulation, decision-making, policy implementation, policy evaluation, and policy termination or policy change as depicted in Fig. 2.1.

The actors, both government and societal actors, pursue their interests constantly in the cycle. Here government refers to the elected officials in the executive branch and the legislative branch and the appointed officials or career bureaucrats in the executive branch. Societal actors include individuals, interest groups, research

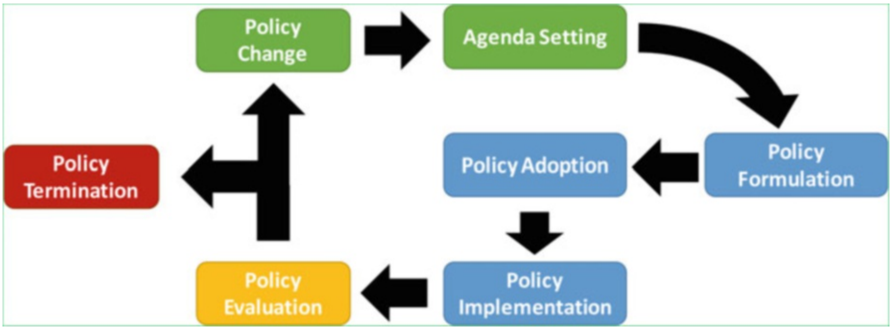


Fig. 2.1 The policy cycle

organizations, mass media, or any entity with interest in affecting the policy process to have their own policy goals or preferences embodied into the policy outcome.

As the stages of policy cycle move toward final decision-making, actors involved change with varying degrees of influence. In agenda-setting, a wide variety of government and societal actors participate to draw attention to their own agenda. In policy formulation, the ones with relevant expertise and connections from both government and society join the process. For policy adoption, only government actors with entitlement to make decisions in the relevant area, the so-called policy makers, participate. For example, in the case of the U.S., these government actors in nuclear waste management (as explained in Sect. 2.4.1) include the Congress (i.e., the Senate and the House of Representatives), Department of Energy, Environmental Protection Agency, Nuclear Regulatory Commission, Department of Transportation, and Department of Interior (also see Example 2.5). State and local government are also be included depending upon the provisions regarding the distribution of authority. The judiciary branch and political institutions such as political parties also play important roles in shaping and sustaining public policy.

2.2.1 Agenda Setting

Agenda-setting is the stage where problems come to the attention of government. While there could be many issues raised by various groups among the public demanding the attention of government, only a small proportion of them passes the screening to get serious attention of public officials. Therefore, government's acknowledgment of the existence of a public problem is agenda setting. The screening will be affected by political, social, cultural and ideological factors as well as the level of awareness of the public on the related issues. While various government and societal actors interact in this stage, outside players such as think tanks, academics, journalists, or citizens can make contributions. The contributions can be in the form of publishing an article or book, by broadcasting a media report, or even personal demonstration. This phase is also strongly affected by occurrence of a sensational event. Oftentimes politicians intentionally create a crisis to set off the stage. Sometimes values and beliefs held by societal actors drive agenda-setting prior to government paying attention.

Publication of the book "Silent Spring" by Rachel Carson in 1962 is an example of how a belief held by a person can be so influential and lead to public policy development. The book awakened the public and the scientific community regarding the environmental damage caused by pesticide (i.e., DDT) uses. This eventually led to the promulgation of National Environmental Policy Act (NEPA) in 1970, marking the beginning of the environmental stewardship era in the U.S. The Environmental Protection Agency (EPA) was established in 1970 as a federal agency to be responsible for protecting the nation's environment from industrial sources.

In the case of the issue of nuclear waste in the U.S., the necessary national policy was not in place at the start of the nuclear industry. While production of nuclear materials and nuclear waste as byproducts began in 1940s and 1950, there was no relevant national policy governing the management of nuclear waste. Although the problem of nuclear waste was recognized, the issue did not reach a stage for serious policy discussions until late 1960s and the 1970s. During this period, a few key people had to make short-term stopgap decisions to deal with continuing buildup of nuclear waste without guidelines from the government. For example, large volume of highly radioactive wastes produced from the treatment of spent fuels at the Hanford Complex in Washington, were stored temporarily in storage tanks without long-term management plan. The spent fuels from plutonium production reactors were reprocessed as part of nuclear weapons program. Many of the storage tanks were later found to be leaking but without any foresight to cope with the problem. The experts in the nuclear sector at that time viewed nuclear waste problem as something that could be readily solved when necessary by applying existing technology. The pressing challenges of nuclear technology development and building enough nuclear weapons stockpile against the Union of Soviet Socialist Republics (U.S.S.R.) at that time were of highest priority. Therefore, management of nuclear waste received relatively little attention from policymakers (OTA 1985). The fire in Rocky Flats plant in Colorado in 1969, as explained in the next subsection, could be considered an agenda setting-off event in this regard for policy making for nuclear waste in the U.S. The event drew enough political attention to the issue of nuclear waste creating a sense of crisis.

In the case of the policy for low level radioactive waste in the U.S., the process for policy development was initiated by an artificial crisis created by the states with operating radioactive waste disposal facilities. As a political movement, three states (South Carolina, Nevada, and Washington) who had operating low-level waste facilities temporarily closed the sites in 1979. These states didn't want to become *de facto* national radioactive waste dump sites with lack of federal attention on low level radioactive waste. During the closure, many of the generators of radioactive waste such as hospitals, research laboratories, universities, and nuclear utilities had to keep the wastes on site for storage with possible closure of facility operations lacking storage space. This incident pushed the Congress to notice the issue and eventually to promulgate the Low Level Waste Policy Act of 1980.

2.2.2 *Policy Formulation*

In the policy formulation stage, government agrees on the existence of a problem and explores policy options and formulate them. The goals to achieve are examined and the related courses of actions to follow are determined. Impacts of the policy are assessed with the considerations of cost implications and reactions from key stakeholders. The process of policy formulation requires the participants' understanding of the causes and ramifications of the problem and searching for solutions. Which actions are considered possible or feasible within the political context or power balance constraints of the system are identified. Options that are believed will not work or perceived to be unacceptable to the powerful actors are eliminated. This stage can be viewed as defining the structure of the policy. The process is often very contentious and subject to a wide variety of pressures as it serves to screen out options with active participation by various interest groups.

As mentioned, occurrence of a fire at the Federal nuclear weapons components facility in Rocky Flats, Colorado, in 1969 was a significant event in the U.S. for nuclear waste management. The facility was a place for the manufacturing of the plutonium metallic core of the implosion type nuclear weapon. The fire left a large volume of low-level, plutonium-contaminated waste (the so-called transuranic waste). The wastes were sent to a site in Idaho for temporary storage (i.e., the National Reactor Test Station). However, the state officials in Idaho thought that their State might end up becoming a dumping ground for waste from Colorado. Senator Frank Church of Idaho demanded Atomic Energy Commission (AEC) Chairman Glenn Seaborg to quickly remove the waste. This pushed AEC to quickly search for a geologic repository site.

Going back in time, AEC earlier began addressing the problem of waste disposal in 1955 through a study by National Academy of Science (NAS). NAS examined a scientific base for the nation's program for nuclear waste management and published a report in 1957. In the report, disposal of high-level nuclear waste was considered technologically feasible through using geological formations and that stable salt formation would be the most promising repository medium. Following this, salt became the geological formation of choice in the U.S. In the 1960s, Project Salt Vault was started in an abandoned salt mine at Lyons, Kansas, to test the effect of waste/heat loading on salt. Experiments were conducted from 1965 to 1967 by using 14 spent fuel assemblies and several electrical heaters. Results showed no measurable effects indicating the adequacy of the salt medium for spent fuel disposal. At that time, there was the atmosphere of goodwill among Federal, State, and local officials. Public tours of the mine were given during the experiment and State and local officials were consulted about various aspects of the experiment.

With the push for finding a disposal site quickly from Idaho, AEC announced in 1970 that the Lyons site had been selected for the first full-scale nuclear waste repository. The decision did not have full endorsement from the state and local government. Confirmatory test results were also pending. State and local officials intensely opposed the announcement. Further investigations found that there were many mining boreholes drilled at the site for resource exploration purposes. Disappearance of a large volume of water flushed into a nearby mine was noted. Subsequently the project was cancelled. The incident became a major milestone event undermining the credibility of federal government to carry out the mission of waste disposal.

By the late 1970s, the problem of isolating nuclear waste drew major attention by the federal government. Government began to allocate substantial personnel and funds while increasingly recognizing the non-technical aspects of the problem. Extensive search for additional repository sites was made. At the same time, efforts for examining the option of building retrievable storage facilities for spent fuel were conducted. Choices between geological disposal and interim storage was discussed at the Congress. It was noted that many of the members of the Congress had conflicts of interests as they were often from the potential host states of the facilities.

Throughout this period, some of the key questions of nuclear waste management were debated. Examples of such questions include,

"*Should the spent fuel be recycled?*"
"*Should a centralized interim storage facility for spent fuel be developed before final disposal?*"
"*How should we go about selecting a site for final waste disposal?*"
"*What should be the balance of authority between federal, state, and local jurisdictions in decision making?*"
"*What is the future use of nuclear energy in the country (as this question is related to the amount of nuclear waste to be produced and disposed of)?*"

In the case of the U.S., most of these questions were answered through the promulgation of national policy on nuclear waste in 1982 (as discussed in the next section).

2.2.3 Policy Adoption

Policy adoption refers to government selecting a particular option or course of action for public policy. This is the decision-making stage of the public policy process. The decision-making involves choices from a relatively small number of alternatives identified from the policy formulation stage. Analytical techniques (e.g., decision analysis, cost benefit analysis) can be used in this stage to assist decision-making. Policy adoption is the most overtly political stage in the policy cycle. Potential solutions to a given problem are winnowed down for final selection with respect to the goal of the policy. Apparently many peoples' interests are colliding and competing in this process. Political competition, bargaining, negotiations, and compromises often play a much bigger role than analytical techniques of decision analysis.

In the management of nuclear waste, the goals can be captured in four categories:

1. Minimize adverse impacts on human health
2. Minimize adverse environmental impacts
3. Minimize adverse socioeconomic impacts
4. Achieve economic efficiency

These objectives are complementary but also competitive. Although achieving the goals can be pursued with varying degrees of priority, human protection takes the highest priority in nuclear waste management. This is reflected in the use of the so-called "health-based" approach in national law, regulations, and standards in nuclear waste management. Other approaches, such as the "technology-based" or "incentive-based" approaches, are rarely considered in dealings with nuclear waste.

Regarding the objective of human protection, the people to be protected includes not only the present day population but also the future generations. Also the present day people includes the general public, people living near the waste management facility, or the workers handling the nuclear waste materials at the facility. The future generations could be the generations in the immediate future or the generations in the remote future. The local public will care more about protecting themselves while environmental groups may equally emphasize protecting the general public and the future generations. The industry may care more about protecting workers.

Adverse environmental impacts can include a very diverse set of possible impacts on the ecosystems. The impacts could include aesthetic impacts in the affected area, adverse archaeological, historical, and cultural impacts, and damages to species of plants or wildlife that are desirable, unique, biologically sensitive, or endangered. Socioeconomic impacts cover potential social or local economic disruptions (e.g., disruptions to existing business patterns), commercial, residential, or agricultural displacement (e.g., impacts of in-migrants on the lifestyle and the values of current resident properties), and adverse impacts on water or other natural resources. Economic efficiency is one of the main goals of public policy as conducting the necessary activities requires spending resources that are limited with the opportunity cost involved.

Adequate guidelines in the political process must be available to arrive at technically robust and socially acceptable decisions. Plus, to assure the outcome as socially acceptable, the actors must gain and maintain public trust.

Passage of Nuclear Waste Policy Act in 1982 is a key milestone of policy adoption in the U.S. While the debates on nuclear wastes continued in 1970s, there was a recognition that the long-term uncertainties and strong doubts about the federal government's capacity to cope with the nuclear waste problem were obstacles to making major progress. Apparently, a comprehensive policy was needed with a focus on solving the final isolation problem of nuclear waste while addressing institutional as well as technical issues. To overcome policy instability with changing administrations, the policy had to be acceptable and credible to all concerned parties.

In 1980, Senate and the House each passed nuclear waste bills but failed to reach agreement in conference negotiations. The Senate bill emphasized development of long-term, monitored retrievable storage (MRS) facilities while the House bill focused on developing mined geological repositories. There were also disagreements about balancing the power between States and the federal government with respect to siting of waste repositories. The environmental and anti-nuclear groups opposed long-term retrievable storage pushing for the solution of permanent disposal.

Nuclear Waste Policy Act (NWPA) of 1982 included provisions for both interim storage and final disposal but emphasized siting, constructing, and operating permanent disposal facilities for high-level waste and spent fuel. The Act implicitly confirmed that spent fuel is nuclear waste. The Act established collection of the nuclear waste fund from the utilities to cover the necessary funding and created the Office of Civilian Radioactive Waste Management within the Department of Energy (DOE) to carry out the program. The Act also included mandatory schedules for siting and licensing two separate geological repositories, with a site for the second to be selected within 3 years of selecting the first disposal facility. DOE had to select three candidate sites for the first national HLW repository.

2.2.4 Policy Implementation

Policy implementation is the process of policies being carried out. Typically, implementation follows ‘top-down’ approach. The top-down approach starts with government’s decision, examines the extent and instruments of policy, and executes the policy dealing with the public and private actors. The alternative ‘bottom-up’ approach starts with the public and private actors involved. Based on identifying and examining their personal and organizational goals and strategies and the network of contacts, ways to move forward are developed to reach policy goals including financing and execution of relevant activities. Policy implementation in nuclear waste management typically follows the top-down approach.

The implementation step involves the use of various policy instruments as actual means of implementation. Policy instruments include laws, regulations, standards, taxes and fees, subsidies, or other economic incentives. Laws are the rules or statues made by government. Regulations are the rules as instructions on how laws are to be enforced. Standards are principles or performance requirements established by government agency to support implementation of laws. Taxes and fees, subsidies, or economic incentives can be used to secure financial resources or to provide economic incentives to push the necessary developments for policy implementation. Which policy instruments are utilized depend on the nature of the problem and social, economic, technological, and political contexts of policy implementation. In the management of nuclear waste, important policy instruments are laws, regulations, standards, and fees.

Policy implementation is also affected by the nature and capabilities (e.g., political and economic resources) of the affected target groups and social actors. Through support or opposition, powerful social groups or the local public can condition the character of implementation. Changes in social and economic conditions of a country can also affect the manner in which the program is implemented. Variations in political circumstances, the status of technology or availability of new technology can also cause changes in the way policies are implemented.

After the NWPA established procedures for the disposal of spent fuel and HLW, DOE nominated five sites, Yucca Mountain in Nevada, Richton Dome in Mississippi, Deaf Smith County in Texas, Davis Canyon in Utah, and Hanford in Washington, in the order of preference. The order of preference was based on the outcome of conducting site investigations and an elaborate decision analysis. Out of these, DOE recommended three sites in 1986 as candidates for the first geologic repository; Yucca mountain in Nevada, Deaf Smith county in Texas, and Hanford in Washington. This choice of three sites was heavily criticized by experts, the affected states, and a congressional committee. Key issue under contention was exclusion of Davis Canyon and Richton Dome and inclusion of the Hanford which was expected to be rejected according to the results of decision analysis. There were suspicions that DOE allegedly distorted the analysis results under political influence. This caused DOE to lose much of its credibility.

For the consideration of the second repository, DOE examined various areas in 17 states including New York, New Jersey, Connecticut, Pennsylvania, Massachusetts, Rhode Island, Vermont, Maryland, South Carolina, Michigan, Maine, New Hampshire, Virginia, North Carolina, Georgia, Wisconsin, and Minnesota. The first 10 states were later eliminated mostly due to oppositions by the states including passing state legislation banning site investigations or disposal of nuclear waste. Public hearings conducted in the remaining seven states faced intense public opposition. This was followed by President's decision (Ronald Reagan) to suspend all site work for the second repository. The decision was accepted enthusiastically by the seven affected states. DOE announced the cancellation of all site-specific work in the candidates' sites.

DOE also recommended Oak Ridge, Tennessee for MRS. However, the Senator of Tennessee (Al Gore) opposed the plan and withdrew the state from the consideration for MRS.

Congress noted that DOE was making political rather than technical decisions and that the stakeholder consensus represented by the NWPA was unraveling. U.S. Congress then decided to amend the NWPA in 1987. The Amendment asked DOE to characterize only the Yucca Mountain site to determine suitability as a repository. It voided DOE's effort to construct a monitored retrievable storage facility at Oak Ridge, TN and establish Office of Nuclear Waste Negotiator to seek a voluntary host site for the MRS facility. It made MRS construction contingent upon the licensing of Yucca Mountain repository. The Amendment asked DOE to report to Congress on the need for second repository between 2007 and 2010.

DOE completed a "viability assessment" of the Yucca Mountain site in December 1998, followed by a draft environmental impact statement (EIS) for the project in

July 1999. The EIS examined the characteristics of the Yucca Mountain site to meet the necessary safety requirement. On August 21, 2001, DOE issued a report stating that Yucca Mountain is acceptable as geological repository meeting the EPA and NRC requirements. EPA issued its final Yucca Mountain standards on June 6, 2001. NRC then revised its repository regulations September 7, 2001. On February 14, 2002, along with the submission of the final EIS, DOE recommended the site to the President as the nation's HLW geological repository. The President (George W. Bush) recommended the site to Congress on February 15, 2002. Nevada Governor (Kenny Guinn) submitted "state veto" April 8, 2002. Overturning the state veto, the House (on May 8) and Senate (on July 9) passed an approval resolution. Then the President signed the resolution on July 23, 2002 finalizing the site selection process.

On June 30, 2008, DOE submitted license application to U.S. NRC with the final repository system performance assessment for Yucca Mountain. U.S. NRC has conducted license reviews with the initial target of authorizing repository construction by September 2011. The corresponding target date for spent fuel receipt was May 2017.

2.2.5 Policy Evaluation

Policy evaluation is the process of examining the outcome of policy implementation by monitoring and evaluating the results. Such monitoring and evaluation are done by both government and societal actors including anyone with interest in the policy. Based on such evaluations, a policy may be labelled a success or a failure, followed by demands for continuation or change. Thus, policy evaluation serves the purpose of active learning on the part of policy actors with the objective of improving the policy. The outcome may either support or change the policy through re-conceptualization. Reconceptualization may include minor changes or fundamental reformulation of the problem, including termination of the policy. The process is concerned with budgets, priorities, efficiencies, expenditures, and any legal issues relating to policy implementation. Such evaluations could involve the judiciary. The judiciary would be concerned with possible conflicts between government actions and constitutional provisions or the established standards of administrative management and individual rights.

In the U.S., development of geologic repository is still on-going and NWPA has not been completely executed. One of the issues noted was annual budget appropriation for the Yucca Mountain project and its impact on the license review process. More prominently, the newly elected Obama administration in 2009 stopped the funding request to the congress for the Yucca Mountain project. As of September 30, 2016, DOE collected $25.4 billion from the utilities as the nuclear waste fund. Including cumulative interest earnings and other revenue (approximately $24.8 billion), the total amount collected becomes $50.2 billion. However only $11.4 billion was actually spent for the Yucca Mountain project (as of September

30, 2016). This leaves $38.8 billion as balance. However, the balance is only in the record and has been absorbed by the federal government to make up for the federal budget deficit. In March 2012, a bill was introduced in the Senate requiring three-fourths of that money to be given back to customers, and the remainder to the utility companies for spent fuel storage.

In August 2013, the U.S. Court of Appeals for the District of Columbia ordered the Nuclear Regulatory Commission to either approve or reject DOE's application for [the] never-completed repository site at Yucca Mountain. The court opinion recognized that a federal law designating Yucca Mountain as the nation's nuclear waste repository remains in effect and noted that the NRC's inaction allows the Obama administration to continue plans to close the proposed waste site. Consensus was building within the Congress and among the nuclear utilities that the repository program needs to be revived.

2.2.6 Policy Change

The policy cycle continues beyond policy implementation and evaluation. Depending upon the outcome of policy evaluation, policy may go through changes or be terminated. These changes may be due to the pressure from interest groups, client complaints, legal challenges, changing financial conditions, or facing the reality that the policy cannot deliver solutions to the problems at hand. The changes can be in the form of replacement by a different but similar policy, consolidation with a similar policy, splitting into smaller pieces, or a drastic change in the approach to the problem.

Policy changes did take place in the U.S. with the cancellation of Yucca Mountain at the nation's geological repository site. In 2006, Senator (Harry Reid) from Nevada became Senate Majority leader who opposed the development of Yucca Mountain. In 2008, both of the leading Democratic presidential candidates (Senators Hilary Clinton and Barak Obama) stated that, if elected, they would end the Yucca Mountain project. This was perceived as part of political deal with the Senate majority leader. When Senator Obama was elected as President, he did indeed cancel Yucca Mountain project in 2010. This represents a non-linear change in policy due to variations in political conditions.

Since 2017, the Trump administration has been asking Congress to approve funding for Yucca Mountain project in an attempt to revive the project. The House of Representative responded with allocation of budget but the Senate refused to allocate any funding arguing that an interim storage facility needs to be developed first. Actual allocation of Yucca Mountain project budget remains to be seen.

The Energy Reorganization Act of the U.S. in 1974 is also considered an example of policy change. The Act replaced AEC with DOE and Nuclear Regulatory Commission (NRC) for promotional and regulatory functions for nuclear energy, respectively. The change addressed the issue of conflict of interest inherent in the governing structure of AEC to guide the use of nuclear technology and to manage its byproduct wastes.

2.2.7 *Policy Termination*

Policy termination is cessation of specific policy. Termination could be through ending a specific government program or abandoning responsibility. In reality, public policy rarely solves a problem completely. Societal actors may not allow the termination. Thus, few programs are ever completely terminated. The Obama Administration's attempt to cancel Yucca Mountain project is an example of policy termination. However, as NWPA remains in effect in the U.S. and as the need for the disposal of spent fuel and nuclear waste persists, policy effort for nuclear waste will continue.

Figure 2.2 captures key milestones in the history of nuclear waste management and related activities in association with national policy development in the U.S.

2.3 Policy Analysis and Decision-Making Models

Policy analysis is the process of examining and evaluating available options to achieve the goals. The analysis can be qualitative or quantitative, or both. Qualitative approach can take the form of interviews or case studies to develop generalizable observations. Quantitative approach includes survey research, statistical data analysis, model-based simulation, cost-benefit analysis, or decision analysis.

2.3.1 *Policy Analysis*

A common practice of policy analysis includes the steps of defining the problem, identifying alternatives, evaluating options, and making a choice. Defining problem is to specify the problem and the goals in the given national context. Identifying alternatives is to lay out the possible options. Evaluating options is based on predicting the consequences of executing each option and using criteria to rank them. Making a choice is to recommend one of them as the best policy option. Conducting such analysis is usually an iterative process.

A case of simple policy analysis for spent fuel management in the ROK is briefly described in Example 2.1.

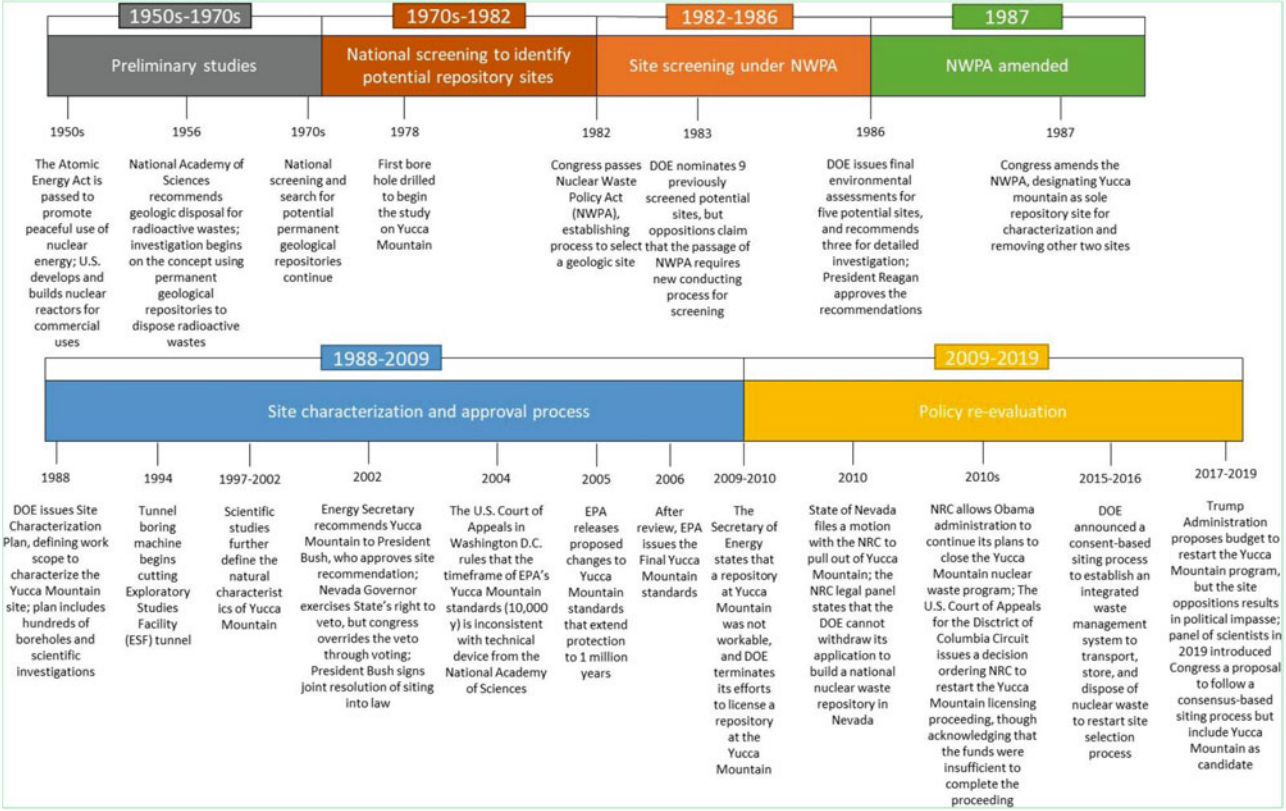


Fig. 2.2 Major timelines of the U.S. Nuclear Waste Management Policy. (Sources of the information: Nevada Legislature 2018 and Eureka County Yucca Mountain Information Office 2019)

Example 2.1: A Problem for Policy Analysis: Spent Fuel Management in the ROK

Define the Problem

The Republic of Korea (ROK) as of 2017 operates 24 nuclear power plant units with 23.1 GWe capacity . This corresponds to ~22% of ROK's total electrical generation capacity and 31.2% of total electrical consumption. Annually, the country produces 750 ton of spent nuclear fuel, totaling 16,000 ton of spent nuclear fuel accumulated from its nuclear power plant fleet.

All of the spent nuclear fuels discharged from the reactors have been in storage in water pools with expectation for permanent disposal in the near future. One exception is that at the Wolsung plant, a dry storage facility, called MACSTOR, is under operation in addition to the wet storage pools. While the country's nuclear power program has grown successfully, the long-term plan for spent fuel management has not been finalized. In the meantime, the spent fuel storage pools at each reactor site are reaching their capacity limits. A plan for spent nuclear fuel management is necessary for sustainable use of nuclear power in the ROK.

How should the country manage its spent nuclear fuels? Developing the necessary plan requires policy analysis.

Identify Alternatives

The options may include expanding the capacity of the storage pools at each reactor site, building separate on-site storage facilities, building a centralized national storage facility off-site, building a permanent disposal facility, or building a treatment facility for recycling of spent nuclear fuel as an alternative to direct disposal. Options such as overseas treatment of spent nuclear fuel or permanent disposal through the use of an overseas facility could also be considered.

Evaluate Options

Each of the options has pros and cons in terms of cost, time required, immediate impact on alleviating storage capacity shortage, public health impact, feasibility of implementation, etc. These pros and cons must be carefully laid out and compared among different options. The comparison could be through quantification of cost and benefits in monetary terms or through establishment of utility measures. The comparison could also be based on simple heuristic analysis. Aggregation of utility measures require weighting of different measures.

Make a Choice

The option that provides the maximum utility or the maximum benefit to cost ratio within the bounds of constraints (e.g., technical or political feasibility) would be selected. The ROK has yet announced the selection of national policy.

In practice, the process of policy decision making can be characterized by using different conceptual models. These include the rational model, the incremental model, and the garbage can model.

The rational model refers to the procedures for decision-making being rational. It is based on the belief that the problem ought to be solved in a 'scientific' or 'rational' manner. Each of the steps of policy analysis is closely followed. The resulting choice among the options represents the most efficient approach to achieve the goals. To support the comparison of alternatives, quantitative techniques such as cost-benefit analysis (CBA) or decision analysis are often employed. Unfortunately, the outcome from applying quantitative techniques are rarely followed due to limitations in practical implementations imposed by the involvement of various stakeholders.

The incremental model refers to decision-making as a political process characterized by bargaining and compromise among the actors. What is rational, in this case, depends on the situations. The choice made represents what is politically feasible rather than desirable. The outcome does not necessarily achieve lofty goals but solves the problem in practical sense.

The garbage can model reflects essentially non-rational process of decision-making. Use of science and rationality is avoided in the process. Goals are defined by the actors and their interactions. Policy decisions are made as the policy makers go along in a process.

2.3.2 Cost-Benefit Analysis

The process of conducting a cost-benefit analysis (CBA) is similar to the steps of policy analysis. Essential component of CBA is in the assessment of impact of each option, both favorable and unfavorable, in monetary terms. The favorable impacts are considered benefits and the unfavorable ones as costs. The net benefit is total benefit minus total cost. The alternative with largest net benefit is the preferred choice.

One practical issue in conducting CBA is that the benefits and costs of options may not occur at the same time or accrue over a period. Thus, consideration of time value of money is necessary. Time value of money indicates that a monetary value in current possession may be worth more than the same amount of monetary value in the future as the money in current possession can earn interest income in the future. The value of investment P_0 at present time (at time period 0) after n years with added interest will become

$$P_n = P_0 \times (1 + r)^n \tag{2.1}$$

where

P_n = principle amount at the end of the nth period
P_0 = original principal amount at period 0

r = rate of interest
n = number of period (or years, if interest is compounded annually)

Thus, the present value of the investment P_0 in terms of the future value including interest becomes

$$P_0 = \frac{P_n}{(1+r)^n} \tag{2.2}$$

For example, $1,331,000 received 3 years from now would be worth $1,000,000 today given an annual interest of 10% (with annual compounding), i.e., $ 1,000,000 × $(1.10)^3$ = $ 1,331,000. The present value can be viewed as the amount that must be invested today to realize a specific value in the future.

When a fixed-amount of cost occurs every year, such amount needs to be combined to determine the total cost. This could be treated as an annuity problem (i.e., money is received each year as a series). If the first payment is received one period from the present and equal payments are made at the end of consecutive periods over a fixed length of time, the annuity is called an ordinary annuity. The present value of an ordinary annuity is the sum of the present values of each annuity received over N years:

$$P_0 = \frac{A}{(1+r)^1} + \frac{A}{(1+r)^2} + \frac{A}{(1+r)^3} + \frac{A}{(1+r)^4} + \ldots + \frac{A}{(1+r)^N} \tag{2.3}$$

where A is the annual payment and r is the interest rate.

Then

$$P_0 = \sum\nolimits_{t=1}^{N} \frac{A}{(1+r)^t} = A \sum\nolimits_{t=1}^{N} \frac{1}{(1+r)^t} \tag{2.4}$$

If different payments are made each year, the present value (P) of all of the payments made each year t up to year T is

$$P = \sum\nolimits_{t=0}^{T} \frac{P(t)}{(1+r)^t} \tag{2.5}$$

where $P(t)$ is the payment at year t. For the cost and benefit occurring every year, the present value of net benefit (benefit minus cost) becomes,

$$NPV\ (net\ preset\ value) = \sum\nolimits_{t=0}^{T} \frac{B_t - C_t}{(1+r)^t} \tag{2.6}$$

where B_t and C_t are the benefit and cost in year t, respectively, and T is the time horizon for the evaluation.

In a discounted cash flow analysis, the analysis to estimate the value of an investment today based on projections of how much money to be generated by the investment in the future, the term, discount rate, is often used in the place of interest rate. Discount rate refers to the interest rate used in so-called discounted cash flow to determine the present value of cash.

From the comparison of cost and benefit, best alternative is the one that maximizes the present value of all benefits minus all costs. This is illustrated in the following example.

Example 2.2: Cost-Benefit Analysis

The following table gives an example of comparing costs and benefits of two different projects. In the case of Project A, there is no initial benefit and initial costs are much higher, but large benefits are accrued in the later years. In contrast, Project B provides constant benefits from the beginning with relatively low initial cost. This example illustrates the comparison of cost and benefit with the consideration of time value of money.

	Project A		Project B	
Year	Benefits	Costs	Benefits	Costs
0	0	15	7	2
1	0	8	7	2
2	0	5	7	2
3	5	3	7	5
4	10	3	7	5
5	60	3	7	5
Total	**75**	**37**	**35**	**21**

If we compare the projects based on simple comparison of total cost and benefit, Project A appears a better option as it gives the total net benefit of $75 - 37 = 38$ compared to $35 - 21 = 14$ from Project B.

With the consideration of time value of money (assuming the discount rate at 10%), the present value of the net benefit is:

$$\mathrm{PV_A} = (0-15) + (0-8)/(1+0.1) + (0-5)/(1+0.1)^2 + (5-3)/(1+0.1)^3 + (10-3)/(1+0.1)^4 + (60-3)/(1+0.1)^5 = 15.27$$

$$\mathrm{PV_B} = (7-2) + (7-2)/(1+0.1) + (7-2)/(1+0.1)^2 + (7-5)/(1+0.1)^3 + (7-5)/(1+0.1)^4 + (7-5)/(1+0.1)^5 = 17.79$$

(continued)

Example 2.2 (continued)

Based on the results, Project B gives higher present value of the net benefit when net benefits are calculated up to Year 5.

Caveat: The example does not present any data beyond Year 5. If the trend observed in the table continues beyond Year 5 and if longer-term gains beyond 5 years are to be considered, Project A could be chosen over Project B. As seen in the results, Project A's benefit is rapidly growing at the end of the evaluation period in the example while Project B's benefit is diminished.

2.3.3 Decision Analysis

Public expenditure decision making is to allocate government resources for public goods. Such decision-making requires explicit representation of the interdependence among the major expenditure components, impacts of the spending, and the applied constraints. Decision analysis (DA) supports such decision-making through quantitative analysis with the consideration of uncertainties. Uncertainties in the decision and its outcomes are captured in a decision tree by using multiple branches with the likelihood of each branch represented by probability. Merits of each decision option are represented through quantification of expected net benefit. The decision whose consequences has the maximum expected (probability weighted) net benefit is recommended as the preferred choice.

The sequence of a decision analysis is as follows.

- For a decision, develop a list of all feasible alternatives.
- Develop a list of all possible outcomes associated with each alternative.
- Make an estimation of the probability associated with each possible outcome.
- Make an evaluation of the consequences (i.e., net benefit or utility) associated with each combination of alternative and outcome.
- Determine the probability weighted utility of each alternative.
- Choose the alternative with the maximum expected net benefit (or utility).

A simple example of decision analysis is given below.

Example 2.3: Decision Analysis

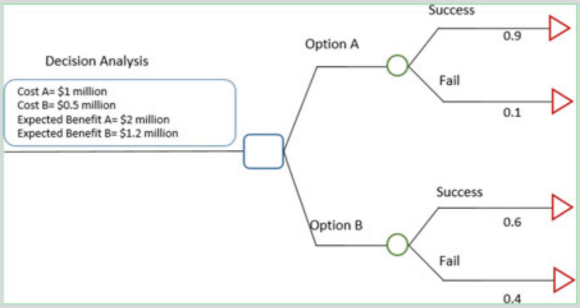
Government has two options (A or B) to solve a public health problem. The probability that option A will be successful is 90% while the probability of success of option B is 60%. The cost of option A is \$1 million while the expected benefit is 2 million if successful. The cost of option B is \$0.5 million with the expected benefit is 1.2 million if successful. Decide which option to choose.

(continued)

Example 2.3 (continued)
Expected net benefit of A
= probability of success*(expected benefit − cost) + probability of failure*
(expected benefit − cost)
= 0.9*(2 − 1) + 0.1*(0 − 1)
= 0.8 million

Expected net benefit of B
= 0.6*(1.2-0.5) + 0.4*(0-0.5)
= 0.22 million

Thus A is preferred.



Difficulties exist in using CBA or DA to support policy development as quantifying impacts of policy options often involves subjective judgments. The quantified uncertainty in DA itself is also uncertain. Conceptualization of the problem by using models to support the analysis may not represent the reality. Nevertheless, CBA or DA offers valuable insight in decision making by forcing the analyst to quantitatively characterize the ramifications of different decisions.

In reality, even if CBA or DA correctly identifies the best option, people may disagree over the findings. In fact, disagreement is inevitable in every policy choice as there will always be losers with the policy choice while some become winners. Some policy decision makers are reluctant to use quantitative approaches and resort to the political process for decision making.

2.4 Development of Laws, Standards, and Regulations

Once government makes up a policy, the policy is announced and put into practice by using policy instruments. Key policy instruments used in the management of nuclear waste are laws, regulations, and standards. These policy instruments are to answer the following questions (as mentioned in Chap. 1).

- Q1 What is nuclear waste?
- Q2 Who are the responsible parties for the management of nuclear waste?
- Q3 How safe is safe? What is the acceptable level of risk?
- Q4 Who should be protected?
- Q5 How long should the prescribed level of safety be provided?
- Q6 What approach should be used to assure safety from nuclear waste?
- Q7 How do we verify safety?
- Q8 How should the environment be protected in comparison with protecting public health?

2.4.1 National Law

The most important policy instrument is national law. Developing national law provides comprehensive and sustainable legal framework to address the challenges of nuclear waste management. National law(s) specifies goals in general terms and the directions to take and answers the first two questions (Q1 and Q2): The law defines nuclear waste and designates entities with specific responsibilities while leaving technical details to administrative agencies. These details become standards/regulations.

Agencies with responsibilities for the development of standards and regulations or development and/or operation of facilities are specified in the law. National law would also define the selection of the method for final disposition of nuclear waste and/or the need for centralized storage of spent nuclear fuels. The related institutional issues covered by the law are as follows:

- Designating agencies with necessary responsibilities and authorities
- Procedures for decision making with timelines (e.g., for activities such as development and operation of final disposal facility)
- Coordination and management of projects (including intergovernmental interactions or local vs. federal government issues)
- Balance of authority among government agencies and between the central and local government
- Provisions of funding (how funding is generated and allocated)
- Process for public involvement
- Any related international activities

Developing national law(s) is the role of the legislative branch (Congress, National Assembly, Parliament, etc.). The legislative branch also controls program implementation by reviewing, authorizing, and appropriating resources.

In the case of the U.S., the Nuclear Waste Policy Act (1982, amended in 1987) provides four cornerstones that form the foundation of a comprehensive program:

1. Detailed schedule milestones for all major decisions related to the federal storage and disposal facilities.

2. Extensive procedures for state and public participation in all major decisions for such facilities.
3. Establishment of the Nuclear Waste Fund.
4. Establishment of a separate office within the DOE, Office of Civilian Radioactive Waste Management, to direct the program.

2.4.1.1 What Is Nuclear Waste (Q1)?

Nuclear waste (or radioactive waste) is defined by national laws as the definition depends on government's position regarding what is considered waste. In the U.S., Nuclear Waste Policy Act (U.S. Code Title 42, Chapter 108, Nuclear Waste Policy) defines various types of nuclear waste as described below (with additional clarifications made by the regulations of the relevant regulatory agency).

Spent Nuclear Fuel (SNF): SNF is what has been withdrawn from a nuclear reactor following irradiation whose constituent elements have not been separated by reprocessing. (Spent fuel is regulated as HLW under 10CFR 60, as described in the next section).

High-Level Waste (HLW): HLW is the highly radioactive material resulting from the reprocessing of spent nuclear fuel, including liquid waste produced directly in reprocessing and any solid material derived from such liquid waste.

Transuranic (TRU) waste: TRU waste is defined as waste material that is contaminated at low level with alpha-emitting isotopes with atomic number $Z > 92$.

Uranium (U) mill tailings: U mill tailings are the residue from the physical and chemical processing of uranium ore after the production of U_3O_8 concentrates.

Low-Level Waste (LLW): LLW is all radioactive wastes other than spent fuel, HLW, TRU waste, and uranium mill tailings (other countries call this class of waste low and intermediate level waste). LLW includes class A, class B, class C, and greater than class C (GTCC) (see Chap. 13).

Mixed waste: Waste that contains both hazardous chemical components and radioactive components.

Naturally occurring or accelerator-produced radioactive materials (NARM): This includes naturally occurring radioactive materials (NORM) and accelerator waste.

The U.S. NWPA has three titles. Title I deals with repositories for disposal of HLW and SNF, an interim storage program, monitored retrievable storage (MRS), and GTCC LLW. Title II deals with research, development, and demonstration related to disposal of HLW and SNF. Title III deals with other provisions related to nuclear waste. The Act also permitted co-disposal of defense HLW in the same repository with civilian spent fuel and HLW.

Many countries along with the U.S. consider spent nuclear fuel a waste as no further use of it is planned or considered. In contrast, several countries (e.g., France and Russia) exclude spent fuel from waste as recycling and reuse of spent fuel is part of the nation's nuclear power program. Classification of nuclear waste in more general terms is made by the International Atomic Energy Agency as discussed in Sect. 6.4.5.

Example 2.4: Distribution of Nuclear Waste in Volume and Activity in the U.S.

Following the definition of nuclear wastes in the U.S., contributions of different classes of nuclear waste (from both civilian and military activities) to the total inventory of radioactivity and volume produced in the U.S. are shown in the following table. The data in the table are based on the cumulative waste produced through 1991. Spent nuclear fuels contain the highest inventory of radioactive materials among all types of nuclear waste. As of 2012, the activity of spent fuel from commercial nuclear power plants in 2014 remained about the same at 851 × 10^{18} Becquerel* (23,000 mega Ci*), compared to 860 × 10^{18} Becquerel (23,245 mega Ci) in 1991 (NWTRB, 2016). In contrast, the volume of spent fuel has increased from 9645 m^3 in 1991 to 90,300 m^3 in 2012. This shows that a large portion of the activity in spent fuel is short-lived and decayed away. Most TRU waste is produced by the processes involving the use of transuranic materials such as the manufacturing and fabrication of nuclear weapons. Uranium mill tailing wastes were generated through uranium milling operations. Low level wastes are generated by utilities, industry, clinical and research laboratories, and government agencies, including those that also generate HLW, TRU, and mill tailings wastes.

Generation of different classes of nuclear waste in the U.S. up to 1991 (ORNL 1992).

		Volume			Activity	
		In 1000 m^3	% (without U mill tailings)	% (with U mill tailings)	Exa (10^{18}) Bq (10^6 Curie)[a]	%
Commercial	Spent fuel	9.546	0.2	8.1×10^{-3}	860.0 (23,245)	95.8
	HLW	1.729	0.04	1.6×10^{-3}	0.962 (26)	0.11
	LLW	1423	29	1.15	0.209 (5.65)	0.02
	U mill tailings	(118,400)	–	96.0	–	0
	Total	1434 (without U mill tailings)	29.3	97.2	861.2 (23,277)	95.9
Military	HLW	395	8.1	0.32	35.9 (971)	4
	LLW	2816	57	2.28	0.497 (13.43)	0.05
	TRU waste	254	5.2	0.21	0.101 (2.72)	0.01
	Total	3465	70.7	2.80	36.5 (987.15)	4.1
Sum Total		**4899**	**100**	**100**	**897.8 (24264)**	**100**

[a] The discussions of radioactivity and its units are given in Sect. 3.1.6

2.4.1.2 Who Are Responsible for the Management of Nuclear Waste (Q2)?

The responsibility for management of nuclear waste is basically based on the "polluter-pays" principle. The "polluter-pays" principle means that those who produce pollution should pay for the costs of managing it. Therefore, management of nuclear waste (including spent fuel) is primarily the responsibility of the waste producer (e.g., the owner of nuclear power plant).

In reality, government takes the ultimate responsibility in nuclear waste management as the role of protecting the public belongs to government: The nuclear power program in any country begins by a government decision. Construction and operation of nuclear power reactors are always under government regulations. Government makes relevant and necessary decisions to ensure safety in all aspects of nuclear power program including the management of its waste.

While government takes such role, the industry as the producer of waste has to take care of the waste until the waste is delivered unto government custody at facilities for centralized storage or final disposal. The industry is also required to pay for the related cost to government. Government may delegate part of the responsibility to local government or private industry under national consensus. This is often the case for low level waste or low and intermediate level waste. Citizens as consumers of electricity bear part of the responsibility by paying electric bills as they benefitted from the services of utility companies. Division or distribution of such responsibility and related rights are specified in the national law. Balance of authority among central government and local government and local municipalities is also one of the important stipulations specified in national law. The national law specifies the extent to which state and local governments and perhaps also the residents can exert their influence on the decision. As an example, the specifics given in the U.S. Nuclear Waste Policy Act is described below.

Example 2.5: Responsibilities for Nuclear Waste Management in the U.S. Under the NWPA, the U.S. government is committed to develop geologic repository as the final phase of managing both civilian and military spent fuel

(continued)

Example 2.5 (continued)
while pursuing the development of interim storage facility. The veto power was granted to the host state for the federal repository. The Congress is allowed to override the state veto through a majority vote within 90 working days.

Overall responsibilities and activities of the US federal agencies in the development of a geological repository are summarized in the following (OTA 1985).

Department of Energy (DOE)

- Site characterization and selection
- Land acquisition
- Technology development
- Application for license
- Repository design, construction, operation, and closure
- Interactions with State/Indian tribes

Environmental Protection Agency (EPA)

- Standard promulgation

Nuclear Regulatory Commission (NRC)

- Regulation promulgation
- Siting review
- License proceedings
- Technology review
- Interactions with State/Indian tribes

Department of the Interior (DOI)

- Site characterization support
- Land access
- Land withdrawal

Under the NWPA, the EPA was asked to develop relevant standards for spent fuel management. For low-level waste, the DOE and states are responsible for site selection, building, and operations of the waste disposal facilities. The industry is required to pay the annual fee to DOE as the nuclear waste fund for the final disposal of spent nuclear fuel at a federal geologic repository.

In 1987, this law was amended by the Nuclear Waste Policy Amendment Act, which redirected the program to phase out site-specific activities at all candidate sites except for the Yucca Mountain site and added 5 years to the schedule for the first geologic repository.

Balance of authority between the central and local government became an issue in the U.S. when the federal government started losing trust from the public in the

1960s. A number of states and municipalities began taking actions in 1970s against federal decisions on siting nuclear waste facilities. Quite a few municipalities restricted transportation of radioactive wastes within city limits by passing laws. In California, construction of additional nuclear power plants was prohibited by a state law in 1976 pending development of a federal program to manage nuclear wastes. A similar actions were taken by several other states. Following the trends, states passed laws preventing site investigations for geological repository development or banning disposal of nuclear waste within the state's territory. In the late 1970s, states and localities were promised to have a right of "consultation and concurrence" on nuclear waste policy by the Carter administration. However, the regulations issued in 1980–1981 by US NRC and Department of Transportation during the Reagan administration prevented localities from blocking transportation of radioactive waste on approved routes. This was slightly changed in the 1982 Nuclear Waste Policy Act, allowing the veto power to a host state for the federal repository development (along with the power to the Congress to revert the veto through a majority vote).

2.4.2 Regulations and Standards

As stated, standards are performance requirements established by a government agency. Establishing standards starts from qualitative general principles related to the safety goals. These principles are translated into quantitative measures to serve as performance targets (i.e., standards).

The standards are adopted by the regulatory agency and translated into specific set of rules as regulations for enforcement. Therefore, regulations are the details of technical specifications or requirements to meet the standards. These regulations are used to protect the citizens and workers as practical regulatory limits.

Through regulations and standards, government controls the detailed practices in nuclear wastes management to achieve overall net social benefit. Specific issues addressed as part of regulations and standards for nuclear waste management are discussed in the following sections.

Example 2.6: Regulations and Standards for Nuclear Waste Management in the U.S.

Under the NWPA, the U.S. Environmental Protection Agency (EPA) was charged with the responsibility for developing standards for spent fuel management and disposal. The corresponding regulations were to be developed by the U.S. Nuclear Regulatory Commission (NRC).

EPA was created in 1970 by the National Environmental Policy Act with responsibility for air and water standards, limits on pollutants, and control of

(continued)

Example 2.6 (continued)
radioactivity. The Atomic Energy Act also gives EPA the power to establish generally applicable environmental standards for protection of general environment from radioactive materials.

The following is the list of EPA Standards with relevance to nuclear waste management.

- **40CFR190**: *Environmental Radiation Protection Standards for Nuclear Power Operation*
- **40CFR191**: *Environmental Radiation Protection Standards for Management and Disposal of Spent Nuclear Fuel, High Level Waste, and Transuranic Radioactive Waste*
- **40CFR192**: *Health and Environmental Protection Standards for Uranium and Thorium Mill Tailings*
- **40CFR193**: *Environmental Radiation Protection Standards for Management and Land Disposal of Low Level Radioactive Waste (Draft)*
- **40CFR197**: *Public Health and Environmental Radiation Protection Standards for Yucca Mountain, Nevada*

The Nuclear Regulatory Commission (NRC) was created in 1974 by the Energy Reorganization Act which split the Atomic Energy Commission (AEC) into two agencies. The NRC licenses and regulates the possession, use, transportation, handling, and disposal of radioactive materials, including wastes under EPA standards. It has the principal power to issue or deny licenses to those who wish to produce, use or possess these regulated materials. In this role, NRC has developed the various sets of regulations that the license holder needs abide by. These regulations can be viewed as the detailed practical interpretations of EPA's standards by NRC. Therefore, if you satisfy the NRC regulations, you are expected to satisfy the EPA's standards automatically. The following is the list of major NRC regulations relevant to nuclear waste management.

- **10CFR20:** *Standards for Protection from Radiation*
- **10CFR51:** *Environmental Protection Regulations for Domestic Licensing and Related Regulatory Functions*
- **10CFR60:** *Disposal of High-Level Radioactive Wastes in Geologic Repositories*
- **10CFR61:** *Licensing Requirements for Land Disposal of Radioactive Waste*
- **10CFR63:** *Disposal of High-Level Radioactive Wastes in a Geologic Repository at Yucca Mountain, Nevada*
- **10CFR71:** *Packaging and Transportation of Radioactive Material*

(continued)

Example 2.6 (continued)

- **10CFR72:** *Licensing Requirements for the Independent Storage of Spent Nuclear Fuel and High-Level Radioactive Waste, and Reactor-related Greater Than Class C Waste*

For example, the dose limit of 50 mSv (5 rem) per year is specified for occupational workers in 10CFR20 (section 1201). Here, dose refers to the energy deposited per mass of the body of a person (see Sect. 5.2.2). The unit of dose is Sv (or rem).

2.4.2.1 How Safe Is Safe? What Is the Acceptable Level of Risk (Q3)?

While the risk from nuclear waste is managed to ensure safety, a related question posed is “How safe is safe?”. If “safe” means absolutely no risk, then none of technological system is “safe”. As a matter of fact, every technology comes with certain level of risk. In fact, the risks we face in daily life cannot be totally eliminated, no matter how much efforts are made. Driving, walking along the street, eating healthy food, or even breathing natural air entails certain level of risk. In this regard, zero- risk or absolute safety is an impossibility.

If “safe” means “reasonably safe” or “safe enough to feel secure”, rather than being absolutely safe, then the question becomes – how safe is “reasonable” or “safe enough”? The answer is related to the concept of acceptable risk level.

De Minimis Risk

One example of defining the level of acceptable risk is to define *De Minimis* risk. *De Minimis* risk originates from Latin words *de minimis non curat lex* which means the law does not concern with trifles. It implies that, below some level of risk, it is not worth the allocation of social or personal resources to address the problem. Therefore *De Minimis* risk is the level of risk below a level of regulatory concern.

Defining such risk level as *De Minimis* was attempted by the US Nuclear Regulatory Commission as part of its effort in setting nuclear safety goals. In 1983, US NRC issued a policy statement that operation of a commercial nuclear power plant should pose, to the public, no more than one tenth of one percent (0.1%) of the background risk to which the public is normally exposed including both acute fatality and latent cancer risks (NRC 1983).

At the time of this announcement, the background individual accidental acute fatality risk in the U.S. was about 1 in 2000 per year (5×10^{-4}) so the goal was 5×10^{-7} per year. Also, background individual latent cancer risk was about 1 in 500 per year (2×10^{-3}) so the goal became 2×10^{-6} per year. In relation to the risks of getting killed in mass transportation accidents, the general public seem to ignore the risk lower than 10^{-5} excess lifetime risk. Accordingly, one in a million risk per year was considered *De Minimis*.

Comparative Risk Approach

Comparative risk analysis (CRA) is a tool to systematically compare risk from different sources for the purpose of effective risk management. An example of such approach was applied in EPA's standards making. The approach was based on controlling the risk from nuclear waste to be less than that from the nature, e.g., the risk of natural uranium. This was adopted in the radionuclide release limits for spent fuel disposal in EPA's 40CFR191. Further explanations of the limits are given in the following example.

Example 2.7: Risk from Uranium Ores
In 40 CFR191, US EPA sets up a qualitative safety goal that the risks from the high level waste repository should be no greater than those if the uranium ore used to supply the fuel for the nuclear power plants had not been mined. Subsequently the EPA developed supporting guidelines to manage the risk of high level waste to be less than that from natural uranium. The details are as follows.

Estimating the health risk from the presence of natural uranium is a very challenging task due to the uncertainties involved. The following summarizes the assumptions made as a way to address the uncertainties (EPA 1980).

- The total amount of spent nuclear fuel produced from the entire U.S. nuclear power program is 100,000 MTHM. The unit, MTHM (metric ton of heavy metal), is explained in Chap. 6, Sect. 6.2.6. The amount of natural uranium required to support such program is 620,000 MT of U_3O_8 (yellow cake). This corresponds to ~600,000 MT (i.e., 6×10^8 kg) of uranium ore.
- Health risk from the presence of uranium ore is realized when the uranium is dissolved and become part of water contamination.
- The probability of release of U and its daughters from U ore per unit time is the same as the average probability of release from earth crust.
- Ra-226 is the dominant contributor to health hazard among the nuclides produced as part of ^{238}U decay chain (see Sect. 3.1.4).
- The depth of soil contributing to the release of ^{226}Ra is 600 m. The total amount of uranium is the U.S. soil down to the depth of 600 m is 3.5×10^{13} kg.
- The average content of ^{226}Ra in the surface water of the U.S. is 2×10^{-13} kg/m^3.
- The total volume of water runoff in the 48 contiguous U.S. states per year is 1.5×10^{12} m^3/year.
- Environmental pathway analysis and human exposure and risk calculation showed that 1 Ci of ^{226}Ra release per year results in 22 cancer deaths per

(continued)

Example 2.7 (continued)
year in the U.S. The unit Ci (curie) is equivalent to the radioactivity level of 3.7×10^{10} decays per second (this will be discussed in the next chapter).

Based on these assumptions, the amount of ^{226}Ra released into rivers from the natural uranium in the U.S. is estimated as,

$$\begin{aligned} &2 \times 10^{-13}\left(\mathrm{kg/m^3}\right) \times 1.5 \times 10^{12}\left(\mathrm{m^3/year}\right) = 0.3\ \mathrm{kg}^{226}\mathrm{Ra\ \ year^{-1}} \\ &\quad = 300\ \mathrm{Ci\ \ year^{-1}} \end{aligned}$$

(1 gm of ^{226}Ra is equivalent to 1 Ci of radioactivity. This is how the unit of curie (Ci) was initially defined by Madame Curie, the discoverer of radium).

The portion of release due to the nuclear power program is,

$$(0.3\ \ \mathrm{kg/year}) * \left(6 \times 10^{8}\ \ \mathrm{kg}\right)/\left(3.5 \times 10^{13}\ \ \mathrm{kg}\right) = 5.14 \times 10^{-6}(\mathrm{kg/year})$$

Then the health risk from natural uranium ore in the U.S. is,

$$\begin{aligned} &\mathbf{5.14 \times 10^{-6}\left(\frac{kg}{year}\right)} \\ &\cdot\left(\mathbf{1000\frac{g}{kg}}\right)\cdot\left(\mathbf{1}\boldsymbol{\frac{Ci}{g}}\right)\cdot\left(\mathbf{22}\boldsymbol{\frac{cancer\ deaths/year}{Ci/year}}\right) = \mathbf{0.11}\ (\boldsymbol{cancer\ death/year}) \end{aligned}$$

Then the health risk over 10,000 years becomes, 0.11 × 10,000 = 1100 deaths.

According to this result, the risk of high level waste to be less than that from natural uranium approximately means no more than 1000 excess deaths over the first 10,000 years of repository existence.

(Note: There is large uncertainty involved in the risk estimation.)

Risk Benefit Analysis

A third way to define the acceptable risk level is through the use of risk benefit analysis (RBA). In this approach, the level of acceptable risk is determined based on quantification of risk and benefits and their tradeoffs. If the benefits outweigh the risk at the given risk level, the risk may be acceptable. According to the RBA approach, the cost associated with risk reduction is compared with the benefit. The risk level where no further risk reduction is beneficial becomes the level of acceptable risk.

This approach was adopted by the U.S. NRC in its consideration for achieving nuclear safety improvements. Both the cost of compliance and the perceived benefits were taken into account. Based on the approach, NRC started using a value of $1000 per person-rem in 1975 (U.S. Federal Register 1975). The person-rem is the unit of radiation dose as the sum of all individual doses among the affected people (the concepts of dose is discussed in Sect. 5.2.2 and also in Sect. 3.2.3 in relation to gamma rays and neutrons).

An interim policy statements on safety goals for operating nuclear power plants published in 1983 (NRC 1983) officially adopted a guideline of using a value of \$1000 per person-rem averted in decisions on safety improvement. This means that the benefit of an incremental reduction of societal mortality risks should be compared with the associated costs on the basis of \$1000 per person-rem averted (in 1983 dollars): If an effort to reduce the risk to humans by 1 person-rem costs more than \$1000, then the effort is not needed. This approach is based on comparing the cost required to avert one cancer death with the assumed value of a statistical life.

This issue was revisited by the NRC in 1995 and the value per person-rem conversion was increased to \$2000 (NRC 1995). If we consider an average inflation rate of 2.68% per year between1983 and 2017, the guideline in 1983 in the year 2017's value would be \$2455 per person-rem. However, accounting simply for the average inflation rate does not reflect other changes happening in the society. For instance, the latest US NRC thinking on the issue is to use \$5200 per person-rem, a much higher value (NRC 2017). According to this new figure and with the assumed probability of fatal cancer per person-rem exposure as 5.1×10^{-4} (National Research Council 2006), the value of a statistical life is \$10.2 million (in contrast to ~\$1 million assumed with \$1000 per person-rem). NRC also recognizes that the assumed value could range between \$2500 and \$7800 per person-rem.

This guideline is intended to encourage efficient allocation of resources in safety-related activities by recommending that the effort for the expected reduction in public risk should be commensurate with the costs of the proposed safety improvements. However, the approach is very controversial in specifying the value of human life in monetary terms. This issue is further discussed in the next section.

The US NRC emphasizes that application of such benefit-cost guideline is not necessary if existing system meets the safety requirements. In case additional measures are needed, use of RBA is recommended to guide the process of improving the system to meet all of the safety requirements.

Comparative Observations

The examples used in defining acceptable level of risk illustrate that risk acceptance may be pursued through CRA or RBA.

Use of RBA as a basis for risk management was demonstrated by Starr in a heuristic analysis (Starr 1969). By using urban dwellers as sample subjects, he examined how an individual performs a crude heuristic optimization of risk benefit tradeoff. For example, an urban dweller may want to move to the suburbs because of a lower crime rate and better schools, at the cost of more time spent driving on highways and a higher probability of traffic accidents. However, if the traffic density increases subsequently, he or she may find the cost (or risk) too high and decide to move back to the city. He assumed that such optimization in balancing the risk and benefit lies behind voluntary human behavior. Then he compared the benefit relative to risk pattern of various voluntary human activities such as hunting, skiing, smoking, riding train/air plane, etc. He also did similar analysis for involuntary activities such as using electric power. It was assumed that the contributions of the activity to the individual's annual income is proportional to its benefit and that the risk benefit tradeoff decision is made by government.

From such comparison, he found that the public would accept risk levels that are approximately 1000 times higher than involuntary risks, as voluntary risks. This could be interpreted as the public accepting involuntary risk levels that are 1000 times lower than voluntary risks. The level of 0.1% of natural background risk as *De Minimis risk* by the US NRC corresponds to this level. These numbers are also in agreement with the target risk values of acceptable risk set by U.S. Environmental Protection Agency or Nuclear Regulatory Commission as discussed earlier.

There are controversies over the ways acceptable level of risk are defined. In the case of using *De Minimis* risk approach, the issue is who decides what is acceptable. Technical experts are equipped to estimate the failure probabilities of the systems that are designed to protect humans. However, making judgment on what is acceptable is not the role of technical experts. Such judgment belongs to each individual according to his/her personal value judgment. Therefore, individual's need for personal decision on what is acceptable must be recognized. A decision by government or scientists on what should be accepted ignores such need and is most likely to face opposition.

In the case of using CRA, while the approach can be useful to prioritize the needs for risk reduction, risk comparisons may not be convincing. For example, if the risk being compared is perceived as unfair, then it is not appropriate to use such risk for comparison with what is perceived as fair. This is also true with unfamiliar risks, involuntary risks, or risks controlled by others (see Chap. 16).

A major problem with the RBA approach is to compare risk with benefits in monetary terms. The core issue lies with the ethical question of "Can the value of human life be translated into economic terms?" Most people would say "no" feeling very uncomfortable with the notion of using monetary value for human life although this is commonly exercised in insurance industry.

Given these controversies, determining the level of acceptable risk requires careful deliberation. Historical anecdotes or the review of social norms that represent collective social judgment would be useful in the deliberation. Table 2.1 shows an example. Although the data are somewhat outdated, the table shows various examples of risk sources along with the estimated annual mortality rate and the degree of social acceptability of the corresponding risk. The table lists some risks that are already "accepted" by the public as *De Facto* or some that are perceived to be high or low. For example, the risk of getting killed in mass transportation accidents is socially accepted. The corresponding risk level is in the order of 10^{-5} or 10^{-6}. Generally, people seem to accept the risk from rare but familiar events. It appears that the 10^{-5} is about the level of risk that seemed to be ignored or accepted by the general public. However, exceptions are also noted. The risk of radiation from nuclear power plant is in the order or 10^{-9} (death per year), yet is not socially

Table 2.1 Examination of risk acceptability for various risk sources (Gotchy 1977)

Risk	Order (death/year)	Risk acceptability?
Meteorite impacts	$\sim 10^{-11}$	*De facto*
Radiation from color-television viewing	$\sim 10^{-9}$	
Radiation risk to U.S. population from nuclear power plant	$\sim 10^{-9}$	Low
Radiation from high-altitude flight	$\sim 10^{-8}$	
Lightning	$\sim 10^{-7}$	*De facto*
Dike flooding (Holland)	$\sim 10^{-7}$	Low
Radiation death from consumer products	$\sim 10^{-7}$	
Steam-boiler explosion	$\sim 10^{-7}$	
Flood, tornadoes, or earthquakes	$\sim 10^{-6}$	*De facto*
Death from cholera or whooping cough	$\sim 10^{-6}$	
Train passenger death	$\sim 10^{-6}$	
Natural background radiation	$\sim 10^{-5}$	
Death from use of contraceptive pills	$\sim 10^{-5}$	
Passenger deaths in aircraft accidents	$\sim 10^{-5}$	Medium
Death from leukemia or tuberculosis	$\sim 10^{-5}$	
Brain damage from whooping cough vaccination	$\sim 10^{-4}$	
Therapeutic use of antidepressant or antihypertensive drugs	$\sim 10^{-4}$	
Overall road accident deaths	$\sim 10^{-4}$	High
Occupational deaths on railways (UK)	$\sim 10^{-4}$	
Death from bronchitis or influenza	$\sim 10^{-4}$	
Occupational death in chemical industry	$\sim 10^{-4}$	
Lung cancer risk from smoking (20 cigarettes/day)	$\sim 10^{-3}$	High
Death from all causes at age 55	$\sim 10^{-3}$	Medium
Occupational pneumoconiosis	$\sim 10^{-3}$	
Death from cancer, stroke, or heath disease	$\sim 10^{-3}$	

accepted. The risk of dike flooding is about 10^{-7} (death per year) but carries low level of social acceptance. When the source of risk incurs emotions or value concerns (as in the cases of nuclear power and dike flooding), the acceptance is low.

In the case of dealings with nuclear power or nuclear waste, defining what is acceptable requires special considerations due to the emotions and values attached to the subject matter. This is further discussed in Chaps. 16 and 17.

The following example explains how radiation protection limits are developed based on the defined acceptable level of risk.

Example 2.8: Interpretation of Acceptable Level of Risk for Nuclear Waste Management Regulations

Based on using the approach of *De Minimis risk* and comparative risk analysis, the risk deemed acceptable to the public is in the order of $10^{-6} \sim 10^{-5}$ per year.

According to the dose-to-risk conversion factor of 5.1×10^{-2} per Sv (as described in Sect. 5.2.5.2), this $10^{-6} \sim 10^{-5}$ per year risk level corresponds to $0.02 - 0.2$ mSv ($2 - 20$ mrem) per year of effective dose.

International advisory body on radiation protection (i.e., International Council on Radiation Protection, ICRP) recommends that radiation doses above background levels to members of the public should not exceed 1 mSv per year (or 100 mrem/year) effective dose for continuous or frequent exposure from radiation sources other than medical exposures (ICRP 1991). This framework is used as a basis for protecting the public health from routine or expected anthropogenic sources of ionizing radiation. It includes any exposures to the public derived from the management and storage of high-level radioactive waste and spent nuclear fuel.

The national authorities responsible for nuclear waste management could apportion, or allocate, a fraction of the 1 mSv per year to establish an exposure limit for nuclear waste disposal facilities (ICRP 1985b). Most countries have endorsed the principle of apportionment of the total allowed radiation dose. Apportionment values for high level waste disposal range from 5% to 30% in different countries. These numbers correspond to radiation doses between 0.05 mSv (5 mrem) and 0.3 mSv (30 mrem) per year. Notice these values also match the $10^{-5} \sim 10^{-6}$ per year risk level discussed above. IAEA is recommending the use of 0.3 mSv per year as its safety standards for geological repositories (IAEA 2011).

Table 2.2 National regulatory approaches to safety in nuclear waste disposal

Country	Regulatory limit	Post-closure periods of safety assessment
Belgium	Expected to be 0.1–0.3 mSv/year	May be as much as 1,000,000 years
Canada	An upper limit of 1.0 mSv/year established; 0.3 mSv per year proposed	Not specified
China	No decision made	At least 10,000 years
Finland	<0.1 mSv/year for normal events; impacts should be comparable to those arising from natural radioactive materials but should remain insignificantly low	~10,000 years
France	<0.25 mSv/year for normal scenarios	10,000 years
Germany	0.01 mSv/year for probable developments; 0.1 mSv/year for less probable developments	1,000,000 years
Japan	No decision made	No decision made
Korea, south	10^{-6}/year for major scenarios, 1 mSv/y for single scenario of low probability natural event or human intrusion	10,000 year
Spain	No decision made	No decision made
Sweden	10^{-5}/year	100,000 year
Switzerland	0.1 mSv per year	10^6 years
UK	10^{-6} per year; source-related dose of <0.3 mSv/year or a site-related dose of <0.5 mSv/year	No timeframe is specified
USA	0.15 mSv/year (up to 10,000 years); 1 mSv/year (after 10,000 years and up to 1 million years)	10^6 years

Sources: NWTRB (2016)

Table 2.2 shows the regulatory limits of various countries for nuclear waste disposal. The adopted values shown in the table are within the range of dose or risk as discussed above.

Besides the regulatory limits for nuclear waste disposal, protection of workers and the public from various activities of handling nuclear waste follows the general radiation protection guidelines. ICRP recommended the use of annual limits of 20 mSv (2 rem) for workers and 1 mSv (0.1 rem) for members of the public (ICRP 1977).

2.4.2.2 Who Should Be Protected (Q4)?

While everyone must be protected from nuclear waste, not all people in a society are expected to personally experience the risk from nuclear waste. It is necessary to specify the individuals or groups of individuals for the purpose of protection from nuclear waste.

The regulations and standards in nuclear waste management define an individual or groups of individuals as "representative" people to protect. Here a "representative" people is an average person among the people affected by the risk source or the most susceptible person who is particularly sensitive to radiation exposure at or near the facility where nuclear waste is handled, stored or disposed of.

In regulations or standards regarding human protection against radiation, this concept of "representative people" is captured by defining a critical group. The critical group is a relatively homogeneous group of people whose location and habit are such that they are representative of those individuals expected to receive the highest doses from the discharges of radionuclides (ICRP 1985a). The critical group dose is defined as that dose received by an average member of the critical group and should be lower than the dose limit specified by the regulation. The regulatory limits given in Table 2.1 are often specified for members of critical group.

Using the critical group approach requires a detailed knowledge of local habits which may not be fixed over the time period of interest. Efforts are needed to derive the habit data from detailed national or regional information based on the characterization of the affected population.

The maximally exposed individual (MEI) is another type individual used for the purpose of human protection. The MEI represents a hypothetical individual who receives the greatest possible projected dose from radiation source in a given population over a specified period of time. Due to the very conservative nature of the assumed scenario, the approach is now seldom used.

The requirement for protecting humans may extend beyond national borders. As the rules and regulations are only under national jurisdiction, such practice requires transnational regulatory system to be in place. The protection of future generations is also a fundamental premise in the management of nuclear waste. Given the inability of obtaining informed consent from the members of future generations, it is the obligation of the present generation to establish a system assuring the protection of future generation. This issue is further discussed in the next section.

2.4.2.3 How Long Should the Prescribed Level of Safety Be Provided (Q5)?

A related issue to defining the person to protect is the time horizon. As nuclear waste contains long-lived radionuclides, efforts to protect the public from nuclear waste must be extended for the future generation. Question then is how long into the future? Do we know enough about future state of human society? How do we form the basis of protection? Human experiences with modern technology are within the time frame of less than 1000 years. Projecting the performance of technology into the long term future beyond the time scale of human experiences is very difficult but needed as part of the activities in nuclear waste management.

From the technical point of view, protection of humans should be based on what is technically defensible or scientifically tractable. Technically defensible approach requires projection of the behavior of the system under consideration. Such projection needs to be made with reasonable degree of confidence. Scientific tractability to predict the behavior of engineered system may be ensured over a few hundred years. Beyond a few hundred years, the uncertainties become large. The related predictions become scientifically questionable. The chance of major changes or disruptions in the environment also rises in such long-term periods. As recorded human history spans less than 10,000 years, going beyond that time span entails significant uncertainties in social development. In this regard, a time period less than 10,000 years is reasonable for the purpose of analysis. Many key nuclides have migrated from the repository region toward the accessible environment within this time frame and their behaviors become descriptive of long times thereafter. At the same time, the likely time for some radionuclides released from nuclear waste to reach the biosphere is longer than 10,000 years. The maximum human exposure to radiation is therefore likely to occur beyond 10,000 years.

This disparity in the time scale between a time period of scientific tractability and a very long period of time to capture the peak dose to future generation demands the use of policy judgment for safety demonstration of nuclear waste.

In the case of the U.S., the U.S. Court of Appeals for the District of Columbia Circuit struck down EPA’s 10,000-year regulatory compliance period in 40 CFR 191 as too short and asked EPA to reconsider the period of regulatory compliance. This led to the current 40CFR193 standards with the regulatory compliance period extended to 1 million years.

Table 2.2 also shows how different countries have come up with the time periods for nuclear waste safety requirement. Note that a number of countries (i.e., Finland, France, and South Korea) have adopted 10,000 years. Several countries (i.e., Belgium, Canada, Germany, U.K., and Spain) did not specify the time frame. Sweden uses 100,000 years. The USA and Switzerland specify up to 1 million years.

2.4.2.4 What Approach Should Be Used to Assure Safety (Q6)?

To assure safety in dealings with nuclear waste, radiation dose limits are established for workers and the public. These limits are accompanied by radiation protection guidelines to support compliance activities.

The general guidelines of radiation protection are based on so-called the principle of time, distance, and shielding. This principle means that protecting humans against radiation are mainly based on minimizing the time of exposure, maximizing the distance from the source, and using shielding to stop/attenuate radiation. Further details of this general principle are discussed in Sect. 5.4.

Application of this principle of time, distance, and shielding to nuclear waste management takes the approach of isolation and containment. Isolation and containment is to minimize the chance of human interactions with nuclear waste.

Isolation is to keep humans away from nuclear waste while the waste remains radioactive. Isolation is typically done through the use of barriers. The barriers can be man-made (engineered) or natural. The man-made (engineered) barriers are used while the waste is being handled and treated before reaching the final disposal phase. The natural barriers are used in the final disposal phase. Combined use of the man-made barriers and the natural barriers is employed to provide long-term isolation of nuclear waste. Segregation as a related concept can be used to support waste isolation by not mixing short-lived radionuclides with long-lived ones or to separate the constituents according to the contained hazards. For example, short-lived nuclides naturally decay away as long as adequate period of storage is provided at the location of generation. By separating the constituents into different waste streams according to the hazard potentials, efficiency in the use of resources in the follow-on radiation protection activities can be enhanced.

Containment is to minimize the mobility of nuclear waste. Stabilization of nuclear waste through immobilization is practiced for this purpose (see Chap. 9). Containment minimizes the possibility of radionuclide release from nuclear waste into the environment. This also enhances the degree of isolation of nuclear waste.

During isolation and containment, the concept of “Let-It-Decay” is always working as the radioactive components in nuclear waste continue to decay away. As all radioactive materials have finite lifetime according to the physical decay characteristics, providing the necessary level of isolation and containment of nuclear waste until the constituents of nuclear waste decay away is one of the primary goals of nuclear waste management.

The approach of isolation and containment can be further elaborated by suggesting specific performance targets (IAEA 2003). These target numbers become so called derived standards. A derived standard is the translation of the fundamental safety criterion into practical measures, such as the total amount of radionuclides passing across a repository boundary expressed in the cumulative amount of radioactivity released over a specified period of time. These derived standards eliminate the need for calculations involved in specifying the exposure pathways and the

Table 2.3 Limits on cumulative release of key radionuclides over 10,000 years as required for HLW disposal according to 40CFR191

Radionuclide	Release limit per 1000 MTHM (curies)	Comments
^{241}Am, ^{243}Am, ^{14}C, ^{129}I, ^{237}Np, ^{238}Pu, ^{239}Pu, ^{240}Pu, ^{242}Pu, ^{226}Ra, ^{233}U, ^{234}U, ^{235}U, ^{236}U, ^{238}U, and any other alpha-emitting radionuclide with a half-life greater than 20 years	100	The limits apply to spent nuclear fuel containing 1000 metric tons of heavy metal (MTHM) exposed to a burnup between 25,000 megawatt-days per metric ton of heavy metal (MWd/MTHM) and 40,000 MWd/MTHM
^{135}Cs, ^{137}Cs, ^{90}Sr, ^{126}Sn, and any other radionuclide with a half-life greater than 20 years that does not emit alpha particles	1000	
^{99}Tc	10,000	
^{230}Th, ^{232}Th	10	

related uncertainty and may facilitate the effort for regulatory compliance demonstration. The related examples utilized in the U.S. are as follows.

Limits on Containment (the Release Limit):
40 CFR 191 of EPA specifies limits on how much of radionuclide inventory in spent fuel can be released to the accessible environment over the post-closure time period of 10,000 years. This represents the limit on total cumulative release of radionuclides. The values of the limits are given in Table 2.3. These limits correspond to the risk of un-mined natural uranium as discussed in Example 2.6.

Limits on Isolation (the Containment Time Limit – Waste Package Lifetime):
U.S. NRC's 10CFR 60 specifies that the amount of release of any radionuclide from the engineered barrier system following the containment period shall not exceed one part in 100,000 per year of the inventory of that radionuclide calculated to be present at 1000 years following permanent closure. 10CFR Part 60 also specifies that waste package for spent fuel or high level waste shall have lifetime between 300 years and 1000 years.

Limits on Isolation (the Radionuclide Transfer Time Limit – Groundwater Travel Time):
10 CFR Part 60 also specifies that geological repository for spent fuel (or HLW) disposal shall be located such that pre-waste-emplacement groundwater travel time along the fastest path of likely radionuclide travel from the disturbed zone to the accessible environment shall be at least 1000 years.

The Supporting Activities
Following the guidelines of isolation and containment of nuclear waste, various activities are conducted in nuclear waste management. The following is the summary.

- Source Control: Source control is to minimize the generation of waste from the production facilities (e.g., nuclear power operation or other nuclear related activities). It also includes waste segregation and the effort to minimize the generation of secondary waste from handling or treatment of the wastes.
- Waste Characterization: The effort to determine the constituent characteristics of the waste by identifying the types and amounts of radionuclides is termed waste characterization (see Sect. 13.3). Such characterization is necessary to understand the hazards involved in the waste and the resulting information is passed on through treatment, packaging, shipment, storage, and disposal of waste. Therefore, the potential hazards contained in the wastes dictate the level of isolation and containment throughout the activities of their management.
- Waste Treatment and Packaging: Nuclear wastes are treated and packaged in a variety of ways to isolate and contain the waste by surrounding it with a low-permeable substance (e.g., concrete, clay, metals, glass, ceramics) and by enclosing it in durability proven containers. The requirements for the treatment and packaging vary depending upon the type and classification of the waste (see Chaps. 9 and 13).
- Waste Transportation: Transportation plays an integral role in connecting various activities of nuclear waste management. To minimize radiation exposure to the public and the risk of sabotage, shipments of nuclear waste must be conducted in a specially controlled way. In the case of HLW or spent fuel, the containers used for shipment must meet strict government regulations and armored security personnel must accompany the transportation (see Sect. 7.5 for spent fuel).
- Waste Storage: Storage is an interim and necessary step in the lifecycle of nuclear waste (see Sect. 7.4 for spent fuel). Storage of nuclear waste allows radioactive decay of nuclear waste constituents and adds flexibility to scheduling of activities before reaching the final disposal facility. For safety and security, the storage facility must be adequately protected during the presence of nuclear waste materials. Retrievability of waste is an important requirement during storage.
- Waste Disposal: Disposal is the final stage and is different from storage in terms of the quantity of radioactivity and the distribution of risks and responsibilities over segments of current and future populations. Disposal of waste must ensure long-term isolation and containment of the wastes while being vulnerable to natural events and human actions (see Chap. 10).

2.4.2.5 How Do We Verify Safety (Q7)?

Success in the use of technology must be demonstrated by showing the performance of the technology to be in compliance with the regulatory limits. Predicting the performance of a system with short time frame requirement (e.g., radiation shielding, transportation, heat-removal) can be readily made while the activities related to safety demonstration of nuclear waste disposal involve very long-term predictions. The related predictions over long periods of time become very challenging. As a caveat, the use of the word, "predicting" in this context is not to predict the future state of things but to provide sufficient technical basis to assure safety in the future (see Chap. 12). The regulatory authority is to provide guidelines on how the exercise of "predicting" should be done for compliance demonstration.

Demonstration of regulatory compliance for nuclear waste disposal requires the use of a special approach called performance assessment. Here, performance of a facility means the potential health, safety, and environmental effects in terms of their magnitudes and likelihoods. In performance assessment, key scenarios that could trigger a radioactive release are catalogued and the projected risk to these individuals according to each of the scenarios is compared to the safety limits. The standard must specify the individual or individuals for whom the risk calculations are to be made. Regulatory agency must provide guidelines as to the treatment of the uncertainty in particular in future site conditions and how the predictive modeling based on today's knowledge should be utilized to provide the basis for regulatory performance of long-term safety. Performance assessment also plays an important role in the design and construction of a geologic repository. The issue related to performance assessment are further discussed in Chap. 12.

2.4.2.6 How Should the Environment Be Protected in Comparison with Protecting Public Health (Q8)?

Nuclear waste shall be managed in a way that provides protection of humans as well as the environment. In other words, in addition to humans, a wide variety of species in the ecological system that may be affected by nuclear waste should be protected.

One common approach in this is to assume that protecting humans is enough to protect the environment. This assumption however may not be true. Some species are more susceptible to damage than humans from radiation. At the same time, protecting the environment and demonstrating such protection is a large challenge. For example, developing a clear concept of "environment" is difficult. The term, environment, implies both quality and quantity including the health and diversity of ecological systems. The life cycles of ecological systems vary greatly in both space and time with complicated life histories of species. Should the focus of protection be only on the population and ecosystem levels with no attention on the individual level? What should be the end point of protection? What are the biological effects of radiation on various species in the ecosystem? These are challenging questions to be answered as part of the discussions for environmental protection.

Protecting the environment may be pursued by identifying the most sensitive or vulnerable species in the affected environment. In that case, protecting these special species serves as the surrogate for environmental protection. In a report on the use of Reference Animals and Plants (ICRP 2007), ICRP suggested ways to address the issue through limited case studies. While this effort provides an example of establishing basis for radiation protection of the ecosystem, any form of ‘dose limits’ for environmental protection has not been proposed.

2.5 Conclusion

Given its special nature, management of nuclear waste is largely a public policy problem requiring decisions by the government. History tells that lack of government policy and leadership led to mistakes and failures in nuclear waste management. This chapter outlined the details of how policy is made and what should be included in the policy for nuclear waste management. Although a plethora of issues having multiple stakeholders with diverse interests are involved, policymaking can settle disputes in nuclear waste management regarding goals, responsibilities, procedures, appropriation of resources, and ethical issues. Policy also guides and supports technical activities including technology developments necessary to effectively support nuclear waste management. In that regard, the importance of good policy to achieve success in the management of nuclear waste cannot be emphasized enough. For any country with nuclear power program should put a priority on developing a good national nuclear waste policy early on.

Homework

Problem 2.1: Based on the review of the history of U.S. nuclear waste management, summarize the lessons learned. Discuss how these lessons could be applied to future nuclear waste management.

Problem 2.2: Describe key technical and institutional issues to be covered in a national policy for nuclear waste management.

Problem 2.3: Suggest your own creative way of defining acceptable level of risk by using an example familiar to you. Compare the results with the levels of acceptable risk discussed in Sect. 2.4.2.1 of the chapter.

Problem 2.4: In 40CFR190, US EPA requires that the health risks from a spent fuel disposal facility should be no greater than the risks from an “equivalent” uranium ore body for the first 10,000 years after spent fuel emplacement. Present any arguments for and against this approach.

Problem 2.5: US NRC has suggested using the guideline of $1000 per person-rem averted be adopted in 1983 dollars based on risk-benefit tradeoff.

(a) What is the assumed value of life in using the risk-benefit guideline in 2020 US dollars?
(b) Discuss any ethical issues involved in the suggestion.

Problem 2.6: This problem is to conduct a very simplified hypothetical decision analysis to support the selection of options for spent nuclear fuel management. The three options considered are (1) building multiple independent spent fuel storage facilities at each nuclear reactor locations, (2) building one centralized spent fuel storage facility (away from the reactors), and (3) building one geologic repository. The total capacity to be provided by each option is 70,000 metric ton of spent fuel storage or disposal. The cost, benefit, and the probabilities of success of each option are given in the following table. Assume the average inflation rate is 2.68% per year between 1985 and 2020. Assume also that the high or low values of each cost item are equally probable.

	Cost		Benefit	Probability of success
	Low	High		
Option 1	5.7 billion (1985 $)	18.2 billion (1985 $)	600 billion (2020 $)	90%
Option 2	3.5 billion (1985 $)	7.7 billion (1985 $)	600 billion (2020 $)	30%
Option 3	6 billion (2010 $)	11 billion (2010 $)	1200 billion (2020$)	20%

Option 1: Building multiple independent spent fuel storage facilities at each nuclear reactor locations
Option 2: Building one centralized spent fuel storage facility (away from the reactors)
Option 3: Building one geologic repository

Decide which option to choose based on the $ value in 2020.

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Chapter 3
Basic Nuclear Science and Engineering

Abstract Understanding the basics of nuclear and radiation science and engineering provides the basis of technical approaches to nuclear waste management. This chapter describes how various types of ionizing radiation are produced and interact with matter, and how nuclear reactor works. The process of nuclear fission as the fundamental step of nuclear energy generation is also described along with its implications in nuclear waste generation.

Keywords Ionizing radiation · Radioactive decay · Radiation interactions · Nuclear reactors · Fission products

Controlling the risk associated with nuclear waste requires understanding of basic nuclear and radiation science and engineering. Understanding of how radiation is produced and how radiation interacts with matter lays the foundation for the control of radiation risk. Understanding how nuclear reactor works provides the basis for the understanding of nuclear wastes characteristics.

3.1 Science of Radiation

3.1.1 What Is Radiation and Why Is It Produced?

Radiation is the energy released from an unstable atom when the atom goes through a process of becoming stable or less unstable. This means there are atoms that are unstable (i.e., having too much energy to be stable) and these unstable atoms have natural tendency to move toward being less unstable. Whenever an atom moves from an unstable state to less unstable or stable states, radiation is released. Therefore, radiation is produced as long as an unstable atom exists: There are more than 1000 unstable atoms known to exist (with many of them of natural origin). It is also noted that radiation is emitted from both natural and man-made materials as unstable atoms are part of both materials. Also, as far as a specific type of radiation is concerned,

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radiation emitted from natural radioactive materials cannot be distinguished from that produced from man-made materials.

Atom

An atom is a basic building block of a material and is composed of the nucleus and the orbital electrons. Atoms are combined together to form a molecule which is the smallest fundamental unit of a chemical compound. The number of atoms existing in the earth is in the order of $10^{49} \sim 10^{50}$. This large number of atoms belongs to one of the 92 elements found in nature. Including the man-made ones, there are a total of 118 known elements (as of 2016). These elements can be further categorized by the composition of their atomic nucleus. Nucleus is the small, dense center of the atom, composed of protons and neutrons. A general term, nuclide, is used to denote any species of atom as characterized by the number of protons and neutrons present in the atomic nucleus.

Nucleus

As protons and neutrons are much heavier than electrons, a nucleus carries most of the mass of the atom. The mass of each proton and neutron is about 1.67×10^{-24} g while the mass of electron is only 9.11×10^{-28} g. In terms of so-called, atomic mass unit (amu), the neutron has a mass of 1.00898 amu, which is slightly greater than the proton mass of 1.00759 amu. The atomic mass unit is a unit of mass equal to one-twelfth of the mass of an atom of carbon-12 (^{12}C) and is equal to approximately 1.66×10^{-24} gm.

The number of electrons of an atom is equal to the number of protons (or the atomic number) for the balance of positive and negative charge within an atom. Therefore, a nucleus also has a positive electric charge equal to an integral multiple Z of the electrical charge $e = 1.6 \times 10^{-19}$ Coulomb. A proton has a single unit of positive charge. A neutron has no charge. Electron also has a single unit of negative charge.

The atomic mass is equal to the total combined mass of individual atoms (thus the mass of protons and neutrons) when the number of atoms reach the Avogadro's number, $N_A = 6.02 \times 10^{23}$, the number of atoms in a mole. One mole is defined as the number of atoms in precisely 12 gm of Carbon-12.

Nuclide

A nuclide refers to a species of atom characterized by the constituents of its nucleus. A radionuclide is a nuclide that emits radiation. Definition of nuclide is represented by a symbol of an atom as $^A_Z X$, where X is the chemical symbol of the atom. Z is the number of protons and is called the atomic number. A is the sum of the number of protons and neutrons and is called the atomic mass number. Nuclides containing the same number of protons but different numbers of neutrons are called isotopes. An element can have many isotopes (e.g., ^{12}C, ^{13}C, ^{14}C, ^{15}C, etc.). Fig. 3.1 shows an example of a stable isotope of carbon $^{12}_{6}C$. The notation of a nuclide is usually given without the atomic number which is known for all nuclides (e.g. ^{12}C instead of $^{12}_{6}C$).

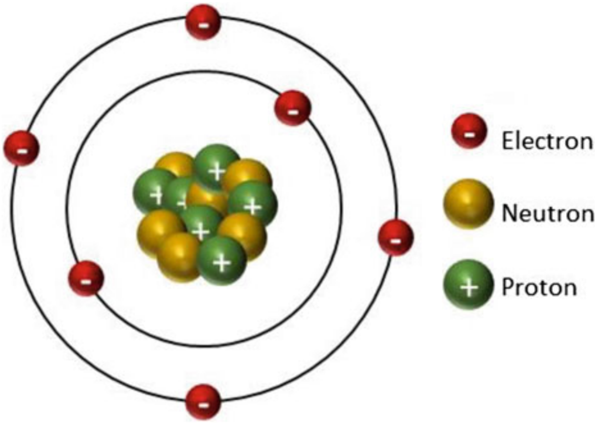


Fig. 3.1 Depiction of a stable isotope, carbon-12, with 6 protons, 6 neutrons, and 6 electrons

There are about 1300 nuclides presently known to exist. About 80% of them are radionuclides. Therefore, only about one-fifth of the existing nuclides are stable (~275 nuclides). The rest of nuclides are unstable. Also, all nuclides with atomic number greater than 83 (i.e., beyond bismuth, Z = 83) are naturally unstable (i.e., radioactive).

Electrons

The atomic nucleus is surrounded by orbital electrons, negatively charged subatomic particles. As stated, the number of orbital electrons in an atom is equal to the number of protons in the nucleus, making the atom electrically neutral. The mass of an electron is approximately 1/1836 of that of the proton. The orbital electrons are bound to the nucleus through the Coulomb force. The Coulomb force is generated through electrostatic attraction or repulsion due to charge versus charge interactions. The force is attractive between the positive and negative electrical charges or repulsive between the same electrical charges.

Electrons that are not bound to an orbit are free electrons. Free electrons are unstable and decay into neutrons with a half-life of 613.9 seconds (definition of half-life is given in Sect. 3.1.6). *Electrons* in atoms exist in spherical shells of various radii, representing different energy levels. The energy levels keep the electrons in the orbit from being attracted to the positively charged nucleus.

Stability

As mentioned, the atomic nucleus has a positive electrical charge due to the presence of protons. Thus an atom with a large number of protons will carry a large amount of positive charge within the nucleus. Presence of large amount of positive charge in the nucleus will create repulsion among protons. The resulting charge-to-charge repulsion (called Coulomb force) becomes the source of instability within the nucleus. This repulsive force between protons can be reduced when neutrons are inserted as buffer between the protons. Therefore, as Z (atomic number) increases, more neutrons may be needed to reduce the repulsive force. There also exists an attractive force between protons and neutrons that tends to hold them together. This

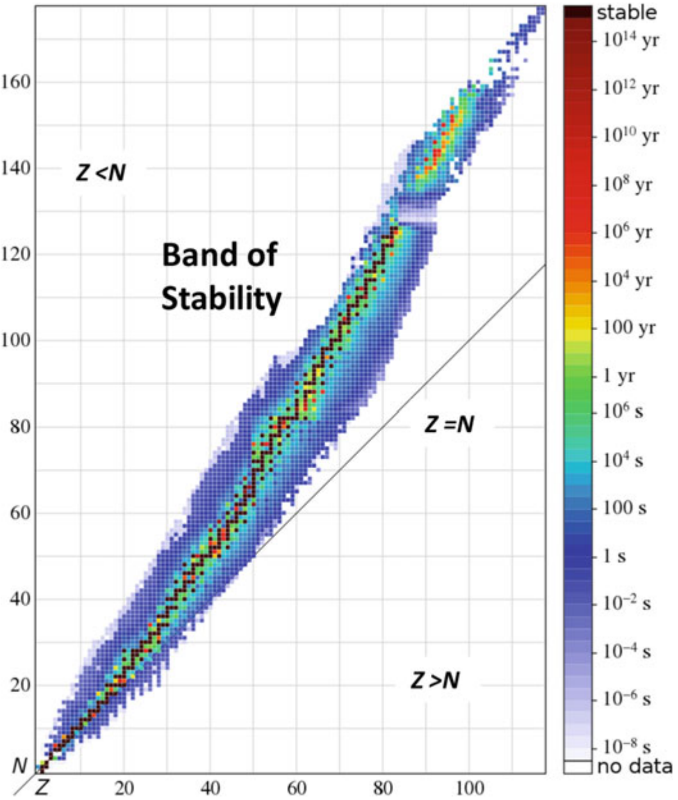


Fig. 3.2 Neutron-proton diagram of stable nuclide (edited from source: Wikipedia Commons, Isotopes and half-life)

attractive nuclear force must be sufficient in stable nuclei to overcome the disruptive Coulomb force. However, for given number of protons, the number of neutrons that can be included in a stable nucleus is limited. When the repulsive force from protons and the mitigating effect by the given number of neutrons are balanced, atom is stable. If there is a net imbalance between the attractive nuclear force and the disruptive Coulomb force, the nuclide is unstable.

Figure 3.2 shows the relationship between the number of protons, Z, and neutrons, N = A-Z, of stable nuclides. There exists a linear relationship between Z and N for low atomic mass numbers (for Z almost up to 20). Thus, light elements are stable when the number of neutrons and protons are approximately equal (N = Z). On the other hand, all stable atomic elements with Z > 20 contain more neutrons than protons. As Z increases, the number of neutrons increases more rapidly. This indicates that for a large element to be stable, a larger number of neutrons than that of protons is needed. There exists a fine balance between the number of protons and neutrons for a nuclide to be stable.

3.1.2 Ionizing Radiation

Ionization is the process through which an electron of the atom or molecule is removed. Ionization takes place when the electron is stripped away from their orbit by being impacted by an incident particle. The ejected electron is rapidly captured by another molecule to yield a negative ion. Thus, the result of ionization is the formation of two ions, one negative and one positive (i.e., the atom that lost one electron), referred to as ion pairs. Such ion pair production and subsequent energy transfer can result in deleterious changes in the interacting medium. Ionization is one of the most immediate and fundamental outcomes of radiation interaction with matter.

Ionizing radiation is any radiation particle with energy greater than the energy required to remove an electron from an atom. It consists of directly or indirectly ionizing particles or a mixture of both. It includes both charged particles and uncharged particles capable of causing ionization by primary or secondary processes. In contrast, when the energy of radiation is not large enough to produce ionization of atoms and molecules with which they interact, the radiation is classified as non-ionizing radiation.

For example, the binding energy of an electron in hydrogen atom is 13.6 eV. However, to remove the electron from hydrogen atom, about two to three times greater than the binding energy is needed. The extra energy needed is to cause excitation of electrons (raising orbital electrons to higher but still bound states) before being stripped off from the atom. Therefore, approximately 34 eV is required to produce an ionization of hydrogen.

As long as radiation carries enough energy to cause ionization, it is an ionizing radiation. Ionizing radiation include ultraviolet lights, x-rays, gamma rays, and other types of high energy particles such as electrons, protons, alpha particles, neutrons, or other cosmic particles. These particles are listed in Table 3.1. The particles released from the decay of unstable atom are all ionizing radiation.

While the photons are collectively called electromagnetic radiation, different names and categories of them can be assigned according to the frequency of photons. This is shown in Table 3.2. Electromagnetic radiation is a wave of energetic photons

Table 3.1 Types of basic ionizing radiation particles and their characteristics

Type	Particle	Symbol	Charge	Rest mass (amu)	Half-life
Neutral	Photon	γ, X	0	0	<15 min (free neutron) Stable (in a nucleus)
	Neutron	n	0	1.009	10.4 min
Electrons	Electron	β^-, e^-	-e	5.486×10^{-4}	Stable*
	Positron	β^+, e^+	+e	5.486×10^{-4}	Stable
Ions	Proton	p	+e	1.007	Stable
	Alpha particle	α	+2e	4.003	Stable

*Free electrons have a half-life of 613.9 seconds

Table 3.2 Different types of photons (electromagnetic radiation) and their energy

Type	Energy (eV)			Ionizing?
Electric wave		<	4.1×10^{-10}	Non-ionizing
Radio waves	4.1×10^{-10}	~	1.24×10^{-10}	Non-ionizing
Infrared (IR)	0.0124	~	1.24	Non-ionizing
Visible	1.77	~	3.1	Non-ionizing
Ultra-violet (UV)	3.1	~	124	Ionizing
X-ray (diagnostic)	12.4 keV	~	124 keV	Ionizing
X-ray (therapy)	124 keV	~	1.24 MeV	Ionizing
X-ray (accelerators)	12.4 MeV	~	1240 MeV	Ionizing
Gamma rays	12.4 keV	~	14.4 MeV	Ionizing
Cosmic photons		>	12.4 MeV	Ionizing

that contain an electric field and a magnetic field traveling at the speed of light (e.g., 3×10^8 m/sec). Therefore, each photon has a wavelength and a frequency. The wavelength is defined as the distance between the two peaks of the electric field contained in the electromagnetic radiation. The frequency of a photon is defined as how many wavelengths a photon propagates per second. Photons exists in a continuous spectrum that extends over many decades of frequency including radiations from the lowest frequencies (radio waves) to the highest (x-rays and gamma rays).

Energy of photons is represented by $E = h\nu$ where h is Planck's constant ($=6.626 \times 10^{-27}$ erg-sec) and v is the frequency of the photon. The wavelength of the photon can also be associated with the frequency of the photon by the relationship, $\lambda = c/\nu$, where λ is wavelength of the radiation. It can be seen that among the photons, visible lights and infrared radiation are non-ionizing radiation as their energy is lower than the level needed to cause ionization of an atom.

Also photons can be called γ-rays or x-rays depending on the way it is produced (not by their energy):

- Gamma rays are the result of nuclear transitions. The energies of gamma rays vary from a few tens of keV to a few (up to 10) MeV.
- Continuous X rays (called bremsstrahlung) are the result of an acceleration of a swiftly moving charged particle and can be produced over the same entire energy range of gamma rays as well.
- Characteristic X-rays result from transitions of tightly bound inner electrons. The maximum energy of characteristic X-rays is about 100 keV.

Definition: What Is X-Ray?

– X-ray is very similar to gamma ray except that x-ray originates from orbital electrons and usually has lower energy. Gamma ray originates from atomic nucleus. X-ray has two distinct types:

1. A continuous spectrum x-ray (or, simply a continuous x-ray)
2. A sharp line spectrum (discrete energy) x-ray.

– A sharp line spectrum x-ray is produced when an electron drops from a higher energy orbit to a lower energy orbit due to the presence of vacancy caused by ejection of electron in the lower energy orbit. The difference in the energy levels between the two orbits is the amount of energy carried by x-ray.

– The continuous spectrum results from the radiation emitted by the electrons which are decelerated in the Coulomb field of force of the nuclei of the target atoms (through bremsstrahlung). Bremsstrahlung (means "breaking radiation" in German) refers to the process of producing continuous x-rays due to deceleration of high-energy electron due to Coulombic interactions.

3.1.3 Types and Characteristics of Ionizing Radiation

Radioactive decay is a random process occurring at the level of single atoms and takes place when an unstable atom loses energy by emitting radiation. Therefore, radioactive decay refers to the spontaneous release of radiation by radionuclides to increase its degree of stability. The type of decay taking place in a nuclide depends on the atomic mass of the nuclide and its proton to neutron ratio.

There are six different types of spontaneous radioactive decay including: (1) β^- decay, (2) β^+ decay, (3) electron capture, (4) α decay, (5) γ decay or isomeric transition, and (6) spontaneous fission. In any of these radioactive decay, mass number, charge, total energy, and linear and angular momentum are conserved.

Beta (β^-) Decay

A "neutron-rich" unstable nuclide, i.e., having the *Z/N* ratio to be low to be stable, goes through β^- decay. In Fig. 3.2, these nuclides are located above/at the left hand-side of the band of stability. This can be described as

$$n \rightarrow p + e^- + \overline{\nu}$$

where p, n, e^-, ν are proton, neutron, electron, and anti-neutrino, respectively. In β^- decay a neutron is converted to a proton and an electron is released along with anti-neutrino. Through this decay, the atomic number of a radionuclide increases by one and the neutron to proton ratio decreases. Examples of β^- decay is ^{14}C becoming ^{14}N, ^{32}P becoming ^{32}S, and ^{129}I becoming ^{129}Xe. Figure 3.3 gives an illustration of β^- decay of ^{32}P with the changes in atomic number and energy. Table 3.3 shows key examples of β^- emitters and the energy of the released electrons from them.

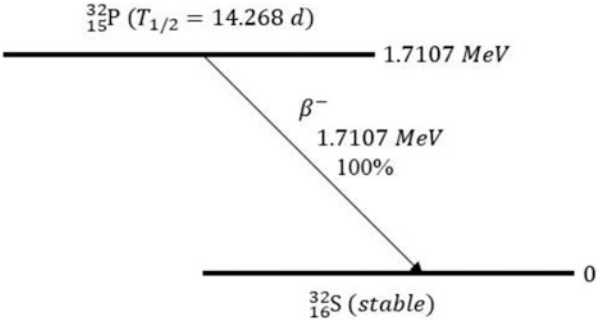


Fig. 3.3 An illustration of β^- decay for ^{32}P

Table 3.3 Examples of beta emitters and their energy (IAEA-NDS ENSDF, Nuclear Structures and Decay Data)

Nuclide	Half-life		Max β-particle energy (MeV)	Average Energy (MeV)
^{3}H	12.32	y	0.0186	0.0057
^{14}C	5700	y	0.1565	0.0495
^{16}N	7.13	s	4. 2892 (66.26%), 10.4191 (28.04%)	1.9412 (66.26%), 4.9798 (28.04%)
^{32}P	14.268	d	1.7107	0.6950
^{60}Co	1925.28	d	1.491 (0.06%), 0.318 (99.92%)	0.606 (0.06%), 0.0958 (99.2%)
^{85}Kr	10.72	y	0.687	0.2514
^{90}Sr	28.79	y	0.5460	0.1959
^{90}Y	4.00	h	2.2801 (99.99%)	0.9337 (99.99%)
^{90}Rb	158	s	6.587 (33%), 5.755 (26%), 2.221 (8.8%), 2.451 (7.7%), 3.204 (5.7%)	2.9922 (33%), 2.5909 (26%), 0.9022 (8.8%), 1.0097 (7.7%), 1.3652 (5.7%)
^{99}Tc	2.111×0^{5}	y	0.2975 (99.9984%) 0.2080 (0.0016%)	0.0846 (99.9984%) 0.0817 (0.0016%)
^{129}I	1.57×10^{7}	y	0.149	0.0400
^{137}Cs	30.08	y	0.5140 (94.7%). 1.176 (5.3%)	0.1743 (94.7%), 0.4163 (5.3%)
^{187}Re	4.33×10^{10}	y	0.002469	0.000618
^{210}Pb	22.20	y	0.0170 (84%), 0.0635 (16%)	0.0042 (94%), 0.0162 (16%)

Positron Emission (β^+ Decay) or Electron Capture

A "proton-rich" unstable nuclide decays by β^+ or electron capture (EC) to become closer to be stable. These nuclides are unstable due to the excess of protons or deficiency of neutrons. They become stable by lowering *Z* and increasing *N* through conversion of a proton to a neutron. This is called β^+ decay, represented as:

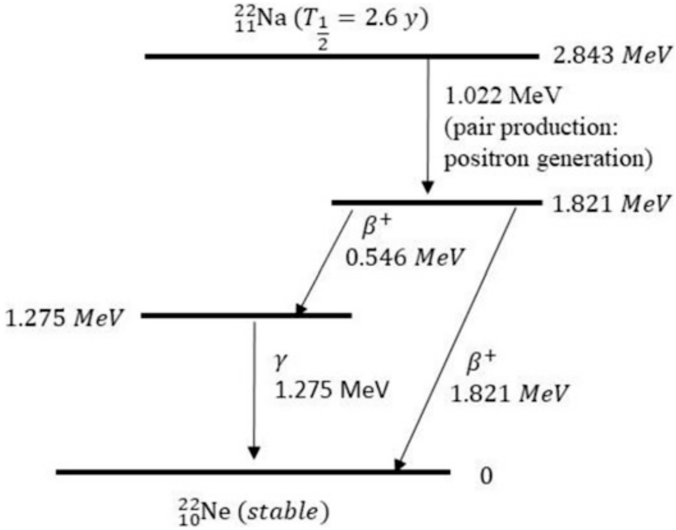


Fig. 3.4 An Illustration of β^+ decay and an electron capture for ^{22}Na

$$p \rightarrow n + \beta^+ + \nu$$

where β^+ and ν are positron (positive electron, or antiparticle of electron) and neutrino, respectively, as a consequence of the process. These nuclides are located at the right-hand side of the *Z/N* curve of the stable nuclide ("band of stability") in Fig. 3.2. An example of β^+ decay is $^{22}_{11}Na \rightarrow {}^{22}_{10}Ne + {}^{0}_{+1}e^+$ where ^{22}Na decays into a stable ^{22}Ne by emitting a positron (Fig. 3.4).

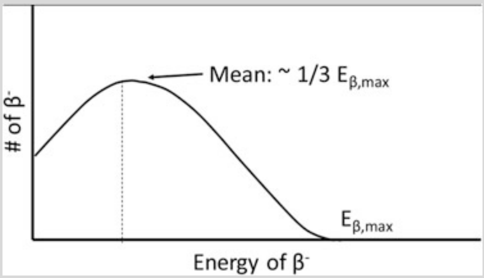
Sometimes, the "proton-rich" unstable nuclides become stable by absorbing an orbital electron to the nucleus (instead of converting a proton to a neutron). This is called electron capture (EC). An example of electron capture is $^{195}_{79}Au + {}^{0}_{-1}e^- \xrightarrow{EC} {}^{195}_{78}Pt$. Positron emission and EC are competing processes for "proton-rich" unstable nuclides with the probability of the latter increasing as the atomic number increases.

All three processes, β^-, β^+, and EC, are designated as beta decay. (The term "beta decay" without any specification usually means β^- decay as this is most common). In beta decay, mass number of the nucleus remains unchanged, but the atomic number changes.

Concept: Why Is the Energy of Electron from Beta β^- Decay Continuous?

Energy released in β^-decay is shared with the neutrino, the electron, and the recoil of the daughter nucleus. Because of its relatively large mass, the recoiling daughter nucleus receives negligible energy and it can be assumed that the total β-decay energy is distributed between neutrino and electron, ranging between zero and the energy corresponding to the difference in the mass of the initial products and the final products.

Thus, the spectrum of the energies of β^- particles is continuous as shown in the following figure.



Typically, the average energy of β^- particles is about the third of the maximum beta energy. Thus,

$E_\beta^{max} = Q$ and $E_\beta^{avg} \approx \frac{Q}{3}$

Alpha (α) Decay

In general, nuclides with a large atomic number are naturally unstable and the short-range nuclear forces holding the protons together in the nucleus are barely able to counterbalance the mutual repulsion of their protons. In this case, as a quick way to increase their stability by reducing the number of protons, alpha particle is often

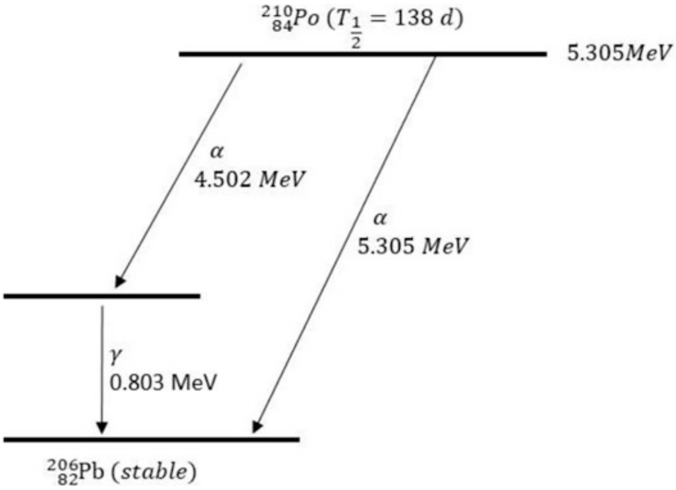


Fig. 3.5 An Illustration of α^-decay for ^{210}Po

released. This is called α-decay, i.e., radioactive decay with alpha particle emission. It can be represented as:

$$ {}_{z}^{m}X \rightarrow {}_{z-2}^{m-4}Y + {}_{2}^{4}He $$

An example of a decay is given in Fig. 3.5 for ^{210}Po to illustrate the changes in atomic number and release of particles.

Alpha decay is observed for elements heavier than lead (Z > 82) and for a few nuclides as light as the lanthanide elements. Lanthanides are typically called rare earths and refer to the elements with atomic numbers from 57 to 71. The first element in the group is lanthanum and as the group exhibit similar chemical properties, they are called the lanthanides. Nuclei with A > 140 always decay by alpha particle emission. Alpha particle consists of two protons and two neutrons bound together into a particle, *which is identical to the nucleus of the helium atom*. Major nuclides with α-decay as examples are shown in Table 3.4.

Gamma (γ) Decay

γ ray is a high energy photon produced from the transition of a nuclei from a high energy (excited) state to a lower energy or ground state. The energy of γ ray represents the difference in energy between the excited state and a lower energy state. This excited state could be an outcome of nuclear reaction or the immediate result of α or β^- decay. Any nuclear reaction or the transformation of an atom with the emission of either an alpha or a beta particle often leaves the product nucleus in an excited state.

Nucleus emitting γ rays does not change its atomic number or mass number as γ ray has no charge or mass. Gamma decay can be represented as:

$$ {}_{z}^{m}X^{*} \rightarrow {}_{z}^{m}X + \gamma $$

where ${}_{z}^{m}X^{*}$ is an excited isotope of ${}_{z}^{m}X$.

An illustration of gamma decay is given in Fig. 3.6 for the case of ^{60}Co where release of two gamma rays along with one electron (β^{-} decay) are depicted. Examples of gamma-ray emitters are also listed in Table 3.5.

Spontaneous Fission

A few very large nuclides reduce the atomic instability quickly by going through spontaneous fission, i.e., splitting of the nucleus into two fragments. Spontaneous fission is observable among nuclear species of mass number 230 or higher. Measurable rates of spontaneous fission can be observed only among the heaviest element such as californium. The nuclides that undergo spontaneous fission also subsequently go through α-decay. In the case of the californium-252, 3.1% of the nuclei undergo spontaneous fission and 96.9% undergo alpha decay as shown below:

Table 3.4 Examples of alpha emitters and their energy (IAEA-NDS ENSDF, Nuclear Structures and Decay Data)

Nuclide	Half-life		α-particle energy in MeV
^{220}Rn	55.6	s	6.2881 *(99.886%)* 5.747 *(0.114%)*
^{222}Rn	3.8235	d	5.4895 (99.92%) 4.986 (0.08%)
^{218}Po	3.098	m	6.0024 (99.98%) 5.181 (0.001%)
^{214}Po	164.3	µs	7.6868 (99.99%) 6.9022 (0.010%)
^{212}Po	0.299	µs	8.7849
^{210}Po	138.376	d	5.3043
^{226}Ra	1600	y	4.7843 (93.84%), 4.601 (6.16%)
^{232}Th	1.40×10^{10}	y	4.0123 (78.2%), 3.9472 (21.7%) 3.8111 (0.069%)
^{238}U	4.468×0^{9}	y	4.198 (79.0%), 4.151 (20.9%), 4.038 (0.078%)
^{237}Np	2.144×10^{6}	y	4.7880 (47.64%), 4.7714 (23.2%), 4.7665 (9.3%), 4.6400 (6.43%), 4.6650 (3.478%)
^{238}Pu	87.7	y	5.4990 (70.91%) 5.4563 (28.98%) 5.3577 (0.105%)
^{239}Pu	24.110	y	5.1566 (70.77%) 5.1443 (17.11%) 5.1055 (11.94%)
^{241}Am	432.6	y	5.4856 (84.8%) 5.4428 (13.1%) 5.388 (1.66%) 5.5445 (0.37%) 5.5115 (0.225%)
^{242}Cm	162.8	d	6.1127 (74.08%) 6.0694 (25.92%) 5.9692 (0.035%)
^{244}Cm	18.1	y	5.8048 (76.9%) 5.7626 (23.1%)

$$^{252}_{98}\mathrm{Cf} \overset{3.1\%}{\rightarrow} FP's + \gamma's + 3.74n$$

$$^{252}_{98}\mathrm{Cf} \overset{96.9\%}{\rightarrow} {}^{248}_{96}\mathrm{Cm} + \alpha$$

where *FP* stands for fission products. On average, 3.74 neutrons are released during a spontaneous fission of californium.

With ^{238}U, spontaneous fission occurs only 5.5×10^{-5} percent of its decays and alpha decay is about 2 million times more probable than spontaneous fission. Further

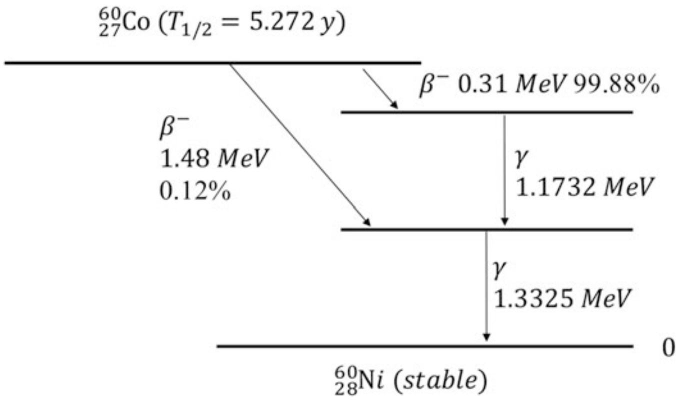


Fig. 3.6 An illustration of ^{60}Co decay

Table 3.5 Examples of gamma ray emitters and their energy (IAEA-NDS ENSDF, Nuclear Structures and Decay Data)

Nuclide	Half-life			γ-ray energy in MeV
^{41}Ar	109.61	m		1.2936 (99.16%) 1.6770 (0.052%)
^{54}Mn	312.20	d		0.8348
^{60}Co	1925.28	d		1.3325 (99.9826%) 1.1732 (99.85%)
^{131}I	8.0252	d		0.3645 (81.5%) 0.6370 (7.16%) 0.2843 (6.12%) 0.0802 2.62%
^{137m}Ba*	2.552	m		0.6617 (89.8%)

*^{137m}Ba is the immediate decay product of ^{137}Cs. Often the gamma ray from 137mBa is considered the outcome of 137Cs decay

discussion of spontaneous fission in comparison to neutron-induced fission is provided in Sect. 3.2.2.

Comparison of the Energy of Radioactive Particles from Decay

The energy carried by radiation varies depending upon the types of decays involved. The amount of energy released in nuclear fission is quite large (~200 MeV) and the fission fragments typically carry several MeV's kinetic energy. The energy of alpha particles is also very high with the kinetic energies of about 4 to 9 MeV. All alpha particles emitted from a particular nuclide decay pathway are mono-energetic, i.e., have the same energies. In contrast, beta particles carry less kinetic energy ranging between few keVs and few MeVs. The beta particles are not mono-energetic and have continuous energy spectrum ranging between zero and the energy corresponding to the difference in the mass of the initial products and the final products. Typically, the average energy of β particles is about the third of the maximum beta energy.

As mentioned in Sect. 3.1.2, the energy of gamma ray typically ranges between a few tens keV and a few MeV (10 keV ~ 7 MeV). In contrast, naturally observed x-ray carries the energy between hundreds of eV and a hundred keV.

Using Einstein's relationship between mass and energy ($E = mc^2$), the decrease in mass in the final products can be calculated into the corresponding release of radiation energy. One atomic mass unit can be shown to be equal to 931 million electron volts (MeV). The electron volt (eV) represents the increase in the kinetic energy of an electron when it falls through an electrical potential of one volt ($1\ \text{eV} = 1.60219 \times 10^{-19}\ (\text{C}) \times 1(\text{V}) = 1.60219 \times 10^{-19}\ (\text{J})$).

3.1.4 Natural Radioactivity

The majority of heavy nuclides are naturally radioactive. These heavy radionuclides decay by a series of α-particle emission and form three different types of decay series. These decay series include so-called 4n (called thorium series), 4n + 2 (called uranium series), and 4n + 3 (called actinium series) decay series as represented in Table 3.6. These decay series start from a long-lived parent and eventually decay to an isotope of lead as stable product. Thorium series starts from thorium-232 (^{232}Th) and ends with lead-208 (^{208}Pb). Uranium series starts with uranium-238 (^{238}U) and ends with lead-206 (^{206}Pb). Actinium series (named for its first-discovered member, actinium-227, ^{227}Ac) starts from uranium-235 (^{235}U) and ends with lead-207 (^{207}Pb).

There is another series called 4n + 1 series (neptunium series) headed by neptunium (^{237}Np) (with half-life of 2×10^5 years). This series does not occur naturally but is artificially produced through nuclear reactions and ends with thallium-205 (^{205}Tl) as the final (stable) product. These four decay series are summarized in Table 3.6.

The most important one among these series is the 4n + 2 uranium series in relation with nuclear waste management due to the ubiquitous presence of ^{238}U in the natural environment and the nuclear fuel cycle. The parent nuclide, ^{238}U, decays through α and β^- particle emissions producing fourteen nuclides in its decay chain. Table 3.7 shows the half-life and the energy of the particles emitted in the series.

Other important naturally occurring radionuclides are tritium (^{3}H), ^{14}C, potassium-40 (^{40}K), and rubidium-87 (^{87}Rb). These nuclides are commonly present in soil, food, and human body through food chains. ^{3}H and ^{14}C are also very common in nuclear waste from various sources including nuclear power plant. They all decay through β^- particle emissions.

Table 3.6 Decay series of actinides

Series	Name	Parent	Half-life (year)	Principle decay products in the series (total number of decay products)	Stable end product
Thorium series (naturally occurring)	4n	${}^{232}_{90}Th$	1.40×10^{10}	^{232}Th-^{228}Ra-^{228}Ac-^{228}Th-^{224}Ra-^{220}Rn-^{216}Po-^{212}Pb-^{212}Bi-^{212}Po-^{208}Tl-^{208}Pb (**11**)	${}^{208}_{82}Pb$
Neptunium series (not naturally occurring)	4n + 1	${}^{237}_{93}Np$	2.14×10^{6}	^{237}Np-^{233}Pa-^{233}U-^{229}Th-^{225}Ra-^{225}Ac-^{221}Fr-^{217}At-^{213}Bi-^{213}Po-^{209}Tl-^{209}Pb-^{209}Bi-^{205}Tl (**13**)	${}^{205}_{81}Tl$
Uranium series (naturally occurring)	4n + 2	${}^{238}_{92}U$	4.47×10^{9}	^{238}U-^{234}Th-^{234}Pa-^{234}U-^{230}Th-^{226}Ra-^{222}Rn-^{218}Po-^{214}Pb-^{214}Bi-^{214}Po-^{210}Pb-^{210}Bi-^{210}Po-^{206}Pb (**14**)	${}^{206}_{82}Pb$
Actinium series (naturally occurring)	4n + 3	${}^{235}_{92}U$	7.04×10^{8}	^{235}U-^{231}Th-^{231}Pa-^{227}Ac-^{227}Th-^{223}Fr-^{223}Ra-^{219}At-^{219}Rn-^{215}Bi-^{215}Po-^{215}At-^{211}Pb-^{211}Bi-^{211}Po-^{207}Tl-^{207}Pb (**16**)	${}^{207}_{82}Pb$

Table 3.7 Decay information of radionuclides in the ^{238}U decay chain

Nuclide	Decay mode	Half-life	Energy released, MeV
^{238}U	α	4.47×10^9 y	4.27
^{234}Th	β^-	24.1 d	0.273
^{234}Pa	β^-	6.7 h	2.19
^{234}U	α	2.46×10^5 y	4.86
^{230}Th	α	7.54×10^4 y	4.77
^{226}Ra	α	1600 y	4.87
^{222}Rn	α	3.82 d	5.59
^{218}Po	α,β	3.098 m	6.11, 0.26
^{214}Pb	β	26.8 m	1.02
^{214}Bi	α,β	19.9 m	5.62, 3.27
^{214}Po	α	164.3 μs	7.83
^{210}Pb	α,β	22.2 y	3.79, 0.063
^{210}Bi	α,β	5.01 d	5.04, 1.16
^{210}Po	α	138.4 d	5.41
^{206}Pb	Stable	–	–

3.1.5 Man-Made Production of Radioactivity

The largest source of man-made radiation is nuclear reactor. In a nuclear reactor, radioactivity is produced through nuclear fission and other nuclear reactions. Nuclear reaction is a process that changes the identity or characteristics of an atomic nucleus through bombardment by an energetic particle (such as neutron or charged particle). In most of nuclear reactions, the interacting nuclei is put in an excited state and releases gamma rays.

Consider an example: ${}^{14}_{7}N + {}^{4}_{2}He \rightarrow {}^{17}_{8}O + {}^{1}_{1}H$. This is the first artificial nuclear reaction performed by E. Rutherford in 1919. This nuclear reaction can also be written as ${}^{14}_{7}N(\alpha, p){}^{17}_{8}O$, where the parenthesis contain the projectile and the released particle(s), in order. In this bombardment reaction, capture of the projectile particle is followed by instantaneous decomposition of the resulting new compound nucleus. Neutrons, with no carrying charge (thus no charge-related repulsion), are very effective in creating nuclear reactions as projectile particles. Charged particles as projectile particles must have a very high energy to overcome electrostatic repulsion to create nuclear reaction.

In 1939, Hahn and Strassman showed that neutron-induced nuclear reaction of uranium-235 resulted in splitting of ^{235}U into lighter nuclides, i.e., fission. When fission reaction takes place, fission products are produced by dividing the mass of the fissioning parent nuclide. An example of typical fission reaction is

$$n + {}^{235}_{92}\mathrm{U} \rightarrow {}^{236}_{92}\mathrm{U}^{*} \rightarrow {}^{144}_{56}\mathrm{Ba} + {}^{89}_{36}\mathrm{Kr} + 3n.$$

where the asterisk indicates an excited state of ^{236}U.

Nuclear fission takes place by a man-made design or through natural processes. As the result of a fission reaction, two or three neutrons, on average, are produced. Production of multiple neutrons enables continuation of fission as a chain reaction (under the presence of a variety of nuclear capture reactions). Capture of neutrons in nuclear reactions makes the new compound nucleus "neutron-rich" unstable, commonly releasing β^- particles (beta- decay). The gamma rays are also emitted whenever a beta decay leaves the nucleus in an excited state.

The radioactivity of the fission products as well as the products of other nuclear reactions constitutes the inventory of radioactivity in a nuclear reactor. Key nuclides and their characteristics (half-life and energy of the particles emitted) in the nuclear reactor fuel are summarized in Table 7.8. Other nuclides produced from nuclear reactions in the structural materials of nuclear reactor and their characteristics are also shown in Table 14.1.

3.1.6 General Description of Radioactive Decay

Radioactive decay refers to the disintegration process of an unstable atomic nucleus to become less unstable or stable by releasing radioactive particles such as α particles, β particles (electrons), or γ rays. Each of the particles released will be an agent of energy transfer to the interacting matter. This will be discussed in the next section (3.1.7).

Eventually all radioactive materials cease to be radioactive and become stable. In other words, when a radionuclide decays, it does not disappear but eventually become a stable nuclide. This means a radioactive material does not remain radioactive forever but eventually becomes non-radioactive.

In discussing radioactive decay, the following questions can be asked.

"How long does a nuclide remain radioactive?"

"How often do radioactive particles get released from the decay?"

"How many radioactive particles are released from a radioactive material?"

To answer these questions, we define a parameter called decay constant. Decay constant (λ) is the probability of decay of a single radionuclide per unit of time. It is an inherent characteristics of a radionuclide as an invariable, constant quantity, independent of time and the radionuclide population.

According to the definition of decay constant, the probability of disintegration of a radionuclide during the time interval Δt is $\lambda \Delta t$. Therefore the number of atoms disintegrated during the time internal Δt is, $\Delta N = \lambda N \Delta t$. For a very small time interval ($\Delta t \ll 1$), the equation becomes, $dN = \lambda N dt$.

Then, time dependent changes in the number of radioactive atoms present then become (the negative sign indicates decrease in number),

$$\frac{dN}{dt} = -\lambda N \tag{3.1}$$

which gives,

$$N(t) = -N(0)\mathrm{e}^{-\lambda t} \tag{3.2}$$

This equation gives the relationship for the number of atoms of radionuclides, $N(t)$, remaining at time t while decaying away from the initial number of atoms, $N(0)$.

The question, "How long does a nuclide remain radioactive?" leads to the definition of half-life (designated as $t_{1/2}$). Half-life is the time required for half of the atoms of a radioactive material to decay away. To be precise, it is the interval of time in which half the atoms of a radionuclide are transformed into other nuclear species by emitting particles and energy (through radioactive decay). Therefore, at t $= t_{1/2}$,

$$N(t) = 1/2N_0 = N_0\mathrm{e}^{-\lambda t_{1/2}}$$

$$1/2 = \mathrm{e}^{-\lambda t_{1/2.}}$$

By taking logarithm on both sides,

$$t_{1/2} = \frac{ln\,2}{\lambda} \approx \frac{0.693}{\lambda} \tag{3.3}$$

Therefore, decay constant, λ, is directly related to half-life, $t_{1/2}$, as.

$$\lambda = \frac{ln\,2}{t_{1/2}} \approx \frac{0.693}{t_{1/2}} \tag{3.4}$$

If the time elapsed is twice or three-times the half-life, the amount remaining is 25% and 12.5%, respectively. If the time elapsed is eight- or ten-times the half-life, the amount remaining is 0.38% or 0.098%, respectively. Therefore, after eight- or ten-times the half-life, less than 0.4% or 0.1% of the initial amount, respectively, remains.

Another question of interest, *"How often do radioactive particles get released from the decay?"* is to understand how often a nuclide decays. As decay constant (λ) is the probability of decay of a radionuclide per unit of time, the larger the decay constant, the higher the frequency of decay. The frequency of radioactive decay is related to a term called *"activity"* of radionuclide. The higher the activity, the higher the frequency of radioactive particle emission from a radionuclide.

Activity is defined as the rate of radioactive decay per time in a given material and is represented as a function of decay constant as follows:

$$A(t) = \lambda N(t) \tag{3.5}$$

where $A(t)$ is the activity at time t and $N(t)$ is the number of atoms of the radionuclide in the source. Activity also represents time dependent changes in the number of radioactive atoms present as represented in Eq. 3.1,

Activity is commonly expressed in the number of disintegrations per second, defined in a unit called Becquerel. The Becquerel (Bq) is the SI unit (International System of units) for radioactivity given in reciprocal seconds, s^{-1}, meaning one Becquerel is equal to 1 disintegration per second (abbreviated as dps).

Another unit, called the Curie, has been widely used to represent the level of activity. The Curie (Ci) is defined as,

$$1\ \text{Ci} = 3.7 \times 10^{10}\ \text{dps} = 3.7 \times 10^{10}\ \text{Bq} \tag{3.6}$$

One Curie was originally defined by the discoverer of radium, Marie Curie as the number of decays per unit time of 1 gram of ^{226}Ra assuming its half-life at 1580 y.

Another related quantity of interest is the specific activity. The specific activity is defined as the number of decays rate per unit mass of a radionuclide.

$$\textit{Specific Activity} = \textit{Activity in unit mass} \tag{3.7}$$

Example 3.1: Calculate the Specific Activity of ^{226}Ra (assuming $t_{1/2} = 1600$ year)

$N =$ number of atoms in 1 gram

$$N = \frac{6.022\times10^{23}\left(\frac{atoms}{mol}\right)}{226\left(\frac{gm}{mol}\right)} = 2.665 \times 10^{21}\left(\frac{atoms}{gm}\right)$$

$$\lambda = \frac{0.693}{1600\ (yr)} = 1.373 \times 10^{-11}(s^{-1})$$

$$\text{Activity} = A = \lambda N = 3.66 \times 10^{10}\left(\frac{atoms}{gm-\ sec}\right) \approx 3.7 \times 10^{10}\left(\frac{dps}{gm}\right) = 1\left(\frac{Ci}{gm}\right)$$

3.1.7 Decay Chains

Decay of a nuclide can result in a stable nuclide but often leads to the formation of another nuclide that also remain radioactive. Thus, disintegration of a parent nuclide leads to disintegration of its daughter nuclide. This creation of new radioactive daughter products can continue as a series.

A series of radioactive decays of different decay products is called the decay chain. The decay chain is a sequential series of transformations of nuclide. The decay series present in the nature (e.g., 4n + 2 series) were described in the earlier section on natural radioactivity (Table 3.6).

A number of fission products are also part of decay chains. The following decay chain is an example.

$$^{90}_{38}Sr \overset{29.1\,yr}{\rightarrow} {}^{90}_{39}Y \overset{64.5\,hr}{\rightarrow} {}^{90}_{40}Zr$$

Strontium-90 (^{90}Sr) decays to Yttrium-90 (^{90}Y) with a half-life of about 29.1 years which in turn decays with a half-life of 64.5 hours to produce a stable zirconium-90 (^{90}Zr). Both radionuclides involve β^- decays. In general, this example represents two nuclides decay chain given as,

$$N_1 \overset{\lambda_1}{\rightarrow} N_2 \overset{\lambda_2}{\rightarrow} N_3$$

The numbers of nuclides involved in this decay chain at any given time can be obtained by solving the following equations together,

$$\frac{dN_1}{dt} = -\lambda_1 N_1 \tag{3.8}$$

$$\frac{dN_2}{dt} = -\lambda_2 N_2 + \lambda_1 N_1 \tag{3.9}$$

$$\frac{dN_3}{dt} = +\lambda_2 N_2 \tag{3.10}$$

Solutions of these equations are obtained as,

$$N_1(t) = N_1(0)\mathrm{e}^{-\lambda_1 t} \tag{3.11}$$

$$N_2(t) = \frac{\lambda_1 N_1(0)}{\lambda_2 - \lambda_1}\left[\mathrm{e}^{-\lambda_1 t} - \mathrm{e}^{-\lambda_2 t}\right] \tag{3.12}$$

$$N_3(t) = N_1(0)\left[1 + \frac{\lambda_2}{\lambda_1 - \lambda_2}\mathrm{e}^{-\lambda_1 t} - \frac{\lambda_1}{\lambda_1 - \lambda_2}\mathrm{e}^{-\lambda_2 t}\right] \tag{3.13}$$

The solutions for this two nuclides decay chain for the example decay chain of ^{90}Sr are represented in Fig. 3.7.

Note that the half-life of the parent is much longer than that of the daughter product, in this example. The results as time-dependent changes in the number of atoms show that the decay of ^{90}Sr leads to the buildup of ^{90}Y and eventually reaches the stage where the activity of ^{90}Sr and its daughter ^{90}Y are at the same level. This is because the number of decay of ^{90}Sr is much less than that of ^{90}Y in a given time period. This situation has a special name and is called “secular equilibrium”.

Under the secular equilibrium, as soon as the decay of the parent nuclide takes place, the subsequent daughter product decay almost immediately follows. This

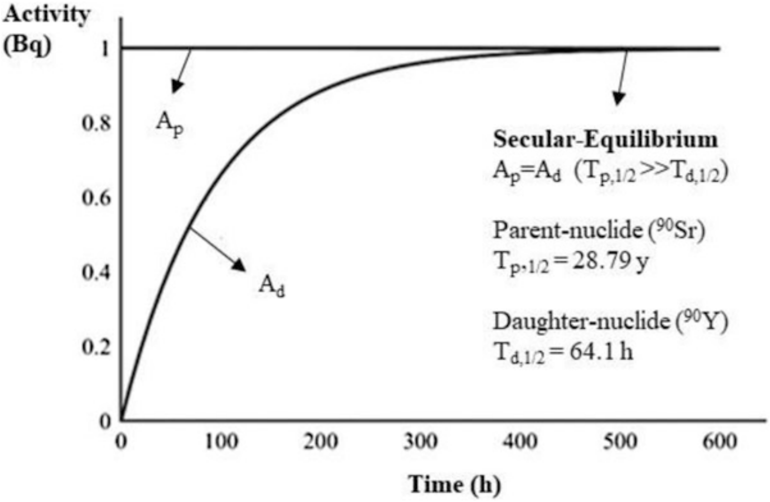


Fig. 3.7 The concept of secular equilibrium with ^{90}Sr decay chain. *(In the figure, the Y-axis is the normalized activity by the existing parent activity)*

means that as time passes, the number of decays of the parent nuclide per unit time eventually determines that of the daughter product, as shown in Fig. 3.8. This "secular equilibrium" situation happens if the half-life of the parent is very much longer than the half-life of the daughter in a decay chain.

As the half-life of the parent is much greater than that of daughter, $T_{p,1/2} >> T_{d,1/2}$, or $\lambda_2 >> \lambda_1$, the equation for the number density of the daughter can be simplified as,

$$N_2(t) = \frac{\lambda_1}{\lambda_2 - \lambda_1} N_1(0)\left[e^{-\lambda_1 t} - e^{-\lambda_2 t}\right] \cong \frac{\lambda_1}{\lambda_2} N_1(0) e^{-\lambda_1 t} \tag{3.14}$$

Thus,

$$N_2(t) = \frac{\lambda_1}{\lambda_2} N_1(t) \tag{3.15}$$

Therefore the activities of the parent and the daughter are the same, i.e. $A_2(t) = A_1(t)$. Under secular equilibrium, the activity of the daughter can be determined solely by the decay characteristics of the parent nuclide.

The 4n + 2 decay series shows the examples of secular equilibrium. While there are 14 decay products in ^{238}U decay chain, after allowing the decay of ^{238}U for a sometime, it can be found that the activities of all of the decay products of ^{238}U reach the same value.

Consider another example of decay series with β^- decay as follows.

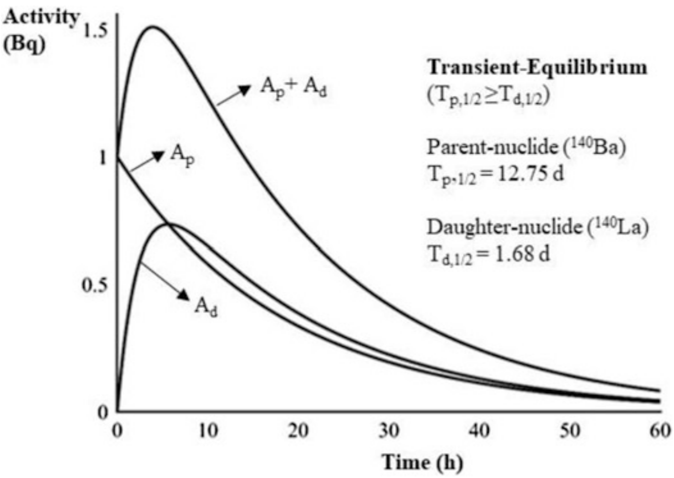


Fig. 3.8 The concept of transient equilibrium

$$^{140}_{56}Ba \overset{12.8\ d}{\rightarrow} {}^{140}_{57}La \overset{40.2\ hr}{\rightarrow} {}^{140}_{58}Ce$$

In this case, barium-140 (^{140}Ba) decays to lanthanium-140 (^{140}La) with a half-life of 12.8 days. ^{140}Ba also decays and produces a stable cerium-140 (^{140}Ce) with the half-life of 40.2 hr. Note the difference in half-life between the parent and the daughter product is not very large. In this case, while the decay of ^{140}Ba leads to the buildup of ^{140}La, the level of the activity of ^{140}Ba and its daughter ^{140}La do not reach the same level. In fact, it appears that there is a constant ratio between the activities of ^{140}Ba and ^{140}La. This is shown in Fig. 3.8.

The situation where the ratio of activity between the parent and the daughter reaches and remains constant is called "transient equilibrium". After such equilibrium is achieved, the activities of parent and daughter are decreasing in a constant ratio.

This "transient equilibrium" situation happens if the half-life of the parent is considerably longer, but not very much longer, than the half-life of the daughter in a decay chain. In this case, the rate of disintegration of not only the parent but also the daughter nuclide affects the number of decays of the daughter product.

Under transient equilibrium, as the daughter nuclide has a shorter half-life than that of its parent nuclide, the daughter nuclide builds up to an amount that remains in constant ratio to the amount of the parent nuclide and the amount of the daughter nuclide decreases at a rate controlled by the half-life of the parent nuclide.

$$N_2(t) = \frac{\lambda_1}{\lambda_2 - \lambda_1} N_1(0)\left[e^{-\lambda_1 t} - e^{-\lambda_2 t}\right] \cong \frac{\lambda_1}{\lambda_2 - \lambda_1} N_1(0)e^{-\lambda_1 t}$$
$$= \frac{\lambda_1}{\lambda_2 - \lambda_1} N_1(t) \tag{3.16}$$

$$\lambda_2 N_2 = \lambda_2 \frac{\lambda_1}{\lambda_2 - \lambda_1} N_1(t) = \frac{\lambda_2}{\lambda_2 - \lambda_1} \lambda_1 N_1 \tag{3.17}$$

Thus,

$$A_2(t) = \frac{\lambda_2}{\lambda_2 - \lambda_1} A_1(t) \tag{3.18}$$

Therefore, the activities of parent (A_1) and daughter (A_2) are maintained at a constant ratio as,

$$\frac{A_2(t)}{A_1(t)} = \frac{\lambda_2}{\lambda_2 - \lambda_1} \tag{3.19}$$

This condition is met when $T_{p,1/2} \geq T_{d,1/2}$, or $\lambda_2 \geq \lambda_1$.

There are also cases where the half-life of the daughter product is longer than the parent half-life such as,

$$^{218}_{84}Po \overset{3.10\,m}{\longrightarrow} {}^{214}_{82}Pb \overset{26.8\,m}{\longrightarrow} {}^{214}_{83}Bi$$

In this case, the activity of the parent will continue to decrease while that of the daughter will increase and then decrease. There is no equilibrium established between them. This is shown in the Fig. 3.9.

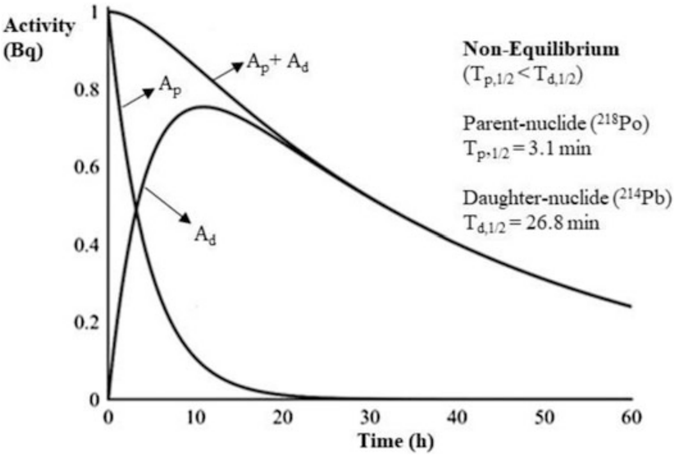


Fig. 3.9 The concept of non-equilibrium

In a multi-member decay chain, the decays occur consecutively through a series of radionuclides until reaching a stable nuclide. This is called batch decay. Batch decay is concerned with the radioactive decay of a given amount of initially pure parent material to produce various decay products. The decay products will build up and subsequently decay away as time progresses.

The general case of a multi-member decay chain can be written as,

$$N_1 \rightarrow N_2 \rightarrow N_3 \rightarrow \cdots \rightarrow N_j \rightarrow \cdots \rightarrow N_i \rightarrow \cdots$$

The amount of N_i of any nuclide present at time t can be written, if $N_1(t=0)\neq 0$, $N_{2,3..}(t=0)=0$,

$$N_i = \lambda_1\lambda_2\ldots\lambda_{i-1}N_1^0 \sum_{j=1}^{i} \frac{e^{-\lambda_j t}}{\prod_{k=1,k\neq j}^{k=i}(\lambda_k-\lambda_j)} \quad (i>1)$$

This equation is known as the Bateman equation. By superposition, the batch decay equation can be further generalized for the case of arbitrary initial amounts N_l^0 of any of the radionuclides in the chain:

$$N_i = \sum_{l=1}^{i-1}\left[\lambda_l\lambda_{l+1}\ldots\lambda_{i-1}N_l^0 \sum_{j=l}^{i}\frac{e^{-\lambda_j t}}{\prod_{k=l,k\neq j}^{k=i}(\lambda_k-\lambda_j)}\right] + N_i^0 e^{-\lambda_i t}$$

Example 3.2: Buildup and decay of radon progeny

When radon-222 (^{222}Rn) decays with a half-life 3.8 days, it forms a number of short-lived radioactive decay products called 'radon progeny' such as polonium-218 (^{218}Po), lead-214 (^{214}Bi), and bismuth-214 (^{214}Bi). All are radioactive isotopes of heavy metal elements and all have half-lives that are much less than that of radon. Radon itself does not contribute much to dose since it is immediately exhaled from the lung before decaying. The main hazard of radon comes from the inhalation of radon progeny in air. Some of the inhaled radon progeny are retained in the lung, with the subsequent alpha decays delivering energy to the surrounding tissues.

Assuming a unit activity of Po-218 is present initially, calculate the activities of Po-218, Pb-214, and Bi-214 after 10 min.

The amount of any of these nuclide at an arbitrary time t is given by the Bateman equation as below;

For $i = 3$ *and* $N_2(t) = N_3(t) = 0$; $N_1(t) = N_1(0)e^{-\lambda_1 t}$ $N_2(t) = \frac{\lambda_1 N_1(0)}{\lambda_2-\lambda_1}\left[e^{-\lambda_1 t} - e^{-\lambda_2 t}\right]$ $N_3(t) = N_1(0)\left[1 + \frac{\lambda_2}{\lambda_1-\lambda_2}e^{-\lambda_1 t} - \frac{\lambda_1}{\lambda_1-\lambda_2}e^{-\lambda_2 t}\right]$	Po-218 ($T_{1/2}$ = 3.1 min) → λ_1 = ln2 / 3.1 = 0.2236 min^{-1} Pb-214 ($T_{1/2}$ = 26.8 min) → λ_1 = ln2 / 26.8 = 0.02586 min^{-1} Bi-214 ($T_{1/2}$ = 19.9 min) → λ_1 = ln2 / 19.9 = 0.03483 min^{-1}

In terms of activity by multiplying λ_1, λ_2, and λ_3 to the above equations, respectively: $(A_1(0) = 1\ Bq)$

$A_{Po-218}(t) = A_1(t) = A_1(0)e^{-\lambda_1 t} = 0.1069\ Bq$

$$A_{Pb-214}(t) = A_2(t) = \frac{\lambda_2 A_1(0)}{\lambda_2-\lambda_1}\left[e^{-\lambda_1 t} - e^{-\lambda_2 t}\right] = 0.08701\ Bq$$

$$A_{Bi-214}(t) = A_3(t) = A_1(0)\frac{\lambda_3}{\lambda_1}\left[1 + \frac{\lambda_2}{\lambda_1-\lambda_2}e^{-\lambda_1 t} - \frac{\lambda_1}{\lambda_1-\lambda_2}e^{-\lambda_2 t}\right] = 0.02195\ Bq$$

After much longer time periods, the activity of these daughters will become the same as the parent, Rn-222, due to the presence of secular equilibrium.

3.2 Interaction of Ionizing Radiation with Matter

When radioactive particles enter matter, they go through interactions involving transfer of energy. This section describes how the transfer of energy affects the interacting system as the basis for understanding biological effects of radiation.

3.2.1 Directly or Indirectly Ionizing Radiation

In terms of the characteristics of radiation interactions with matter, ionizing radiation can be classified into directly ionizing or indirectly ionizing radiation.

Directly ionizing radiation are electrically charged (positively or negatively) particles causing ionization through direct charge-to-charge interactions with the orbital electrons. These charged particles include electrons (negatively charged), protons (positively charged), alpha particles (positively charged), and any other charged particles. Indirectly ionization radiation is uncharged particles (γ-rays, x-rays, and neutrons) but can liberate electrons or other charged particles through interactions with the target (the nucleus or orbital electrons). Thus indirectly, the radiation produces directly ionizing radiation.

Usually, electrons play the key role in energy transfer in a medium with both directly and indirectly ionizing radiation. The electrons, while being slowed down, can produce a large number of ionizations and excitations. The resulting damage will depend on the number and spatial distribution of these ionizations and excitations.

3.2.2 Interaction of Directly Ionizing Radiation with Matter

The directly ionizing (charged) particles continuously lose their energy in the interacting medium through impulses exerted as electrical forces between the charge of the particles and orbital electrons. Such charge versus charge (of the electrons) interactions are called Coulombic interactions. In this case, the charged particle does not need to hit the interacting targets for the interaction to take place as long as impulses are exerted. Therefore, the probability of a charged particle passing through a layer of matter without any interaction is nil as the layer of matter is always full of electrons.

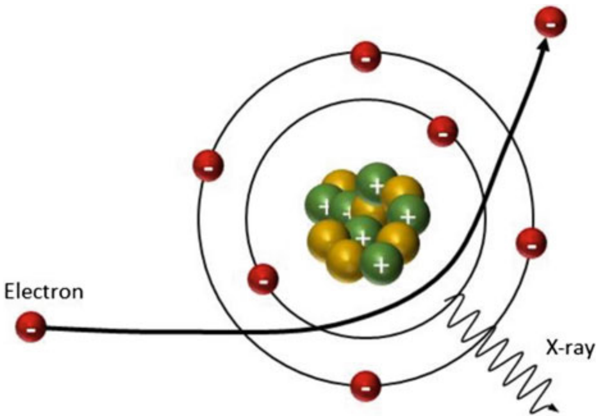
Most of these coulombic interactions individually transfer only a very small fraction of the incident particles' kinetic energy. In fact, charged particles participate in 10,000 ~ 100,000 or up to 1000,000 interactions before being completely stopped. Therefore, loss of the kinetic energy of the incident charged particle is gradual and can be considered a continuous slowing-down like process. The energy transferred in each interaction may be sufficient to knock an electron out of an atom and thus ionize it, or it may leave the atom in an excited, non-ionized state.

Stopping Power and Range

While both electrons and heavy charged particles lose energy to atomic electrons via charge vs. charge interactions, the details of the energy loss characteristics are different. For example, electrons are much more penetrating than heavy charged particles because the electrons are much lighter and their speeds are higher than heavy charged particles with the same kinetic energy. Heavy charged particles travel much less distance before losing all of the kinetic energy and being stopped.

Two parameters, i.e., stopping power and range, are used to represent the distance travelled by a directly ionizing particle and the rate of energy loss. The stopping power of a medium is the expected value of the rate of energy loss per unit path length by a charged particle (e.g., MeV/cm) at any point along their track in the medium. Stopping power generally is a function of energy, and the reciprocal of stopping power gives the distance traveled per unit energy loss. The range of a particle is defined as the expected value of the path length (in one direction) that a charged particle follows until it comes to rest. Range of a particle varies as a function of the energy of the particle. For example, the range of a 1-MeV electron in water is about 0.43 cm compared with about 7.2 μm for an α particle with the same energy. Thus, the distance travelled by a directly ionizing particles before being stopped can be very different depending on the particle type.

Fig. 3.10 An illustration of bremsstrahlung



It should be noted that electrons can also produce indirectly ionizing particles through a radiative energy transfer process called bremsstrahlung (see Fig. 3.10). This radiative energy transfer process occurs as high electron (~ MeV) decelerates around the target nucleus. In this case, the energy loss is through an x-ray emission. As electrons lose the kinetic energy through both collision and the radiative process (through bremsstrahlung production), the total stopping power of electrons in a medium has two components, i.e., collisional and radiative components (this is also true for other charged particles).

The collisional stopping power is the average energy loss by the charged particle per unit path length due to Coulomb collisions with bound atomic electrons of the medium. This collisional stopping power leads to ionization and excitation of atoms in the medium and is often called linear energy transfer (LET). The radiative stopping power is the average energy loss per unit path length due to the emission of bremsstrahlung x-ray in the electric field of the atomic nucleus and of the atomic electrons. This radiative component typically does not contribute to energy deposition in the interacting medium.

The two components of the stopping power of a charged particle in a medium are represented as.

$$Stopping\ Power = \left(\frac{dE}{dx}\right)_{collision} + \left(\frac{dE}{dx}\right)_{radiative} \tag{3.20}$$

where $\left(\frac{dE}{dx}\right)_{collision}$ is the collisional stopping power) and $\left(\frac{dE}{dx}\right)_{radiative}$ is the radiative stopping power.

Sometimes, mass stopping power (MeV cm^2/gm), which is the stopping power divided by the density of the absorbing medium, is used for its practical utility. Use of mass collisional stopping power removes the need to consider density.

LET, the collisional stopping power, is an important parameter in describing the damage caused by a charged particle. LET varies significantly depending upon the type of the particle. The following equation show the expression for LET for a heavy charged particle.

$$-\left(\frac{dE}{dx}\right)_c = \frac{4\pi k_0^2 z^2 e^4 n}{m_0 c^2 \beta^2}\left[\ln\frac{2m_0 c^2 \beta^2}{I} - \ln(1-\beta^2) - \beta^2\right] \tag{3.21}$$

where,

$k_0 = 8.99\text{x}10^9$ N m^{2}C^{-2}
m = rest mass of electron
e = magnitude of the electron charge
z = atomic number of the incident particle
ze = charge of incident particle
n = number of electrons per unit volume in the medium (= $N*Z$)
c = speed of light in vacuum
$\beta = V/c$ = speed of particle relative to c

V = velocity of incident heavy charged particle
N = number of absorber atoms per unit volume
Z = atomic number of the absorber material
I = the mean excitation energy

The equation shows that LET increases with the atomic number of the target and the charge of incident particle but decreases quickly with increase in the velocity of incident particle.

This qualitative trend also applies to the LET of electrons. However, an electron moves much faster and can lose a large fraction of its energy in a single collision with an atomic electron.

The LET for electron can be written

$$(-\frac{dE}{dx})_{col} = \frac{4\pi k_0^2 e^4}{mc^2 \beta^2} NZ \left[\ln \beta \frac{T + mc^2}{I} (\frac{I}{mc^2})^{1/2} - \frac{1}{2}\beta^2 \right] \tag{3.22}$$

where T is the electron's kinetic energy ($T = E - mc^2 = mc^2\left(\frac{1}{\sqrt{1-\beta^2}} - 1\right)$).).

At 1 MeV of the incident particle energy, the LET of an electron and an alpha particle in water is about 1.87 and 1410 MeV/cm, respectively. This shows three orders of magnitude difference and indicates that the ionization events in the medium are very densely located within the alpha particle track while the ionization event from electron is sparsely located. Therefore, the distance between ionization is very short with alpha particles implying highly concentrated energy transfer. However, the average distance between ionizations with electrons is much longer (almost three orders of magnitude) resulting in a much lower rate of energy transfer (i.e., much less concentrated energy transfer). Please also refer to Fig. 5.3 for graphic illustration of this point.

As shown in (Eq. 3.21), stopping power is inversely proportional to the square of the velocity of a charged particle, implying that the stopping power increases as the particle energy becomes lower. The radiative stopping power is negligible for the kinetic energy of the particle below 200 keV but makes a noticeable contribution to the total stopping power above 1 MeV. Also for a heavy charged particle like an alpha particle, the radiative stopping power is negligible. This is because the intensity of bremsstrahlung radiation produced is inversely proportional to the square of particle's mass. With alpha particle whose mass is more than 7000 times larger than the mass of electron, the chance of alpha particle producing bremsstrahlung is $(7000)^2$ or 4.9×10^7 times less than that of electron.

Accordingly, radiation yield (the contribution from bremsstrahlung to energy loss) is zero with alpha particle but non-negligible with electrons at energies above 1 MeV (radiation yield at ~0.5%) and increases to several percent at above 4 MeV. This also implies that shielding of high energy β-rays becomes problematic due to the production of bremsstrahlung x-rays.

For an electron, the following approximate formula gives the ratio of radiative and collisional stopping powers for particles of different energy in different absorber medium. Here E is the kinetic energy of the electron, expressed in MeV, and Z is the atomic number of an element of the medium.

$$\frac{(-dE/dx)_{radiative}}{(-dE/dx)_{collision}} \cong \frac{ZE}{800} \tag{3.23}$$

In the case of heavy charged particle, such as alpha particle, as the particle is very much heavier than the interacting electrons in the medium, each interaction transfers very small amount of energy. Therefore, for heavy charged particles to slow down, a very large number of interactions takes place in the medium. This situation is similar to a bullet being slowed down in water with straightness of the paths. All heavy charged particles exhibit the similar behavior of straight path movement while being slowed down.

In contrast, the electrons as incident particle will transfer comparatively much larger energy in a single interaction with electrons in the medium. The electrons may also be deflected in large angles. This causes the actual path of electrons to be not straight and random. The electron can also be scattered backward. Therefore, the thickness of the medium required to stop an electron is different from the path length traversed by the electron.

Range, by convention, only considers the uni-directional distance of the particle's movement ignoring any multi-directional traveling path of the particle. Thus, the fluctuations in the directions of the particles movements are ignored in the definition of range.

Table 3.8 gives example values of range, LET (i.e., collisional stopping power), and radiative stopping power of electrons and alpha particles at different energy levels. It can be seen that alpha particles in general have a much higher value of LET and smaller value of range compared to electrons. Accordingly, alpha particles (and other heavy charged particles such as protons) are called high LET radiation and

Table 3.8 Comparison of the range and LET of electrons and α particles in water

Particle	Energy (MeV)	LET (collisional stopping power) (MeV/cm)	Radiative stopping power (MeV/cm)	Radiation yield	Range in water (cm)
Electron	0.01	23.2	–	0.0001	0.00025
	0.1	4.20	–	0.0007	0.0140
	0.2	2.84	0.006	0.0012	0.0440
	0.5	2.06	0.010	0.0026	0.174
	1.0	1.87	0.017	0.0049	0.430
	4.0	1.91	0.065	0.0168	2.00
	10.0	2.00	0.183	0.0416	4.88
Alpha	1.0	1410	–	–	0.00072
	5.3*	474	–	–	0.0047

* Energy of alpha particle from ^{210}Po

Ratio of the mass of an alpha particle to an electron is 7340. The charge of the particle is +2 for an alpha particle and 1 for electron electrons are called low LET radiation. The LET values of electrons are almost constant (~ 2 MeV/cm) at energies above 0.5 MeV.

Although not explicitly described in the table, the range of an alpha particle is uniquely determined by its energy. Typical range of an alpha particle in water is close to the dimensions of a single cell in biological system (~ few tens of μm). Therefore, alpha particles cannot penetrate the skin and do not pose external hazard to humans. However, alpha particles can be very harmful through internal exposure (via inhalation or ingestion) as alpha energy will be deposited locally within a very short distance or small volume of internal organs. The range of electrons is larger than 0.1 mm at above 100 keV which is the average thickness of the epidermis and reaches a value close to a centimeter above 1 MeV. Thus, beta emitters can damage internal organs as well as the external part of body such as skin and eyes.

3.2.3 *Interaction of Indirectly Ionizing Radiation*

The interaction process of indirectly ionizing radiation (x-rays, gamma rays, and neutrons) is quite different from that of charged particles. While charged particles lose energy continuously through Coulombic interactions with the electrons in the medium, the indirectly ionizing particles lose energy in the medium “by chance”. This chance is the chance of incident particle directly hitting the “target”. This process of hitting the target is through either physical contact or a very close encounter with the target. Therefore, characterizing the probability of hitting the target is essential in describing the interactions of indirectly ionizing radiation in matter.

3.2.3.1 Cross Sections

To describe the interaction between indirectly ionizing radiation and matter ‘by chance’, a quantity called *cross section* is defined to represent the probability of interaction.

Consider an interaction involving an incident particle and a number of targets (electrons) as depicted in Fig. 3.11. We draw a spherical surface of radius r around each target. If the incident particle happens to pass through one of the targets represented by the sphere, the interaction occurs. If it does not pass through the spheres, the interaction does not occur. We call the planar area of each sphere, πr^2, the cross section, denoted by a symbol σ. Use of the word cross section here is to represent the concept of an area for interactions.

The number of targets and the cross section of each target can be used to determine the probability that a particle will interact with the targets in the region. For a particle (e.g., a gamma ray) incident perpendicularly to a thin slab of area A and thickness Δx (see Fig. 3.11), the probability of collision with the electrons in the slab is obtained by dividing the area blocked out by all of the targets by the total area of the slab, A:

$$\text{Probability of Collision} = \frac{A \cdot \Delta x \cdot N \cdot \sigma}{A} = \Delta x \cdot N \cdot \sigma \tag{3.24}$$

where, σ is the cross section of electron target, N is the number of targets per unit volume (i.e., cm^3). The total number of targets in the slab is $A \cdot \Delta x \cdot N$. The area blocked out by the targets is $A \cdot \Delta x \cdot N \cdot \sigma$ (assuming no overlapping of the targets).

Then the probability of collision per unit distance is given as,

$$\text{Prob of collision per unit distance} = \frac{\Delta x \cdot N \cdot \sigma}{\Delta x} = N\sigma \tag{3.25}$$

This probability of collision per unit distance is the product of the number of targets in unit volume and the cross section. Therefore, cross section, although as a literal geometric term, represents the probability constant for interaction as shown in the equation.

The unit of cross section is called the barn (1 barn $= 10^{-24}\ \mathrm{cm}^2$). The term, barn, was first introduced by physicists at the Los Alamos National Laboratory during World War II. They found that certain neutron cross sections became as large as $10^{-24}\ \mathrm{cm}^2$ which was perceived “as big as a barn” (i.e., big enough for the intended nuclear reaction). So the usage of the term, barn, started. Today the barn is an internationally accepted unit of cross section for nuclear reactions. It turns out that 1 barn, i.e., $10^{-24}\ \mathrm{cm}^2$, is typically the area of nuclei (indicating the barn is not an arbitrary unit of area).

When the indirectly ionizing radiation interacts in a medium, it might be absorbed, scattered, or even disappear ‘by chance’. Characterizing such chance of different interactions requires the use of relevant cross sections. Cross sections for the interactions of indirectly ionizing particles normally range from thousandths of a barn (milli-barns) to hundreds of barns. Some interactions may have extremely high

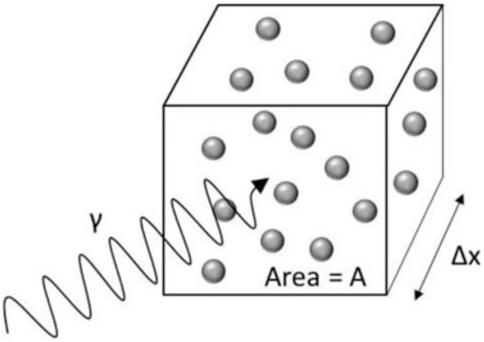


Fig. 3.11 Schematics of radiation interactions in a slab with target (electrons)

cross sections, of the order of a million barns. When several types of independent interactions are involved, a total cross section is the sum of the cross sections for each of the contributing interactions.

3.2.3.2 Interactions of Photons with Matter

Photons (x-rays or gamma rays) lose energy through chance encounters. The probability of the encounter depends on the energy of the photons and the number and size of targets (i.e., cross section). This cross section is not a fixed quantity but varies by the kinetic energy of the incident photon, depending upon the type of interactions. For example, the cross section increases dramatically with decrease in the photon kinetic energy for the interaction of photoelectric effect. In the case of the interaction of pair-production, the cross section increases with increase in photon kinetic energy. These are further described below. Also, x-rays or gamma rays of the same energy interact in a medium in exactly the same way.

Attenuation Coefficient

As cross section describe the probability of interaction, it directly affect the distance of travel of photons in a medium. The distance travelled by photon in a medium is also controlled by the composition of the medium.

As defined in Eq. 3.25, the probability of photon interaction while travelling unit distance in a medium is $N\sigma$. Such probability is also designated as the (linear) attenuation coefficient, μ. Thus,

$$\text{The attenuation coefficient} = \mu = N\sigma \tag{3.26}$$

Consider the attenuation of a photon beam in a slab. The number of photons colliding within thickness Δx of the slab is determined as the product of the number of incident photons and the probability of collision: # colliding = (number of incident photons)*(probability of collision), or

$$\Delta I = -I\Delta x \cdot \mu \tag{3.27}$$

where I is the number of the incident photons (typically as the number per area).

If the thickness of the slab is very small (i.e., $\triangle x < <1$), then the Eq. 3.27 can be re-written as,

$$\frac{dI}{dx} = -I \cdot \mu = \mu I \tag{3.28}$$

which can be solved as

$$I(x) = I(0)e^{-\mu x} \tag{3.29}$$

Therefore, the changes in the intensity of the photons in the slab is described by an exponential law, characterized by cross sections or an attenuation coefficient μ, and the thickness of the target. This is very similar to the exponential relationship of radioactive decay (Eq. 3.2).

Note that $e^{-\mu x}$ is the probability (i.e., $I/I(0)$) that a normally incident photon will traverse a slab of thickness x without an interaction. This factor $e^{-\mu x}$ then describes the fraction of "un-collided photons" that go through a material up to the distance x. Using the factor, we can also define so-called half-value layer (HVL) as the thickness of a material for which photons have a 50% probability of penetration without interaction, i.e., $e^{-\mu x} = 0.5$, $x = -\ln(0.5)/\mu = 0.693/\mu$. In 5 half-value layers, the number of photons will be reduced to 3.1% of the incident number.

The concept of a range as discussed in the discussion for charged particles is not used or applicable for indirectly ionizing radiation, as range is defined based on the assumption of continuous encounters. Instead, the distance between interactions of photons is defined as the mean free path. The mean free path (mfp) is the average distance that a photon moves between collisions and is determined as,

$$\text{mfp} = \int_0^\infty x \cdot p(x)dx$$

where, $p(x)dx$ = the probability that a photon will have its first collision in dx in the neighborhood of x = (the probability that the photon survives up to x without collision)*(the probability that it does in fact collide in the additional distance dx)

$$= \left(\frac{I(x)}{I(0)}\right) \cdot (\mu \cdot dx) = e^{-\mu x} \cdot \mu \cdot dx$$

Therefore, $\text{mfp} = \mu \int_0^\infty x e^{-\mu x} dx = \mu \left\{ -\left[\frac{xe^{-\mu x}}{\mu}\right]_0^\infty + \int_0^\infty \frac{e^{-\mu x}}{\mu} dx \right\} = \mu\left(0 + \frac{1}{\mu^2}\right) = \frac{1}{\mu}$

Unlike the case of charged particles, the interactions of indirectly ionizing particles can change the direction of travel, with or without loss of energy. Also, the amount of energy loss in a single event of interaction can be very small or none or very significant. It turns out that x-rays and gamma rays lose a small amount of energy in a single interaction event while neutrons can lose a significant amount. Accordingly, x-rays and gamma rays are classified as low LET radiation and neutrons as high LET radiation.

Types of Interactions

There are different types of interactions of photons in a medium depending upon the energy of the incident photon. These interactions include Compton scattering, photoelectric effect, pair production, photonuclear interactions, and Rayleigh (coherent) scattering.

Among them, the first three are most important with their relative importance varying with the energy of the photons:

1. The photoelectric effect is dominant at the lower photon energies (in the energy range of photon below 0.5 MeV);
2. Compton scattering dominates in the energy range from 0.5 to 5 MeV (in which a majority of the gamma rays occur), and;
3. Pair production dominates in the energy range above 5 MeV.
4. Photonuclear interactions are only significant for photon energies above a few MeV (more practically 10 MeV).
5. Rayleigh scattering occurs at low energy (about <1 MeV) with very small cross section.

In the photonuclear reaction, an energetic photon enters and excite a nucleus, which then emits a proton or neutron. While the produced proton or neutron contribute to energy transfer in the medium, the contribution is small. Thus this is commonly neglected in the discussions of health effect. Rayleigh scattering is an elastic scattering as the photon loses essentially none of its energy. The photon is usually redirected through only a small angle. It does not involve energy transfer thus not relevant to discussing radiation health effect.

Importance of photoelectric effect, Compton scattering, and pair production as a function of the energy of the incident photons is captured in Fig. 3.12. As shown, the atomic number Z of the interacting medium, in addition to the energy of photons, also affects the relative importance. The curve indicates the region of predominance in Z and E for each interaction. The curve also shows where two kinds of interactions are equally probable. For low-Z media (e.g., carbon, air, water, and human tissue) the region of Compton scattering dominance is very broad, extending from ≅20 keV to ≅30 MeV. For higher Z-media, dominance of Compton scattering becomes narrows at around 1 to a few MeV.

In the photoelectric effect (see Fig. 3.13), the incident photon is absorbed in the struck atom releasing photoelectron. Thus, the photoelectric effect is an absorption interaction with the atom as the target. The incident photon is absorbed by the atom raising the atom to an excited state. Then the atom de-excites by ejecting one of its orbital electrons, usually one of the tightly bound (lower energy level) electrons. The energy of the ejected electron is equal to the energy of the photon minus the electron's binding energy.

Sometimes, electron ejection causes the emission of a second electron called Auger electron. When the tightly bound electron is released, the vacancy left is filled by another electron from a higher energy orbit falling into the vacancy. This usually results in a release of energy as characteristic x-ray. Instead of releasing characteristics x-ray, the energy of the photon can also be transferred to an outer electron, which is ejected from the atom. The electron is called an *Auger electron*. In this case, the kinetic energy of Auger electron is discreet as the energy of the photon minus the binding energy of the Auger electron. The probability of Auger electron release decreases with atomic number. The probability of Auger electron release is about the same as the release of characteristic x-ray at $Z = 30$.

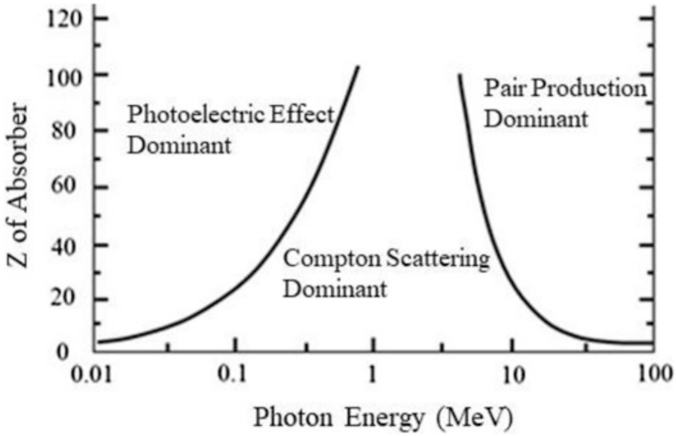


Fig. 3.12 Relative importance of the three major types of photon interactions

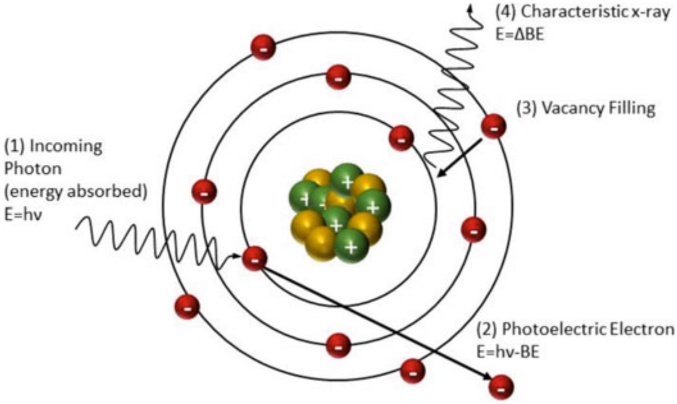


Fig. 3.13 Events involved in the photoelectric effect process

In Compton scattering (Fig. 3.14), the incident photon has an elastic collision with a free orbital electron (or an electron with small binding energy compared to the energy of incident photon). The incident photon strikes an electron, and the interaction produces ejection of an energetic electron and a scattered photon.

In pair production, the incident photon interacts with an atom (in the fields of the nucleus) and is completely absorbed. In the place of absorbed photon, a positron-electron pair is created and released. Thus pair-production is an absorption process in which a photon disappears and gives rise to an electron and a positron. However, the positron is quickly annihilated and produces two photons at 0.511 MeV(see Fig. 3.15). Therefore, the minimum energy of the incident photon to produce pair production is the sum of the rest mass energy of two photons which is equal to 1.022 (0.511 × 2) MeV.

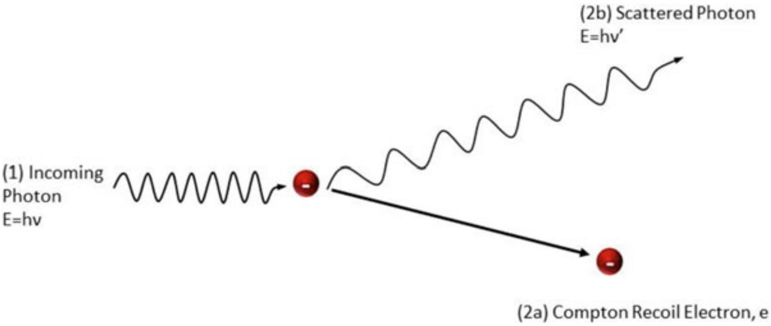


Fig. 3.14 A schematic of Compton scattering

The linear attenuation coefficient, i.e., the probability of interaction per unit distance, can be determined for each of the photon interactions. Assume that the absorber material consists of N atoms per cm^3, the linear attenuation coefficient (cm^{-1}) for the photoelectric effect (τ), Compton scattering (σ), and pair production (κ) is defined, respectively, as,

$$\text{For the photoelectric effect,} \qquad \tau = \mathrm{N} \cdot {}_a\tau \tag{3.30}$$

$$\text{For Compton scattering,} \qquad \sigma = \mathrm{N} \cdot Z \cdot {}_e\sigma = \mathrm{N} \cdot {}_a\sigma = n \cdot {}_e\sigma \tag{3.31}$$

$$\text{For pair production,} \qquad \kappa = \mathrm{N} \cdot {}_a\kappa \tag{3.32}$$

where, N is atoms/cm^3, Z is atomic number, ${}_a\tau$ is the cross section for photoelectric effect per atom, ${}_e\sigma$ and ${}_a\sigma$ are the cross section for Compton scattering per electron and per atom, respectively, and ${}_a\kappa$ is the cross section for pair production per atom. Here the subscript a and e denotes atom and electron, indicating the target of the respective interactions.

The cross section per atom for the photoelectric effect is known to increase sharply with the increase in the atomic number of the target material. For high Z elements, the dependency is approximately Z^3 and for low Z elements, the dependency is $Z^{3.8}$. The cross section also decreases sharply with the energy of photons (hv). The dependency is approximately $1/(hv)^3$ at low energy (~0.1 MeV) and gradually changes to $1/(hv)$ at high energy (~ 5 MeV).

In the low energy region (~ 0.1 MeV) where the photoelectric effect is most dominant, the approximate cross section per atom for photoelectric effect can be represented as,

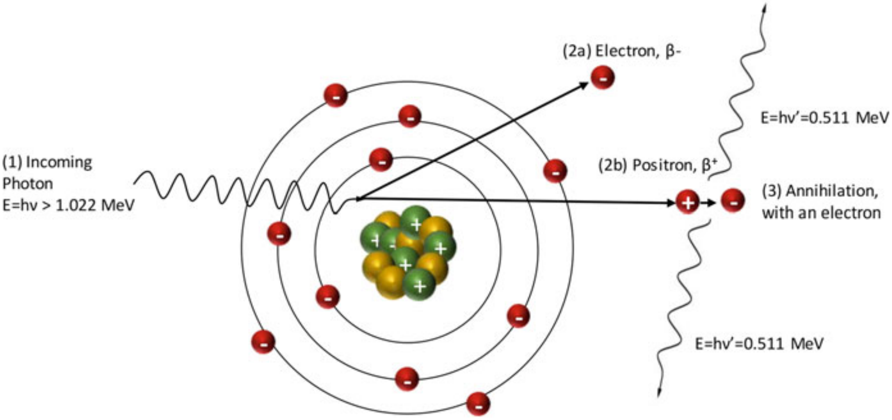


Fig. 3.15 A schematic of the pair production process

$$
{}_{a}\tau \propto \frac{Z^4}{(hv)^3}\left(\frac{cm^2}{atom}\right) \tag{3.33}
$$

In contrast, the cross section per electron for Compton scattering is almost independent of atomic number and decreases as the photon energy increases.

$$
{}_{e}\sigma \propto 1/(hv)
$$

The cross section per atom for pair-production ($({}_{a}\kappa)$) increases as Z^2 and logarithmically with photon energy hv.

$$
a^k \propto z^2 \cdot \ln (hv)
$$

The probability of a photoelectric effect per length dx is τdx. Likewise, the probability of a Compton scattering and pair production per length dx is σdx and κdx, respectively. To consider the combined effect of all three interactions, the probabilities of each of the three interactions are combined. Accordingly, the probability of photon interaction for all three interactions per unit distance traveled becomes

$(\tau + \sigma + \kappa)dx$. This quantity is given a special name of total linear attenuation coefficient (ignoring Rayleigh scattering and photonuclear interaction).

$$
\textit{Total linear attenuation coefficient} = \mu = \tau + \sigma + \kappa \quad \left[cm^{-1}\right] \tag{3.34}
$$

Mass attenuation coefficient is another term widely used to describe photon interactions in matter. Mass attenuation coefficient is defined as the total linear

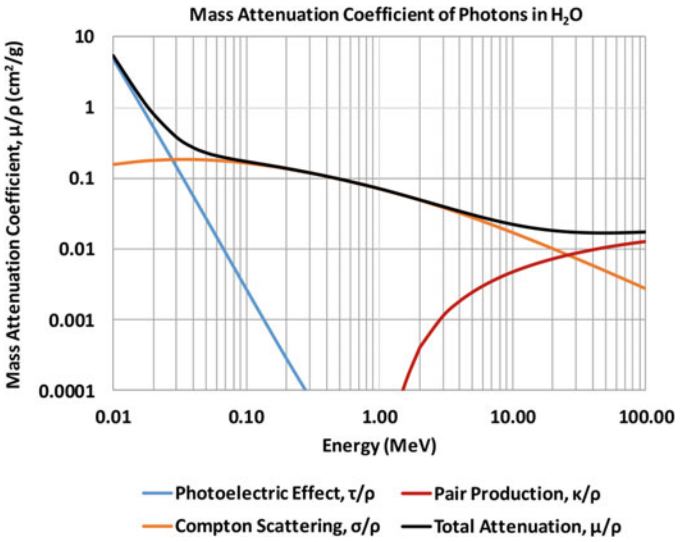


Fig. 3.16 Mass attenuation coefficient of photons in water (data source: NIST XCOM Database)

attenuation coefficient divided by the density of the interacting medium, ρ. Then the total mass attenuation coefficient of the medium is defined as,

$$\text{Total mass attenuation coefficient} = \frac{\mu}{\rho} = \frac{\tau}{\rho} + \frac{\sigma}{\rho} + \frac{\kappa}{\rho} \quad \left[\frac{cm^2}{g}\right] \tag{3.35}$$

Figure 3.16 shows the contribution of each term to the total mass attenuation coefficient in water as an example.

Figure 3.17 gives the mass attenuation coefficients of photons for several materials of interest (i.e., water, muscle, aluminum, concrete, lead). The energy range covered is from 0.010 MeV to 100 MeV. At low photon energies, the mass attenuation coefficient is mainly the result of photoelectric effect with the values being higher with higher Z materials. In the case of lead, the so-called k-edge effect is shown which represents a sudden increase in the cross section of photoelectric effect when the energy of the photon is just above the binding energy of the innermost electron shell of the atoms interacting with the photon. This sudden increase reflects the resonance effect (the frequency of electron circulation around a nucleus matches the frequency of the incident photon) giving the electron the greatest chance to be separated from the atom. This effect is clearly shown with high atomic number targets but disappears with low Z targets.

The curves also indicate the values of the mass attenuation coefficient decreasing rapidly with photon energy increase. Then in the photon energy range of several hundred keV to a few MeV, Compton scattering mainly controls the value of mass attenuation coefficient. The difference between absorber materials is small as the

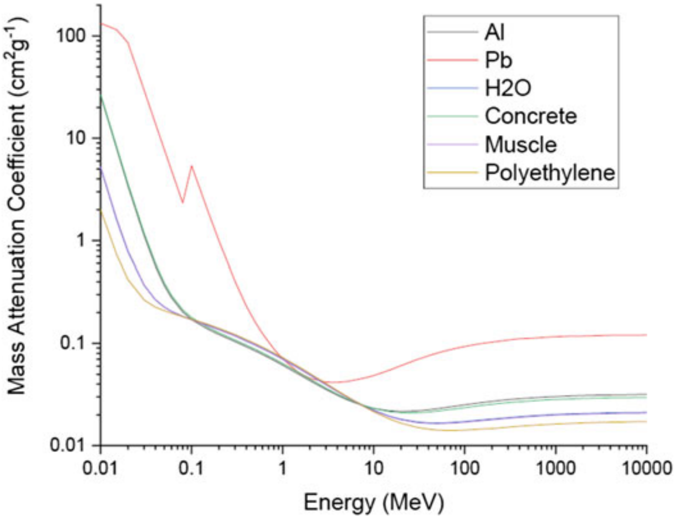


Fig. 3.17 Mass attenuation coefficients for various materials

elements (except hydrogen) contain about the same number of electrons per unit mass (*NZ/A*, where *Z/A* is almost constant, ranging between 0.4 and 0.5). Pair-production starts playing a role at above its threshold energy of 1.022-MeV but its effect becomes important above ~10 MeV with enhanced effect with higher Z absorber materials.

Also, the probability of survival of photons within a medium is of interest in terms of photon's penetrating capability. The probability of photons having no photoelectric interaction in a distance x is $e^{-\tau x}$. Also, the probability of no Compton Effect or no pair production is $e^{-\sigma x}$ and $e^{-\kappa x}$, respectively. Then the probability of no interaction of any kind is the probability of no interaction of all three of the three interactions. Such probability is the product of the probabilities of the incident particle surviving each photon interaction mechanism as,

$$Probability\ of\ Survival = e^{-\tau x}e^{-\sigma x}e^{-\kappa x} = e^{-(\tau+\sigma+\kappa)x} = e^{-\mu x} \tag{3.36}$$

Energy Transfer and Energy Absorption

One of the key issues in describing radiation interactions in a matter is to understand how the interactions affect biological systems through energy absorption. In the case of photons, absorption of their energy in a medium leads to production of photo-electrons, Compton scattered electrons, and electrons released from pair-production. These electrons then go through Coulombic-force interactions with electrons in the medium for energy deposition.

The energy of electrons released from the photoelectric effect is the initial photon energy minus the binding energy of the ejected electron (*hv-BE*). In Compton

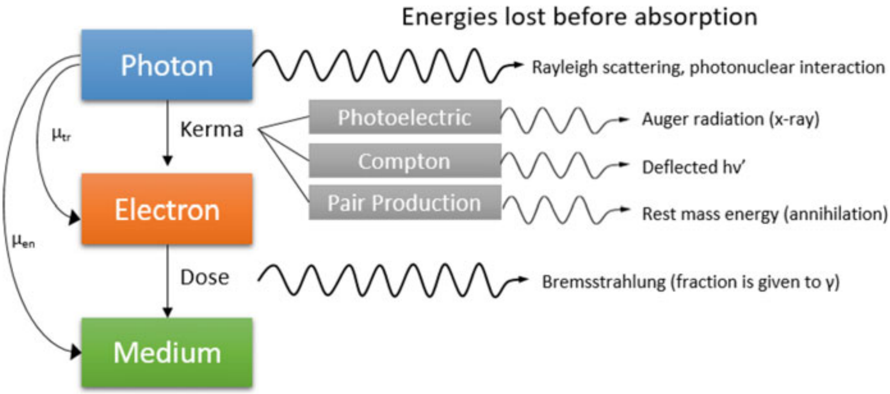


Fig. 3.18 The concept of *kerma* and *absorbed dose* for photons

scattering, the scattered electron carries the energy of incident photon minus the energy of the deflected photon (hv-hv'). In pair-production, the energy of the ejected electrons is the energy of the initial photon minus the rest mass energy of the two electrons released (hv-$2m_0c^2$). In a biological system, as the range of these electrons is rather short, almost all of the electron energy is absorbed in a small volume. An exception to this is the energy used for bremsstrahlung x-ray production. The resulting x-rays are likely to move away from the system without depositing much of its energy in the local area.

We can view energy deposition from the incident photons into the target medium as two step process. The first step is the process of energy transfer from the photons to the electrons. The amount of energy transferred from the incident photons to the electrons in the medium per unit mass is called *kerma*. As an acronym, *kerma* stands for "kinetic energy released per unit mass". *Kerma* is the sum of the initial kinetic energies of all the electrons released through photon interactions in a matter, divided by the mass of the matter.

The second step is the process of energy absorption by the medium through Coulombic interactions with the electrons. The amount of energy absorbed by the medium per unit mass is called *dose* or *absorbed dose*. A explained in Chap. 5, absorbed dose is the key parameter of interest in describing biological effects of radiation.

These two stages of energy absorption is depicted in Fig. 3.18. *Kerma represents the energy given to electrons from the incident photons and absorbed dose represents the energy given to the medium from the electrons*. In reality, absorbed dose is very close to kerma. The difference between the two exists only if the energy of the ejected electrons are lost without being absorbed, such as through bremsstrahlung x-ray production. *The unit for both quantities is Gray (Gy), as the energy deposited per unit mass, i.e., joule per kg. 1 Gy is equal to 1 J/kg.*

Example 3.4 Calculation of Kerma and Absorbed Dose

A 10-MeV gamma-ray enters a volume V of a homogeneous material and undergoes pair production, thereby disappearing and giving rise to an electron and positron of equal energies. The electron spends half its kinetic energy in collision interactions before escaping from V. The positron spends half of its kinetic energy in collisions in V before being annihilated in flight. Assuming that,

– the actual number of events in the given mass is 10^4 times what is described above with each photon going through exactly the same reaction,

– the density of the homogenous material in V is 1.1 (g/cm^3).

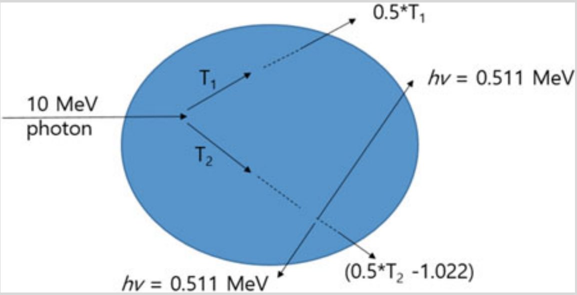
– the total mass in V is 10^{-4} g,

– the mass of the homogeneous material is 24 g/mole.

Determine the kerma and the absorbed dose in V in rad and Gy

Answers:

(a) Determine the kerma and the absorbed dose in V in Gy (1 Gy = 6.24×10^9 MeV/g)



The photon disappears and releases two electrons (T_1 and T_2) in the target

The energy transferred to the two electrons are the energy of the incident photon minus the rest mass energy of photons = 10 MeV – 2*(0.511) MeV = 8.98 MeV

Therefore, $T_1 = T_2 = 8.98/2 = 4.49$ MeV

Kerma = 8.98 (MeV) × $10^4/10^{-4}$ g = 8.98 × 10^8 MeV/g = 0.144 Gy

Incident photon energy absorbed in the target from the electrons = 8.98 MeV – 0.5*T1 – (0.5*T2–1.022) – 1.022 (this is from positron annihilation) = 8.98–0.5*T1–0.5*T2 = 4.49 MeV

Absorbed dose = 4.49 (MeV) × $10^4/10^{-4}$ g = 4.49 × 10^8 MeV/g = 0.072 Gy

---

Kerma and absorbed dose can be quantitatively determined by using the concept of the linear energy transfer coefficient and the linear energy absorption coefficient, respectively. These two quantities are the variations of the linear attenuation coefficient. The linear energy transfer coefficient reflects the loss of energy from the incident photon to electrons in the linear attenuation coefficient. The linear energy absorption coefficient further reflect the loss of energy by bremsstrahlung x-ray production.

The linear energy transfer coefficient can be seen as the weighted sum of the linear attenuation coefficient for photoelectric effect, Compton scattering, and pair-production, where the weights are the ratio of the energy of the incident photon to the energy of the ejected electron. This is shown in the following.

$$\mu_{tr} = \tau_{tr} + \sigma_{tr} + \kappa_{tr} = \tau\,[h\nu - Be] + \sigma\left[\frac{h\nu - h\nu'}{h\nu}\right] + \kappa\left[\frac{h\nu - 2m_0c^2}{h\nu}\right] \tag{3.37}$$

Therefore, the linear energy-transfer coefficient (μ_{tr}) is the probability of photon interaction per unit distance traveled in a matter transferring energy to electrons per unit distance traveled. This represents the interaction probability per unit distance contributing to kerma.

The linear energy absorption coefficient is the linear energy transfer coefficient times the ratio of the energy absorbed in the medium to the energy transferred to electrons from photons. That ratio of energy absorption is one minus the fraction of the energy lost in radiative transfer.

$$\mu_{en} = \mu_{tr}(1 - G) \tag{3.38}$$

where, G is the average fraction of the initial kinetic energy transferred to ejected electrons that is subsequently emitted as bremsstrahlung. Therefore, the linear energy absorption coefficient (μ_{en}) is the interaction probability per unit distance contributing to absorbed dose. For low values of *Z* and *hv, G* approaches zero. For increasing Z or *hv, G* increases gradually. For example, in Pb with $h\nu = 10$ MeV, *G* is 0.26.

The mass energy transfer coefficient (μ_{tr}/ρ), and the mass energy absorption coefficient (μ_{en}/ρ) in a medium are also often used for the calculation of kerma and absorbed dose as,

$$\frac{\mu_{tr}}{\rho} = \frac{\tau_{tr}}{\rho} + \frac{\sigma_{tr}}{\rho} + \frac{\kappa_{tr}}{\rho} = \frac{\tau}{\rho}\left[\frac{h\nu - BE}{h\nu}\right] + \frac{\sigma}{\rho}\left[\frac{\overline{T}}{h\nu}\right] + \frac{\kappa}{\rho}\left[\frac{h\nu - 2m_0c^2}{h\nu}\right] \tag{3.39}$$

$$\frac{\mu_{en}}{\rho} = \frac{\mu_{tr}}{\rho}(1 - G) \tag{3.40}$$

As examples, Table 3.9 shows the values of mass attenuation coefficient, mass energy transfer coefficient, and mass energy-absorption coefficient in water and lead. In water, the effect of bremsstrahlung is very small for photon energies less than ~10 MeV. In lead, a high Z material, the bremsstrahlung effect becomes prominent accounting for a significant difference between the mass energy-transfer coefficient and mass energy-absorption coefficient.

The total energy absorbed in a matter per unit distance traveled by the photon can be represented as the product of the energy of the incident photon, the number of photons per area, and the energy absorption coefficient, as shown in the following equation.

Table 3.9 Photon mass attenuation, mass-energy transfer, and mass-energy absorption coefficients (cm^2/g) in Water and Lead (RSIC 1988)

	Water			Lead		
Energy (MeV)	μ/ρ	μ_{tr}/ρ	μ_{en}/ρ	μ/ρ	μ_{tr}/ρ	μ_{en}/ρ
0.01	5.33	4.95	4.95	131	126	126
0.10	0.171	0.0255	0.0255	5.55	2.16	2.16
0.50	0.0969	0.0330	0.0330	0.1588	0.0945	0.0898
1.0	0.0708	0.0311	0.0310	0.0710	0.0389	0.0379
2.0	0.0494	0.0262	0.0260	0.0453	0.0257	0.0230
3.0	0.0397	0.0230	0.0228	0.0417	0.0259	0.0225
5.0	0.0303	0.0195	0.0191	0.0423	0.0305	0.0253
10.0	0.0222	0.0163	0.0157	0.0497	0.0418	0.0325
100.0	0.0173	0.0167	0.0122	0.0931	0.0918	0.0323

$$\begin{aligned} Total\ energy\ absorbed &= \Phi h\nu\mu_{en} \\ &= \Phi h\nu\frac{\mu_{en}}{\rho}\rho \text{ (as energy absorbed per volume)} \end{aligned} \tag{3.41}$$

where, Φ is the number of incident photons per unit area (this is called the fluence) and *hv* is the kinetic energy of the photons. If the number of photons passing per unit area per time (this is called flux or flux density) is used instead of using fluence, the result is the rate of energy absorption per unit mass, i.e., the absorbed dose rate. Similarly, the product of the fluence, the energy of photon, and the energy transfer coefficient is the total energy transferred to the electrons, i.e., kerma. Using the flux instead of fluence gives the kerma rate in the medium.

Example 3.5 Energy Transfer from the Photons

Consider a beam of 3-MeV gamma-rays perpendicularly incident on an Fe foil that is very thin in comparison with the range of the secondary electrons.

(a) Calculate the expected value of the energy transferred to charged particles per unit mass of the foil in Gy.

(b) Calculate the expected value of the energy imparted to matter per unit mass of the foil in Gy.

- ${}_a\tau$, the microscopic cross section for photoelectric effect *per atom* = 10.3 barn.
- ${}_e\sigma$, the Klein-Nishina cross section *per electron* = 0.168 barn.
- ${}_a\kappa$, the microscopic cross section for pair-production *per atom* = 0
- B_e, the average binding energy of the photoelectrons = 88 keV
- E, is the average kinetic energy of the Compton scattered photons = 1.255 MeV
- *G*, the average fraction of the initial kinetic energies of the electrons that is lost radiatively = 0.028
- ρ, the density of the material = 7.86 g/cm3
- A, the atomic weight = 55.847
- Z is the atomic number = 26

- N_a is the Avogadro's number

(a) *Calculate the expected value of the energy transferred to charged particles per unit mass of the foil in Gy.*

$$\mu_{tr} = \tau_{tr} + \sigma_{tr} + \kappa_{tr} = N \cdot {}_a\tau\left[\frac{h\nu - BE}{h\nu}\right] + NZ \cdot {}_a\sigma\left[\frac{\overline{T}}{h\nu}\right] + N \cdot {}_a\kappa\left[\frac{h\nu - 2m_0c^2}{h\nu}\right]$$

$$N = \frac{\rho N_a}{A} = 8.47 \times 10^{22}\,\frac{atoms}{cm^3} = 0.0847 \times 10^{24} cm^{-3}$$

(to be later canceled with unit "barn")

$$\mu_{tr} = 0.0847 \times 10^{24} cm^{-3} \times 10.3 \times 10^{-24} cm^{-2}\left(\frac{3-0.088}{3}\right) + 0.0847 \times 26 \times 0.168 \left(\frac{3-1.255}{3}\right) + 0.0847 \times 0\left(\frac{3-1.02}{3}\right) = 1.06 cm^{-1}$$

$$\frac{\mu_{tr}}{\rho} = \frac{1.06 cm^{-1}}{7.86\ g/cm^3} = 0.135 cm^2/g$$

The energy transferred to charged particles per unit fluence =
3 MeV · 0.135 (cm^2/g) = 0.405 (MeV/g per 1 particle per cm^2)
= 6.5 × 10^{-11} Gy (1 Gy = 6.24 × 10^9 MeV/g)

(b) *Calculate the expected value of the energy absorbed in the matter per unit mass of the foil in Gy.*

The expected value of the energy imparted to matter = 6.5 × 10^{-11}·(1-G) = 6.5 × 10^{-11}· (1 − 0.028) = 6.3 × 10^{-11} Gy

3.2.3.3 Interactions of Neutrons with Matter

Neutrons are another type of indirectly ionizing radiation. As an electrically neutral particle, neutron interacts with matter only when it collides with a target. With regard to energy transfer, the target of neutron interactions is nucleus not electrons.

Neutrons interact with atomic nuclei via nuclear force. The nuclear force is very short ranged (~10^{-15} meter) and neutrons have to move close to a nucleus to be able to interact. Because of small size of the nucleus (~ 10^{-13} cm), neutrons have low probability of interaction.

In nuclear reaction, the incident neutron strikes the target nucleus and coalesces with it to form a compound nucleus. The compound nucleus is unstable and may release the neutron back or goes through emission of other particles including gamma rays. The case of neutron being released back is termed scattering. When other particles are released, the reaction is termed absorption. Therefore, interaction of a neutron with nucleus results in either absorption or scattering of neutrons.

Neutron scattering can appear as neutrons being deflected after the interaction and moving in different directions. If the scattered neutron carries the same amount of energy as before the interaction, it is called elastic scattering. This is represented as (n, n) reaction. In elastic scattering, energy of neutron is too small to change the energy of nucleus and neutron cannot transfer kinetic energy to a nucleus.

If the neutron loses some amount of energy from the interaction, it is called inelastic scattering. Inelastic is represented as (n, n') reaction. When the scattering is inelastic, the nucleus absorbs some energy internally and is left in an excited state. Therefore, inelastic scattering does not occur unless the neutron has sufficient energy to place the target nucleus in its first excited state. Except for the lightest nuclei, hydrogen, all nuclei have excited states. Also, in a nucleus, there is a state of lowest energy, the ground state, and the excited states. A nucleus in an excited state can lose its excitation energy by emitting γ-rays (or in special cases through releasing electrons).

In neutron absorption, the unstable compound nucleus relieves its energy in a variety of ways. If the compound nucleus goes through emission of one or more gamma rays, this is called the neutron capture reaction. This capture reaction is represented as (n, γ) reaction. There are also cases where the compound nucleus may release other forms of particles such as charged particles like proton (i.e., (n, p) reaction), alpha particle (i.e., (n, α) reaction), or two or more neutrons (i.e., (n, 2n) or (n, 3n) reaction). Reactions of (n,2n) and (n,3n) occur with energetic neutrons. These reactions (i.e., (n, p), (n, α), (n, 2n), (n, 3n)) are also called neutron transfer reactions.

Sometimes, in the case of very heavy nucleus, nuclear fission occurs producing two or more smaller parts (lighter nuclei called fission fragments), represented as (n, f). In nuclear fission, a large amount of energy is released along with two or more free neutrons and gamma rays. This fission reaction becomes the basis of fission nuclear reactors.

As discussed in Sect. 3.2.3.1, a cross-section, σ, defined for a neutron for particular reaction represents the effective 'target area' of a nucleus with an incident neutron. It has a unit of barn (10^{-24} cm^2), and it can be interpreted as the probability of particular interaction between an incident neutron and a target nucleus. This cross section is also called microscopic cross section.

For neutron interactions, the linear attenuation coefficient defined in photon interactions (Eq. 3.26) is given a different name as the macroscopic cross section. A macroscopic cross section is the product of the microscopic cross section and the number of target nuclei in a unit volume (this is called the number density) as,

$$\Sigma = \sigma N \tag{3.42}$$

The number density, N (nuclei density) is given in the unit of the number of nuclei/cm^3. The macroscopic cross-section Σ has the unit of cm^{-1} and represents the probability of interaction per unit distance. The microscopic cross-section of neutron depends on target nucleus (hydrogen, boron, uranium, etc.) and neutron energy (thermal neutron, epithermal neutron, fast neutron, see the definitions in 3.3.1). Typically the microscopic cross section is much larger at low energies than at high energies. Examples of the neutron cross-sections of the common nuclides inside a nuclear reactor are given in Table 3.10.

To describe possible impact on humans, the main targets of neutron interactions are hydrogen, oxygen, carbon, and nitrogen as the main elements in the human tissue as shown Table 3.11.

In terms of energy transfer of neutrons in a medium, for fast neutrons, the recoil protons resulting from elastic scattering of hydrogen nuclei mostly contribute to the deposition of energy in tissue. As the energy of neutrons become lower, the cross section for capture increases, i.e., epithermal and thermal neutrons have a high probability of being captured in the tissue. For these neutrons, the two principal

Table 3.10 Neutron cross-section data of common materials used inside a nuclear reactor (in barn, 10^{-24} cm^2)

		Thermal neutron			Fast neutron		
		Scattering	Capture	Fission	Scattering	Capture	Fission
Moderator	H-1	20	0.2	–	4	0.00004	–
	H-2	4	0.0003	–	3	0.000007	–
	C-12	5	0.002	–	2	0.00001	–
Structural materials Others	Zr-90	5	0.006	–	5	0.006	–
	Fe-56	10	2	–	20	0.003	–
	Cr-52	3	0.5	–	3	0.002	–
	Ni-58	20	3	–	3	0.008	–
	O-16	4	0.0001	–	3	0.00000003	–
Absorber	B-10	2	200	–	1	0.4	–
	Cd-113	100	30	–	4	0.05	–
	Xe-135	400	2000,000	–	5	0.0008	–
	In-115	0.0201	100	–	4	0.02	–
Fuel	U-235	10	29,900	583	4	0.09	1
	U-238	39	2	0.00002	5	0.07	0.3
	Pu-239	8	269	748	5	0.05	2

Table 3.11 Composition of Human Body Tissue (Weight Percent) (source: ICRU 1977)

Element	Muscle (general)	Skeletal Muscle	Whole Body
H	10.2	10.06	10.5
O	72.9	75.48	67.7
C	12.3	10.78	18.7
N	3.5	2.77	3.1
Other	1.1	0.91	
Total	**100**	**100**	**100**

reactions of neutron capture in tissue are ($^{1}H(n,\gamma)^{2}H$) with hydrogen and ($^{14}N(n,p)^{14}C$) with nitrogen. The (n,γ) reaction releases a 2.22-MeV gamma ray. The $^{14}N(n,p)^{14}C$ reaction releases the kinetic energy of the recoil carbon (0.04 MeV) and proton (0.58 MeV), i.e., 0.626 MeV. The energy of gamma ray mostly escapes the body depositing only a fraction of its energy. The energy of the proton and the recoil carbon nucleus is deposited in the immediate vicinity of the capture site, leading to absorbed dose.

Example 3.6: Calculation of the absorbed dose

Calculate the absorbed dose in a 1-g sample of muscle tissue exposed to a fluence of 10^8 cm^{-2} thermal neutrons. Assume one gram of muscle tissue contains approximately 0.102 g of hydrogen, 0.123 g of carbon, 0.740 g of oxygen, and 0.035 g of nitrogen.

Solution:

Number of atoms per gram of muscle tissue:

– Hydrogen: 6.022×10^{23} (atoms/mol)· (0.102 (g/g-muscle)/1.008 (g/mol)) = 6.09×10^{22} atoms/g-muscle

– Carbon: 6.022×10^{23}*(0.123/12.01) = 6.17×10^{21} atoms/g

– Oxygen: 6.022×10^{23}*(0.740/16.0) = 2.79×10^{22} atoms/g

– Nitrogen: 6.022×10^{23}*(0.035/14.01) = 1.50×10^{21} atoms/g

Density of muscle tissue ~1.06 g/cm^3

The dose from $^1H(n,\gamma)^2H$ is negligible as the size of the target is small. Therefore, the energy absorption is through the $^{14}N(n,p)^{14}C$ reaction. In this case, each capture event by nitrogen-14 results in the deposition of energy E = 0.626 MeV, which will be deposited in the sample. The thermal neutron capture cross section = 1.70 b. N-14 is ~99.6% abundant.

Dose = $10^8\ cm^{-2} \times 1.50 \times 10^{21}$ atoms/g $\times 0.996 \times 1.70 \times 10^{-24}\ cm^2$
$\times$ 0.626 MeV = 1.59×10^5 MeV/g = 2.6×10^{-5} Gy

3.3 Nuclear Reactors

Nuclear reactor is a device to produce energy by using nuclear fission. Through nuclear fission, nuclear energy stored in the nuclear fuel is released and converted into thermal energy. This thermal energy is used to heat up the coolant (e.g., water or helium). Then the energy stored in the coolant is used to produce steam. The produced steam in turn drives the turbine. The mechanical energy derived by driving the turbines is then converted into electrical energy through connection to electric generators.

As far as being an energy generating device, nuclear reactor performs a similar function as a coal-fired power plant. In a coal-fired power station, chemical energy stored in the fuel (i.e., coal) is converted into thermal energy, followed by its conversion to mechanical energy and then electrical energy. In a coal-fired power station, oxygen (i.e., chemical energy) is used to keep the fuel burning. In a nuclear reactor, neutrons are used to keep the burning of fuel that is uranium by nuclear fission. While the coal-fired power plant produces a large volume of effluents (including CO_2) and ashes as waste, nuclear power plant produces spent nuclear fuel as waste at much smaller volume.

The concept of nuclear reactor was first proposed by Leo Szilard in his 1934 patent (British patent no. 630726, application first filed on June 28, 1934 and published on Sep. 28, 1949). The first man-made nuclear reactor was built and operated in December 1942 in Chicago, Illinois, by Enrico Fermi as part of Manhattan Project.

A natural nuclear reactor also existed and operated at Oklo in Gabon, Africa about two billion years ago. Existence of such natural reactor was due to the presence of a uranium-rich mineral deposit flooded with water as neutron moderator. Such combination started fission chain reactions which were sustained for hundreds of thousands of years. Presence of unnatural fission products and abnormal depletion of ^{235}U content in the soil remains providing the indication of sustained nuclear fission reactions.

Nuclear reactors are quite different from nuclear weapons in terms of the possibility of explosion. While nuclear weapons use highly enriched uranium (typically more than 80–90% of the uranium used is ^{235}U) or plutonium (^{239}Pu), most nuclear reactors use low-enriched uranium with the portion of ^{235}U at less than 5%. As ^{238}U makes up the remaining majority (more than 95%) of fuel, the process called Doppler broadening in ^{238}U prevents the process of power excursion in nuclear reactors. Doppler broadening is a negative feedback mechanism to decrease the power level when the temperature of the system rises from energy generation. This negative feedback doesn't exist in nuclear weapons. Also in nuclear reactors, neutrons are produced not only promptly (within 10^{-14} seconds) from nuclear fission but also from the beta decay of fission products as delayed neutrons (after a few milliseconds or up to a few minutes after the fission). Presence of such delayed neutrons (at less than 1% of total neutrons) enables control of nuclear reactors because, if only prompt neutrons are utilized, a nuclear reactor is in a subcritical state (i.e., not able to sustain fission chain reactions). The lacking portion of neutrons becomes available when the fission reactor is about to die out as delayed neutrons, just in time to sustain the chain reaction. The time scale governed by the delayed neutron production is slow enough allowing the control of the system. Such control is not possible in nuclear weapons due to extremely rapid development of the nuclear detonation process.

3.3.1 Types of Nuclear Reactors

The neutrons generated from fission have energies ranging from a few hundred keV to about 10 MeV. The average energy of fission neutrons is 1.98 MeV and the most probable energy is 0.73 MeV. Probability of interaction is strongly affected by energy of the neutrons. If the energy is 10 keV or upward, the neutron is referred to as "fast". Neutrons with energies below 10 keV but above 0.5 eV are called "epithermal" (or intermediate-energy) neutrons. All neutrons with energies below 0.5 eV are usually called "thermal" neutrons. Thermals neutrons have lost most of its initial energy and have come to thermal equilibrium with their surroundings. In the case of thermal equilibrium at 20 °C, the corresponding neutron energy is about 0.025 eV.

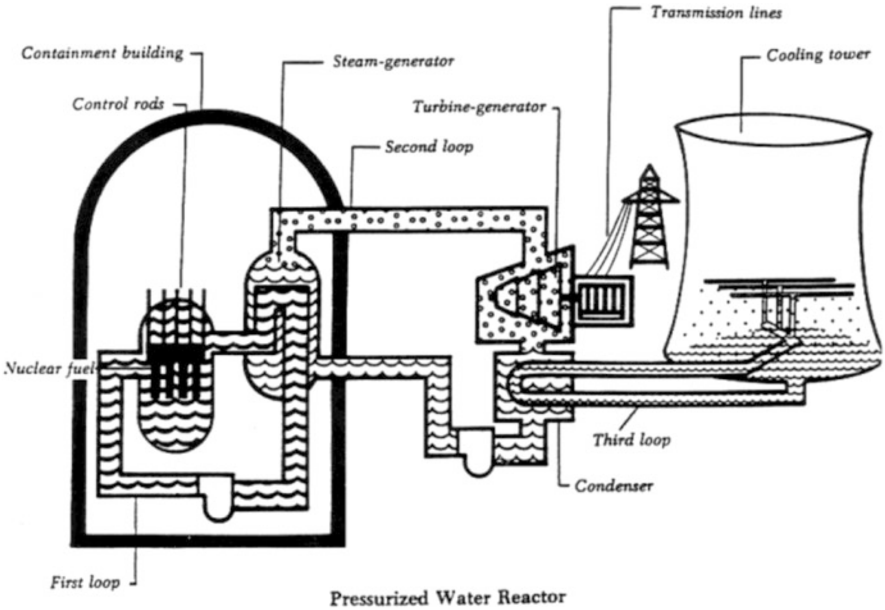
Although the concept of utilizing nuclear fission for energy generation is well-known, how the concept is actually implemented can vary depending upon the specifics of the design. For example, if the fission is sustained in the reactor by fast neutrons (i.e., the neutrons produced from fission without going through slowing down), the reactor is called fast reactor. If the fission is sustained by thermal neutrons (by having fast neutrons go through slowing down), the reactor is called thermal reactor. Most of the commercially operating nuclear reactors in the world are thermal reactors. This is because the cross section for nuclear fission reaction is much larger for thermal neutrons than for the fast or epithermal neutrons. The process of neutron slowing down to increase the probability of fission is called moderation. The material used for moderation is called a moderator. In most reactors, the material used for moderation is also used as coolant to extract thermal energy.

Depending upon the coolant material used, nuclear reactor can also be classified. If light water is used as coolant, the reactor is called light water reactor (LWR). LWR includes a pressurized water reactor (PWR) and a boiling water reactor (BWR). If the coolant is heavy water (D_2O), the reactor is called a heavy water reactor (HWR). This HWR is also called CANDU ($\underline{\text{Can}}$adian $\underline{\text{D}}$e$\underline{\text{u}}$terium) reactor developed by Canada. If the coolant is liquid metal, it is called a liquid metal cooled reactor (LMR). All of these reactors use the coolant also as moderator. Sometimes, the coolant material and the moderator material can be different. This is the case with gas cooled reactor (GCR), a reactor cooled by gas (helium or CO_2) but moderated by graphite. In Magnox reactor or advanced gas-cooled reactor (AGR), the CO_2 gas and graphite are used as coolant and moderator, respectively. There is also a light water cooled, graphite moderated reactor (LWGR) such as the RBMK reactor.

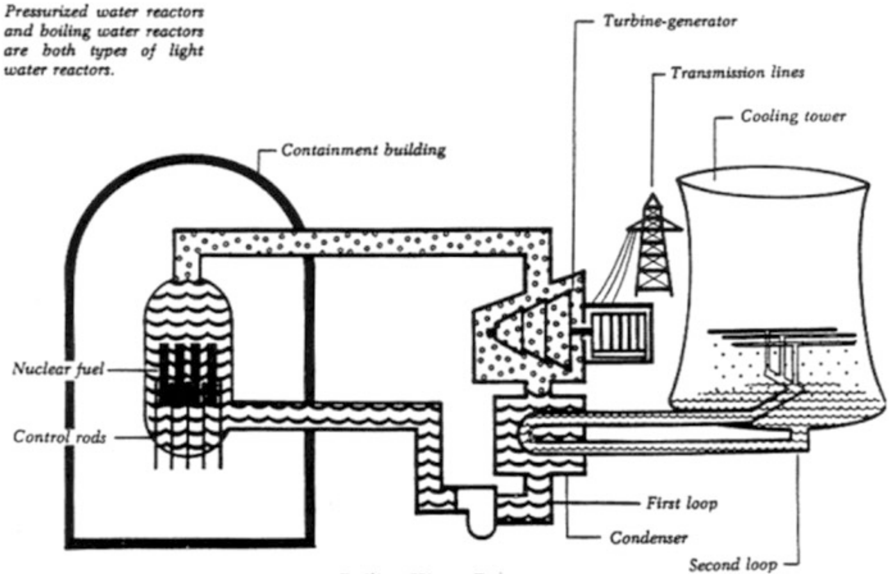
Among 450 commercially operating nuclear reactors in the world (as of 2016), 291 are PWRs, 78 are BWRs, 49 are HWR. The remainders are 14 GCRs, 3 LMR, and 15 LWGR. Therefore, more than 80 percent of commercially operating nuclear reactors in the world are LWRs, as either PWRs or BWRs. Also LMRs are employed for the fast reactor applications while all other commercially operating nuclear reactors are for thermal reactor applications. The fast reactors using liquid metal coolant for the purpose of breeding fissile materials is called liquid metal fast breeder reactor (LMFBR). The concept of breeding fissile is explained in the next section. There is also the gas (helium)-cooled fast reactor currently under development (see subsection 15.1.1.3).

Figure 3.19 illustrates energy generation from both a pressurized water reactor (PWR) and a boiling water reactor (BWR). While light water (H_2O) is used as coolant in both PWR and BWR, water in a PWR is kept under very high pressure to prevent boiling but in a BWR water is allowed to boil to produce steam directly within the reactor. In PWR the system pressure used to prevent water boiling is ~150 atm (~2000 psi). The system pressure for BWR is about 72 atm.

In a PWR, a separate system to produce steam is employed apart from the cooling water circulation system. The cooling water circulation system, called the primary loop, is where the heat from nuclear fission is extracted by water and routed to a steam generator. The secondary loop refers to a separate water supply system for the purpose of steam production. The steam generator provides the connection between the primary and secondary loop. In the steam generator, the heat in the primary loop coolant is transferred to the water in the secondary loop to produce steam. The high pressure steam drives the turbines through expansion and is collected and condensed and recirculated in the second loop.



Pressurized Water Reactor



Boiling Water Reactor

Fig. 3.19 Schematic diagrams of PWR (Left) and BWR (Right) (source: U.S. NRC Library)

In a BWR, the steam produced from the stored thermal energy in the coolant from nuclear fission directly goes through turbine (i.e., there is no secondary loop) to drive electric generators and is then condensed back to water to be recirculated.

Table 3.12 shows the summary characteristics of different reactor types.

Table 3.12 Types of reactors and a summary of key characteristics of some advanced reactor designs

	PWR	BWR	HWR	GCR	LMFBR	SMR
Acronym	AP-1000	ABWR	ACR	GTHTR300C	BN-1200	SMART
Thermal power (MW_{th})	3400	3926	3200	600	2800	300
Neutron	Thermal	Thermal	Thermal	Thermal/fast	Fast	Fast
Coolant	H_2O	H_2O	H_2O	Helium	Liquid Na	H_2O
Moderator	H_2O	H_2O	D_2O	Graphite	None	H_2O
Fuel (U enrichment %)	UO_2 (3–5%)	UO_2 (3–5%)	UO_2 (Natural U)	UO_2 and MOX	Nitride or MOX	UO_2 (4.8%)
Primary system Pressure (MPa)	15.513	7.07	11.6	7.0	0.54	15
Core power density (MW/m^3)	109.7	49.2	18	5.4	–	62.62
Coolant flow (kg/s)	14,300	14,502	13,100	439	–	2090
Coolant temperature (°C)	279.4–324.7	278–288	275–319	587–850	410–550	295.7–323
Thermal efficiency	32	34.4	36.5	47	40.7	30.3

Data source: IAEA Advanced Reactors Information System

PWR: *Pressurized Water Reactor (**AP-1000**: Westinghouse Advanced Passive PWR)*

BWR: *Boiling Water Reactor (**ABWR**: General electric-Hitachi Advanced Boiling Water Reactor)*

HWR: *Heavy Water Reactor, (**ACR-1000**: Atomic Energy of Canada Limited's (AECL) Advanced CANDU Reactor)*

GCR: *Gas Cooled Reactor,* (**GTHTR300C**: *Japan Atomic Energy Agency (JAEA) Gas Turbine High Temperature Reactor)*

LMFBR: *Liquid Metal Fast Breeder Reactor, (**BN-1200**: JSC "Afrikantov OKBM" Russian Federation)*

SMR: Small Modular Reactor, (***SMART***: *Korean System integrated Modular Advanced Reactor)*

MOX: mixed oxide

3.3.2 Fuel for Nuclear Reactors

Nuclear fission takes place, as long as a sufficient amount of excitation energy exists in the atomic nucleus. Nuclear reactor is based on using neutrons as projectile to add such excitation energy to the atomic nucleus to induce fission. However, only a few nuclides can be utilized for such purpose, and such nuclides (called fissile, in the case of thermal neutron induced fission) are used as fuel for nuclear reactor.

Figure 3.20 illustrates the point that the neutron-induced fission reaction occurs in a nuclide if the reaction can overcome the energy barrier inherently present in the nucleus. The energy level, E_0, at $x = 0$ is the energy level of an original nucleus at the ground state. For this nucleus to go through fission, the energy level of the nucleus must be lifted (through excitation) above the level represented as $E_{threshold}$. This $E_{threshold}$ is the threshold energy, i.e., the energy barrier that prevents fission reaction from happening in most nuclides. This threshold energy depends on the composition

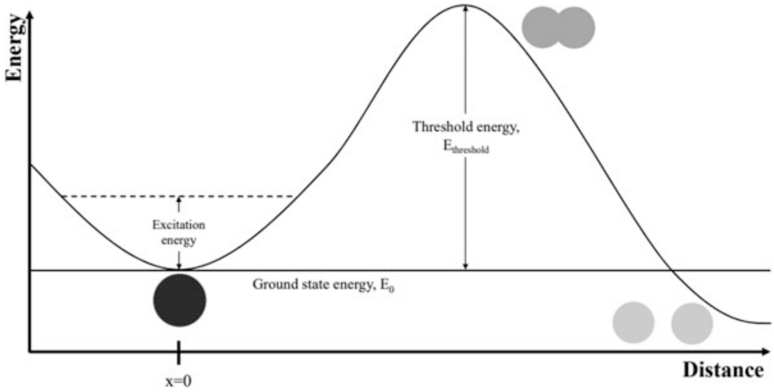


Fig. 3.20 Threshold energy for fission

and structure of the nucleus. If addition of neutron can provide the necessary excitation energy to overcome the threshold energy, nuclear fission occurs.

The energy added by a neutron to a nucleus depends on the nuclides involved and can be determined by examining the differences in the total mass of the reactants and products. For example, let's consider the following reaction of ^{235}U.

$$^{235}_{92}U+^{1}_{0}n \rightarrow ^{236}_{92}U^{*}$$

In this case, the differences in the total mass of the reactants and products is,

$$\begin{aligned}\Delta m &= [mass\ of\ reactants] - [mass\ of\ products] \\ &= [235.0439 + 1.008665] - 236.0457 = 0.006865\ amu\end{aligned}$$

The energy corresponding to this mass difference is = 0.006865 (amu) × 931.5 (MeV/amu) = 6.4 MeV. This is the energy given to the nucleus of ^{235}U as the excitation energy when a (zero kinetic energy) neutron is added to the nucleus. Thus adding a neutron to ^{235}U will provide 6.4 MeV of excitation energy. The threshold energy (E_{critical}) of ^{235}U for fission is 5.3 MeV. As the energy gained by adding zero-kinetic energy neutron is larger than the threshold energy for fission, adding a neutron to ^{235}U automatically lead to fission. This calculation can also be made for other nuclides. The results are shown in Table 3.13.

In the case of ^{238}U and ^{232}Th, the excitation energy gained by adding a zero-kinetic energy neutron to the nucleus is not enough to overcome the threshold energy. In this case, providing an additional energy is needed to overcome the energy threshold barrier. For example, the ^{238}U nuclides can go through fission if additional energy, such as 1 MeV, is given. This additional energy can be provided by hitting the ^{238}U nucleus with a neutron with high kinetic energy, i.e., a fast neutron. These nuclides that require high energy neutrons for fission to take place are

Table 3.13 Comparison of threshold energy and excitation energy through neutron addition for heavy radionuclides

Reaction	Threshold energy (MeV)	Excitation energy (MeV) gained by adding a neutron to the nucleus
${}^{233}_{92}U + {}^{1}_{0}n \rightarrow {}^{234}_{92}U$	4.6	6.6
${}^{235}_{92}U + {}^{1}_{0}n \rightarrow {}^{236}_{92}U$	5.3	6.4
${}^{238}_{92}U + {}^{1}_{0}n \rightarrow {}^{239}_{92}U$	5.5	4.9
${}^{232}_{90}Th + {}^{1}_{0}n \rightarrow {}^{233}_{90}Th$	6.5	5.1
${}^{239}_{94}Pu + {}^{1}_{0}n \rightarrow {}^{240}_{94}Pu$	4.0	6.4

called “fissionable”. These fissionable nuclides are utilized as nuclear fuel in fast reactors (using fast neutrons).

As seen in the table, with ^{233}U, ^{235}U, and ^{239}Pu, the excitation energy gained by adding a neutron to the nucleus of the respective nuclides is greater than the threshold energy. These nuclides are called fissile. A fissile is a nuclide for which fission is possible with the addition (i.e., reaction) of neutrons with zero kinetic energy or any energy including a thermal neutron.

The fissile, ^{235}U, ^{233}U and ^{239}Pu are good nuclear fuel materials for thermal reactors. In a fast reactor, the fissionable, ^{232}Th or ^{238}U, can also be used as fuel since the neutrons with high kinetic energy are available in a fast reactor.

^{238}U and ^{232}Th can also be changed into a fissile through neutron capture eventually becoming ^{239}Pu and ^{233}U, respectively, through the reactions shown below

$$ {}^{238}_{92}U + {}^{1}_{0}n \rightarrow {}^{239}_{92}U \rightarrow {}^{239}_{93}Np + {}^{0}_{-1}e \rightarrow {}^{239}_{94}Pu + {}^{0}_{-1}e $$

$$ {}^{232}_{90}Th + {}^{1}_{0}n \rightarrow {}^{233}_{90}Th \rightarrow {}^{233}_{91}Pa + {}^{0}_{-1}e \rightarrow {}^{233}_{92}U + {}^{0}_{-1}e $$

These nuclides are also called “fertile”.

Nuclear fission can also occur spontaneously (as discussed in Sect. 3.1.3) with heavy elements such as uranium and plutonium. Occurrence of spontaneous fission is due to quantum tunneling. Quantum tunneling refers to a particle penetrating through a potential energy barrier that is higher than the kinetic energy of the particle through quantum mechanical effect. Through quantum tunneling, the heavy nuclide penetrates the potential energy barrier and splits the nucleus.

3.3.3 The Process of Fission in Thermal Nuclear Reactors

Once neutrons are produced by fission, the neutrons collide with other nuclides in the system. These collisions result in various nuclear reactions such as capture, fission, scattering, or an outcome of neutrons moving out of the system (i.e., leaked out). The probability of each outcome depends on the energy of the neutron and the composition and structure of the system.

In a thermal reactor, as neutrons initially produced from fission are fast, they are slowed down by collision with atomic nuclei of the moderator to reach down to the region of thermal energy (~0.025 eV). Most fission takes place in the thermal energy region. Therefore, we can think of a neutron cycle starting from a fast neutron produced by fission, slowed down to thermal energy and used to produce fission creating the next generation of fast neutrons.

Assume that a neutron life cycle in a thermal reactor begins with a total of N_0 fast neutrons, as the initial number. The number slightly increases as fast fission takes place due to the presence of both ^{235}U and ^{238}U. This increase is represented by a factor ε, called the fast-fission factor. The system now has $N_0\varepsilon$ fast neutrons. Some of these fast neutrons can leak out of the system if the direction of their movements is toward outside the system and not back-scattered. This reduction by leakage in the fast neutron energy region is called the fast-neutron leakage probability, represented by P_{FL}. Then the non-leakage probability of fast-neutron, P_{FNL}, is $1\text{-}P_{FL}$.

The remaining neutrons, $N_0\varepsilon$ $(1\text{-}P_{FL})$, may be absorbed (i.e., captured) in non-fission reactions before reaching the thermal energy region. Most of this non-fission absorption takes place in the so-called "resonance" region. This resonance region is where neutrons have energies between 1 keV and 1 eV. In this region, the neutron capture cross-section is very high, reaching peaks more than 100 times the base value. This resonance happens when the kinetic energy of a neutron approaching the target nucleus matches the energy level of one of the excitation states of the compound nucleus. In this case, the incident neutron has a much higher chance of being captured by the target nucleus, creating a peak in the absorption cross section.

Therefore, the probability of neutron capture significantly exceeds the probability of fission in the resonance region. The probability of neutrons surviving the resonance region without being captured is called the resonance escape probability, p. So the number of neutrons survived the resonance region becomes, $N_0\varepsilon(1\text{-}P_{FL})p$ and enters the thermal energy region (1 eV – 0.025 eV).

The thermal neutrons can still escape from the system through leakage. This is termed thermal leakage and its probability is represented by P_{TL}. Thus the thermal non-leakage probability, P_{TNL}, becomes $1\text{-}\ P_{TL}$. Then the number of remaining neutrons in the thermal region becomes, $N_0\varepsilon(1\text{-}P_{FL})\mathrm{p}(1\text{-}P_{TL})$.

The remaining thermal neutrons can be absorbed either in the fuel or in other materials (e.g., moderator). The fraction of the neutrons that are absorbed in the fuel is represented by f, thermal utilization factor, which is the ratio of the number of neutrons absorbed in the fuel to the total number of neutron absorption in other non-fuel part of the system. Then the number of neutrons that are absorbed in the fuel as thermal neutrons is $N_0\varepsilon(1\text{-}P_{FL})\mathrm{p}(1\text{-}P_{TL})f$. From this neutron absorption in fuel, new neutrons will be generated by fission. The number of fission neutrons produced per neutron absorbed in the fuel is called the thermal fission factor, η . Then the number of fission neutrons starting a new generation becomes $N_0\varepsilon(1\text{-}P_{FL})\mathrm{p}(1\text{-}P_{TL})f\eta$, or $N_0\varepsilon P_{FNL}\mathrm{p}P_{TNL}f\eta$.

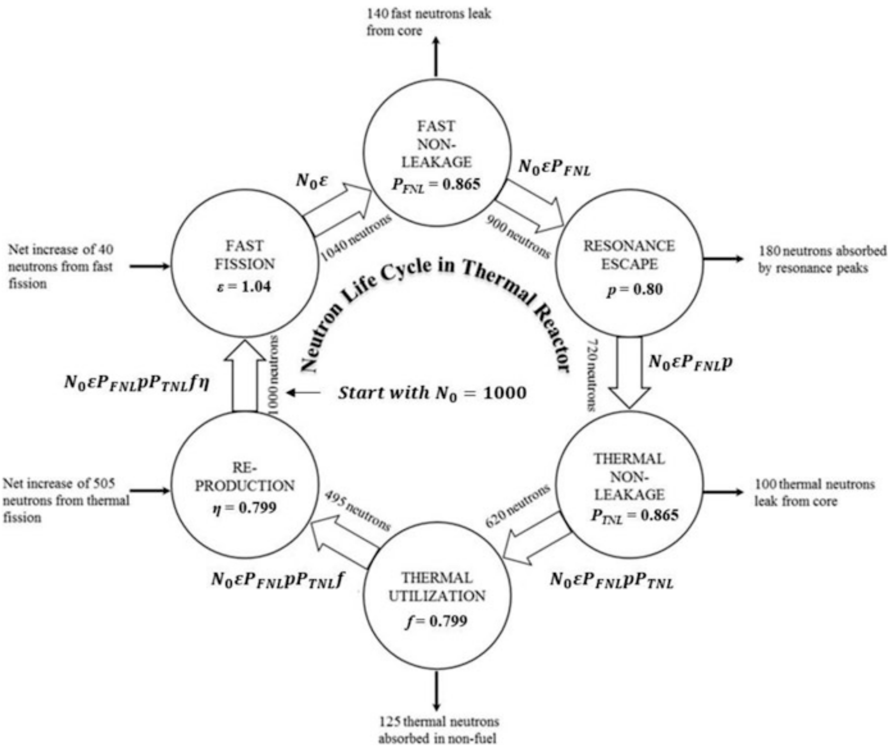


Fig. 3.21 Neutron life cycle in a thermal reactor (reproduced from source: DOE 1993)

The lifecycle of a neutron in a thermal reactor starts as a fast neutron produced by fission goes through the slowing down phase leading to producing next generation of neutrons. This is shown in Fig. 3.21.

Now, compare the number of neutrons we started with in the previous cycle with that at the beginning of a new cycle. For a nuclear reactor to maintain steady operating condition, the number at the beginning of a new cycle should be equal to the number starting the previous cycle. This condition is called “critical”. The condition of criticality is maintained by keeping the ratio of the number of the neutrons at the beginning of a new cycle to that starting the previous cycle to be exactly one. Such ratio is defined as the effective multiplication factor, k_{eff}, and is represented as:

$$k_{eff} = \frac{\mathrm{N}_0 \varepsilon P_{FNL} f p P_{TNL} \eta}{\mathrm{N}_0} = \varepsilon P_{FNL} f p P_{TNL} \eta \tag{3.43}$$

The effective multiplication factor, k_{eff}, is represented by the combination of six factors which is called, the six factor formula. Table 3.14 shows the summary of each of the parameters of six factor formula. When k_{eff} is equal to 1, the system is critical. If $k_{eff} < 1$, the system is subcritical and if $k_{eff} > 1$, the system is supercritical. Under

Table 3.14 The parameters of six-factor formula

Symbol	Name	Meaning
p	The resonance escape probability	Fraction of fission neutrons that manage to slow down from fission to thermal energies without being absorbed.
ε	The fast fission factor (Epsilon)	$\frac{\text{total\#of fission neutrons}}{\text{total\#of fission neutrons from just thermal fissions}}$
P_{FNL}	The fast non-leakage probability	The probability that a fast neutron will not leak out of the system.
p	The resonance escape probability	Fraction of fission neutrons that manage to slow down from fission to thermal energies without being absorbed.
P_{TNL}	The thermal non-leakage probability	The probability that a thermal neutron will not leak out of the system.
f	The thermal utilization factor	Probability that a neutron that gets absorbed in the thermal region does so in the fuel material.
η	Reproduction Factor (Eta)	The number of fission neutrons produced per absorption in the fuel.

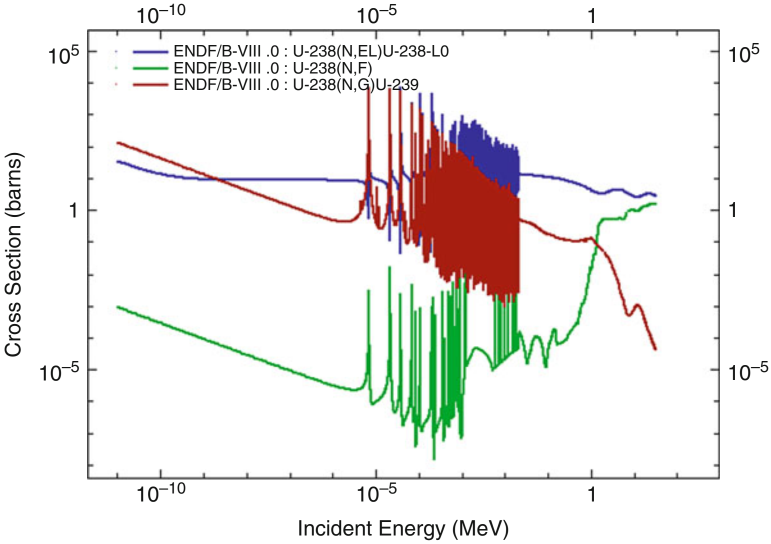


Fig. 3.22 ^{238}U cross-section at different energies of incident neutron (data source: Evaluated Nuclear Data File (ENDF))

subcritical condition, the fission chain reaction will not be sustained in the system and a nuclear reactor will die out. Under supercritical condition, the neutron production will continue to increase producing more power which in turn increases neutron production and power production. So a steady level of power population cannot be sustained but power excursion continues.

For the above described neutron interactions, the corresponding cross sections vary as a function of energy. For example, as shown in Fig. 3.22 for ^{238}U, the cross

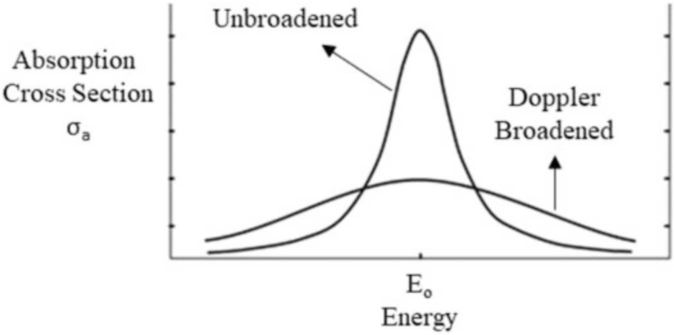


Fig. 3.23 The concept of Doppler broadening

section for neutron absorption in the fuel is very small for fast neutrons and becomes larger as the energy decreases to a thermal region. The figure also shows that in the intermediate energy region, the cross section varies rapidly as the energy of the neutron changes. This is the region of resonance absorption as discussed earlier. Beyond the resonance region, the absorption cross section of ^{238}U increases significantly toward the lower thermal region. In particular, this thermal neutron energy region is called the 1/v region as the absorption cross section of neutron increases with the reciprocal of the velocity (kinetic energy) of the neutron.

One interesting phenomenon to note in the resonance absorption region is Doppler broadening. Doppler broadening, as mentioned at the beginning of this section, refers to the broadening of the cross section peak with the increase in the temperature of the fuel (see Fig. 3.23). This broadening is due to the Doppler Effect with fuel temperature increase. The Doppler Effect takes place as neutron cross section depends on the relative velocity between neutron and nucleus. As mentioned, neutron absorption in the resonance region can happen with sharp peaks when the energy of neutron matches the energy levels of the excitation states of the compound nucleus. With fuel temperature increase, nuclei in the fuel have increased thermal motion. This changes the relative energy between the target nuclei and the neutrons resulting in the absorption of neutrons over a wider distribution of energies around specific excitation energy levels. This effectively increases absorption of neutrons in this resonance region. This increase in cross section with fuel temperature increase also happens with fission. However, as the number of fissile (^{235}U) nuclei in the fuel is small (compared to ^{238}U), the overall effect of Doppler broadening is negative feedback, i.e., increase in fuel temperature results in increase in neutron absorption in ^{238}U, i.e., reduction in the number of neutrons available in the thermal region for fission.

This Doppler broadening plays an important role for nuclear safety as the increase in fuel temperature automatically leads to reduction in power generation. This Doppler broadening also prevents nuclear reactors from behaving like nuclear weapons (where no mechanisms of negative feedback exists).

3.3.4 Products of Nuclear Fission

In an experiment performed in 1934, Fermi and his co-workers bombarded uranium with neutrons as an attempt to produce elements of atomic number greater than 92. They found the resulting products having four beta-ray activities with different half-lives. They incorrectly interpreted the outcome as the formation of one or more transuranic elements. In reality, what they observed was the formation of smaller products of fission. In 1939 after a series of very careful chemical experiments, Hahn and Strassmann found that one of the elements formed by bombarding uranium with neutrons was an isotope of the element barium, Z = 56. Another radioactive element found was lanthanum, Z = 57, which is the beta decay product of barium. Hahn and Strassmann suggested that the beta-ray activities observed in Fermi and his co-workers' experiments were probably from radioactive isotopes of elements of lower atomic number. They explained the formation of such lower atomic number nuclides as the result of the splitting of uranium nucleus: After being bombarded by neutrons, uranium nucleus split into two nuclei of medium atomic masses. This finding marks the discovery of nuclear fission. What Hahn and Strassmann observed in their experiment was the production of barium and krypton as fission fragments. The fission fragments were excessively neutron rich and thus went through β^- decays converting neutrons to protons as observed in the experiments.

As shown in Hahn and Strassmann's experiments, the split of uranium nucleus typically produces the fragments of substantially different masses. In fact, plotting the masses of fission fragments produces "double-humped" curve as shown Fig. 3.24 (for ^{235}U fission). This asymmetric distribution is the characteristic of the masses of

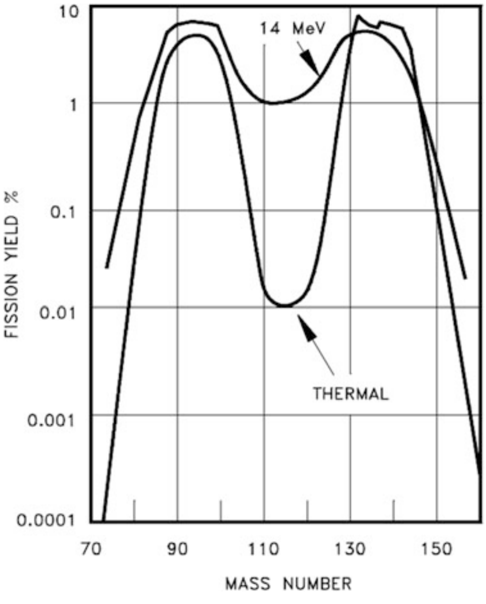


Fig. 3.24 Fission product yields of thermal and 14 MeV fission neutrons in ^{235}U (from Ram 1977)

Table 3.15 Cumulative yields of fission products of U-235 for thermal and fast neutrons (data source: IAEA, Fission Yield Data: Cumulative Fission Yield)

Nuclide	Thermal yield	Fast yield	Nuclide	Thermal yield	Fast yield
$^{1}H_{1}$	0.00171	0.00269	$^{54}Xe_{130}$	0.000038	0.000152
$^{1}H_{2}$	0.00084	0.00082	$^{54}Xe_{131m}$	0.0313	0.0365
$^{1}H_{3}$	0.0108	0.0108	$^{54}Xe_{133}$	6.6	6.61
$^{2}He_{3}$	0.0108	0.0108	$^{54}Xe_{133m}$	0.189	0.19
$^{2}He_{4}$	0.1702	0.17	$^{54}Xe_{135}$	6.61	6.32
$^{35}Br_{85}$	1.304	1.309	$^{54}Xe_{135m}$	1.22	1.23
$^{36}Kr_{82}$	0.000285	0.00044	$^{55}Cs_{134}$	0.0000121	0.0000279
$^{36}Kr_{85}$	0.286	0.286	$^{55}Cs_{137}$	6.221	5.889
$^{36}Kr_{85m}$	1.303	1.307	$^{56}Ba_{140}$	6.314	5.959
$^{38}Sr_{90}$	5.73	5.22	$^{57}La_{140}$	6.315	5.96
$^{40}Zr_{95}$	6.502	6.349	$^{58}Ce_{141}$	5.86	5.795
$^{41}Nb_{94}$	4.2×10^{-7}	2.90×10^{-8}	$^{58}Ce_{144}$	5.474	5.094
$^{41}Nb_{95}$	6.498	6.345	$^{59}Pr_{144}$	5.474	5.094
$^{41}Nb_{95m}$	0.0702	0.0686	$^{60}Nd_{142}$	6.30×10^{-9}	1.70×10^{-9}
$^{42}Mo_{94}$	8.70×10^{-10}	0	$^{60}Nd_{144}$	5.475	5.094
$^{42}Mo_{96}$	0.00042	0.000069	$^{60}Nd_{147}$	2.232	2.148
$^{42}Mo_{99}$	6.132	5.8	$^{61}Pm_{147}$	2.232	2.148
$^{43}Tc_{99}$	6.132	5.8	$^{61}Pm_{148}$	5.00×10^{-8}	7.40×10^{-9}
$^{44}Ru_{103}$	3.103	3.248	$^{61}Pm_{148m}$	1.04×10^{-7}	1.78×10^{-8}
$^{44}Ru_{106}$	0.41	0.469	$^{61}Pm_{149}$	1.053	1.064
$^{45}Rh_{106}$	0.41	0.469	$^{61}Pm_{151}$	0.4204	0.431
$^{50}Sn_{121m}$	0.00106	0.0039	$^{62}Sm_{148}$	1.49×10^{-7}	2.43×10^{-8}
$^{51}Sb_{122}$	3.66×10^{-7}	0.0000004	$^{62}Sm_{150}$	0.000061	0.0000201
$^{51}Sb_{124}$	0.000089	0.000112	$^{62}Sm_{151}$	0.4204	0.431
$^{51}Sb_{125}$	0.026	0.067	$^{62}Sm_{153}$	0.1477	0.1512
$^{52}Te_{132}$	4.276	4.639	$^{63}Eu_{151}$	0.4204	0.431
$^{53}I_{129}$	0.706	1.03	$^{63}Eu_{152}$	$3.24x\ 10^{-10}$	0
$^{53}I_{131}$	2.878	3.365	$^{63}Eu_{154}$	$1.95\ x\ 10^{-7}$	$4.00\ x\ 10^{-8}$
$^{53}I_{133}$	6.59	6.61	$^{63}Eu_{155}$	0.0308	0.044
$^{53}I_{135}$	6.39	6.01			

fission fragments. Typically these fission fragments are called fission products. The percent of the fission products produced with a given mass number is denoted as fission product yield. The fission product yields of various fission products from ^{235}U as a function of atomic mass number are shown in Fig. 3.24 and Table 3.15 for both thermal neutrons and fast (14 MeV) neutrons. As the figure is on the logarithmic scale, the fission-product yield distribution is severely asymmetrically distributed.

The most prevalent range of atomic mass of fission products is roughly in 90–100 and 135–145. There exists a very small probability (about 0.01%) of producing equal-mass fragment at $A \approx 117$. The fission also sometimes produces three fragments, which is called ternary fission. Ternary fission releases tritium, ^{3}H, as one of the fission fragments. The probability of ternary fission is about 1 out of 400.

3.3.5 Nuclear Criticality Control

In general, *criticality* refers to the condition of neutron balance maintaining the status of self-sustaining chain reaction of neutrons in a system of nuclear materials. Therefore, under the condition of criticality, the number of neutrons produced in a system is equal to the number of neutrons lost in the system. When a nuclear reactor is operating normally, it is in a state of *"criticality."* Thus the number of neutrons produced by fission in the fuel matches exactly the number of neutrons leaked out of the system or absorbed in the system while going through neutron life cycle.

In activities handling fissile materials (e.g., spent fuel management), nuclear criticality control is strictly imposed to maintain the effective multiplication factor, k_{eff}, to be always below 1.0. In fact, for a margin of safety, the effective multiplication factor is maintained below 0.95 under the normal operating conditions. Under an abnormal or accident condition, the effective multiplication factor should be kept at less than 0.99.

Nuclear criticality control is achieved by controlling the factors that influence the neutron balance in the system. The system could be a nuclear reactor, a spent fuel storage pool, transportation casks, or a nuclear waste disposal facility. These factors are the ones directly or indirectly control the parameters in the six factor formula. Therefore, considerations include those affect the absorption, fission, leakage, or back-scattering of neutrons in the system. Other considerations also include the changes in mass or concentration of fissile or fertile, geometry and/or size of the neutron containing system, type and concentration of moderators, nature and thickness of neutron reflectors surrounding fissile material, and nature and concentration of neutron-absorbing poisons.

The shape or geometry of the system containing fissile material affects the leakage of neutrons from the system through the differences in the surface area. A shape with a large surface area for the same volume of material is the high leakage geometry such as long, small-diameter columns or thin flat slab (in comparison to a more compact shape such as cylinder or sphere). When geometric control is not feasible, control of fissile mass, enrichment, moderation, reflection, and neutron poisons is employed. Changing the density or concentration of fissile or neutron poisons in a given volume will also change the probability of fission. Use of effective neutron moderation scheme using materials like water, heavy water, and graphite, increases the probability of fission. Use of back-scattering of neutrons escaping the system through the implementation of neutron reflectors will decrease leakage and increase the probability of fission. As reflectors, materials such as hydrogen, beryllium, carbon, lead, uranium, water, polyethylene, concrete, tungsten carbide and steel can be used. Degree of interactions between two or more regions containing fissile material also affects criticality through collaborative utilization of neutrons leaking from the neighboring units. In this case, the distance between the units or use any material between them can control the degree of neutron utilization. Presence of neutron poisons as absorbers is effectively utilized in nuclear criticality control. The commonly used neutron poisons include boron, cadmium, and gadolinium. Specifics of criticality control in spent fuel management and reprocessing are discussed in Sects. 7.3 and 8.2.5.

3.4 Conclusion

Generating electricity using nuclear power requires operation of nuclear reactors. At the same time, operation of nuclear reactors produces nuclear waste. As discussed in the chapter, the materials used for nuclear fission, the necessary provisions to sustain nuclear fission reactions, and the amount of energy extracted from nuclear fuels provide the basis of describing the characteristics of nuclear waste from nuclear reactors. Through radioactive decays of unstable nuclei, nuclear waste releases ionizing radioactive particles and these particles go through various interactions with matter and transfer their energy to the host medium. The amount of energy absorbed in the host medium becomes the basis of describing biological effects of radiation. Roughly over the twentieth century through the advancements of modern physics and chemistry, understanding these processes, interactions, and the related characteristics of radioactive particles has been pursued. Today these understandings help to devise ways to manage nuclear waste in a safe manner.

Homework

Problem 3.1 How many Curies and Becquerels are there in 1 gm of ^{3}H, ^{14}C, ^{60}Co, ^{99}Tc, ^{129}I, ^{137}Cs, ^{226}Ra, ^{238}U, ^{239}Pu? This amount is called specific activity. Compare the specific activity of these radionuclides and explain the differences observed.

Problem 3.2 The total volume of uranium mill tailings waste accumulated up to 1991 in the U.S. is 118,700,000 m^{3}. Estimate the total amount of radioactivity within this volume of waste. Make appropriate assumptions as needed.

Problem 3.3 A 3.0-MeV photon enters a volume V of a homogeneous target material producing a 1.5 MeV Compton electron at point A. The electron emits a 0.8-MeV bremsstrahlung photon at B before coming to rest at C within the target. The bremsstrahlung photon escapes from the target material without interaction. The photon scattered at A is scatted again at D, producing one 1.0-MeV Compton electron, which escapes the target material after losing 0.4-MeV in the target. The scattered photon at D escapes the target. Assuming the mass of the target is 10 g, determine the kerma and the absorbed dose in V in Gy.

Problem 3.4 For the problem given Example 3.4, (a) determine the linear energy transfer coefficient in V, if the applied photon fluence is 10^9 cm^{-2}, and (b) determine the cross section of the atom in the homogeneous material for the pair production reaction.

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Chapter 4
Basic Chemical Science for Nuclear Waste Management

Abstract Chemistry controls stability and durability of materials in nuclear waste management as well as the fate and transport of radioactive materials in the environment upon their release from nuclear waste. This chapter describes the basics of chemical science such as role of electrons in chemical reactions, types of chemical bonds, various chemical reactions of importance, and the role of parameters such as free energy, pH, and oxidation potential in chemical reactions.

Keywords Electrons · Thermodynamics · Free energy · Equilibrium constant · Chemical reactions

Understanding how materials interact with natural forces provides the basis of using technologies for the primary goal of nuclear waste management, i.e., isolation and containment of nuclear waste. These interactions are either physical or chemical. While physical interactions do not *per se* change the properties of materials, chemical interactions can drastically change the properties of materials. As the product of chemical reactions, chemical changes affect stability and durability of materials in nuclear waste management. If radioactive materials are released from the system of nuclear waste management, their eventual fate will also largely be dependent on chemical reactions in the surrounding environment (e.g., in water, air, soil, or other material). This chapter describes the basics of chemical science for such understanding.

4.1 Chemical Properties

4.1.1 Electron Energy Levels

In an atom, electrons rotate the nucleus in orbits with specific energy levels. Thus each electron possesses a particular energy. Also, according to the so-called Pauli Exclusion Principle, only a specified number of electrons have the same energy in an

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https://doi.org/10.1007/978-94-024-2106-4_4

Table 4.1 The energy levels and the number of assigned electrons in each energy level as patterns in atoms

Shell	Orbital, *s* (l = 0)	Orbital, *p* (l = 1)	Orbital, *d* (l = 2)	Orbital, *f* (l = 3)	Orbital, *g* (l = 4)	Orbital, *h* (l = 5)
K (n = 1)	2					
L (n = 2)	2	6				
M (n = 3)	2	6	10			
N (n = 4)	2	6	10	14		
O (n = 5)	2	6	10	14	18	
P (n = 6)	2	6	10	14	18	22

atom. Therefore, electrons of an atom are grouped in different energy levels surrounding the nucleus of the atom. As the number of electrons in an atom increases, electrons are added to different energy levels with discrete differences in energy.

The energy levels to which each electron belongs are determined by four sets of quantum numbers. These quantum numbers together describe the unique quantum state of an electron. The first quantum number n, called the principal quantum number, determines the quantum shell to which the electron belongs, indicating the energy level or the distance from the nucleus of the electrons. These quantum shells are also assigned a letter. The shell for $n = 1$ is designated K, for $n = 2$ is L, for $n = 3$ is M, for $n = 4$ is N, and so on. The K shell represents the inner most shell of electrons near the nucleus. As n increases, the electrons are farther away from the nucleus and at a higher potential energy and are less tightly bound to the nucleus.

The number of energy levels within each quantum shell is determined by the angular momentum quantum number l and the magnetic quantum number m. The angular momentum quantum number l determines the total number of energy sub-levels in a given energy level and varies from 0 to $n - 1$, being designated as s ($l = 0$), p ($l = 1$), d ($l = 2$), f ($l = 3$) orbitals, and so on. Here, orbital refers to the region in space where the electron is likely to be found the most around the nucleus (thus represents the probability picture of an electron).

The magnetic quantum number m determines the number of the orbitals available and their orientation within the energy sub-levels. The value of m_l can range from $-l$ to + l, including zero. The fourth quantum number, s, is called the spin quantum number corresponding to the direction of electron spin, i.e., positive (s = +1/2) or negative (s = −1/2). The number of electrons that can be accommodated at each energy level in a given orbital is determined by so-called Pauli Exclusion Principle. That is up to 2 for s orbitals, 6 for p orbitals, 10 for d orbitals, and 14 for f orbitals. The energy levels and the number of electrons assigned to each energy level of atoms are shown in Table 4.1.

The shorthand notation to denote the electronic structure of an atom uses the numerical value of the principal quantum number (n), the lower case letter notation for the angular momentum quantum number (l), and a superscript showing the number of electrons in each orbital. For example, the electronic structure of germanium with Z of 32 is:

$1s^2\ 2s^2\ 2p^6\ 3s^2\ 3p^6\ 3d^{10}\ 4s^2\ 4p^2$. (i.e., two electrons in 1*s* orbital, two electrons in 2*s* orbital, six electrons in 2*p* orbital, etc.)

4.1.2 Types of Elements and the Periodic Table

In an atom, the chemical properties are largely determined by the few highest-energy electrons, i.e., the ones in the outermost shell: The ability of the atom to form chemical combinations with other atomic elements is often determined by the number of electrons in the *s* or *p* orbitals of the outermost shell as these electrons are involved in chemical bond formation, determining the combining power of an atom.

The number of electrons in the *s* or *p* orbitals in the outermost shell is called valence. An atom with valence of 1 or 2 is called monovalent or divalent, respectively. Valence is a measure of the combining power of an atom. i.e., chemical properties.

The valence is also the basis of the periodic table. The group 0 elements have a valence of zero. They are found on the right hand side of the periodic table and are so-called **noble gases**. Noble gases are characterized by a completed outer shell of electrons, thus are chemically stable (very unreactive as the most inert elements). The elements in this group include helium (He), neon (Ne), argon (Ar), krypton (Kr), xeon (Xe), and radon (Rn). With low boiling points (ranging from -60 °C to -270 °C), these elements exist as gas under ambient conditions.

The group I elements have only one electron in the *s* orbital as the outermost electron. These are called alkali metals (e.g., Li, Na, K, Cs). One exception is hydrogen which has one electron in the outermost shell but is a nonmetal. The outermost electron of the group I element can be rather easily removed from the atom. The elements in this group are highly reactive and tend to lose one electron and form positive ions in chemical reactions. This property is called "electropositive". These elements are monovalent. Removal of a second electron from them requires a much higher energy involving separation from the inner orbital or shell.

In the group II elements, there are two outer electrons. They tend to lose two electrons (divalent) and form double charged positive ions (electropositive) in chemical reactions. They are called alkaline earth metals (e.g., Be, Mg, Ca, Sr).

Similarly group III and IV elements have three and four outer electrons, (valences of +3 and + 4) respectively. They are less electropositive than the group I and II elements. The group V elements have two outer electrons in the *s* orbital along with three electrons in the *p* or *d* orbitals. They typically exhibit either a + 3 or + 5 valence.

The group VI and VII elements have valence of 6 and 7 and contain 4 and 5 electrons in the outer *p* orbital, respectively. For these elements, addition of one or two electrons form stable monovalent (the VII elements) or divalent (the VI elements) negative ions (e.g. F^- or O^{-2}). The reactivity of the element in these groups is caused by the desire to fill its outer energy level by accepting one (the group VII) or two (the group VI) electrons. The group VII elements are referred to as halogens (e.g., fluorine (F), chlorine (Cl), bromine (Br), and iodine (I)).

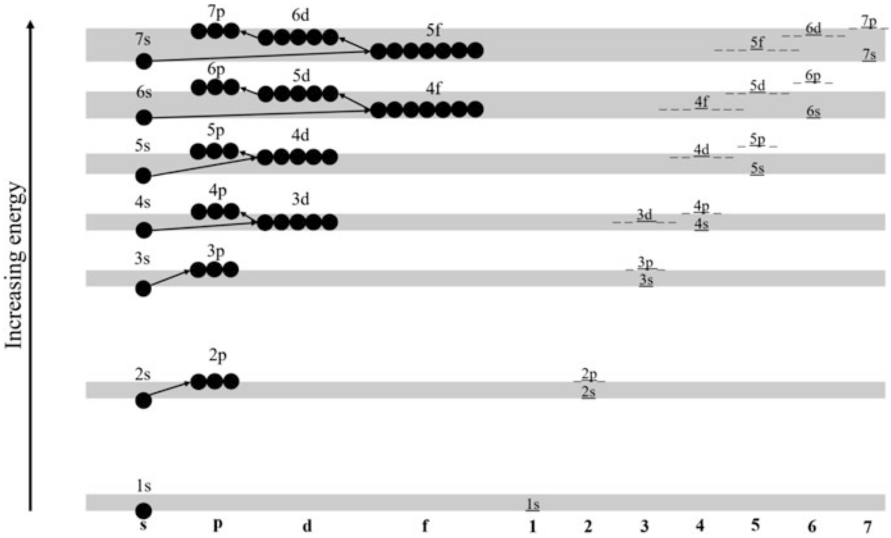


Fig. 4.1 The energy levels of electrons of an atom in association with the principal (n) and orbital (l) quantum numbers

As the atomic number becomes large, the relative stability of energy levels of different orbitals becomes similar. Then electrons fill in the orbitals in the order 1*s*, 2*s*, 2*p*, 3*s*, and 3*p*. Yet, the 4*s* orbital becomes more stable and is filled before the 3*d*. For example, we would expect the electronic structure of iron, with Z = 26, to be:

$$1s^2\ 2s^2\ 2p^6\ 3s^2\ 3p^6\ 3d^8.$$

Instead, the actual structure is:

$$(\mathrm{Fe})1s^2\ 2s^2\ 2p^6\ 3s^2\ 3p^6\ 3d^6\ 4s^2.$$

The order of orbital filling follows the order of energy increase. This order is shown in Fig. 4.1 as the electron occupancy energy levels. As the energy level of 3*d* is higher than that of 4*s*, the 4*s* orbital gets filled before 3*d*.

The elements showing this behavior of incomplete *d* shell are called the transition metals (e.g., titanium (Ti), chromium (Cr), manganese (Mn), iron (Fe), cobalt (Co), nickel (Ni), copper (Cu), and zinc (Zn)). This unfilled 3*d* level causes the magnetic properties of the transition metals. Transition metals have similar chemical properties.

With larger atomic number elements, incomplete 4*d* and 5*d* shell also occurs in other series of transition elements. As an example, technetium has the electronic structure as,

$$(\mathrm{Tc})1s^2 2s^2 2p^6 3s^2 3p^6 3d^6 4s^2 3d^{10} 4p^6 5s^2 4d^5$$

Among the bigger elements, when the inner 4*f* shell is not completely filled, a similar and even more pronounced effect of transition metals occur. Elements in this group are called rare earth elements (also called lanthanides, e.g., lanthanum (La), cerium (Ce), neodymium (Nd), samarium (Sm), and europium (Eu)). For example, the electronic structure of cerium is as follows.

$$(\mathrm{Ce})1s^2 2s^2 2p^6 3s^2 3p^6 3d^6 4s^2 3d^{10} 4p^6 5s^2 4d^{10} 5p^6 6s^2 4f^2$$

Any of the elements from atomic number 89 (actinium) to 103 (lawrencium) are called **actinides** (e.g., actium (Ac), thorium (Th), U, Pu). The actinides are extremely unstable heavier elements and thus all radioactive. Actinides are the one with the inner 5*f* shell not completely filled. Uranium, as an example, has the following electronic structure. In this case, the energy of 5*f* and 6*d* electrons are close to each other. Instead of filling 5*f*, electrons enter into the 6*d* orbital.

$$(\mathrm{U})1s^2 2s^2 2p^6 3s^2 3p^6 3d^6 4s^2 3d^{10} 4p^6 5s^2 4d^{10} 5p^6 6s^2 4f^{14} 5d^{10} 6p^6 7s^2 5f^3 6d^1$$

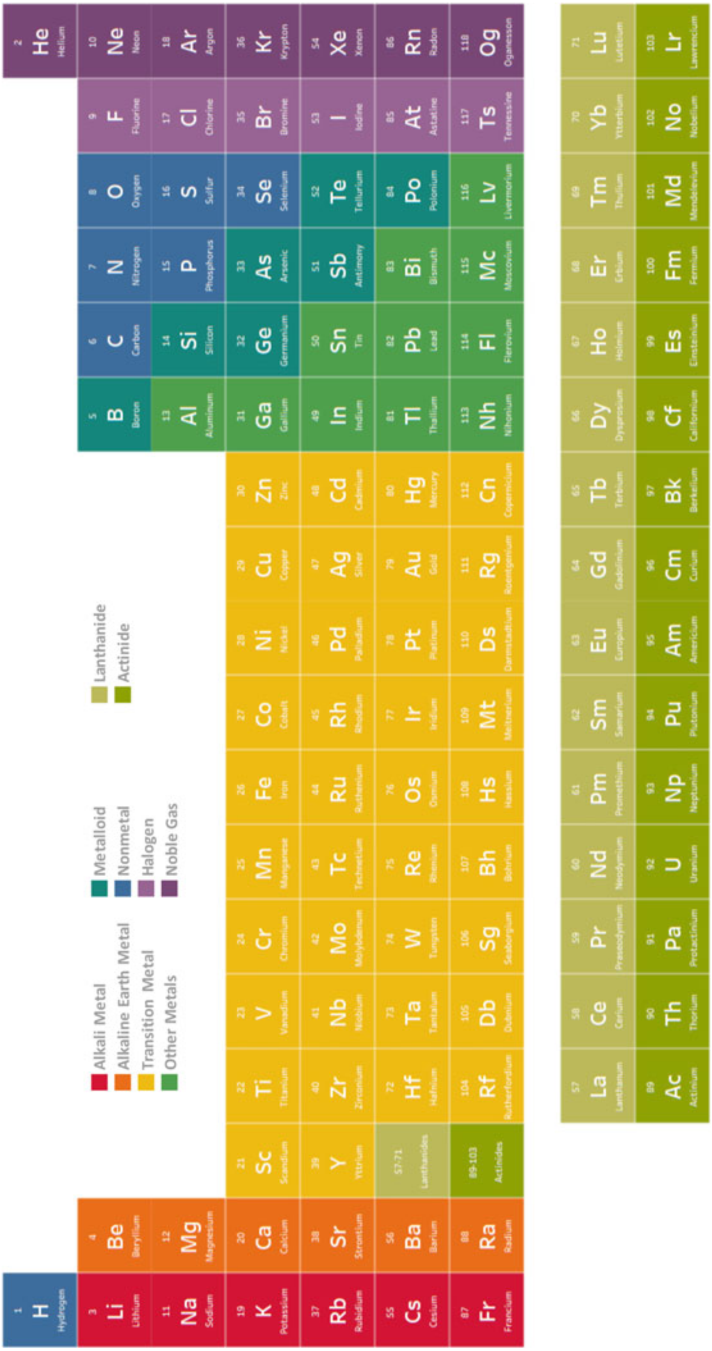
Those elements with incomplete d shell of f shell (as transition metals, rare earth elements, or actinides) have variable (multiple) valences causing chemical reaction characteristics of the elements omplex (e.g., see Sect. 4.3.4). Those elements that lack metallic attributes are simply called **non-metals**. Non-metals can exist as gas (hydrogen (H), nitrogen (N) and oxygen (O)) and solids (including carbon (C), phosphorous (P), sulfur (S), and selenium (Se)). These non-metallic solids are incapable of conducting electricity or heat.

The elements whose properties are intermediate between those of metals and non-metals are called metalloids (e.g., boron (B), silicon (Si), germanium (Ge), arsenic (As), antimony (Sb), and polonium (Po)). These metalloids are electrical semiconductors.

There are also a set of elements in the periodic table located between the transition metals (to the left) and the metalloids (to the right) in groups 13, 14, and 15. These are called **other metals** (e.g., aluminum (Al), gallium (Ga), indium (In), tin (Sn), thallium (Tl), lead (Pb), ad bismuth (Bi)). Their valence electrons are only in the outer shell with oxidation numbers of +3, ±4, or −3. These other metals are good conductors of electricity and heat.

Figure 4.2 shows the periodic table with ten named groups represented in the table in different color: alkali metals, alkali earth metals, rare earths, actinides, transition metals, other metals, metalloids, non-metals, halogens, and noble gases. As mentioned, elements with similar properties (i.e., by the number of valence electrons) are categorized into groups.

The periodic table can also be organized into four blocks according to the subshell in which the last electron resides. As shown in Fig. 4.3, the *s* block includes alkali metals and alkali earth metals (groups 1 and 2). The *p* block contains the last six groups (groups 13 to 18), the *d* block contains groups 3 to 12, and the *f* block has the



1 H Hydrogen																	2 He Helium
3 Li Lithium	4 Be Beryllium											5 B Boron	6 C Carbon	7 N Nitrogen	8 O Oxygen	9 F Fluorine	10 Ne Neon
11 Na Sodium	12 Mg Magnesium											13 Al Aluminum	14 Si Silicon	15 P Phosphorus	16 S Sulfur	17 Cl Chlorine	18 Ar Argon
19 K Potassium	20 Ca Calcium	21 Sc Scandium	22 Ti Titanium	23 V Vanadium	24 Cr Chromium	25 Mn Manganese	26 Fe Iron	27 Co Cobalt	28 Ni Nickel	29 Cu Copper	30 Zn Zinc	31 Ga Gallium	32 Ge Germanium	33 As Arsenic	34 Se Selenium	35 Br Bromine	36 Kr Krypton
37 Rb Rubidium	38 Sr Strontium	39 Y Yttrium	40 Zr Zirconium	41 Nb Niobium	42 Mo Molybdenum	43 Tc Technetium	44 Ru Ruthenium	45 Rh Rhodium	46 Pd Palladium	47 Ag Silver	48 Cd Cadmium	49 In Indium	50 Sn Tin	51 Sb Antimony	52 Te Tellurium	53 I Iodine	54 Xe Xenon
55 Cs Cesium	56 Ba Barium	57-71 Lanthanides	72 Hf Hafnium	73 Ta Tantalum	74 W Tungsten	75 Re Rhenium	76 Os Osmium	77 Ir Iridium	78 Pt Platinum	79 Au Gold	80 Hg Mercury	81 Tl Thallium	82 Pb Lead	83 Bi Bismuth	84 Po Polonium	85 At Astatine	86 Rn Radon
87 Fr Francium	88 Ra Radium	89-103 Actinides	104 Rf Rutherfordium	105 Db Dubnium	106 Sg Seaborgium	107 Bh Bohrium	108 Hs Hassium	109 Mt Meitnerium	110 Ds Darmstadtium	111 Rg Roentgenium	112 Cn Copernicium	113 Nh Nihonium	114 Fl Flerovium	115 Mc Moscovium	116 Lv Livermorium	117 Ts Tennessine	118 Og Oganesson

57 La Lanthanum	58 Ce Cerium	59 Pr Praseodymium	60 Nd Neodymium	61 Pm Promethium	62 Sm Samarium	63 Eu Europium	64 Gd Gadolinium	65 Tb Terbium	66 Dy Dysprosium	67 Ho Holmium	68 Er Erbium	69 Tm Thulium	70 Yb Ytterbium	71 Lu Lutetium
89 Ac Actinium	90 Th Thorium	91 Pa Protactinium	92 U Uranium	93 Np Neptunium	94 Pu Plutonium	95 Am Americium	96 Cm Curium	97 Bk Berkelium	98 Cf Californium	99 Es Einsteinium	100 Fm Fermium	101 Md Mendelevium	102 No Nobelium	103 Lr Lawrencium

Fig. 4.2 The periodic table of elements. (Image from Flerlage and Flerlage 2016)

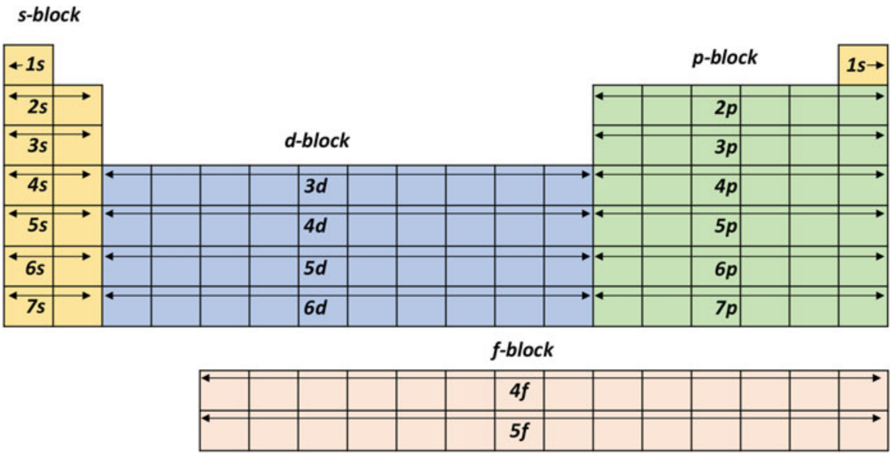


Fig. 4.3 The periodic table in s-, p-. d-, and f-blocks

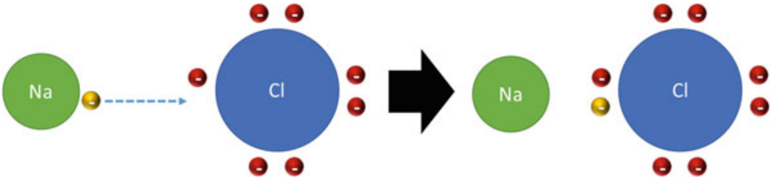


Fig. 4.4 An example of ionic bond (NaCl)

lanthanides and actinides usually located in two rows at the bottom of the period table. Blocks help when writing out the electron configuration of an element.

4.1.3 Chemical Bonds

Atoms are combined together to form a molecule as the smallest fundamental unit of a chemical compound. Connection of atoms in a molecule is possible through chemical bonds. The mechanism for the formation of chemical bonds depends on the way how electrons are utilized and configured in the *s* or *p* orbital of the outer shell for bonding. These mechanisms include mainly four different types, i.e., the ionic bond, the covalent bond, the metallic bond, and the Van der Waals bond.

Ionic bond is formed by electrostatic attraction between particles with opposite charges (positive and negative). In this bond, one or more electrons are removed and attached to anther atom. The ionic bond usually occurs between metallic ions and nonmetallic ions (e.g., LiF, LiBr, NaCl, NaI). The ions are trapped by attraction forces between opposing charges. As shown in Fig. 4.4 as an example, sodium has one electron in the outermost shell while chlorine has seven electrons in the

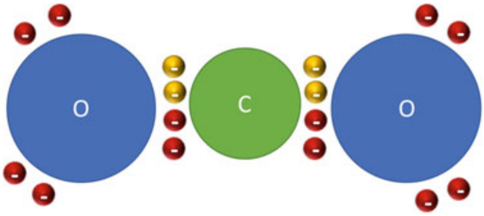


Fig. 4.5 An example of covalent bond (CO_2)

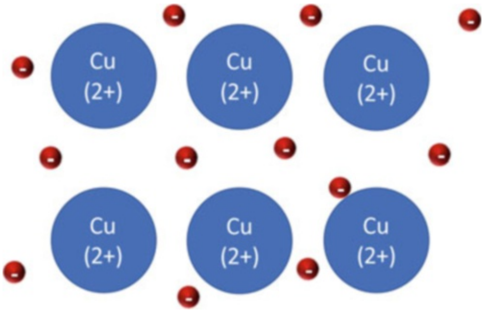


Fig. 4.6 An example of metallic bond (metal copper)

outermost shell. So chlorine needs one electron to fill the outermost shell which can be provided by the excess of sodium. When sodium donates an electron to the chlorine, it becomes positively charged while chlorine becomes negatively charged. The positively charged sodium is attracted to negatively charged chlorine to form NaCl.

Covalent bond refers to the connection of atoms through sharing of electrons. The sharing can be through one or more pairs of electrons by two atoms to fill the *s* or *p* orbital of the outermost shell of each atom. These shared electrons are also called bonding electrons. The molecules formed by covalent bonding are stabilized by the balance of attraction between the atomic nucleus and the electron pair and repulsion between atoms. The bonding is very strong. If a compound has unshared electron pairs, covalent bonds can be formed. Non-metals form compound through covalent bond by sharing electrons (e.g., H_2O, CO_2, NH_3). An example of covalent bond is shown for CO_2 in Fig. 4.5. By sharing electrons, both oxygen and carbon occupy eight electrons in the outermost shell giving them stability.

The metallic bond is found in metals, metalloids, and alloys (a substance made by melting two or more elements together). The bond refers to electrostatic attraction between the positively charged atoms of metallic elements and the “sea” of electrons surrounding the atoms. The “sea” of electrons is formed when the metallic elements, with a low electronegativity, give up their valence electrons, leaving behind a positively charged atom core (i.e., an atom without valence electrons consisting of the nucleus and inner electrons). The valence electrons released can move freely within the electron sea in the metal and become associated with the atom cores. Fig. 4.6 shows an example of metallic bond where positive Cu atom cores are surrounded by a sea of electrons (i.e., the bonding of the metal copper).

The van der Waals bond is the weak electrostatic attraction that joins molecules or groups of atoms. This electrostatic attraction results from a transient shift in electron density which causes one region or another in a molecule become electrically positively or negatively charged. Electrically positive regions of a molecule would be attracted to the electrons of another molecule. This bond caused by attraction between molecules quickly vanishes at longer distances between interacting molecules. The van der Waals bond is usually a secondary one in addition to the covalent or ionic bonds. Examples are the intermolecular forces between HCl molecules or H_2O molecules.

4.2 Basics of Chemical Reactions

Chemical reaction is a process in which the chemical properties of a substance are changed by the rearrangement of the molecular or ionic structure of a substance. In comparison to nuclear reactions where rearrangement of nuclear structures takes place, chemical reactions occur by the rearrangement of the orbital electrons. In a chemical reaction, reacting species (called "reactants") collide and interact with each other (and come close enough) to break bonds between atoms in the reacting species. As a result, the atoms rearrange to form new bonds and produce new substances. These newly created substances are called products.

Understanding why chemical reactions take place requires the understanding of bonding in molecules, how molecules interact, what determines whether an interaction is favored or not, and what the outcome will be. Thermodynamics plays important role in this determination. Thermodynamics allows to identify reactions that are possible, to calculate the equilibrium composition of the products, and the minimum energy needed for the reaction to proceed. Equilibrium indicates a state of balance where the rate of the forward reaction by the reactants and that of the backward reaction by the products are equal. While thermodynamics deal with the final expected chemical composition after the chemical reaction equilibrium, it does not, however, answer the question of "How long does it take for the reaction to reach equilibrium?". This question is answered by chemical kinetics which addresses the issues related to the rate and mechanism of chemical reactions.

Many chemical reactions taking place in the natural environment are typically very fast (e.g., within minutes or hours). Even the reactions that are quite slow are within the period of a few years. Therefore, the rates of chemical reactions are usually not of significance in the discussions of nuclear waste management where the time periods of interest are beyond hundreds and thousands of years,. The primary interest in the discussion of chemistry in relation to the study of nuclear waste management is about equilibrium characterized by thermodynamics.

Definitions

First law of thermodynamics

The first law of thermodynamics is the law of conservation of energy. In an isolated system, the total energy is constant while energy can be transformed from one form to another. Thus the energy cannot be destroyed or created in the system.

Second law of thermodynamics

The second law of thermodynamics is the law of disorder which is represented by so-called entropy. Entropy represents the disorder or randomness of the system. In an isolated system, the total entropy can never decrease over time.

The kinds of chemical reactions occurring in a system will depend on the substances (the types of reactants) involved and the conditions or the medium provided for the reaction. For example, consider a case when two substances are dissolved in water. If one substance gives free hydrogen while the other one yields OH^-, the so-called acid-base reaction can take place. If two substances are connected through a transfer of electrons in water or through a circuit, the substances are engaged in an oxidation and reduction reaction. If the substances change phases between solids and the aqueous state through a chemical reaction, dissolution and precipitation would be involved. If two dissolved species together form a new soluble species, the complexation reaction is happening in this case. If a chemical species is attached to the surface of solids through a chemical reaction, the reaction is called sorption.

The above listed chemical reactions are not mutually exclusive as they may identify specific aspects of the same phenomenon.

Regardless of the specific chemical reactions involved, there are governing principles behind chemical reactions. These governing principles are described by the use of certain devices of chemistry. These devices include free energy and equilibrium constant.

4.2.1 Free Energy in Chemical Reactions

All chemical reactions require breaking of existing chemical bonds in a substance and through new chemical bonds a new chemical substance is formed. However, regardless of the exact nature of the chemical bonds involved in a chemical reaction, the direction of a chemical reaction and the final equilibrium composition are controlled by energy.

Consider the following example, the production of water.

$$H_2 + \frac{1}{2}O_2 \rightleftarrows H_2O$$

We assume that the species are under a so-called standard condition, i.e., 1 atm (atmospheric pressure) and 25 °C.

If the reactants (i.e., H_2, O_2) have enough energy (requiring no extra energy to the system) for the production of products (H_2O), the reaction will occur. On the other hand, if the reaction requires additional energy beyond what is available from the reactants, it will not proceed. This can be explained by using the concept of the free energy (or Gibbs free energy).

It turns out, combination of hydrogen gas and oxygen to produce water does not take place. This is because the reaction requires extra energy. In other words, in the currently considered system, the so-called free energy needed for the reaction to proceed is not available.

The free energy is the energy available to perform "useful work" and can be represented by the following equation.

$$G = H - T \cdot S \tag{4.1}$$

where, G is the free energy [J], H is enthalpy [J], S is entropy [J/K], and T is absolute temperature of the system [K]. It is assumed that the system is a closed system (i.e., there is no exchange of energy or matter with the outside of the system).

Enthalpy is a measure of the total energy of compounds in a system, referring to the energy associated with the bonds and attractions within molecules and between molecules (i.e., the combination of intramolecular forces and intermolecular forces) in the system. Entropy is a measure of disorder or randomness in a system, referring to the degree of "dissociation" of the system. The energy related to the amount of randomness is always greater than zero, and is not available as free energy. Entropy is expressed in units of energy per Kelvin. The equation indicates that the total free energy in a system is the enthalpy in the system minus the product of entropy and temperature which represents the energy related to the amount of randomness in the system that is not available for useful work.

As the total free energy of the reactants, hydrogen and oxygen gas, are less than that of the products that is water, the reaction for the production of water will not proceed.

Chemical reactions proceed toward a composition that minimizes the total free energy through the combination of maximizing disorder and minimizing enthalpy (see Fig. 4.7). The actual pathway taken does not affect the final equilibrium state. In other words, the criterion for equilibrium is that the total free energy of the system is a minimum (at a given temperature and pressure). The total entropy of the system, the disorderliness of the system is at its maximum at equilibrium.

Let us examine the following hypothetical reversible chemical reaction taking place at a constant temperature.

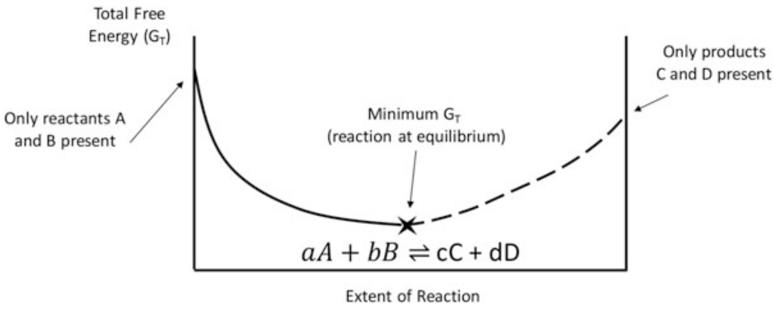


Fig. 4.7 Variation of Gibbs free energy for the chemical reactions showing the state of equilibrium (at the far left and right side, only reactants and products are present, respectively)

$$\mathrm{aA + bB \rightleftharpoons yY + zZ} \tag{4.2}$$

The reactants A and B combine to form the products Y and Z. In this reaction, *a* moles of A combine with *b* moles of B to form *y* moles of Y and *z* moles of Z.

If A and B were added to a reaction vessel as reactants and the total free energy of the system were calculated as a function of extent of reaction as the reaction proceeded, the free energy of the system decreases until it reaches the minimum value. Then the reaction is at equilibrium.

Also, if the chemical reaction requires no input of free energy to proceed in the forward direction, approaching equilibrium is spontaneous. Non-spontaneous reactions require addition of free energy to go forward.

For the given chemical reaction, the total Gibbs free energy of the compounds involved can be represented as

$$G_T = n_A\overline{G}_A + \mathrm{n_B}\overline{G}_B + \mathrm{n}_Y\overline{G}_Y + \mathrm{n}_Z\overline{G}_Z \tag{4.3}$$

where, n_A, n_B, n_Y, and n_Z are the number of moles of A, B, Y, and Z that are present and $\overline{G}_A$, $\overline{G}_B$, $\overline{G}_Y$, and $\overline{G}_Z$ are the free energy per mole of each substance.

As stated, the equilibrium condition of the reaction is achieved when G_T reaches a minimum. In general, reaction in the direction that decreases G_T occurs spontaneously. In contrast, reaction in the direction that increases G_T is not spontaneous or will not occur in a closed system. Also as any chemical reaction proceeds in an incremental fashion, the change in G_T can be represented by the incremental changes in free energy ΔG as,

$$\Delta G = \left(\sum_i v_i\overline{G_i}\right)_{products} - \left(\sum_i v_i\overline{G_i}\right)_{reactants} \tag{4.4}$$

where v_i is the stoichiometric coefficients (e.g., a, b, y, z in the previous chemical reaction) and $\overline{G_i}$ is the free energy per mole.

For a given value of ΔG, the following observations are made for a chemical reaction.

(a) When ΔG is <0, the reaction may proceed spontaneously – reaction may proceed spontaneously to the right (G_T decreases as the reaction proceeds)
(b) When ΔG is >0, the reaction cannot proceed spontaneously as written unless energy is supplied from an external source (G_T would increase if the reaction were to proceed).
(c) $\Delta G = 0$ (i.e., G_T is at a minimum): the reaction is at equilibrium and will not proceed spontaneously in either direction.

ΔG can also be represented as $\varDelta G = \varDelta H - T\varDelta S$, where $\varDelta H$ and $\varDelta S$ is a difference in sums of individual enthalpies and entropies, respectively, for products and reactants (T is absolute temperature).

Values of ΔG for a reaction can be determined by using the relationship

$$\varDelta G = \varDelta G^0 + RTln\frac{[Y]^y[Z]^z}{[A]^a[B]^b} \tag{4.5}$$

where

$$\varDelta G^0 = \left(\sum_i v_i\overline{G_i^0}\right)_{products} - \left(\sum_i v_i\overline{G_i^0}\right)_{reactants} \tag{4.6}$$

$[X]$ = activity of X
$\overline{G_i^0}$ free energy per mole of species i at 25 °C and 1 atmosphere pressure (standard free energy per mole of species i)
R = gas constant (8.314510 J/K-mol)
T = temperature (K)

Activity refers to a measure of the effective concentration of a species under non-ideal (e.g., concentrated) conditions. It determines the real chemical potential for a real solution rather than an ideal one. A related term, concentration, of a species is simply a measure of how much it is dissolved in a liquid. For dilute solutions, activity and concentration are the same. However, in concentrated solutions, they are different. The value of the activity of a substance is dependent on the choice of standard state conditions. The activity generally is assumed to be 1 (as a dimensionless quantity) for the dilute solutions or for pure substances in condensed phases (solids or liquids).

Table 4.2 Thermodynamic constants for species of interest[a] (condensed from Robie et al. 1979)

Species	ΔH°_f KJ/mole	ΔG°_f KJ/mole
Al_2O_3 (s)	−1653.517	−1562.702
B_2O_3 (s)	−1273.500	-1194.325
Ca^{2+} (aq)	−542.830	−553.540
$CaCO_3$ (s), calcite	−1207.370	−1128.842
CaO (s)	−635.089	−603.487
$Ca(OH)_2$	−986.085	−898.408
CH_4 (g)	−74.810	−50.708
Cl^- (aq)	−167.080	−131.270
Co^{2+} (aq)	−58.200	−54.400
Co^{3+} (aq)	92.000	134.000
CO_2 (g)	−393.510	−394.375
CO_2 (aq)	−412.919	−386.225
CO_3^{2-} (aq)	−677.140	−527.900
HCO_3^- (aq)	−691.990	−586.850
H_2CO_3 (aq)	−699.650	−623.170
Cs^+ (aq)	−258.040	−283.625
CsOH (s)	−416.726	−370.690
Cu^+ (aq)	71.670	50.000
Cu^{++} (aq)	64.770	65.520
CuO (s)	−157.320	−129.564
Cu_2O (s)	−168.610	−146.030
F^- (aq)	−335.350	−281.705
Fe^{2+} (aq)	−89.000	−78.870
Fe^{3+} (aq)	−48.500	−4.600
Fe_2O_3 (s)	−824.640	−742.683
Fe_3O_4 (s)	−1115.726	−1012.566
$Fe(OH)_3$ (s)	−824.248	−694.544
H^+ (aq)	0	0
H_2O (g)	−241.814	−228.569
H_2O (l)	−285.830	−237.141
I_2 (g)	62.420	19.329
I^- (aq)	−56.900	−51.915
K_2O (s)	−363.171	−322.087
KOH (s)	−424.676	−378.932
MgO (s)	−601.490	−569.196
NaOH (s)	−425.800	−379.651
NO_3^- (aq)	−207.400	−111.500
NH_3 (g)	−45.940	−16.410
NH_3 (aq)	−80.290	−26.600
NH_4^+ (aq)	−133.260	−79.457
HNO_3 (aq)	−206.572	−110.499

(continued)

Table 4.2 (continued)

Species	ΔH°_f KJ/mole	ΔG°_f KJ/mole
OH^- (aq)	−230.025	−157.328
P_2O_5 (s)	−1470.000	−1337.897
PO_4^{3-} (aq)	−1259.550	−1001.550
PbO (s)	−219.409	−189.202
SiO_2 (s)	−910.700	−856.288
SO_4^{2-} (aq)	−907.510	−741.990
Sr^{2+} (aq)	−545.800	−559.440
SrO (s)	−590.490	−560.353
TiO_2 (s)	−944.700	−889.446
U^{3+} (aq)	−514.600	−520.500
U^{4+} (aq)	−613.800	−579.100
ZrO_2 (s)	−1100.560	−1042.790

[a] For a hypothetical ideal state of unit molality, which is approximately equal to that of unit molarity

Example 4.1: Gibbs Free Energy

We cannot measure absolute values of free energy but can measure changes in free energy as relative values using convention.

At standard state, every element is assigned, by convention, a free energy of *zero* per mole. For example, H_2 (g), O_2 (g), $C_{graphites}$ (s) are all assigned free energy values of 0 kcal/mole. Also, to establish a baseline for ionic substances in solution, H^+ at a concentration of 1 mole/l in an ideal solution and at standard state conditions has been assigned a free energy of zero.

Assigning a value of zero to elements at standard state allows us to measure the free energy change involved in forming compounds at standard state. This free energy change is called the standard free energy of formation, ΔG°_f (from their component elements at standard state). Table 4.2 presents a summary of thermodynamic constants including the ΔG°_f values for some substances commonly encountered in water chemistry.

For example, we can determine that the formation of calcite ($CaCO_3$ (s)) from carbon, calcium, and oxygen has the following free energy,

(continued)

Example 4.1 (continued)

$$C\ (graphite) + \frac{3}{2}\ O_2(g) + Ca(s) \rightarrow CaCO_3(calcite)(s); \qquad \Delta G^{\circ}_{f} = -1,128.842 \frac{kJ}{mole}$$

where, $C(graphite)$, $O_2(g)$, and $Ca(s)$ all have free energy of *zero* by convention.

The minus sign indicates that free energy is released or given off. We can also deduce that breaking down of 1 mole of calcite into 1 mole of Ca metal, 1 mole of graphite, and 3/2 moles of oxygen would require the input of +1128.842 kJ of free energy per mole (as going in the opposite direction of the chemical reaction in this example).

4.2.2 Equilibrium Constant

Consider an example of chemical reaction (Eq. 4.2), $aA + bB \rightleftharpoons yY + zZ$.

In the reaction, the reactants A and B interact with each other and produce the products Y and Z. If the reactants A and B were introduced into a reaction vessel, the concentrations of A and B will decrease until they reach values that do not change with time (while the concentrations of Y and Z increase from zero to time-invariant values). The state of equilibrium represents a balance between the forward process and the backward process going at equal rate: At equilibrium, the forward rate of reactions is equal to the rate of the reverse reaction. Therefore at equilibrium, the driving force of forward reaction is equal to the driving force of reverse reaction while two reactions are still going on, i.e., $k_1[A]^a[B]^b = k_2[Y]^y[Z]^z$.

The ratio of concentrations of the products to the concentrations of the reactants is the so-called equilibrium constant, *K*.

$$K = \frac{k_1}{k_2} = \frac{[Y]^y[Z]^z}{[A]^a[B]^b} \equiv \text{equilibrium constant} \tag{4.7}$$

The equilibrium constant defines the equilibrium condition for a particular reaction showing the balance between reactants and products. Because pure solids and liquids do not affect the reactant amount for the reaction equilibrium, their concentration values are set to be 1 when calculating the equilibrium constant.

Note that adding or removing reactants or products in the reaction will not change the equilibrium constant. For example, if the concentration of the reactants are increased, the equilibrium of the reaction will force the forward reaction. Therefore, the reactants are consumed and their concentration is decreased. This phenomenon of any disturbance (e.g., concentration, pressure) of chemical equilibrium being counteracted to reestablish an equilibrium is called Le Chatelier's principles.

<Conventions>

For a chemical reaction of

$$\mathbf{aA} + \mathbf{bB} + \ldots \rightleftharpoons \mathbf{yY} + \mathbf{zZ} + \ldots$$

the equilibrium constant for such particular chemical reaction is given as follows:

$$K = \frac{k_1}{k_2} = \frac{[Y]^y[Z]^z\ldots}{[A]^a[B]^b\ldots}$$

- K has a definite value for *a particular chemical reaction*, not for a particular substance.
- Values of K are given for 25 °C and 1 atm total pressure (standard temperature and pressure, STP).
- Concentrations of gases, in the equilibrium constant formula, are expressed as partial pressure in *atm.*
- Concentrations of solutes in aqueous solutions are expressed as *mole/liter* of solution.
- Concentrations of pure solids/liquid do not appear in the formula (i.e. their values are equal to 1).

From the discussions of free energy in the previous section, the reaction is at equilibrium when the free energy change is zero, i.e., $\Delta G = 0$. There is no difference in free energy between the reactants and the products in equilibrium.

From Eq. 4.5,

$$\Delta G = \Delta G^0 + RTln\frac{[Y]^y[Z]^z}{[A]^a[B]^b} = 0$$

$$\Delta G^0 = -RTln\frac{[Y]^y[Z]^z}{[A]^a[B]^b} = -RTln\,K$$

Therefore, the equilibrium constant K becomes,

$$K = e^{\left(-\frac{\Delta G^0}{RT}\right)} \tag{4.8}$$

Example 4.2: Equilibrium Constant −1

(a) Derive the equilibrium constant and ΔG^0 for the following chemical reaction, in which liquid H_2O dissociates to H^+ and OH^- (thus the symbol *aq*) at standard temperature and pressure (25 °C and 1 atm).

$$H_2O\ (l) \rightarrow H^+(aq) + OH^-(aq)$$

(b) Does this reaction proceed as written above (from left to right) when $[H^+] = 10^{-6}$ M and $[OH^-] = 5 \times 10^{-8}$ M?

Solutions:

(a) For the given standard state conditions and neglecting the ionic strength effects, K can be written as follows for the dilute solution,

$$K = \frac{1 \cdot [H^+] \cdot 1 \cdot [OH^-]}{1} = [H^+][OH^-]$$

This equilibrium constant is designated as K_w. This value is about 10^{-14} at STP.

From Table 4.2

	ΔG_f°
$H_2O\ (l)$	−237.141
H^+	0
OH^-	−157.328

From Eq. 4.6,

$$\Delta G^0 = \left(\sum_i v_i \overline{G_i^0}\right)_{products} - \left(\sum_i v_i \overline{G_i^0}\right)_{reactants}$$

$$\Delta G^0 = [(1)(0) + (1)(-157.328)] - [(1)(-237.141)] = +79.813\ kJ$$

b) From Eq. 4.7

$\Delta G = \Delta G^0 + RT\ ln\ [H^+][OH^-]$
$\Delta G = 79.813 + 8.314510 \times 10^{-3} \times 298.15 \times \log[(10^{-6})(5 \times 10^{-8})]$
$\Delta G = +4.3$ kJ

Because $\Delta G > 0$, the reaction is not spontaneous as written and can proceed spontaneously only in the opposite direction, i.e. H^+ and OH^- are combining to form H_2O molecules.

4.3 Types of Chemical Reactions

4.3.1 Acid-Base Reactions

A well-known example of acid-base reaction is acid rain causing damage to structures. The dilute sulfuric acid in the rain, for example, comes into contact with calcite (the base) in the structural materials and changes its chemical form through the following chemical reaction.

$$\mathrm{CaCO}_{3(\mathrm{s})} + \mathrm{H}_2\mathrm{SO}_{4(\mathrm{aq})} \rightleftharpoons \mathrm{CaSO}_{4(\mathrm{s})} + CO_{2(\mathrm{g})}$$

Acid/base reactions involve the transfer of the proton (H^+) between two species. Therefore, they are the basis for the determination of pH of natural water or any solution. The pH is one of the key parameters controlling various chemical reactions in water. It also affects migration behavior of contaminant in groundwater or surface water. The pH is defined as the negative logarithm of the H^+ concentration,

$$pH = -\log\ [H^+] \tag{4.9}$$

The value of pH represents the balance between acid (H^+) and alkaline (OH^-) in a solution and indicates how acidic or alkaline the solution is. If there are equal quantities of H^+ and OH^- ions, the solution has a pH of 7 (neutral). If the pH value decreases below 7, the solution is acidic as there are more H^+ ions than OH^- ions. If the pH value increases above 7, the solution is alkaline (as there are more OH^- ions than H^+ ions).

Acids are formed when oxide compounds of nonmetals dissolve in water. For example, CO_2 forms HCO_3^- upon dissolution in water. The term base or alkali is used for a substance that neutralizes an acid.

Definitions

Acid.	A substance containing hydrogen, which gives free hydrogen when dissolved in water (any substance whose molecules can react with unattached electron pairs in other compounds is also acid).
Base.	A substance that neutralizes an acid by containing the OH group but does not dissolve in water
Alkali.	A base that dissolves in water and produces OH^-
Alkali metal.	Any metal of the group IA: lithium, sodium, potassium, rubidium, cesium.
Alkaline-earth metal.	

(continued)

One of the group IIA: calcium, strontium, barium (including magnesium, beryllium and radon).

It is often assumed that more presence of Cl^-, SO_4^{2-}, F^-, Br^-, $H_2BO_3^-$ is acid environment.

A solution containing large amount of Na^+ and K^+ is commonly referred as alkaline. Nonmetallic oxides are sometimes called “acid oxide” whether they dissolve in water or not. For example, silica (SiO_2) can be called “acid oxide”. Any metal oxide, regardless of whether it dissolves in water, is called “basic oxide”.

Acid-base reactions provide basis for other chemical reactions and can be classified into solid dissolution and precipitation, aqueous complexation, or sorption reactions.

4.3.1.1 The Concept of pH

As stated, pH is a measure of the concentration of H^+ in solution. In reality, a hydrogen ion (H^+) cannot exist in water; it is in the form H_3O^+, the hydronium ion, formed by the combination of water with the hydrogen ion. However, it will be represented as H^+, for convenience.

The concentration of H^+ originates from the dissolution of water. In fact, water is always slightly dissociated. The following reaction proceeds until H^+ and OH^- have equal concentrations of 10^{-7} M, at which point the solution is neutral.

$$OH^- + H^+ \rightleftarrows H_2O \tag{4.10}$$

For all water solutions, the product of $[H^+][OH^-]$ is constant as 10^{-14}. This means that H^+ is present even in strongly basic solutions and OH^- in strong acids. Therefore, the acidity or alkalinity of a solution can be specified by giving the concentration of one of H^+ or OH^-.

Example 4.3: pH of a Solution
What is the pH of the following solution?

(a) A solution made up to contain 0.001 M HCl.
(b) A solution containing 0.001 M NaOH

Solutions:

(continued)

Example 4.3 (continued)

(a) The solution has $[H^+] = 10^{-3}$ M. By definition, the pH as the negative log of 10^{-3} is 3.

(b) The solution has $[OH^-] = 10^{-3}$ M. As $[H^+][OH^-] = 10^{-14}$, Then $[H^+] = 10^{+3} \times 10^{-14} = 10^{-11}$, pH = 11.

The concentration of hydrogen ion in an aqueous system is closely related to the chemistry of carbonate species in exchange with atmospheric CO_2 and the chemical equilibrium of other chemical species in the same aqueous environment. In fact, the equilibrium of calcium carbonate in contact with natural water is one of the most important chemical reactions in natural water system. Neutral water exposed to CO_2 in the atmosphere will allow dissolution of CO_2 at the equivalent level of CO_2 partial pressure in the atmosphere.

The concentration of CO_2 in water can be determined in conjunction with Henry's law: The amount of a given gas dissolved in a given volume of liquid is directly proportional to its partial pressure above the liquid,

$$k = \frac{C}{P_{gas}} \tag{4.11}$$

where k is Henry's law constant, C is the concentration of the gas, and P_{gas} is the partial pressure of the gas.

In addition, the principle of charge balance also applies,

$$\sum_{i}^{cations} \left(\frac{mol}{l}\right)_i - \sum_{j}^{anions} \left(\frac{mol}{l}\right)_j = 0 \tag{4.12}$$

The CO_2 will react with H_2O to form H_2CO_3, a weak acid, which will dissociate to form H^+ and HCO_3^-. HCO_3^- also dissociates to produce H^+ and CO_3^{2-}. The resulting balance of species determines the pH of the solution. The following example illustrates these points.

Example 4.4: Determination of pH of Natural Water

As the concentration of carbon dioxide is far greater than that of the other weak acids, the most important acid, in describing pH of natural water, is carbonic acid (H_2CO_3), a weak acid.

Natural water has the following reactions:

(continued)

Example 4.4 (continued)

$$CO_2(g) + H_2O \rightleftarrows H_2CO_3$$

$$H_2CO_3 \rightleftarrows H^+ + HCO_3^-$$

$$HCO_3^- \rightleftarrows H^+ + CO_3^{2-}$$

$$H^+ + OH^- \rightleftarrows H_2O$$

and the equilibrium constants for the chemical reactions, assuming STP, are,

$$K_H = \frac{[H_2CO_3]}{P_{CO_2}} = 10^{-1.47}$$

$$K_{H_2CO_3} = \frac{[H^+]\left[HCO_3^-\right]}{[H_2CO_3]} = 10^{-6.35}$$

$$K_{HCO_3^-} = \frac{[H^+]\left[CO_3^{2-}\right]}{\left[HCO_3^-\right]} = 10^{-10.33}$$

$$K_{H_2O} = K_W = \frac{[H^+][OH^-]}{[H_2O]} = 10^{-14}$$

Determine the pH of natural water when the atmospheric CO_2 concentration is at 400 ppm.

Solution:

The CO_2 concentration is 400 ppm, meaning the number of CO_2 molecules in the air is 400 per million air molecules. Since the gas pressure is proportional to the number of molecules, it means the partial pressure of CO_2 is (1 atm) × (400×10^{-6}), or $P_{CO_2} = 400 \times 10^{-6} = 10^{-3.40}$atm.

So far, we have four equations for five unknowns, $[H^+]$, $[OH^-]$, $\left[HCO_3^-\right]$, $\left[CO_3^{2-}\right][CO_3{}^{2-}]$, $[H_2CO_3]$, since $[H_2O]$ is 1.

Thus, one more equation is needed to solve this problem. This equation comes from the principle of charge balance (from Eq. 4.12):

$$[H^+] - 2\left[CO_3^{2-}\right] - \left[HCO_3^-\right] - [OH^-] = 0$$

Using Henry's law (Eq. 4.11) and the value of K_H,

$$[H_2CO_3] = K_H P_{CO_2} = 10^{-1.47} \cdot 10^{-3.40} = 10^{-4.87}[\text{mol/l}]$$

By using the information given in the problem,

(continued)

Example 4.4 (continued)

$$\left[HCO_3^-\right] = \frac{10^{-6.35}[H_2CO_3]}{[H^+]} = \frac{10^{-6.35} \cdot 10^{-4.87}}{[H^+]} = \frac{10^{-11.22}}{[H^+]}$$

$$\left[CO_3^{2-}\right] = \frac{10^{-10.33}\left[HCO_3^-\right]}{[H^+]} = \frac{10^{-10.33}}{[H^+]} \cdot \frac{10^{-11.21}}{[H^+]} = \frac{10^{-21.54}}{[H^+]^2}$$

$$[OH^-] = \frac{10^{-14}}{[H^+]}$$

By using the above terms in the charge balance equation,

$$[H^+] - 2\frac{10^{-21.54}}{[H^+]^2} - \frac{10^{-11.22}}{[H^+]} - \frac{10^{-14}}{[H^+]} = 0$$

By solving this, we get $[H^+] \cong 10^{-5.65}$, thus the pH value of the water is 5.65.

It is noted that the *pH* of natural water is determined by a balance between the dissolution of the weakly acidic CO_2 and that of basic rocks. More precisely, the *pH* is determined by the extent of dissolution of the dissolved carbonic acid, and other weak acids through the balancing of negative charge (HCO_3^-, CO_3^{2-}, OH^-) from the acids with positive charge (e.g., Na^+, K^+, Ca^{2+}) from the strong mineral bases coming from key rock forming minerals. In groundwater studies, soil CO_2 from organic decomposition is another source of importance. As calcite (calcium carbonate, $CaCO_3$) and dolomite (calcium magnesium carbonate, $CaMg(CO_3)_2$) are soluble in acid solution, even rainwater will dissolve carbonate rocks. Likewise, a change in pH can also result in a precipitation of $CaCO_3$ from a solution that was at equilibrium prior to pH change.

4.3.2 Dissolution-Precipitation Reactions

Dissolution and precipitation reaction is another important chemical reaction in natural water system. It controls the phase changes between solids and the aqueous phase. These reactions are the basis of rock dissolution through weathering and control the composition of natural water and the sediments. Through dissolution and precipitation, the equilibrium between solids and the aqueous phase in natural water is maintained, explaining the concentration of various species in surface and groundwater. The degree of dissolution of a compound in water is controlled by solubility. Solubility is the maximum concentration of a chemical dissolved in water at equilibrium.

Table 4.3 Characterization of oxidation and reduction in the changes of hydrogen, oxygen, and electron

	Oxidation	Reduction
Hydrogen	Loss	Gain
Oxygen	Gain	Loss
Electron	Loss	Gain

Dissolution is a primary mechanism of releasing radionuclides from waste form into adjacent environment. The converse, precipitation, is the process of adding contaminants from solution to solids. Large amounts of precipitate can also alter the pore structure of the soil and cause a decrease in its permeability. Precipitation of dissolved species causes retardation in radionuclide transport.

4.3.3 Oxidation-Reduction Reactions

4.3.3.1 Basic Definitions

An oxidation-reduction reaction refers to any chemical reaction that involves a transfer of electrons between two chemical species. These electron transfers take place in two parts of half (oxidation or reduction) reactions. Furthermore, these two half reactions are always coupled by electrons: Electrons are generated in an oxidation reaction and consumed by reduction. In slightly different words, oxidation is a chemical reaction in which a substance donates (gives away) electrons and reduction is a reaction in which a substance accepts (or gains) electrons.

Oxidation-reduction reactions control the process of material degradation in the environment. In fact, the geochemical cycles of elements in nature are driven in part by oxidation and reduction reactions. Photosynthesis which drives energy flow in ecosystems is oxidation-reduction reaction. Life on earth in general is sustained through oxidation-reduction processes. The fate of many chemicals and radionuclides in natural environment is also controlled by oxidation-reduction process as the reaction affects the solubility and migration characteristics in surface or ground water. Oxidation-reduction reactions can be classified as either aqueous complexation, precipitation, or adsorption reactions.

Example 4.5: Photosynthesis

Photosynthesis is a reduction of inorganic carbon to organic matter while producing oxygen. This is shown below.

$$6\mathrm{CO_2(g)} + 6\mathrm{H_2O} \rightarrow \mathrm{C_6H_{12}O_6} + 6\mathrm{O_2(g)}, \quad \Delta G = +2872\ \mathrm{kJ\ mol^{-1-}}$$

In the reaction, carbon is reduced and oxygen is oxidized. As ΔG is >0, the reaction cannot proceed spontaneously. The energy needed for the reaction to proceed is provided by solar energy.

Some differences between acid-base reactions and oxidation-reduction reactions are noted. Acid-base reactions in solution occur fast whereas oxidation-reduction reactions are slow. Also, for the oxidation-reduction reactions, the value of the equilibrium constants is often extremely large or extremely small. That is not the case for acid-base reactions.

Transfer of electrons between species in oxidation and reduction is represented by the changes in the oxidation number of the chemical species (atom, ion, or molecule) involved in the reaction. For the species receiving electrons, the oxidation number is decreased (called ‘reduction’). For the species donating electrons, the oxidation number is increased (called ‘oxidation’). Here oxidation number means the electrical charge assigned to the chemical species. Oxidation can also be defined as gaining of oxygen by a substance. Reduction is then loss of oxygen by a substance. This is summarized in Table 4.3

An oxidation-reduction reaction is also called ‘redox’ reaction and any pair of species comprising the same element in different oxidation states is a redox couple.

$$\textit{oxidized state} + n \cdot e^{-} \rightleftharpoons \textit{reduced state} \tag{4.13}$$

Definition: Oxidation State

Oxidation number (or oxidation state (OS) or valence state) is defined as the electrical charge assigned to an atom. It is obtained directly from the compositional formula as explained below.

1. A free and uncombined element (Li, H in H_2, O in O_2, etc.): The OS is zero.
2. Hydrogen in compounds: The OS is usually +1 (except in the case of the metallic hydrides, where it is -1)
3. Oxygen in compounds: The OS is usually -2 (except in peroxides, where it is -1, or in fluorine compounds, where it may be positive).
4. Alkali metals, alkaline earth metals, and halogens: The OS is usually +1, +2, and $-$ 1.
5. The algebraic sum of the positive and negative oxidation states of all atoms in a neutral molecule is zero.
6. The algebraic sum of the positive and negative oxidation states of all atoms in an ion is same as the charge of the ion.

Oxidation number is different from valence as valence is the number of electrons present in the outermost shell of a particular element while oxidation number relates to the number of electrons lost or gained by the element.

Example 4.6: Determine the Oxidation State(s) of the Followings

(a) H_2

(b) CO_2

(continued)

(c) BF_3
(d) S_2O_3
(e) H_2SO_4
(f) $Na_2Cr_2O_7$
(g) HPO_4^{2-}

Solution:

(a) $H_2 = 0$ (elemental state)
(b) $O = -2$; $0 = C + 2(-2) \rightarrow$ Solving for C gives: $C = +4$, $O = -2$
(c) $F = -1$; $0 = B + 3(-1) \rightarrow B = +3$, $F = -1$
(d) $O = -2$; $0 = 2S + 3(-2) \rightarrow S = +3$, $O = -2$
(e) $H = +1$, $O = -2$; $0 = 2(+1) + S + 4(-2) \rightarrow S = +6$, $H = +1$, $O = -2$
(f) $Na = +1$, $O = -2$; $0 = 2(+1) + 2(Cr) + 7(-2) \rightarrow Cr = +6$, $Na = +1$, $O = -2$
(g) $H = +1$, $O = -2$; $-2 = (+1) + P + 4(-2) \rightarrow P = +5$, $H = +1$, $O = -2$

4.3.3.2 Half-Reactions

As explained, oxidation is an increase in oxidation state (number), giving away of electrons, such as,

$$Zn \rightarrow Zn^{2+} + 2e^-$$

Reduction is a decrease in oxidation state (number), gaining electrons, such as,

$$Cu^{2+} + 2e^- \rightarrow Cu$$

Addition of the two half-reactions becomes the following:

$$Zn + Cu^{2+} \rightarrow Zn^{2+} + Cu$$

Each of the processes that occurs as electrons are produced or consumed at an electrode is called half-reaction. Therefore, addition of two separate half-reactions, an oxidation and a reduction, gives the complete oxidation and reduction reaction.

4.3.3.3 Log of Electron Activity "pe" and Oxidation ("Redox") Potential

Discussions in the previous section on acid-base reaction showed that the pH value in a solution indicates the tendency for accepting or transferring hydrogen ions (H^+). Similarly, another parameter, *pe*, is defined to represent the relative tendency of a solution to accept or transfer electrons through chemical reactions. In parallel to the definition to pH, *pe* is defined as the negative logarithm of the electron activity or concentration, i.e.,

$$pe \equiv -log\ [e^-] \tag{4.14}$$

Thus, *pe* represents the electron activity or redox intensity in redox reactions as e^- is being exchanged in the reaction. (Here activity refers to concentration of species after taking into account the effect of electrostatic forces between ions dissolved in the water, as mentioned in Sect. 4.2.1.

The value of *pe* can be calculated for any half redox reaction as a measure of the "reducing power" of the corresponding redox couple in the system of interest. Low *pe* (i.e., high [e^-]) indicates high reducing power (i.e., low tendency for oxidation and high tendency for reduction), and high pe (i.e., low [e^-]) indicates the opposite. Thus a value of *pe* indicates the tendency for oxidation reduction reactions. Calculation of *pe* in a redox reaction is illustrated below.

$$K = \frac{[Products]}{[Reactants]} = \frac{[Reductants]}{[Oxidants]}\frac{1}{e^n} \tag{4.15}$$

$$log\,K = log\,\frac{[Reductants]}{[Oxidants]} - n\ log\ e^- \tag{4.16}$$

$$-log\,[e^-] = \frac{1}{n}\left[log\,K - log\,\frac{[Reductants]}{[Oxidants]}\right] \tag{4.17}$$

$$pe^0 = \frac{1}{n}\,log\,K \tag{4.18}$$

$$pe = pe^0 - \frac{1}{n}\,log\,\frac{[Reductants]}{[Oxidants]} \tag{4.19}$$

where, *n* is the number of electrons involved in the reaction. As shown, pe^0 is the log of the equilibrium constant for the reaction divided by the number of electrons involved in the reaction (shifting from one kind of atom to another) involving one electron. It represents the electron activity when all species are at unit activity.

Also, *pe* is related to the Gibbs free energy change in the reaction according to the following,

$$pe = \frac{-\Delta G}{2.3nRT} \tag{4.20}$$

where, ΔG is the Gibbs free energy change, *n* is the number of electrons involved in the reaction, *R* is gas constant, 8.314 J-mol^{-1} K^{-1}, and *T* is temperature in K.

Table 4.4 Examples of effective equilibrium constants of aquatic redox couples

	pe°	$pe_w{}^\circ\ (pH = 7)$
$\frac{1}{2}H_2O_2 + H^+ + e^- = H_2O$	+ 30.0	+23.0
$\frac{1}{4}O_2(g) + H^+ + e^- = \frac{1}{2}H_2O$	+20.75	+13.75
$\frac{1}{5}NO_3^- + \frac{6}{5}H^+ + e^- = \frac{1}{10}N_2(g) + \frac{3}{5}H_2O$	+21.05	+12.65
$\frac{1}{2}MnO_2(s) + 2H^+ + e^- = \frac{1}{2}Mn^{2+}H_2O$	+20.8	+9.8[a]
$\frac{1}{2}NO_3^- + H^+ + e^- = \frac{1}{2}NO_2^- + \frac{1}{2}H_2O$	+14.15	+7.15
$\frac{1}{8}NO_3^- + \frac{5}{4}H^+ + e^- = \frac{1}{8}NH_4^+ + \frac{3}{8}H_2O$	+14.9	+6.15
$\frac{1}{6}NO_3^- + \frac{4}{3}H^+ + e^- = \frac{1}{6}NH_4^+ + \frac{1}{3}H_2O$	+15.2	+5.8
$\frac{1}{2}O_2(g) + H^+ + e^- = \frac{1}{2}H_2O_2$	+11.5	+4.5
$Fe(OH)_3(am) + 3H^+ + e^- = Fe^{2+} + 3H_2O$	+16.0	+1.0[a]
$\frac{1}{6}SO_4^{2-} + \frac{4}{3}H^+ + e^- = \frac{1}{48}S_8(col) + \frac{2}{3}H_2O$	+5.9	−3.4
$\frac{1}{8}SO_4^{2-} + \frac{5}{4}H^+ + e^- = \frac{1}{8}H_2S(g) + \frac{1}{2}H_2O$	+5.25	−3.5
$\frac{1}{8}SO_4^{2-} + \frac{9}{8}H^+ + e^- = \frac{1}{8}HS^- + \frac{1}{2}H_2O$	+4.25	−3.6
$\frac{1}{8}HCO_3^- + \frac{9}{8}H^+ + e^- = \frac{1}{8}CH_4(g) + \frac{3}{8}H_2O$	+3.8	−4.0
$\frac{1}{8}CO_2(g) + H^+ + e^- = \frac{1}{8}CH_4(g) + \frac{1}{4}H_2O$	+2.9	−4.1
$\frac{1}{16}S_8(col) + H^+ + e^- = \frac{1}{2}H_2S(g)$	+3.2	−3.8[b]
$\frac{1}{16}S_8(col) + \frac{1}{2}H^+ + e^- = \frac{1}{2}HS^-$	−0.8	−4.3
$\frac{1}{6}N_2(g) + \frac{4}{3}H^+ + e^- = \frac{1}{3}NH_4^+$	+4.65	−4.7
$H^+ + e^- = \frac{1}{2}H_2(g)$	0	−7.0
$\frac{1}{4}HCO_3^- + \frac{5}{4}H^+ + e^- = \frac{1}{4}{''}CH_2O{''} + \frac{1}{2}H_2O$	+1.8	−7.0
$\frac{1}{4}CO_2(g) + H^+ + e^- = \frac{1}{4}{''}CH_2O{''} + \frac{1}{4}H_2O$	−0.2	−7.2
$\frac{1}{2}HCO_3^- + \frac{3}{2}H^+ + e^- = \frac{1}{2}CO(g) + H_2O$	+2.2	−8.3
$\frac{1}{2}CO_2(g) + H^+ + e^- = \frac{1}{2}CO(g) + \frac{1}{2}H_2O$	−1.7	−8.7

[a] In the reaction of reductive dissolution of metal oxides, the concentrations of the dissolved metals (Mn^{2+} and Fe^{2+}) have been fixed at 1 μM to more accurately reflect their relative redox properties

[b] This reaction is listed out of order so as not to spate it form the reaction of formation of the bisulfide ion HS^-

Example 4.7: Determination of *pe* from pH and Oxygen Partial Pressure

The major oxidant in "oxic" natural water is oxygen. Its tendency to oxidize other compounds can be given by the *pe* of the oxygen/water couple. Determine the value of *pe* of oxic natural water for the following conditions.

1. pH = 7 and O_2 atmospheric concentration of 20% ($P_{O_2} = 0.20$ atm)
2. pH = 7 and $P_{O_2} = 0.30$ atm
3. pH = 7 and $P_{O_2} = 0.15$ atm
4. pH = 6.5 and $P_{O_2} = 0.20$ atm
5. pH = 8.5 and $P_{O_2} = 0.20$ atm

(continued)

Example 4.7 (continued)

Solution:

1. pH = 7 and O_2 atmospheric concentration of 20% (P_{O_2} = 0.20 atm = $10^{-0.7}$ atm)

$\frac{1}{4}O_2(g) + H^+ + e^- = \frac{1}{2}H_2O;$

$pe^0 = 20.75$ (from Table 4.4, given later in this section)

$$[O_2] = P_{O_2}^{1/4}[H^+]$$

$$[H^+] = 10^{-7}$$

$$n = 1$$

$$pe = pe^0 - \frac{1}{n} log \frac{[Reductants]}{[Oxidants]} = pe^0 - log \frac{1}{P_{O_2}^{1/4}[H^+]}$$

$$= 20.75 - log \frac{1}{(0.2)^{1/4} \cdot 10^{-7}} = 13.58$$

2. pH = 7 and P_{O_2} = 0.30 atm

(in the same way as above)

$$pe = 20.75 - log \frac{1}{(0.30)^{1/4} \cdot 10^{-7}} = 13.62$$

3. pH = 7 and P_{O_2} = 0.15 atm

$$pe = 20.75 - log \frac{1}{(0.15)^{1/4} \cdot 10^{-7}} = 13.54$$

4. pH = 6.5 and P_{O_2} = 0.20 atm

$$pe = 20.75 - log \frac{1}{(0.20)^{1/4} \cdot 10^{-6.5}} = 14.08$$

5. pH = 8.5 and P_{O_2} = 0.20 atm

(continued)

Example 4.7 (continued)

$$pe = 20.75 - \mathit{log}\ \frac{1}{(0.20)^{1/4} \cdot 10^{-8.5}} = 12.08$$

We can see that *pe* is insensitive to the changes in the partial pressure of oxygen and is more sensitive to the changes in pH. In nature, *pe*'s in the range of 12–14 are the characteristic of oxic natural water.

Oxidation (or "redox") potential, E_h, is another expression for electron activity. E_h is also used as a measure of the tendency for oxidation or reduction and can be experimentally determined. Measurement of E_h is explained in the next subsection (4.3.3.4).

E_h is related to *pe* as following

$$pe = \frac{F}{2.3RT} E_h \tag{4.21}$$

$$\begin{aligned} E_h &= \frac{2.3RT}{F} pe \\ &= \frac{2.3RT}{F} \left\{ pe^0 - \log \frac{[Reductants]}{[Oxidants]} \right\} \\ &= \frac{2.3RT}{F} \left\{ \frac{1}{n} \log K - \frac{1}{n} \log \frac{[Reductants]}{[Oxidants]} \right\} \\ &= \frac{2.3RT}{nF} \left\{ \log K - \log \frac{[Reductants]}{[Oxidants]} \right\} \end{aligned} \tag{4.22}$$

Here *F* is the Faraday constant = 96,485 coulombs (the electrical charge of 1 mole of electrons), and the term, 2.3 *RT/F*, is equal to 0.059 V at 25 °C. While *pe* is a dimensionless scale (like *pH*), E_h (redox potential) is measured in volts. A value of E_h is derived based on thermodynamics to represent potential for transfer of electrons.

The oxidation potential of a given system can be represented by using the standard oxidation potential as follows.

$$\mathrm{E_h} = \mathrm{E_h^0} - \frac{2.3\mathrm{RT}}{\mathrm{nF}} \log \frac{[\mathrm{Reductants}]}{[\mathrm{Oxidants}]} \tag{4.23}$$

$$= \mathrm{E_h^0} - \frac{0.059}{\mathrm{n}} \log \frac{[\mathrm{Reductants}]}{[\mathrm{Oxidants}]} \tag{4.24}$$

where

$$E_h^0 = \frac{2.3RT}{F} pe^0 = \frac{2.3RT}{nF} \log K \tag{4.25}$$

Here E_h^0 is used to represent the standard oxidation potential of redox reactions corresponding to unit activities of products and reactants. The above Eq. 4.24 is the thermodynamic relationship of the oxidation potential to the composition of the solution, known as the Nernst equation. The equation relates the oxidation potential of a redox reaction to the standard electrode potential and activities of the chemical species undergoing oxidation and reduction.

We can also define pe^0 as the effective equilibrium constants for redox couples that corresponds to the value of E_h^0 to represent the electron activity when all species other than the electron are at unit activity.

$$pe^0 = \frac{F}{2.3RT} E_h^0 \left(= \frac{1}{n} \, log \, K \right) \tag{4.26}$$

The values of pe^0 are listed in Table 4.4 for major aquatic redox couples.

Example 4.8: Equilibrium Redox Speciation

From the measured redox potential of the dominant redox couple in a given aquatic system, one can calculate the equilibrium redox speciation of minor species. For the following reaction involving iron in a rock, the measured electrode potential was 260 mV.

$$Fe^{3+} + e^- \rightleftharpoons Fe^{2+}$$

Determine the ratio of $[Fe^{2+}]$ to $[Fe^{3+}]$ at equilibrium with the oxygen of the atmosphere and water at neutral pH.

Solution:

The *pe* of the oxygen/water couple has been calculated in Example 4.7.

Iron in a rock

$$Fe^{3+} + e^- \rightleftharpoons Fe^{2+}$$

$$pe = pe^\circ - log \frac{[reductants]}{[oxidatants]} = pe^\circ - log \frac{[Fe^{2+}]}{[Fe^{3+}]}$$

$$pe^\circ = \frac{1}{n} \, log \, K = \frac{1}{1} \, log \left(\frac{[Fe^{2+}]}{[Fe^{3+}]} \frac{1}{e^-} \right) = 13.0$$

(continued)

Example 4.8 (continued)

The measured electrode potential was 260 mV.

$$E_h = \frac{2.3RT}{F} pe = 59.2\, pe\ (mV) = 260\ \text{mV}$$

$$pe = \frac{260}{59.2} = 4.4$$

$$4.4 = 13.0 - \ log\ \frac{[Fe^{2+}]}{[Fe^{3+}]}$$

$$\text{Therefore,}\quad \frac{[Fe^{2+}]}{[Fe^{3+}]} = 10^{8.6}$$

4.3.3.4 Measurements of Oxidation Potential

As oxidation and reduction processes involve transfer of electrons, the charge separation resulting from electron movements can be quantified and measured in voltage, i.e., electric potential. The oxidation potential, E_h, is the measure used for such quantification. It can be determined experimentally by using electrodes, which are electrical conductors through which a current enters or leaves an electrolyte. An electrolyte is any conducting medium such as water, acid solution, molten salt, in which the flow of current takes place due to the movement of ions and electrons.

Consider the oxidation-reduction process, $Zn + Cu^{2+} \rightarrow Zn^{2+} + Cu$. This represents the sum of two half reactions, occurring when a zinc electrode is immersed in an electrolyte and a copper electrode is placed in the electrolyte and connected through a circuit. In the solution, zinc will be oxidized liberating two electrons with the formation of Zn^{2+} ions.

$$Zn \rightarrow Zn^{2+} + 2e^- \qquad \text{(a)}$$

In the same way, copper will also be oxidized to form copper^{2+} ions

$$Cu \rightarrow Cu^{2+} + 2e^- \qquad \text{(b)}$$

The tendency for the first reaction to occur is greater than that for the second reaction and the second reaction is actually driven backwards:

$$Cu^{2+} + 2e^- \rightarrow Cu \qquad \text{(c)}$$

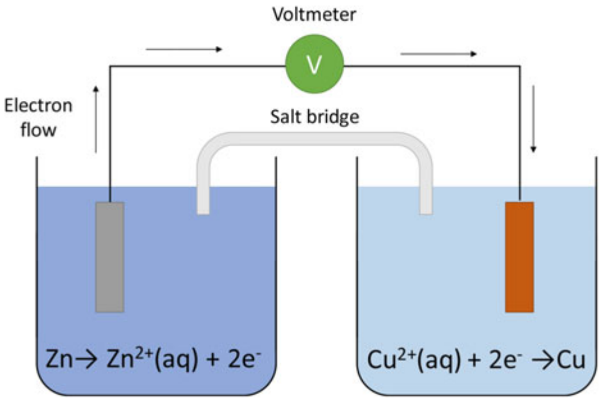


Fig. 4.8 Example of a zinc-copper galvanic cell

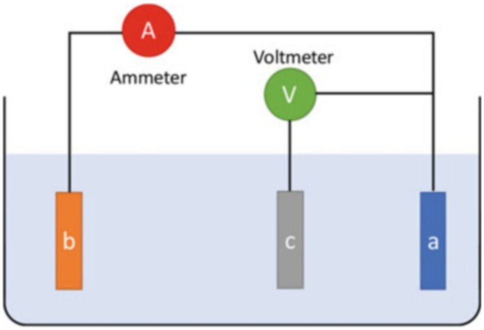


Fig. 4.9 Three-electrode setup for measurement of electrode potential in a galvanic cell, where *a* is working electrode, *b* is counter electrode, and *c* is reference electrode)

The fact that the electrons flow from the zinc electrode to the copper electrode indicates that the tendency for the reaction (a) to occur is stronger than that for the reaction (c), and the two dominant reactions in the electrolyte become zinc oxidation and copper reduction.

If we form a circuit, like what is depicted in Fig. 4.8, the electrons flow from the zinc electrode to the copper electrode. In other words, in the outer circuit, electrons liberated from the zinc electrode travel into the copper electrode where they react with Cu^{2+} ions to form metallic copper. The compartments are connected by a solution to allow electricity conduction but prevent bulk mixing of the electrolytes. The magnitude of the electromotive force (*emf*) developed in the circuit can be measured as the relative tendencies of the two processes as potential difference.

The potential difference between the electrodes is determined by measuring the relative potential of the each half-reaction and comparing them. The potential for a half reaction is a measure of the tendency for a given half-reaction to occur. It is also an indication of how far the reaction mixture is from equilibrium. If the measured value is a negative voltage, the reaction takes place voluntarily. If positive, the reaction requires outside energy. The measured oxidation potential is a relative quantity, not an absolute one, defined relative to a reference electrode. A reference electrode is an electrode with a stable and well-known electrode potential.

Table 4.5 Standard electrode potentials of half-reactions at 25 °C vs. SHE

Reducers	Oxidizers	Half-reaction	Electrode potential (Volts)
Lithium	Li^+	$Li^+ + e^- \rightarrow Li$	−3.03
Potassium	K^+	$K^+ + e^- \rightarrow K$	−2.92
Calcium	Ca^{2+}	$Ca^{2+} + 2e^- \rightarrow Ca$	−2.87
Sodium	Na^+	$Na^+ + e^- \rightarrow Na$	−2.71
Magnesium	Mg^{2+}	$Mg^{2+} + 2e^- \rightarrow Mg$	−2.38
Aluminum	Al^{3+}	$Al^{3+} + 3e^- \rightarrow Al$	−1.71
Manganese	Mn^{2+}	$Mn^{2+} + 2e^- \rightarrow Mn$	−1.04
Zinc	Zn^{2+}	$Zn^{2+} + 2e^- \rightarrow Zn$	−0.76
Iron	Fe^{2+}	$Fe^{2+} + 2e^- \rightarrow Fe$	−0.41
Cobalt	Co^{2+}	$Co^{2+} + 2e^- \rightarrow Co$	−0.28
Nickel	Ni^{2+}	$Ni^{2+} + 2e^- \rightarrow Ni$	−0.23
Lead	Pb^{2+}	$Pb^{2+} + 2e^- \rightarrow Pb$	−0.13
Iron	Fe^{3+}	$Fel^{3+} + 3e^- \rightarrow Fe$	−0.036
Lead (Pb)	Pb^{2+}	$Pb^{2+} + 2e^- \rightarrow Pb$	−0.13
H_2	$2H^+$	$2H^+ + 2e^- \rightarrow H_2$	0
Copper	Cu^{2+}	$Cu^{2+} + 2e^- \rightarrow Cu$	+0.34
OH^-	O_2	$O_2 + 2H_2O + 4e^- \rightarrow 4OH^-$	0.40
Copper	Cu+	$Cu^+ + e^- \rightarrow cu$	0.52
Silver	Ag^+	$Ag^+ + e^- \rightarrow ag$	+0.80
Mercury	Hg^{2+}	$Hg^{2+} + 2e^- \rightarrow Hg$	+0.85
OH^-	H_2O_2	$H_2O_2 + 2e^- \rightarrow 2OH^-$	0.88
Platinum	Pt2+	$Pt^{2+} + 2e^- \rightarrow Pt$	+1.20
H_2O	O_2	$O_2 + 4H^+ + 4e^- \rightarrow 2H_2O$	+1.23
$2Cl^-$	Cl_2	$Cl_2\ (g) + 2e^- \rightarrow 2Cl^-$	+1.36
Gold	Au^{3+}	$Au^{3+} + 3e^- \rightarrow Au$	+1.52
H_2O	H_2O_2	$H_2O_2 + 2H^+ + 2e^- \rightarrow 2H_2O$	+1.78
$2F^-$	F_2	$F_2 + 2e^- \rightarrow 2F^-$	+2.87

An example of using electrodes for oxidation potential measurement is shown in Fig. 4.9. The electrodes are connected by an electrolyte. The electrode *a* is called the sensing electrode or the working electrode. This is where the half-reaction of interest occurs. Depending on the type of the reaction, the working electrode is a cathode (if the reaction is a reduction) or anode (if the reaction is an oxidation). The electrode *b* is the counter electrode or the auxiliary electrode. This is used as part of a galvanic cell (an electrochemical cell that derives electrical energy from spontaneous redox reactions taking place within the cell) to allow electrons flow between the two electrodes. If the working electrode is an anode, the counter electrode is a cathode. The electrode *c* is the reference electrode with the known electrode potential whose use is to measure the relative potential of the working electrode.

An example of a reference electrode is the standard hydrogen electrode (SHE). The hydrogen electrode is based on the following half redox reactions:

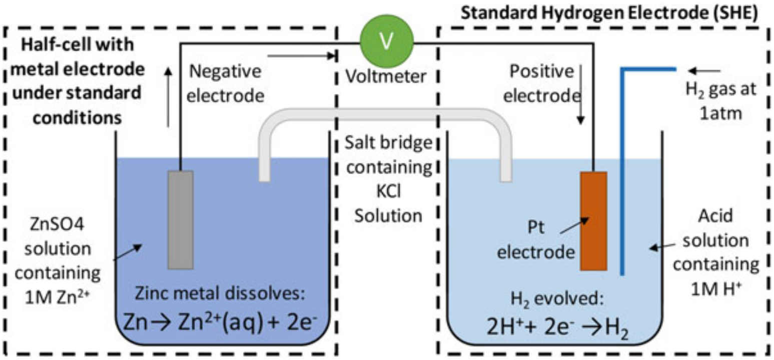


Fig. 4.10 An example of using standard hydrogen electrode to measure the standard oxidation potential

$$2H^+(aq) + 2e^- \rightarrow H_2(g)$$

$$H_2 \rightarrow 2H^+ + 2e^-$$

Under these reactions, a sufficiently high current can be passed through the electrode. The SHE provides the basis of the thermodynamic scale of oxidation-reduction potentials with its absolute electrode potential estimated at 4.44 V (at 25 °C).

To form a basis for comparison with other electrode reactions, the standard electrode potential (E^0) of the reference electrode is set to be zero volts at all temperatures. The standard potential refers to the potential differences for reactions that take place at 25 °C with all substances at unit activities (i.e., gases at 1 atm pressure and dissolved substances at 1 mol/liter). In the case of using SHE as reference electrode, the standard potentials of other elements in comparison with that of the SHE at the same temperature are shown in Table 4.5. Please note that, for the half-reactions shown in the table, when the measured potential is negative, the oxidizer is stable. When the measured potential is positive, the reducer is stable.

For the two half-reactions depicted in Fig. 4.8, the oxidation potential of each of the half-reactions can be measured by using a reference electrode (as shown in Fig. 4.9). The half-cell represented in Fig. 4.10 is for the reaction $Zn \rightarrow Zn^{2+} + 2e^-$ with SHE as a reference electrode. If we measure the oxidation potential for the reaction, $Zn \rightarrow Zn^{2+} + 2\,e^-$, the value represents the tendency of the process to occur with respect to the reference electrode (SHE). Under a 1 mole solution of Zn^{2+} ions, the measured potential was −0.76 volt as shown in Table 4.5 (this is the standard oxidation potential, Eh^0). The fact that the zinc electrode is negative with respect to the hydrogen electrode means that electrons flow in the outer circuit from the zinc electrode to the hydrogen electrode. Therefore, there is a greater tendency for the occurrence of oxidation in zinc.

We can also set up the half-cell for the Cu/Cu^{2+} system with a reference electrode. In this case, the measured potential is (+0.34 V) which is less than for the Zn/Zn^{2+}

(−0.76 V). This means that the reaction, $Zn \rightarrow Zn^{2+} + 2e^-$, has a stronger tendency to occur than $Cu \rightarrow Cu^{2+} + 2e^-$. If two half-cells are connected together, the second reaction is driven backwards by the first one. The potential difference of the cell, when the Zn^{2+} and Cu^{2+} concentrations are both 1 mole, is equal to the difference between the oxidation potentials measured through setup in Fig. 4.10, i.e., −0.76 – (0.34) = −1.10 volts.

Example 4.9: Galvanic Cell Potential

Determine the open circuit potential of the following electrochemical reaction by using standard potentials and molarity for ion concentrations.

$H_2O_2 + Ni \rightarrow H_2O + Ni^{2+}$

Solution:

The half reactions are:

(Anode) $Ni \rightarrow Ni^{2+} + 2e^-$ (with standard potential: −0.23 V vs. SHE, from Table 4.5)

(Cathode) $H_2O_2 + 2H^+ + 2e^- \rightarrow 2H_2O$ (with standard potential: 1.78 V vs. SHE, from Table 4.5)

Total reaction: $H_2O_2 + 2H^+ + Ni \rightarrow 2H_2O + Ni^{2+}$

Cell potential: $E_{cathode} - E_{anode} = 1.78 - (-0.23) = 2.01$ V

4.3.4 Complexation Reactions

Complexation (or, complex formation) is a reversible chemical reaction between two dissolved species. It can be considered an example of acid-base reaction. Through a complexation reaction, a new soluble species, called a complex (sometime called, coordination complex or coordination compound) is formed.

A coordination complex is a combination of a central atom or ion and a surrounding array of bound molecules or ions. The central atom or ion is usually metallic and is called the coordination center. The surrounding array of bound molecules or ions are called ligands or complexing agents.

$$coordination\ center + ligand \leftrightarrow complex$$

A ligand is a molecule or anion containing at least one pair of electrons to donate to a metal atom/ion. A ligand can be inorganic or organic. Inorganic ligands include common anions in the natural water systems such as OH^-, Cl^-, CO_3^{2-}, SO_4^{2-}, PO_4^{3-}. Organic ligands include synthetic organic complexing agents such as EDTA (ethylene diamine tetra acetic acid) or molecules associated with natural humic substances. Humic substances are organic polyelectrolyte macromolecules derived from decayed vegetable matters or other organic substances. They are nutrients for plants and increase the ability of soil to retain water (more descriptions of humic substances are in Sect. 10.2.2.2).

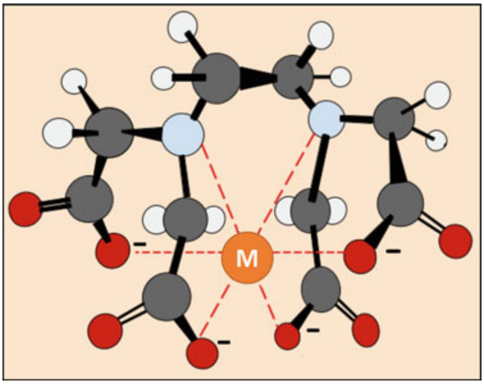


Fig. 4.11 Chemical structure of EDTA, having more than one site for bonding which enables multiple bonds with a single metal

One of the applications of complexation reaction is the use of chelating agent. Chelating agents are organic ligands that can form several bonds to a single metal ion. Thus with a chelating agent, there is more than one site for bonding enabling multiple bonds with a single metal. EDTA is an important example of a chelating agent. It can form four or six bonds with a metal ion increasing the stability of the EDTA complex. EDTA is frequently used in soaps and detergents, and as a stabilizing agent in the food industry or as an anticoagulant for stored blood in blood banks. EDTA is also widely used for decontamination of the radioactively contaminated surfaces in nuclear power plants (Fig. 4.11).

Another important role of complexation is the determination of the speciation of aqueous species in surface water or groundwater. Complexation affects solubility and also controls biological uptake of dissolved chemical species (thus affecting toxic effect of a chemical) and sorption of metals from solution. Surface properties of solids can be modified through complexation.

In the case of metallic elements, occupancy of electrons in their electron orbitals control the tendency for complexation. For example, the alkali metals (e.g., Li, Na, K) have only one electron in the outermost shell. Therefore the energies needed to remove an electron from the metal is the lowest and these metals react violently with water and produce hydroxides as strong bases. In contrast, most transition metals and the actinides have variable valence (e.g., iron (Fe) with OS +2 and + 3) and their complexation characteristics varies with valence state changes. Such changes in complexation behavior directly affect the sorption properties or solubility of metals in water. This is further discussed in Sect. 11.4.

4.3.5 Sorption

Sorption refers to the attachment of chemical species on the surfaces of minerals or other solids. The attachment can be through mechanisms such as adsorption, absorption, or surface precipitation.

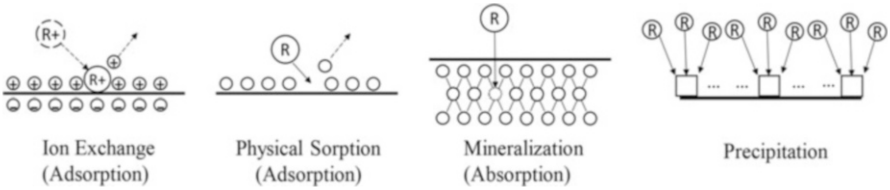


Fig. 4.12 Schematic representations of possible sorption mechanisms as a result of interaction between a solid surface and a species ('R' represents the radionuclide species)

Adsorption is a surface process in which the attachment is through weak van der Waals force (this is called physical adsorption) or through attraction of opposite charges (this is called electrostatic adsorption) or through stronger covalent or ionic chemical bonding. Due to the presence of various cations and anions in natural water systems, adsorption at the surface is competitive as different ions in the solution compete for adsorption at a limited number of sites on the substrate. Such competition can be described as an ion exchange process, in which one adsorbed ion on the surface trades places with another ion in the solution. Ion exchange becomes important with high contents of clays and organic matter in the soil system.

Absorption is a bulk process in which atoms or molecules cross the surface and enter the volume of the material through mass transport (e.g., diffusion) in pores or through defects. Absorption is typically accompanied by adsorption.

Surface precipitation refers to the formation of the product of precipitation on the surface. The product of precipitation becomes the sorption layer simply through transformation of aqueous components into solids attached to surface. The sorption layer could be made up of a crystalline material in which two or more elements or compounds are mixed sharing a common lattice. Mixture of two crystalline solids that coexist sharing a common lattice is called solid-solution.

Differences among the mechanisms of sorption are shown graphically in Fig. 4.12. The term sorption is commonly used as a general term if the detailed mechanism of uptake of a species from solution is unknown.

Sorption of a species depends on a range of chemical conditions such as pH, E_h, species concentration, composition of major ions, and site specific parameters like the geometric restrictions imposed by the surfaces of solid including the available reacting surfaces. This implies the influence of oxidation and reduction on sorption. Sorption plays an important role in controlling the contaminant transport rate in groundwater.

4.3.6 Biodegradation of Organic Matter

Biodegradation refers to chemical disintegration of organic materials by the action of microbial organisms. Through biodegradation, complex molecules are broken into simpler molecules, such as water, carbon dioxide, and soil organic matter called humus which can be readily utilized by microorganisms as their energy source.

Hydrolysis is an important step in biodegradation as organic compounds become more susceptible to biodegradation through the step. In hydrolysis an organic molecule reacts with water and is broken down through splitting of a bond and addition of the hydrogen cation and the hydroxide anion of water. After hydrolysis, organic material can be degraded aerobically (with oxygen) or anaerobically (without oxygen), mediated by microorganism. Microorganisms use the chemical reactions to derive energy for metabolic processes or as source materials for biosynthesis.

Consider the following example.

$$CH_2O + H_2O \rightarrow CO_2(g) + 4H^+ + 4e^-; \quad pe^0 = 1.20$$

As most organic materials contain carbon in a more reduced state than the one in carbon dioxide (which is in +IV oxidation state), when organic materials are degraded through oxidation, energy is released. The released energy is used by microbes for their metabolic processes. In the reactions, the microbes play the role of catalyst by accelerating the process. Those microbes that use the energy from the oxidation of organic compounds are called aerobes. Thus the aerobes use oxygen to drive metabolic processes as oxidation reactions. The microorganisms that derive energy from reduction are called the anaerobes. The anaerobes are also able to get energy by oxidation. However, when oxygen is depleted, anaerobes derive energy from the reduction of oxidized carbon compounds or carbon atoms.

Factors influencing the rate of microbially mediated chemical reactions include: water temperature, pH, number and species of microorganisms present, oxidation potential, concentration of the contaminant, and the presence of nutrients or microbe toxicants.

4.3.7 Role of Temperature

Temperature is known to have large influence on chemical reactions: Increase in temperature increases chemical reaction rates. Therefore, large variations in system temperature can have a strong impact on chemical reactions. This role of temperature is explicitly represented by an empirical relationship called, the Arrhenius law. According to the Arrhenius law, the reaction rate constant is a function of temperature as shown below.

$$k = Ae^{(-E_a/RT)} \tag{4.27}$$

where

A = the pre-exponential factor, a constant that is independent of temperature for a particular reaction

E_a = the activation energy, treated as a constant for a particular reaction

R = the ideal gas constant, 8.314 J mol^{-1} K^{-1}
T = the temperature in K (RT ~ 2.5 kJ/mol at 298 K)

4.4 Conclusion

The chemical processes caused by radioactive particles as part of their interactions with matter determine the outcome of those interactions in the biological system. The chemical processes a material go through in a given physical and chemical environment of a system determine the fate of the material in the system. This chapter provided an overview of basic chemistry as a basis for the discussion of biological effects of radiation as well as the behaviors and fate of materials used in nuclear waste management. The chemical reactions also determine the concentrations of radionuclides in various environmental media and the migration characteristics of radionuclides through groundwater. Details of these chemical reactions in specific materials or environments will be discussed in Chap. 9 (material corrosion and radionuclide leaching) and Chap. 11 (sorption, solubility, and migration of radionuclides in groundwater). Careful exercise of the understanding of chemistry will be critically important in analyzing and demonstrating safety in nuclear waste management, in particular, during the step of final disposal of nuclear waste.

Homework

Problem 4.1: Write the electronic structure of neptunium and plutonium and explain possible implications of such structure in chemical reactions.

Problem 4.2: For the following chemical reaction, determine if the reaction is spontaneous under the standard condition. Estimate the equilibrium constant for the reaction and explain the result.

$$\mathrm{Fe(s)} \rightarrow \mathrm{Fe^{2+}(aq)} + 2\mathrm{e^-}$$

Problem 4.3: For the groundwater within the basalt rock medium, the following compositions (mg/l) were observed at 25 °C.

$$\mathrm{Ca^{2+}}\,1.3;\mathrm{Mg^{2+}}\,0.04;\mathrm{Na^{+}}\,250;\mathrm{K^{+}}\,1.9;\mathrm{Cl^{-}}\,148;\mathrm{SO_4^{2-}}\,108;\mathrm{CO_3^{2-}}\,46;\mathrm{SiO_2}\;145;\mathrm{F^{-}}\,37$$

Calculate the pH value of the groundwater.

Problem 4.4: Determine the oxidation number of the elements in each of the following compounds.

(a) $Fe(OH)_3$, Fe_2O_3, Fe_3O_4
(b) I_2, I^-, IO_3^-, IO_4^-
(c) $Tc, TcO_4^-, TcO_2, TcO^{2+}$
(d) $U_3O_8, UO_2, UO_2(OH)_2, UO_2^{2+}$
(e) NpO_2, NpO_2^+, NpO_2OH
(f) $PuO_2, PuO_2^+, PuO_2^{2+}, PuOH^{2+}, PuO_2OH^+, Pu(OH)_2^{2+}$

Problem 4.5: For the copper reduction reaction, $Cu^{2+} + 2e^- \rightarrow Cu$, at 25C and 1 atm, determine the following.

(a) The Gibbs free energy change
(b) The equilibrium constant
(c) The copper ion (Cu^{+2}) concentration

Further Reading

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Chapter 5
Science of Risk and Radiation Protection

Abstract Providing appropriate level of safety in nuclear waste management requires understanding of biological effects of ionizing radiation. Starting with discussing radiation interactions in biological system, this chapter describes the process of energy transfer from ionizing radiation to biological targets and how such energy transfer leads to critical effects of concern such as cell death, mutation, and cancer. The chapter also discusses how the concept of "dose" is defined to characterize the risk of radiation among the exposed individuals.

Keywords Ionization and free radicals · DNA damage and repair · Dose · Cancer risk · Radiation protection

Most physical destructive agents – heat, noise, and physical objects – can be sensed by human body allowing humans to take protective measures. In contrast, human body cannot sense the presence of ionizing radiation unless at very high dose levels. Therefore, intentional measures of protection must be in place when people handle radioactive materials. Providing appropriate level of safety is foremost important in all phases of nuclear waste management and such effort requires understanding of radiation-related health risks. We will address the following questions in this chapter.

"How does radiation affect human body?"

"How do we utilize the knowledge of radiation health effect to protect humans?"

5.1 Biological Effects of Radiation

5.1.1 Interaction Mechanisms in a Biological System

Ionizing radiation is any radiation with enough energy to eject an orbital electron from an atom (as discussed in Sect. 3.2.1). The ejected electron is rapidly captured by another molecule to yield a negative ion. Thus an ionization event results in

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formation of ion pairs, the negative ion with the ejected electron and the positively charged atom that lost one electron. The energy of ionizing radiation may also be given to the medium through the process of excitation. Although excitation is not as damaging as ionizations, excitation can lead to breakage of chemical bonds.

Ionization and excitation can directly damage biologically important molecules in the cell. This is called the direct action of radiation. In this case, the energy of ionizing radiation is directly deposited in the target molecule without involving any intermediate steps. The resulting ionization and excitation right away initiate the chain of events that leads to a biological change.

In an alternative way of causing biological damage, the formation of ion pairs may not directly damage the target molecules but occurs in cellular water. The ion pairs formed in water then form free radicals which in turn initiate a chain of chemical reactions in the targets leading to ultimate biological damage. This is the indirect action of radiation. Thus, free radicals are intermediaries between ion-pairs and chemical products. A free radical is a free (not combined) atom or molecule carrying an unpaired orbital electron in the outer shell, i.e., an electrically neutral atom or molecule that has an unpaired electron. Due to the presence of an unpaired electron, free radical is chemically very reactive. Free radicals can diffuse to reach and induce chemical changes at critical sites in biological systems. Therefore, in the case of indirect action, radiation damage occurs through the actions of free radicals. The toxic effects of a number of chemical carcinogens and mutagens, as well as of certain tumor promoting agents, are also the outcome of free radicals activities and interactions. Because the process of free radical diffusion controls the damage to the targets, if the diffusing free radical species are scavenged before reaching the targets, damages by the indirect action of radiation can be reduced.

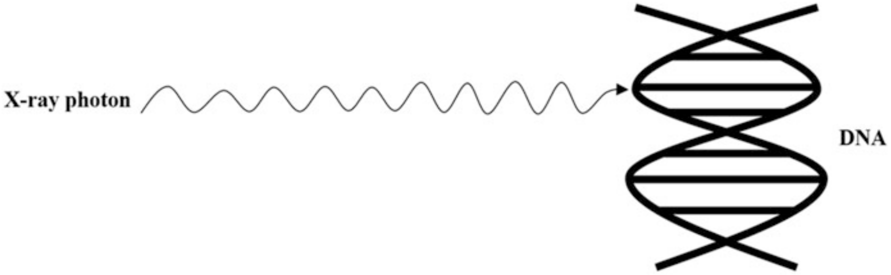
Direct effects are thought to cause about 20% of the damage, while indirect effects account for the remainder (80% of the target is water). Importance of direct effect is much more pronounced when high linear energy transfer (LET) radiation, such as alpha particles or neutrons are involved (Fig. 5.1).

5.1.2 Stages in Radiation Interaction with Biological Systems

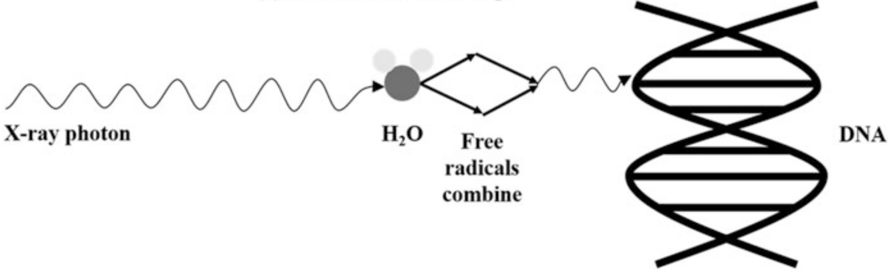
When radiation interacts with important biological molecules, i.e., the critical targets in the cells, the atoms of the target itself may be ionized or excited, initiating the chain of events that leads to a biological change. The sequence of events as radiation passes through a biological system is as follows.

The first stage of the events is the **physical stage**. This stage occurs within 10^{-15} s of the interaction and includes the occurrences of excitations and ionizations of the target molecules through the ejection of electrons. The energy of the incoming radiation is transferred to the biological molecules or the interacting medium through ionizations (or excitation).

Consider the case of radiation interaction with water: An electron is ejected from a water molecule producing a free electron and a positively charged molecule as,



(a) Direct action or damage



(b) Indirect action or damage

Fig. 5.1 Schematic of direct and indirect biological effect of radiation

$$H_2O + radiation \rightarrow H_2O^+ + e^-$$

The released electron is most likely to be captured by another water molecule converting it into a negative ion as following:

$$e^- + H_2O \rightarrow H_2O^-$$

Therefore, the products of ionization of water are H_2O^+, H_2O^- and free electrons.

The next **pre-chemical stage** lasting from 10^{-15} to 10^{-11} s after the initial interactions follows the physical stage. In this stage, the ion pairs produced from the physical stage last only a very short time (less than 10^{-10} sec) and undergo reactions to form free radicals.

For example, the unstable positive ion (H_2O^+) and a negative ion (H_2O^-) formed from the physical stage decompose into an ion and a free radical as follows

$$H_2O^+ \rightarrow H^+ + OH^0$$

$$H_2O^- \rightarrow H^0 + OH^-$$

A free radical is a free atom or molecule, thus not combined, and carries at least one unpaired orbital electron in the outer most shell. In an atom or molecule with an even number of electrons, the electrons spin around its own axis either clockwise or counterclockwise resulting in paired spins and stability. In contrast, presence of unpaired electron with unpaired spin in the outer most shell of a free radical gives a high degree of chemical reactivity. The free radical's unpaired electron has a strong tendency to pair with a similar one in another radical. The odd unpaired electron may also be eliminated by the atom in an electron-transfer reaction. In the end, the products of the pre-chemical stage are free radicals, electrons, and other products of free radical interactions. In the case of water as target molecule, the chemical species produced include OH^0, H^0, H_2, H_3O^+, and e^-_{aq}. Here, e^-_{aq} is called an aqueous electron in water which is formed after an electron has lost most of its kinetic energy and can no longer produce any further ionization in the medium.

The newly formed free radicals will further interact with each other (assuming pure water) to form H_2, H_2O, and H_2O_2. This also indicates that there may be recombination within the track of free radical movements either to produce new molecules such as hydrogen peroxide or to go back to the original molecule, i.e., water. Reunion of the original products are called recombination while the joining of formerly unassociated radicals is called radical combination.

$$H^0 + OH^0 \rightarrow H_2O$$

$$H^0 + H^0 \rightarrow H_2$$

$$OH^0 + OH^0 \rightarrow H_2O_2$$

In a biological system, production of a new molecule is considered a damage. Therefore, there may be no damage in the case of recombination. However, formation of a new molecule through radical formation is a damage and may markedly influence the system. For example, a small alteration in a single molecule may result in a change in genetic information which could affect an entire organism.

The H_2O_2 or H_2 formed from above reactions can also be eliminated by the reactions with other free radicals such as,

$$H^0 + H_2O_2 \rightarrow OH^0 + H_2O$$

$$OH^0 + H_2 \rightarrow H^0 + H_2O$$

In an actual biological system, a variety of molecules and ions are present. Therefore, the free radicals can also react with these molecules and ions, such as oxygen. Oxygen is extremely reactive and combines with a free radical to form the peroxide radical. The peroxide radical is a very reactive form and represents a new damaging molecule to the system.

The third stage is the **chemical stage** lasting from 10^{-11} to 10^{-3} s. In this stage, all molecular products of ionization including free radicals diffuse around and interact with biological molecules in the cell. The average lifetime of a radical is about 10^{-6} s and the free radicals move around during this period and find targets. Most radical-induced reactions would be complete by 10^{-3} s.

Here the average lifetime is the expected lifetime of the free radical and is related to the half-life as,

$$average\ life = \frac{half\ life}{ln\,2} \tag{5.1}$$

where the half-life is the time required for the half of the free radicals to become stable. The half-life of free radicals can be as short as 10^{-9} s (in the case of hydroxyl free radical (OH)) and could be up to several days (in the case of a certain *radical* present in tobacco smoke).

During its lifetime, a free radical will move to various locations of the target, diffusing about 5 nm distance on average and create damages to biological molecules. Efforts are made by the affected cell to repair the damages created by free radicals through simple chemical restitution. This chemical restitution may involve radical exchange but does not involve enzymes (enzymes are any complex proteins working to catalyze specific biochemical reactions). In this radical exchange, the affected radical compound interacts with a molecule and restores itself while converting the other molecule to a radical compound.

The last stage in radiation interaction with biological systems is the **biological** (biochemical and physiological) **stage**. In this stage, enzymatic repair processes take place to repair the damage. This stage (*biochemical stage*) may take ~minutes. If the repair is successful, there is no further damage. If the repair is not successful, the damage as the change in the biochemical structure is fixed. The fixed change may be expressed in the progeny through cell proliferation. The time scale involved (*physiological stage*) in the expression of the biochemical damage in the biological systems can range from hours to days or even years depending on the end effect. For example, effects on cell division may be manifested within several hours. Effects as cancer induction or genetic effect take years. Progression of such effect continue at the sub-cellular, cellular and tissue levels leading to the ultimate outcome, such as cell death or cancer.

In the case of cancer, from the initial damage till the manifestation of solid tumor as the outcome, there exists a time gap. Such time gap is called latency or the latent period and the latent period is minimum 10 to 15 years and on average 25 years (or more). For leukemia, the latent period is minimum 2 to 5 years but on average about 10 years. Fig. 5.2 shows the summary of the stages of radiation interactions with biological systems (Table 5.1).

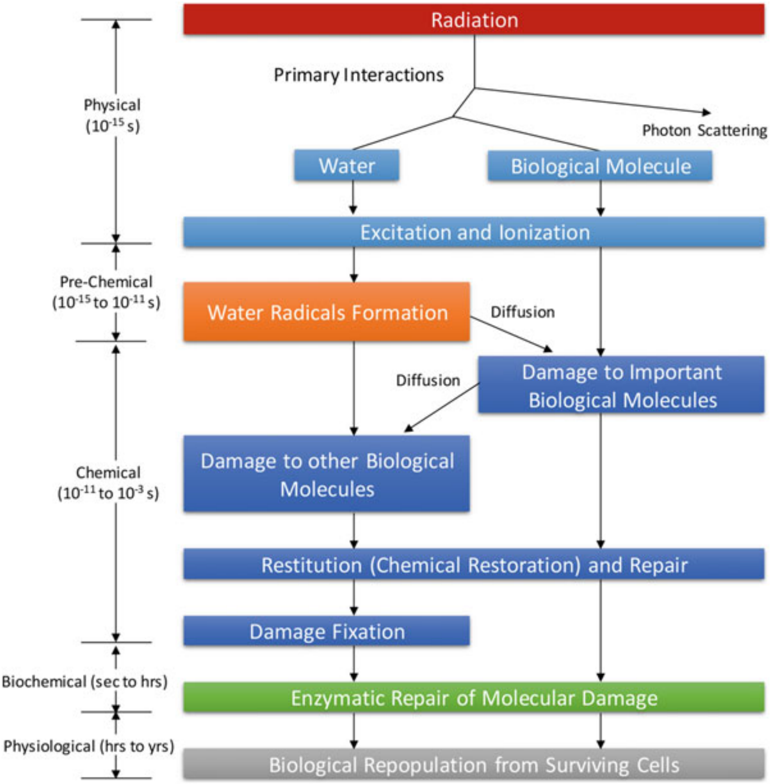


Fig. 5.2 The processes involved from energy transfer to final biological damage

Table 5.1 Stages of radiation interaction with biological systems

Stage	Summary	Time scale
Physical	Energy transfer through ionization and excitation	About 10^{-15} sec
Pre-chemical	Free radical formations and interactions	About $10^{-15} \sim 10^{-11}$ sec
Chemical	Free radical diffusion and interactions with bio-molecules formation, chemical restitutions and repairs	10^{-11} to 10^{-3} sec The average lifetime of a radical is about 10^{-6} s diffusing about 5 nm distance on average
Biological – biochemical	Enzymatic repair of molecular damage, fixation of damage	Sec – hrs
Biological – physiological	Cell killing (hours to days) Oncogenic (years) Mutagenic (years – many generations)	Hrs – yrs

Definition: Latency for Cancer Development

- Latency (or the latent period) is the time gap between the irradiation of the subject and

the ultimate appearance of the tumor.

- For most **solid tumors**, latent period is considerably longer:
  - **Min 10-15 yrs**
  - **Mean 25 yrs or longer** (as long as 35-40 yrs)
- For **leukemia**, latent period is relatively short:
  - **Min 2-5 yrs**
  - **Mean 10 yrs**
- Latent period is possibly related to age at irradiation.
- The existence of a long latency indicates that a number of changes are necessary for ultimate tumor development.

5.1.3 Interactions of Radiation with Biological Targets

As described, the immediate result of the interaction of any radiation with matter is deposition of energy. If the deposited energy is large, the events of ionization tend to be very closely located. If the deposited energy is small, the resulting ionization events are sparsely located. The extent of these ionizing events and the resulting outcome depend on the physical characteristics of the radiation as well as the targets of interactions.

The target of radiation interactions in a biological system, in a simplistic sense, is a cell. In a living organism, a cell is the basic structural, functional, and biological unit of a biological system. However, as the cell includes various biomolecules within the cell structure, these biomolecules are the actual targets of radiation interactions. Therefor the size of these biomolecules and their availability within the tracks of ionization (and excitation) directly affect biological effects of radiation.

5.1.3.1 Interactions of Different Particles in Tissues

Heavy charged particles such as protons, α particles, or heavy ions interact directly with the target atoms or molecules through Coulombic (charge v. charge) interactions. Also as high LET radiation, these particles create a very dense population of ion pairs within the almost straight particle trajectory. As the rate of energy deposition is high, the distance they travel before losing all kinetic energy in tissue is also quite short. For example, 1 MeV α particles travel only a few tens of micrometers in tissue. Thus energy deposition in the target is highly localized. For α particle, the average distance between ionization in tissue ranges between a few Å and up to 15 Å.

In contrast, low LET radiation (i.e., electrons, x-rays, and gamma rays) has low density of ionization and the average distance between ionization is longer than several hundred Å. Table 3.9 in Sect. 3.2.2 shows the comparison of LET and the range in water of major charged particles. Note that the range in water is expected to be close to that in tissue. Figure 5.3 shows the graphical representation of the

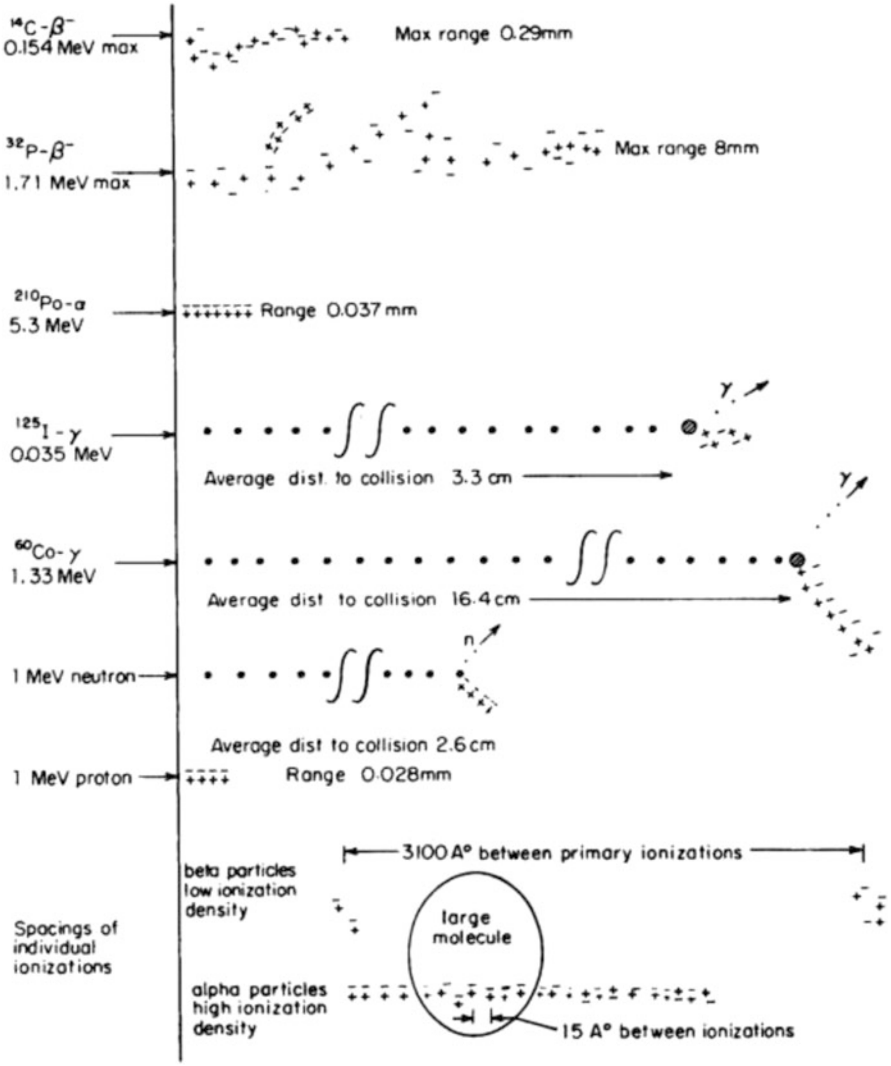


Fig. 5.3 Graphical representation of the tracks of ionization and the range in tissue for various ionizing radiation. (Source: Shapiro 1972)

ionization tracks and range (or average distance between interactions) of different types of ionizing radiation in tissue.

Electrons, compared to heavy charged particles, lose the kinetic energy at much lower rate and travels much longer distance before being stopped, while going through the Coulombic interactions. Thus electrons are more penetrating than heavy charged particles and the distance between ionization for electron is much longer (~3000 Å between ionization). Thus the energy of electrons is distributed over a much longer distance and the resulting ionization density is much smaller compared to the one by a heavy charged particle. Electrons also easily change the direction of their movement and can lose energy by emitting bremsstrahlung photons which may escape the region of immediate vicinity of the tissue. In the case of a high energy electron, its interaction in the target can also produce a secondary electron with enough energy to have its own track creating further ionizations. Such secondary electron is called delta ray (Delta rays are also shown in Fig. 5.3 as the secondary tracks of ionization for the high energy electrons from ^{32}P disintegration, the second figure from the top).

In the case of photons, their interactions with matter depend on the atomic number of the target and the energy of the photons. The atoms in tissue are mainly low Z elements such as hydrogen, carbon, and oxygen. For a typical photon which has the energy of a few hundred keV or about one MeV, the interaction with these targets is mostly through Compton scattering. The photons that are scattered can also interact further before being absorbed or escape the tissue. The average distance between ionization in tissue is 200–300 Å for photons. Thus the energy deposition is not localized. While the intensity of photons decreases exponentially with depth of the target, photons can also reach deep into tissues.

For neutrons, the interaction depends on the types and number of target atoms and the energy of the neutrons. Since living tissue has a high density of hydrogen atoms (in water), interaction with hydrogen becomes most important. For fast neutrons, the interaction with hydrogen results in elastic scattering releasing recoil protons. The emitted protons, as high LET radiation, will result in high density ionization track. The recoil protons deposit energy as heavy charged particle. For epithermal and thermal neutrons, there is a high probability of neutron capture. In this case, the principal capture reactions in tissue are neutron capture by hydrogen (^{1}H(n,p)^{2}H) or by nitrogen (^{14}N(n,p)^{14}C). The emitted gamma ray, as a penetrating radiation, could deposit a fraction of energy while traveling through the body. On average, the distance between ionization for neutrons is few centimeters.

Initial Interactions of Different Particles in Biological System

Heavy charged particles (α particles and protons):

The average distance between ionization events is a few~15 Å. These particles interact directly with the tissue, losing energy as described by the

(continued)

Bethe-Bloch formula (Eq. 3.29), primarily colliding with electrons. The density of ionization is very high and varies with the square of the charge on the particle for a given speed. It reaches a peak near the end of the range following so-called the Bragg curve. In tissue, 1 MeV α particles travel only a few tens of micrometers and thus the energy deposition is very localized.

Electrons

The average distance between ionization events is ~3000 Å. The electron's energy is distributed within a much greater volume and the resulting ionization density is much smaller than that caused by a heavy charged particle.

Photons

The average distance between ionization events is 200-300 Å. Tissue contains mainly low Z elements (C, H, and O) and Compton scattering is the most important interaction process. The scattered photons may interact further by a second Compton scattering or by photoelectric absorption, depending on the energy. Overall energy deposition is not localized. It varies approximately exponentially with depth and can reach deep into tissue.

Neutrons

The average distance between collisions is few cm. Low energy neutrons (0.025 – 100 eV) lose energy primarily through (n,γ) capture reaction. Since living tissue has a high density of hydrogen atoms, mostly in the form of water, the main capture process releases a 2.2 MeV photon. At high energies (above keVs), neutrons lose energy through elastic collision. With high hydrogen concentration, elastic scattering causes mostly the release of protons.

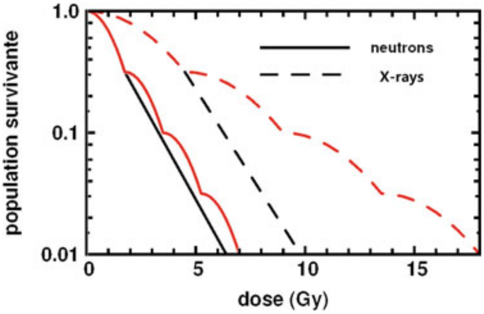
5.1.3.2 Factors Influencing Biological Effects

Effects of radiation within biological systems vary depending upon factors associated with the interactions. These factors include total dose, dose rate, LET, oxygen effect, and the timing of irradiation in the cell cycle.

Total dose refers to the total amount of energy deposited in the target mass. As the increase in energy deposition increases the number of ionization events in the target, higher total dose increases the related biological effects in general.

Dose rate refers to the rate of energy deposition in the target. Total dose to the target system could be delivered instantaneously or over a time period. Even if the total dose delivery is the same, the resulting effect may be different depending upon the rate of dose delivery. It has been found that, for low LET radiation, lowered dose-rate yields higher cell survival for the same total dose. However, such dose rate effect on cell survival was smaller with high LET radiation. This is shown in Fig. 5.4 where the difference in biological effect between the single dose delivery case and the fractionated dose delivery case was shown. In the figure, radiation biological effect

Fig. 5.4 Differences in RBE (relative biological effects) between single dose delivery and fractionated dose delivery for x-ray and neutrons. (Reproduced from Hall 1994)



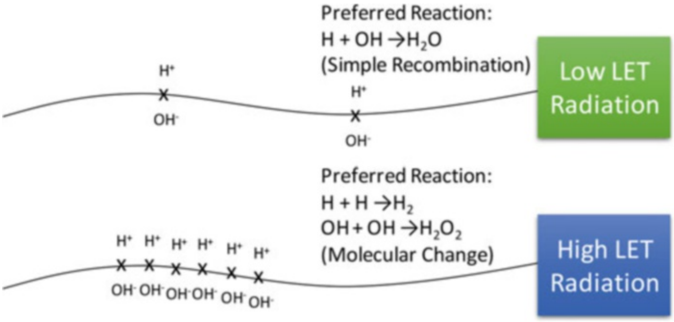


Fig. 5.5 Differences in the fate of the free radicals from ionization of water between the low LET and high LET radiation

was captured through cell survival fraction. For x-rays (a low LET radiation), the dose needed for the 1% cell survival fraction was about 10 Gy with single dose delivery while 18 Gy was needed for the fractionated delivery case. On the other hand, for neutrons (a high LET radiation), the dose needed to result in 1% survival was about 6 Gy and 7 Gy, respectively, for single delivery and fractionated delivery.

For the mutation effect, however, it was found that dose-rate effects may be minimal or absent in human cells. For the effect related to cancer, the dose rate effect was found to be complex (either decrease or increase) depending upon the type of radiation.

As already discussed, LET, as a surrogate measure of the density of ionizations within the track of radiation movement, is an important physical factor in determining biological effects of radiation. Such importance is manifested in both direct and indirect actions of radiation. Importance of LET in radiation biological effects through direction action will be discussed in association with the direct targets of radiation such as DNA in Sect. 5.1.4.1. For example, Fig. 5.15 illustrates how high LET radiation causes critical damage to a DNA through the direct action.

In the case of indirect action of radiation, the key question is whether the results of free radical formation from ionization of water lead to irreversible damage to the biological system. This point is illustrated in Fig. 5.5. The free radicals produced

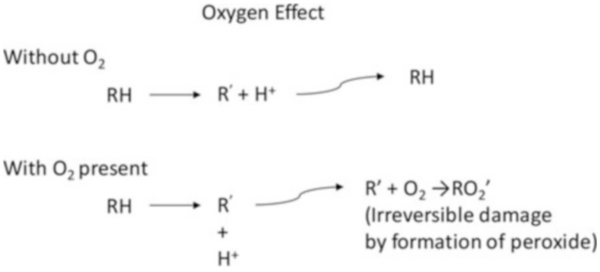


Fig. 5.6 Oxygen effects on the fate of free radicals from ionization

from low LET radiation are sparsely located from each other (200 ~ 3000 Å apart). As the average distance of free radical diffusion during their lifetime (~ 10^{-6} s) is about 5 nm (50 Å), the more likely chemical reactions are the recombination of H^o and OH^o free radicals to form water. In this case, there is no change in chemical forms and no biological damage occurred. However, the free radicals produced from high LET radiation are so closely located from each other. This allows the recombination such as $H^o + H^o$ and $OH^o + OH^o$, through short-distance diffusion of free radicals, readily producing H_2 or H_2O_2. The results are alteration of the original chemical molecule, thus the damage is fixed.

This observation can lead to the following statement. Biological effects of different types of radiation would be the same if LET is the same.

Presence of oxygen also affects biological effects of ionizing radiation: If the medium of radiation interactions is oxygenated, biological effects is increased. It has been shown that the sensitivity of cells to x-ray irradiation decrease about three-fold if irradiation was carried out under anoxic as compared with oxygenated conditions. This oxygen effect is due to the reaction of O_2 molecule with the free radicals (R’) produced from irradiation to produce peroxides ($R' + O_2 \rightarrow RO_2'$). Producing peroxides fixes the damage preventing repair reaction. This point is illustrated in Fig. 5.6. However, this oxygen effect was shown mainly with low LET radiation and was mostly unobserved for high LET radiation.

Cell cycle is also an important factor in affecting radiation effects. This issue will be explained later in the discussions of radiation induced health effects in Sect. 5.1.5.3.

5.1.3.3 Targets of Radiation Interactions

As the smallest unit of life that can replicate independently, a cell is the basic structural, functional, and biological unit of a biological system. There are approximately ~10^{14} cells in a human body with various sizes and shapes (on average, the size of a typical human cell is about between 10 and 30 μm). These sizes and shapes are closely tied to the function of the cell type. For example, red blood cells are small in size in order to pass through narrow capillaries but in the shape of a biconcave disk to maximize the surface area to volume ratio. Neuron cells are long (over a meter)

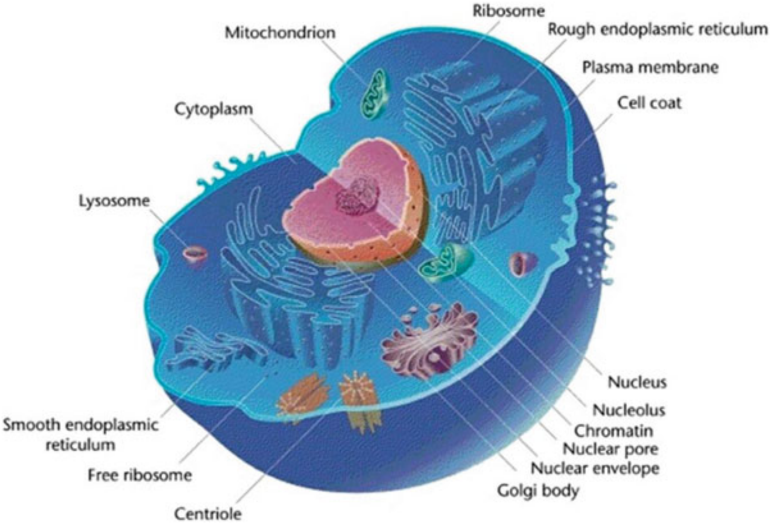


Fig. 5.7 Structure and composition of a cell (Wikimedia Commons 2012)

and narrow (only about 10 μm) to transport signals from the brain to all parts of the body. As to the constituents, however, cells are the same.

When radiation enters a biological system, it has a chance to interact with a variety of cells as the cell components are presented to the radiation as targets of interaction. In reality, cell is such a big target compared to the size of radioactive particle. Therefore, various components of a cell become the actual targets of interaction.

A cell is consisted of cell membrane (also called plasma membrane), nucleus, and various biomolecules surrounded by intracellular fluid, called cytosol. These biomolecules are called cell organelles. The interior region of a cell enclosed by the cell membrane, except nucleus, is also called cytoplasm (see Fig. 5.7).

Cell membrane (also called plasma membrane) is the structural boundary of a cell made of double lipid layer and proteins. It regulates the passage of substances into and out of the cells. Oxygen, CO_2, and water can pass through the double lipids layers through diffusion. Passage of other molecules or ions is, however, controlled by selective transport based on detecting chemical messengers arriving at the cell membrane. Such selective transport is through channels created by embedded proteins in the membrane. The membrane also provides links to adjacent cells.

Cell organelles include various biomolecules that are necessary for the survival and functions of a cell. They include mitochondria, ribosomes, endoplasmic reticulum, Golgi apparatus, lysosomes, peroxisomes, and cell support structure called cytoskeleton.

Mitochondrion is where cell generates energy to perform any necessary functions. It is the power house of the cell. There are as many as 1000 mitochondria present in a cell depending upon the level of activity performed by the cell.

Ribosome is where proteins are made from amino acids using genetic information. The genetic information is stored in DNA (Deoxyribonucleic acid) in the nucleus and is transmitted through a messenger RNA to ribosome. Ribosomes are either ‘free’ in the cytoplasm or bound to the endoplasmic reticulum. There can be as many as ten million ribosomes in a cell. The synthesized proteins are packaged in granular type endoplasmic reticulum that are bound to ribosomes. After being sorted out by Golgi apparatus, the synthesized proteins are released into cytosol. The Golgi apparatus distributes the proteins into vesicles that will be delivered to various part of the cell where these proteins are needed to perform various functions. Most cells have a single Golgi apparatus, but some cells have several of them. The smooth agranular endoplasmic reticulum performs a different function as the site for lipid synthesis. These lipids are major constituents of cell membranes and the principal form of stored energy in the cell. Endoplastic reticulum are found throughout the cytoplasm. Lysosomes play the role of a highly specialized intracellular digestive system where bacteria, the debris from the dead cells taken into the cell, or damaged cell organelles are broken down. Several hundred of them are present in a typical cell. Peroxisome is the site where fatty acids are oxidized and toxic products such as hydrogen peroxide are destroyed. Peroxisomes also break down uric acids and amino acids. All of these organelles are membrane enclosed, i.e., surrounded by cell organelle membranes that work as selective transport channels controlling selective movement of substances between the organelles and the cytosol. There are also filaments of various sizes as non membrane-enclosed organelles in the cell. These filaments make up the cell’s cytoskeleton which maintains and changes the shape of a cell and produces cell movements. The intracellular fluid, cytosol, surrounds all of these cell organelles and nucleus.

Nucleus is the site of storage for genetic information and is surrounded by the nuclear envelope/membrane. The genetic information is used for self-replication, cell biochemical renewal, and cell division. A nucleus includes 46 DNA molecules enclosed with nuclear membrane. Each DNA molecule is a polymer of two polypeptide chains to form double helix coiled around each other and is packaged by special protein (histones) to form a chromatin. The chromatin is further condensed as a packet of compressed and entwined DNA to form a chromosome. These 46 chromosomes are contained in the nucleus of a human cell as 23 pairs. Exception to this is the reproductive cells (gametes) which include only 23 chromosomes and mature red blood cells (with no chromosome). Figure 5.8 shows the process of DNA replication for a cell to undergo division where the old DNA strands open up allowing the new DNA strands to be added.

Each strand of DNA consists of repeating subunits, called nucleotide. Each nucleotide has three components: a phosphate group, a sugar (a monosaccharide, called deoxyribose), and a ring of carbon and nitrogen atoms referred to as a base. The phosphate group is linked to the sugar and the sugar is attached to the phosphate group of the adjacent nucleotide. This sequence continues to form a backbone of nucleotides. The bases are attached to the sugar sticking out to the side of the phosphate-sugar backbone. There are four different types of nucleotides depending on the type of a base attached. The four nucleotide bases are divided into two classes:

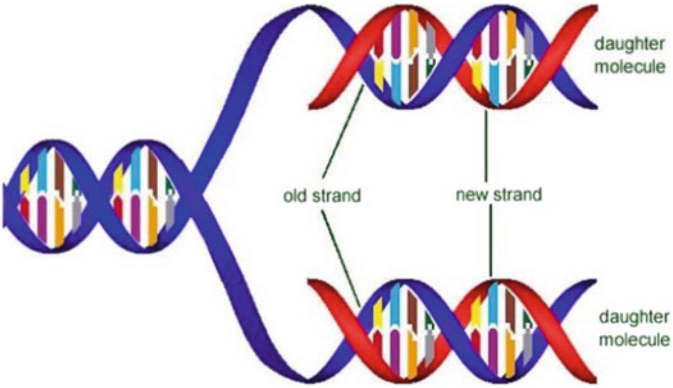


Fig. 5.8 Schematic representation of DNA replication for cell division (source: Phelbs 2016)

(1) the purine bases, adenine (A) and guanine (G), which have double (fused) rings of nitrogen and carbon atoms, and (2) the pyrimidine bases, cytosine (C) and thymine (T), which have only a single ring. Thus, different nucleotides correspond to the four different bases that can be linked to the sugar deoxyribose.

The coded sequence of four different nucleotides determines the amino acid sequences of a protein. Proteins are complex macromolecules formed of chains of amino acids. Typically, each sequence of three nucleotide bases (which is called codon) provides the information for one amino acid. Human body contains 20 different amino acids. These amino acids are then combined to form proteins according to the amino acid sequence of a protein represented in one gene. Each messenger RNA contains the information enough for one gene that is used to replicate one protein. Each of the genes is a particular area of the DNA as the units of hereditary information. Other regions in the DNA that is not covered by genes regulate the process of protein production.

A single molecule of DNA contains many genes (about few hundreds to thousands genes). One human cell contains about 23,000 protein-producing genes in 23 unique DNA strands. These proteins act as structural components in cells while others act as organic catalysts, i.e., enzymes of the cell's biochemical processes.

A complete DNA sequence of one set of chromosomes is referred to as the genome of an organism. All of genes in a DNA make up the genome. A typical human gene is a sequence of approximately 20,000 base pairs. Less than 1 billion base pairs constitute the human genome.

The double helix chains of a DNA are linked to each other through connections of the bases by hydrogen bonds. The way in which bases in each side of the chain are connected have specific pattern, called base-pairings: The purine guanine and the pyrimidine cytosine are always connected through forming three hydrogen bonds (G-C pairing), while the purine adenine and the pyrimidine thymine are connected through forming two hydrogen bonds (A-T pairing). As a result G is always paired with C, and A with T. This base pairing between purine and pyrimidine is maintained at a constant distance between the sugar phosphate backbones of the two chains. The structure of DNA and base pairing specificity are shown in Fig. 5.9. Detailed chemical structure of a DNA strand is captured in Fig. 5.10.

This specificity in base pairing between DNA double helix provides an important basis of DNA protection mechanism, the redundancy of the stored genetic information. This built specificity allows duplicating and transferring genetic information and repairs of damages in the case of DNA damage. At the same time, redundancy in DNA is very limited as the loss of both bases from the double stand chains can take place. Thus simultaneous loss of both bases can lead to an irreversible damage and may also lead to loss of genetic coding that is vital to cellular function and survival.

While the DNA molecule does not have a special or unique sensitivity to radiation damage, it is the only molecule in a cell able to duplicate itself without information from some other cell component. The total genome is unique in each cell and as a result, DNA is the most critical target in the cell.

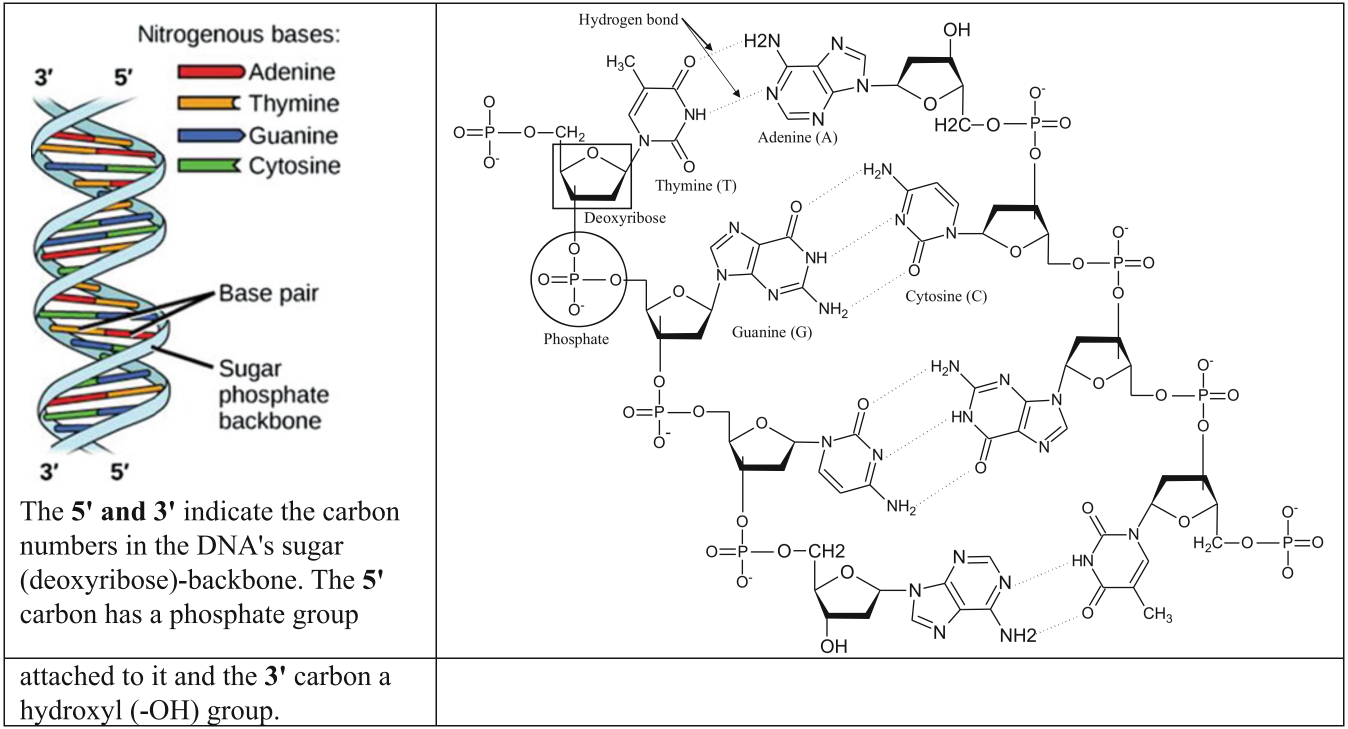
The nucleus that contains DNA is surrounded by the nuclear membrane (also called nuclear envelope) which is another important target for radiation interaction in a cell. The nuclear membrane includes a network of pores covering its surface. These pores play a role in nucleus-cytoplasm communication and the chromatin distribution within the nucleus. When the nuclear membrane is irradiated, a delay in development of the nuclear envelope occurs.

Radiation damage of other biomolecules in the cell can lead to their deactivation. Even so, due to the large degree of redundancy built in the cell, biological activity of the cell can be sustained even with loss of a large number of these biomolecules without serious functional deficit. This is in part due to the fact that damaged molecules are continuously replenished through the synthesis of proteins, and lipids.

It is known that the radiation dose required to kill cells depends on what part of the cell is damaged. For example, the dose required to kill a cell is much lower when the nucleus is irradiated compared to the cytoplasm. The dose that can be tolerated by the nucleus before cell death is less than one-hundredth of that for the case of cytoplasm irradiation.

Damage to the cell from radiation will also depend on the size of the target: Bigger targets presents higher chance of being hit by radiation. While the sizes vary (see Table 5.11), typical sizes of the biomolecules in the cell are 10 nm for cell membrane thickness, 11 nm for ribosome, 200 nm (200 to 500 nm) for lysosomes, 200 nm (200 to 500 nm) for peroxisomes, 3 μm for mitochondrion, and 3 to 10 μm (with a mean of 6 μm) for nucleus. Although nucleus is the largest target in the cell, the sizes of DNA or chromosomes are much smaller than the size of the nucleus.

Note also that DNA molecules, when uncoiled, are 1.8 to 8.5 cm long (about 5 cm on average). The combined total length of all DNAs in one cell becomes about 2 meters. Still, when the DNA is coiled to form a chromosome, it becomes a shorter, thicker fiber in the form of loops with the width of about 700 nm. Within these highly coiled and compressed structures, each DNA double helix has about 2 nm diameter and the distance between outer edges of the bases in DNA double helix is about 1.1 nm, and the distance between base-pairs in DNA is about 0.34 nm (3.4 Å) (Table 5.2).



The **5' and 3'** indicate the carbon numbers in the DNA's sugar (deoxyribose)-backbone. The **5'** carbon has a phosphate group attached to it and the **3'** carbon a hydroxyl (-OH) group.

Fig. 5.9 DNA structure and base pairing

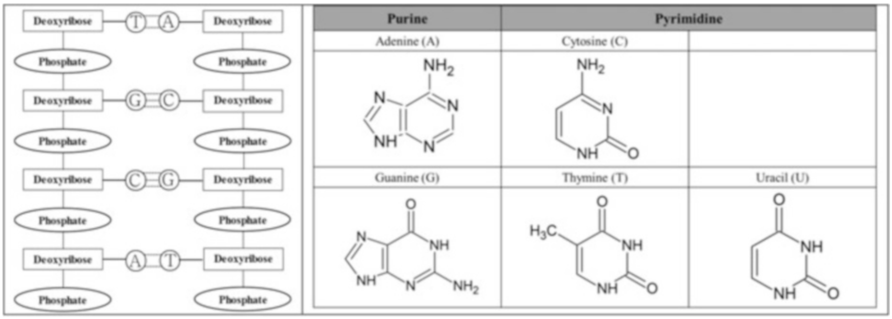


Fig. 5.10 Chemical structure of a DNA and RNA nucleotide, where if X = H then it is a base and if X = ribose phosphate then it is a nucleotide

Table 5.2 Relative sizes of biological molecules and cells

Length	Example
0.1 nm	Diameter of a hydrogen atom
0.8 nm	Amino acid
0.34 nm	Distance between base-pairs in DNA
1.1 nm	Distance between outer edges of the bases in DNA double helix
2 nm	Diameter of a DNA double helix
4 nm	Globular protein
6 nm	Microfilaments
10 nm	Cell membrane thickness
11 nm	Ribosome
25 nm	Microtubule
50 nm	Nuclear pore
100 nm	Large virus
150-250 nm	Small bacteria such as mycoplasma
200-500 nm	Lysosomes
200-500 nm	Peroxisomes
1 μm	Diameter of human nerve cell process
2 μm	E.coli – a bacterium
3 μm	Mitochondrion
5 μm	Length of chloroplast
6 μm (3-10 μm)	Nucleus
9 μm	Human red blood cell
10 – 30 μm	Most eukaryotic animal cells
10 – 100 μm	Most eukaryotic plant cells
90 μm	Small amoeba
100 μm	Human egg

5.1.4 *Radiation Effects on DNA*

5.1.4.1 DNA Damage

Radiation damage to DNA are from biochemical damages in the irradiated DNA molecule through breaking of chemical bonds or modifications of the chemical structure. These damages could mean chain breakage in the sugar-phosphate backbone or loss or changes in the purine or pyrimidine base or deoxyribose sugar. These can be grouped to four categories: nucleotide damage, single-strand breaks, double-strand breaks, and crosslinking (see Figs. 5.11, 5.12 and 5.13).

Nucleotide damage is the most common type of DNA damage and includes depurinations (removal of A or G bases from the backbone), depyrimidinations (removal of C or T bases from the backbone), and base alterations (e.g., deaminations, i.e., removal of amine group from the base). Examples of these are shown in Fig. 5.11. Chemical structure of key chemical groups as examples are also given in Fig. 5.12.

As many as 10,000 depurinations could take place in a cell per day. Depyrimidation occurs at much lower frequency (e.g., 500 per cell per day). Base alterations such as deaminations may occur at the rate of 200 per cell per day. Once depurination or depyrimidination occurs, it becomes so called AP (apurinated or apyrimidinated) site. These alterations in the base sequence mean alteration in the genetic information that could result in a mutation. Fortunately, base damage is readily repaired as explained in the next section.

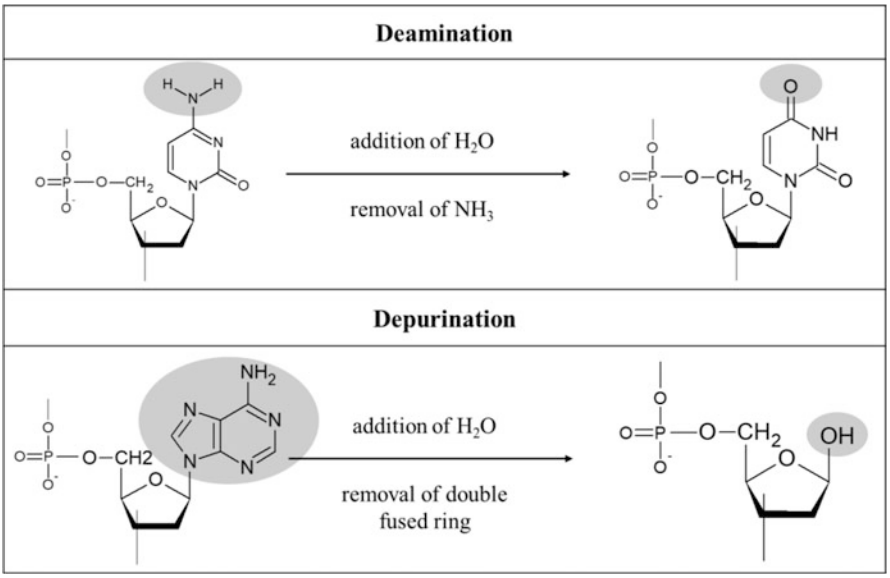


Fig. 5.11 Example processes of depurination or deamination as DNA damage

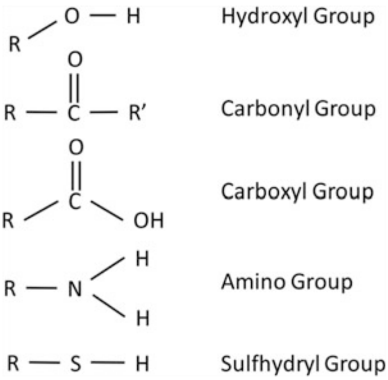
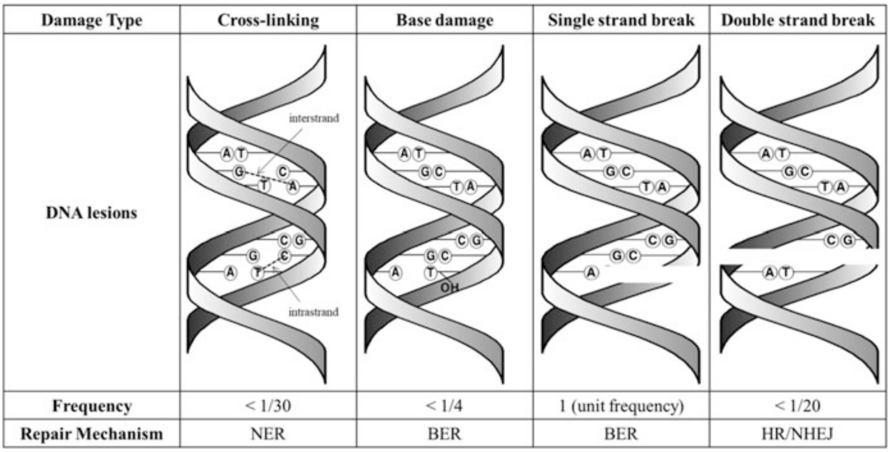


Fig. 5.12 Chemical structure of simple chemical groups



Damage Type	Cross-linking	Base damage	Single strand break	Double strand break
DNA lesions				
Frequency	< 1/30	< 1/4	1 (unit frequency)	< 1/20
Repair Mechanism	NER	BER	BER	HR/NHEJ

Fig. 5.13 Graphical representation of DNA damages (i.e., single strand break, double strand breaks, cross-linking, etc.)

Single-strand breaks (SSBs) are detachments in the sugar-phosphate backbone at only one side of the DNA double helical strands. The detachment can be through phosphate ester bond rupture or sugar carbon-carbon bond rupture. This defect can also be easily repaired as the other side of double helix remains intact.

Double-strand breaks (DSBs) occur when two such breaks occur on both sides of helical strands, either at the same base pair site or a few bases apart. This results in severing of the DNA molecule. DNA DSBs are generated when the two complementary stands of the DNA double helix are broken simultaneously at sites that are sufficiently close to one another that base-pairing and chromatin structure are insufficient to keep the two DNA ends juxtaposed. This could occur when a single particle with high LET produces a break in both strands (with a break in each of the two strands less than about 5 nucleotide units apart) or two single breaks happening side by side (approximately one out of 70 random single breaks leads to double breaks this way). As a consequence, the two DNA ends generated by a DSB are liable to become physically dissociated from one another. This makes ensuing repair process difficult and provides the opportunity for inappropriate recombination with other sites in the genome. These types of breaks are considered to be the most serious type of damage suffered by the DNA. Chemical agents may hold the DNA fragments in place for a while for possible repair. However, if the fragments drift apart, the chance of properly rejoining them becomes very low.

Crosslinking refers to chemical bonds occurring between nucleotides in the DNA. The cross-links within a DNA strand is called intra-strand crosslink while the cross-links between the two DNA double strands are called inter-strand crosslink. The formation of crosslinks can probably be attributed to the formation of reactive sites at the break. Two such reactive sites will join if contact is made between them. Crosslinking within the helix of a DNA molecule may also occur between the bases of nucleotides. This produces dimers in which pyrimidine or purine bases join together to form a dimer linkage that is more stable than a hydrogen bond. Inter-strand cross-links can lead to cell death by blocking DNA replication and/or DNA transcription.

Creations of single lesion in DNA can be in the form of single-strand break (SSB) or base damage. Creation of double lesions in DNA can result in double-strand break (DSB). As an overall average, SSBs are produced four times more frequently than base damages, 20 times more frequently than DSBs, and 30 or more times more frequently than cross-links.

If DNA DSBs remain open, then at the time of mitosis, chromosomes appear broken. When two different DNA double-strand breaks (occurring in the same or different chromosomes) are joined, (thus misjoin to each other), this chromosome rearrangement leads to so-called chromosome aberrations. Chromosome aberration (i.e., imperfection) could be manifested in various forms such as dicentrics (having two centers), rings, reciprocal translocations, and inversions.

In general, the levels of radiation-induced lesions increase linearly with dose. In other words, as the amount of energy deposited to the target becomes larger, the number of ionization produced in the target is larger resulting in a densely populated ionization sites. These densely populated (or closely located) ionization sites can become the sites of the lesions in the DNA.

As the average distance between ionization is about several angstroms for alpha particles and as the distance between the DNA double helix is about 20 angstroms (2 nm), these alpha particles are capable of creating more than single lesion in one DNA in their track. In contrast, with low LET radiation (e.g., electrons or photons), the average distance between ionization is more than hundreds of angstroms implying that a single particle in the low LET radiation category cannot create more than one lesion in a single DNA molecule. Therefore, DNA double strand breaks by high LET radiation are much more likely than low LET radiation. This is also depicted in Fig. 5.14.

As shown in the figure, α-particle can readily create DSB in a single trajectory while β-particle is incapable of creating DSB in a single particle trajectory. Creation of DSS by a β-particle may be possible if multiple β-particles are hitting the same area or the trajectory of random movements of a β-particle overlaps in the same DNA

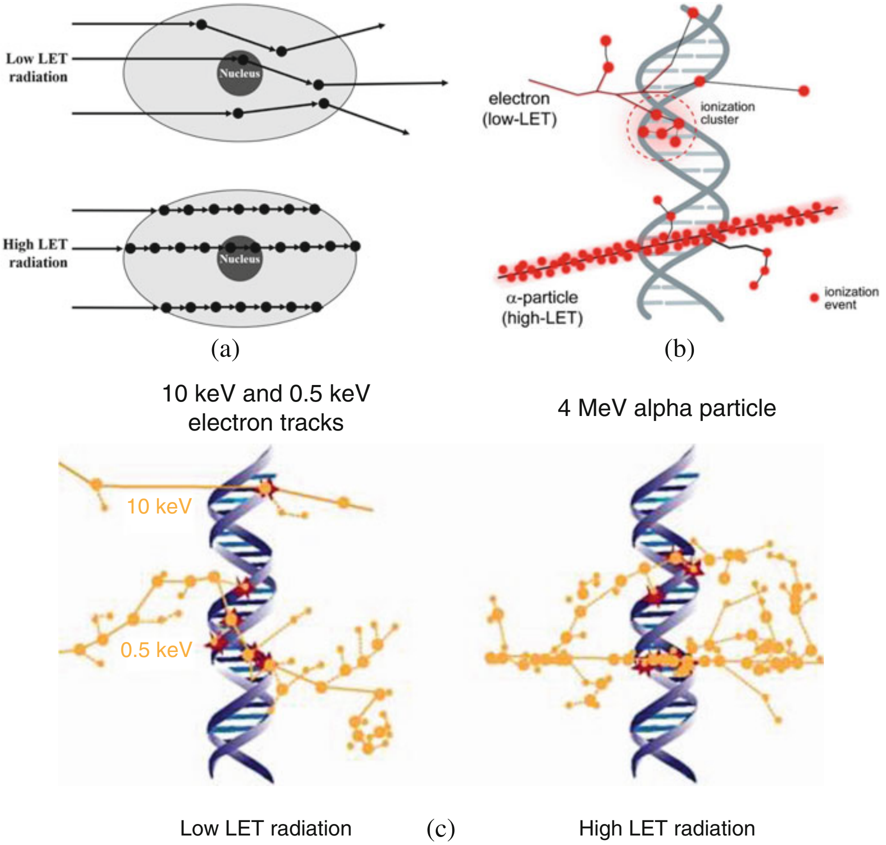


Fig. 5.14 Differences in radiation interactions with cell and DNA between high LET and low LET radiation (Sources: b-Iliakis G et al (2019); c-Schipler and Iliakis 2013)

region. Therefore, if the damage from low LET radiation is localized in a region resulting in clusters ionizations (referred to as blobs or spurs), probability of producing a DSB increases.

It is unlikely that the free radicals produced along the track of a low LET radiation damage the same DNA site as the diffusion distance of the radicals is about 5 nm. In contrast, the free radicals produced from high LET radiation have chance to attack the same DNA site through diffusion. Therefore, the damage from high-LET radiation is not easily repaired. Please also refer to the discussions in Sects. 5.1.2 and 5.1.3.2.

5.1.4.2 DNA Repair Mechanism

As stated, a cell experiences a large number of DNA damages daily from natural causes. At the same time, these damages are routinely repaired properly to allow the cells to continue to survive. This indicates that cell's DNA repair mechanisms are very effective. In fact, the damages such as base damage or SSB are easily repaired. However, repair of DSB is difficult and inherently error-prone. Sometimes repair of cross-linking is error prone.

Cell's DNA repair mechanisms against radiation-induced damages include base excision repair (BER), nucleotide excision repair (NER), homologous recombination (HR), non-homologous end joining (NHEJ), mismatch repair (MMR), and translesion synthesis.

Base damage, single strand breaks, or intra-strand cross-links are repaired by base excision repair or nucleotide excision repair. Inter-strand cross-links are repaired by homologous recombination or translesion synthesis. Homologous recombination (HR) or non-homologous end joining (NHEJ) are used to repair double strand breaks. Mismatch repair is performed whenever new DNA is synthesized as a way to remove and replace mismatched base inserted.

Base excision repair is to repair non-bulky specific lesion. In this repair, only the damaged base is removed and replaced by the correct base. Such base damage may be caused by oxidation (i.e., addition of hydroxyl group, OH, to the base), deamination (i.e., loss of amino group, NH_2, from the base), alkylation (e.g., addition of CH3 to the base) or loss of single base (i.e., depurination or depyrimidation). Various enzymes are involved in the repair including a DNA glycosylase (to remove the damaged base), a AP (apurinated or apyrimidinated site) endonuclease (to recognize "missing tooth"), a DNA polymerase (to resynthesize the missing piece), and a DNA ligase (to seal the gap in the backbone). Figure 5.15 (a) shows an example of base excision repair when the base damage was caused by deamination of cytosine (C) producing uracil (U).

Nucleotide excision repair is to repair bulky lesions (i.e., bulky DNA adducts such as thymine dimer, see Fig. 5.15 (b)). In this case, the damage is recognized and the backbone containing the damage is cleaved on both sides of the bulky adduct. Then a piece of a nucleotide (called an oligonucleotide) containing the damaged bases in a single strand helix is excised, removed, and replaced by a new intact form of the oligonucleotide. Then the gap is filled by the repaired nucleotide closing the backbone back to the position by the action of DNA polymerase. A DNA ligase seals the backbone. An example of nucleotide excision repair (repair of a thymine dimer formation) is shown in Fig. 5.15 (b). We note that several nucleotides are replaced in nucleotide excision repair whereas only single nucleotide is replaced in base excision repair.

Double-strand breaks are mainly repaired through nonhomologous end joining (NHEJ) or homologous recombination (HR). NHEJ is the most common type of DSB repair mechanism. In NHEJ, the broken parts of DNA double strands are cleaned and simply joined together in both strands. So the repair is likely to be incorrect. HR is based on finding a similar (or hopefully identical) sequence of nucleotide in a neighboring DNA and using them to replace the missing parts of DNA thus recombining the double broken DNA.

Mistakes also occur in DNA repairs. These mistakes are rare but occur through polymerase enzyme inserting wrong nucleotide into a sequence. Error rates for

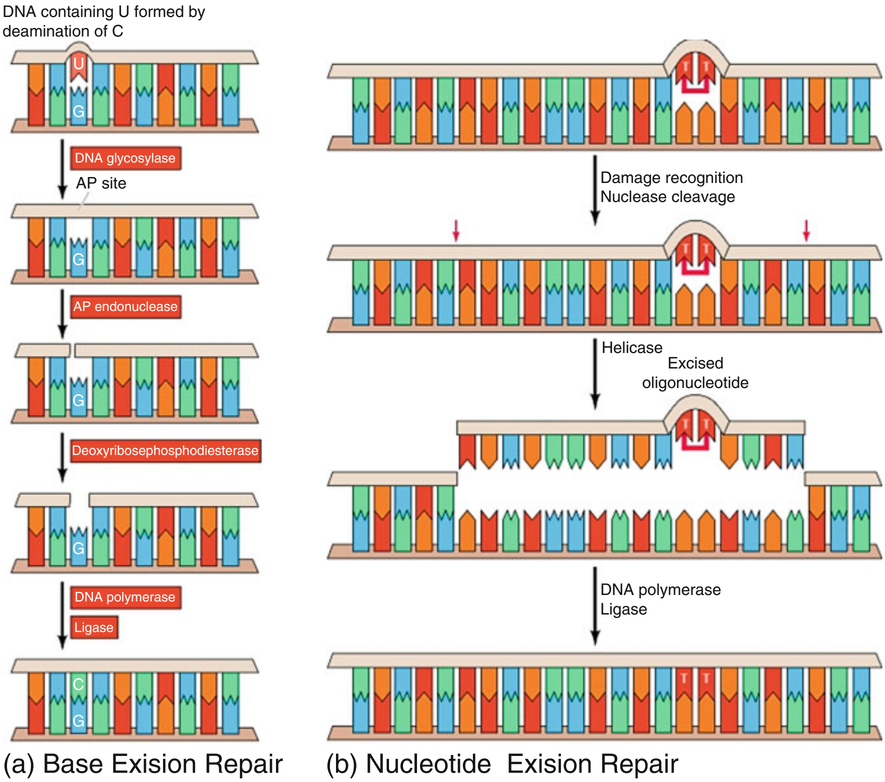


Fig. 5.15 Illustration of Base Excision Repair and Nucleotide Excision Repair through examples. (Source: Cooper et al. 2000)

excision repair are about ~one per 10^7 -10^{11}. Mismatch repair (MMR) is used to correct the error following the process of base excision repair. Therefore, the repair targets mismatched base pairs in the newly replicated DNA. After polymerase adds a nucleotide as part of DNA synthesis, MMR is to check and see if addition of the new bases are correctly done. If not, exonuclease activity is used to remove mismatch. In mismatch repair, only single nucleotide is replaced. MMR is efficient but not perfect. Incorrectly repaired DNA following MMR becomes permanent mutations after the next cell division.

Translesion synthesis is another post-replication repair (see Fig. 5.16). It is applied during the process of DNA replication in the DNA synthesis phase. When a damaged DNA template base is encountered in the replication process, the progression of the replication fork (the structure formed with two branching "prong" during replication) can be stalled or completely blocked. This eventually leads to cell death. To avoid this, cell uses translesion synthesis as a DNA damage tolerance process, allowing the DNA replication machinery to bypass the lesion and continue replication downstream. This results in a gap in the newly synthesized

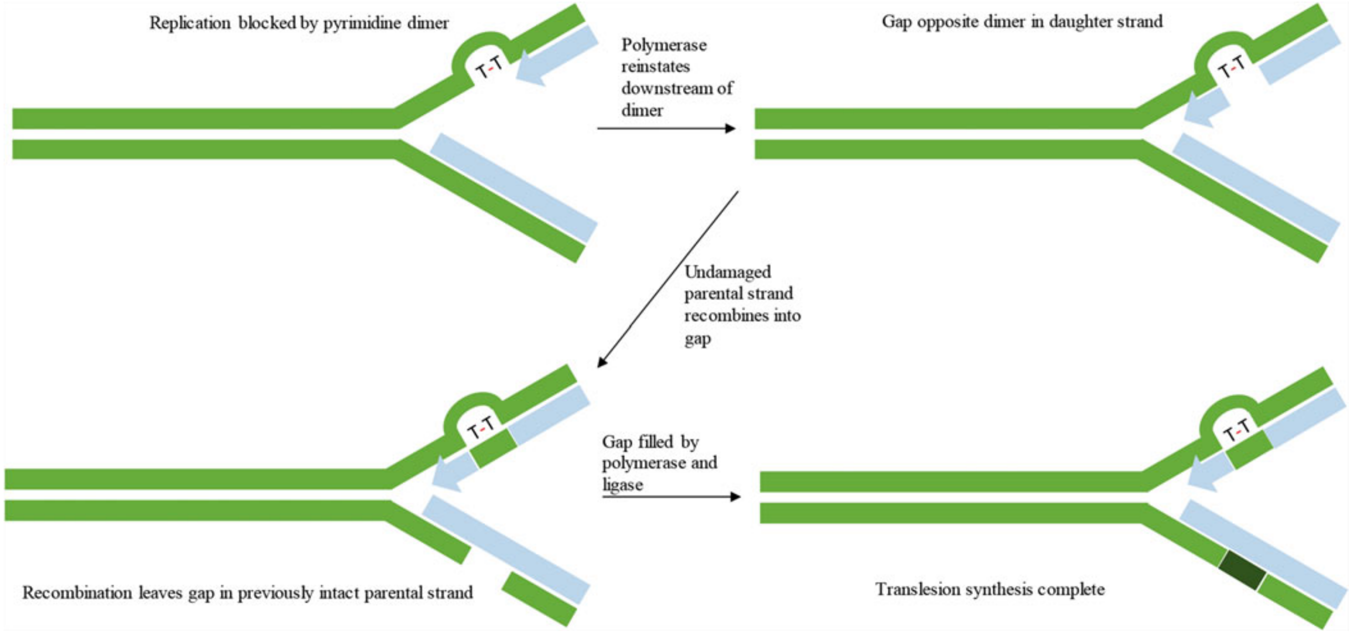


Fig. 5.16 Depiction of translesion synthesis

DNA strand. The gap can be filled by using the template in the other undamaged parent strand. The remaining lesion is removed and repaired by excision repair.

Consider the dose of 1 mGy per year from low LET radiation. This level of dose is expected to cause less than one track of ionizing radiation in the nucleus in a year. As the diameter of the DNA molecule at about 2 to 3 nm within 6 μm diameter nucleus, the probability of this radiation creating more than one tracks hitting the same area of the DNA molecule is extremely low. Therefore, the resulting damages are in the form of easily repairable SSB or base damage. Humans are exposed to the natural background radiation at the level of 3-5 mGy per year as part of daily living. The health effect from these natural background radiation is very low compared to other health effects from other hazardous materials present in our lives.

Example 5.1: Number of DNA Damages from Radiation

If a person is exposed to a natural background radiation at the dose level of 0.5 Gy (500 mrad) per year, what would be the approximate number of SSB and DSB occurring in the body in a year? Make any necessary assumptions and state them clearly.

Estimate the approximate number of SSB/DSB occurring per cell per day according to these estimates. Compare these numbers with the naturally occurring SSB and DSB per cell per day.

Given: The average number of naturally occurring SSB and DSB (per cell per day) in human body is 10,000 ~ 55,000 SSB and 10 ~ 50 DSB.

Solution:

Assumptions:

1. Background doses are from isotropic low LET radiation
2. The number of cells in the body = 3.7×10^{13}
3. SSB are produced 4 times more frequently than nucleotide base damages, 20 times more frequently than DSBs, and 30 times more frequently than cross-links
4. Radiation exposure rate of 1 mGy/y will result in 1 track/y in the nucleus (meaning 1 damage/y/nucleus)

From assumption 1,

→ 500 mrad/y = 5 mGy/y.

From assumption 3,

$$N_{SSB} = 4 \cdot N_B$$

$$N_{SSB} = 20 \cdot N_{DSB}$$

(continued)

Example 5.1 (continued)

$$N_{SSB} = 30 \cdot N_C$$

The sum of SSB, base damage, DSB, or cross-links becomes 100%.

$N_{SSB} + N_B + N_{DSB} + N_C = 100$ (%)

$$N_{SSB} + \frac{N_{SSB}}{4} + \frac{N_{SSB}}{20} + \frac{N_{SSB}}{30} = 100$$

$\rightarrow N_{SSB} = 75$ %, $N_B = 18.75$ %, $N_{DSB} = 3.75$ %, $N_C = 2.5\%$

(there are 75%, 18.75%, 3.75%, and 2.5% chance of SSB, base damage, DSB, and cross-links, respectively, per one radiation-induced DNA damage,)

From assumption 2 and 4

$$N_{damage} = \frac{5mGy}{y} \cdot \frac{1\ damage/nucleus}{mGy/y} \cdot 3.7 \times 10^{13} cells$$

$$N_{damage} = 1.85 \times 10^{14}$$

Thus, the approximate number of SSB and DSB in the whole body:

$$\rightarrow N_{SSB} = 0.75 N_{damage} = 1.4 \times 10^{14} \text{ (per year)}$$

$$\rightarrow N_{DSB} = 0.0375 N_{damage} = 6.9 \times 10^{12} \text{ (per year)}$$

In terms of the number per cell per day, these number become ~3.8 SSB and ~ 0.2 DSB.

The number of natural background radiation induced SSB and DSB is about $4 \times 10^{-4} \sim 7 \times 10^{-4}$% and 0.4 ~ 1.8% of naturally occurring SSB and DSB, respectively.

5.1.5 Radiation Effects on Cells

If the DNA repair mechanism does not properly fix the changes in the DNA, the damage remains permanent. These changes include point mutations affecting single genes or chromosome aberrations that may involve multiple genes. Consequences of such permanent change are manifested as biological effects through cell killing, mutation, or malignant transformation.

When the molecules damaged by radiation are essential for the normal functioning of a cell, the cell suffers injury and may die. If the affected molecule contains genetic information, the resulting changes in genetic information lead to mutation of the cell. Also, if the mutation occurs to genes that are involved with cell growth control, malignant transformation could start to the cell leading into cancer.

5.1.5.1 Cell Killing

A cell is regarded as dead when it loses its ability to proliferate, i.e., to divide. Cell death could result from radiation through either interphase death or mitotic death. Interphase death means cells are lethally damaged by radiation and die soon after the exposure without undergoing any cell division. This occurs when cell receives very high radiation doses. Mitotic death refers to cell death resulting from inhibition of the cell's ability to proliferate indefinitely. In this case, irradiated cells may undergo one or a few cell divisions prior to dying. Mitotic death occurs mainly in cells that are actively dividing or have the potential to divide. Cell killing is also the basis for the use of radiation to treat cancer through mitotic cell death.

There is a strong evidence that cell killing is connected with DNA double-strand breaks and chromosome aberrations. Cells with mis-repaired DNA double-strand breaks are hypersensitive to both radiation-induced cell killing and induction of radiation-induced chromosomal aberrations. It can be thus said that unrepaired DNA double-strand breaks lead to cell death through the production of chromosome aberrations.

Manifestation of the cell killing effect depends on radiosensitivity of a cell. In general, the sensitivity of of a cell is higher if the cell (1) is undifferentiated, (2) has a greater proliferative capacity, and (3) divides more rapidly. Typically the most sensitive cells to radiation-induced cell killing are hematopoietic stem cells, intestinal crypt cells, basal cells of the epidermis, and lymphocytes. The least sensitive cells are nerve cells, muscle fibers, and most mature hematopoietic cells.

Cell killing leads to the acute somatic effects of radiation through inhibiting cell renewal or vascular effects. Typically affected cell renewal systems include skin, gonads, cornea of eye, bone and cartilage, and blood forming tissues. Vascular effects include morphologic changes in capillaries, ischemia (compromised blood supply), severe permeability changes of blood vessels at very high doses, and vascular damage to capillary bed. If the cell killing occurs to the fetus through utero irradiation, it leads to teratogenic effects (congenital malfunction) or developmental effects.

When a large dose of radiation is delivered to the entire body, acute radiation syndromes can occur. Such effects are expected in the rare case of an accident at dose level higher than 0.25 Gy. Acute radiation syndromes are observed from such exposure incidents including nausea, vomiting, diarrhea, and fatigue. Table 5.3 shows various symptoms of acute radiation syndrome at different levels of dose for gamma radiation. At dose level of 4.5 Sv, 50% of an exposed population are expected to die. This level of dose is called LD50 (the lethal dose level where 50% of the exposed people die).

Table 5.3 Acute radiation syndrome for gamma radiation dose

Dose (Gy)	Symptoms	Remarks
0-0.25	None	No clinically significant effects
0.25-1	Mostly none. A few persons may exhibit mild prodromal symptoms, such as nausea and anorexia	Bone marrow damaged; decrease in red and white blood-cell counts and platelet count lymph nodes and spleen injured; lymphocyte count decreases
1-3	Mild to severe nausea, malaise (a general feeling of discomfort, illness, or uneasiness), anorexia (loss of appetite), infection	Hematologic damage more severe. Recovery probable, though not assured.
3-6	Severe effects as above, plus hemorrhaging, infection, diarrhea, removal of hair, temporary sterility	Fatalities will occur in the range 3.5 Gy without treatment
More than 6	Above symptoms plus impairment of central nervous system; incapacitation at doses above ~10 Gy	Death expected

5.1.5.2 Induction of Mutations

A change in the genetic message, known as mutation, occurs as the sequence of bases is altered in DNA. Mutation affects all copies of the protein synthesized using the encoded genetic sequence. A mutation in a cell leads to one of three different outcomes: (1) No noticeable change in the cell's functioning, (2) modification of cell function with continued growth and replication, or (3) cell death.

Mutation can occur either in somatic (non-reproductive) cells or germ (reproductive) cells. If mutation involves somatic cells, it will not be inherited but can cause diseases including abnormality or cancer. Cancer can occur if special genes in the somatic cells are mutated. Abnormality becomes manifest after many generations of cell replication leading to long-term somatic effects

If mutations involve germ (sperm or egg) cells in the gonads (testes in the male and the ovaries in the female), mutation can be inherited leading to heritable genetic effects which are manifested in the offspring of the irradiated individual.

Genetic effect of radiation was one of the key concerns of radiation health effects in 1930s through 1960s. This was based on the observation from Herman Mueller's experiments in 1927 showing genetic mutations among the fruit flies irradiated with x-rays. However, studies of the A-bomb survivors in Japan showed that genetic effect of radiation is a minor concern among humans as no statistically significant heritable effects were observed among the children of the survivors. The observation from the A-bomb survivors study was that cancer is the main health effect of concern among the survivors from radiation exposure.

Table 5.4 Comparison of characteristics of normal cells and cancer cells

	Normal cell	Cancer cell
Shape	Regular	Irregular
Nucleus	Proportionate to the size of the cell	Large, darker
Growth	In control, systematic	Out of control
Reaches maturation	Yes	No
Communicates with other cells	Yes	No
Visibility to immune cells	Yes	No
Requires oxygen	Yes	No
Energy efficiency	Very high (95%)	Very low (5%)
Cell environment	Alkaline	Acidic

5.1.5.3 Malignant Transformation of Cells

A large number of DNA damages occurs (~10,000 per cell per day) among an average individual out of normal living. At the same time, human body has a very successful defense system against these damages through DNA repair mechanisms. The most significant type of DNA damage is double strand breaks for which no successful repair mechanism exists. Mis-repaired DNA damage becomes the site of mutation. If such mutation occurs in the genes of DNA controlling the cell growth mechanism or in the DNA repair genes, malignant transformation (conversion of normal cell to a cancer cell) can be initiated representing the initial step in the induction of cancer.

Cancer is a general name for a group of diseases with malfunction in cell growth control. Normal homeostatic cellular control is lost in a cancer cell. The cancer cell grows continuously, invading, crowding, and overwhelming the surrounding normal cells. If this unregulated growth remains unchecked, the result can be the death of the organism. Comparison of normal cells and cancer cells is summarized in Table 5.4

In order for cells to grow unregulated, genes which regulate cell growth must be damaged. These genes include proto-oncogenes and tumor suppressor genes. Proto-oncogene promotes cell growth and mitosis while tumor suppressor gene slows down cell division or inhibit uncontrolled growth. Tumor suppressor gene also tells cells when to die (according to so-called programmatic cell death). Proto-oncogenes are very active during the early stage of development but become inactive in many mature cells. If a proto-oncogene is mutated, it becomes an oncogene and permanently turned on driving abnormal proliferation. When a tumor suppressor gene is mutated, it gets turn off and inactivated leading to the loss of function as negative growth regulator. Studies showed that inherited abnormalities of tumor suppressor genes could cause certain types of cancer to run in families. Important examples of tumor suppressor gene are p53 and Rb. In particular, ~60% of cancers is associated with absence or damage of p53. p53 is also known to be involved with initiating DNA repairs as a DNA repair gene.

Cancer is a multi-step stochastic process including initiation, promotion, and progression, thus takes long time for its development. As discussed in Sect. 5.1.2, this long-term characteristics is represented by long latency of cancer. For most solid tumors, the minimum latency is about 10–15 years with 25 years as the average. For leukemia, the minimum latency is 2–5 years with about 10 years as the average.

Cancer initiation is transformation of a normal cell into a cancer cell through the activation of one or more proto-oncogenes and/or inactivation of tumor suppressor genes. Thus initiation of cancer typically requires a series of several mutations to tumor suppressor genes or proto-oncogenes. Mutational inactivation of DNA repair mechanisms also leads to cancer initiation. Cancer initiation is preceded by an accumulation of the enough number, combination, and types of aberrant alterations in one cell. Typically, a series of several mutations to tumor suppressor genes or proto-oncogenes are required before a normal cell transforms into a cancer cell.

Cancer promotion refers to the promoting actions of certain agents on the 'initiated' cell toward selective growth enhancement. This appears to involve non-mutational effects on the rate of growth of cells. Thus through cancer promotion, the initiated cancer cell and its progeny survive and grow fast expanding in number while being exposed to a promoting agent. Causes of cancer promotion include defects in terminal differentiation (i.e., proliferation as only one cell type), defects in growth control, resistance to cytotoxicity (i.e., resistance to cell death by certain chemicals or natural killer cells), or defects in programmed cell death (a genetically regulated process of cell suicide for homeostatis).

Examples of such promoting agent include reactive oxygen species (ROS), phorbol esters (e.g. tetradecanol phorbol acetate (TPA)), polycyclic aromatic compounds (e.g. dioxin), electromagnetic field, and some hormones like estrogen. Cancer promoting agent cannot induce cancer by itself but in combination with an initiating agent can synergistically enhance the process of malignant transformation. The promotional stage is known to be susceptible to modifying effects of various factors and thus is reversible. The modifying factors include genetic constitution, gender, age at initiation, physiological state, smoking habits, drugs, and a variety of other physical and chemical agents. The agents that may work as inhibitors of cancer promotion are vitamin A, vitamin C, vitamin E, beta carotene, and antipain (a protease inhibitor). There could be long delays between initiation and promotion.

Cancer progression denotes the process by which transformed cells proliferate with increased growth speed and invasiveness. Thus cancer cells take on an aggressive malignant character overwhelming the growth of other cells leading to metastasis (i.e., spreading). Some tumors are called **benign** because they do not spread beyond their initial location. Benign tumors are not considered to be cancerous but still can cause serious health problems by putting pressure on the surrounding cells through a large growth. The presence of multiple genetic alterations in cancer cells is required for progression. Hormones also can have a large effect on progression. Gender and age will affect this process as hormonal status is dependent on gender and age. Progression is an irreversible process.

Ionizing radiation as universal carcinogen can cause all three phases of cancer development, i.e., initiation, promotion and progression. Normal living of a person also presents opportunities for DNA damage and potential promotion of the cancer due to the ubiquitous presence of carcinogens in our living (food, environment, tobacco smoking, etc.). These damages slowly accumulate through the life of an individual that may lead to cancer occurrence. Thus cancer normally occurs late in life. At the same time, some cancers such as leukemia and lymphoma often occur at an early stage of human life. This may be due to specific chromosomal translocations that lead to transcriptional activation (DNA information is copied into a new molecule of messenger RNA) of an oncogene. The presence of a single copy of the activated oncogene in a cell could then dominate the process of leukemia or lymphoma development.

Exposure to a low dose of radiation may cause non-lethal damage to cells that is "remembered" and expressed after many generations of cell division by the progeny of the irradiated cells. This is called genomic instability. Genomic instability occurs when a gene responsible for the stability of the genome and/or the fidelity of replication is mutated. This mutation could lead to what has been referred to as a mutator phenotype (the increase in mutation rate of cancer cells). Genomic instability may just remain as gene damage not developing into cancer but increases the risk of developing cancer by adding one more step at some point in the sequence of events occurring toward cancer.

Sensitivity of a cell to radiation induced damage depends on the timing in a cell cycle. The cell cycle refers to the series of events that take place in a cell leading to its division and DNA duplication (Fig. 5.17). The cell cycle includes four phases: mitosis (denoted as M), pre-DNA synthesis phase (denoted as G1, i.e., Gap 1)), DNA synthesis phase (denoted as S), and post-DNA synthesis phase (denoted as G2, i.e., Gap 2)). The shortest cell cycle time could be 9 to 10- hours (observed in crypt cells in the intestinal epithelium), but some stem cells could have long cell cycle time up to 200 hours. The difference in cell cycle time mainly depends on the length of G1, the pre-DNA synthesis phase.

Sensitivity of cells to a radiation induced damage during cell cycle depends mainly on the level of sulfhydryl compounds in the cell. Sulfhydryls are a scavenger of free radicals, known as one of the radioprotectors. Presence of sufhydryl

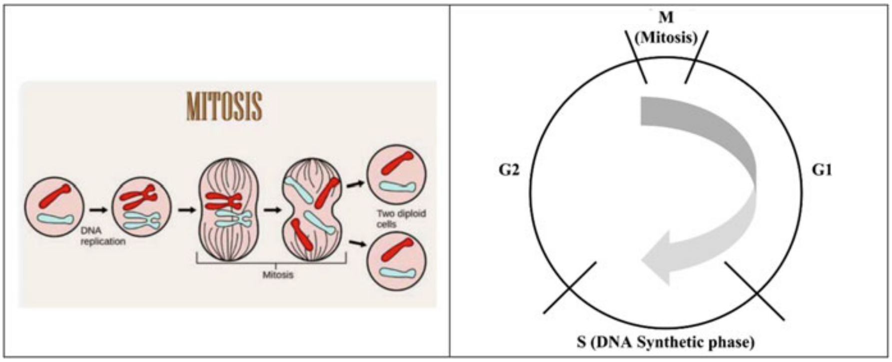


Fig. 5.17 The cell cycle. (Sources: left – Bio Explorer 2019)

compounds can minimize the damages caused by chemical reactivity of free radicals in the cell. The level of sulfhydryls tends to be highest in S and lowest near M. In terms of cell killing, cells are most radiosensitive in M and G2 and most resistant in late S. In terms of mutagenesis, G1 is the most sensitive phase.

5.2 Risk Assessment of Radiation Exposure

Instituting proper measures of human protection against ionizing radiation requires understanding of the risk of radiation exposure. Quantification of such risk is necessary to establish limits on the exposure to radiation as well as to inform members of the public about the significance of their exposure.

5.2.1 *Cancer Risk Estimation for Human Radiation Exposure*

In quantitative assessment of risk (hereafter called risk assessment) of radiation exposure, cancer is the main focus. This is because cancer represents the most important outcome of health effects. In addition to cancer, radiation exposure has been demonstrated to increase the risk of some benign tumors or other diseases, such as cardiovascular disease. However, direct evidence of increased risk of noncancer diseases has not been fully understood at low dose levels with lack of data to support risk quantification. Therefore presently radiation risk assessment focus only on cancer.

Risk assessment is based on the use of relevant data. Ideally the data should come from humans who were exposed to radiation. However, obtaining such data is very problematic or not possible for practical and ethical reasons. Use of animal data could be considered, but animal data does not necessarily represent how radiation may affect biological and physiological responses in human body. Using animal data may be exercised only when relevant human data is not available.

Human data on radiation induced cancer are available from unfortunate events of radiation exposure in the past. The largest body of such data comes from the atomic bomb survivors' study in Japan. This study is called the Atomic Bomb Survivor Study (ABSS). The other data come from various episodes of occupational radiation exposure or medical treatments with radiation. Studies of the people exposed to hazardous agent to determine adverse health outcomes are called epidemiology. The epidemiological studies conducted to people exposed to high levels of ionizing radiation are summarized in Table 5.5.

The ABSS is the most important source of data for developing risk estimates of ionizing radiation. While the study is an outcome of a very tragic wartime event, the developed data includes a large, relatively healthy population at the time of exposure

Table 5.5 Important epidemiological studies with quantitative estimation of dose to specific organs and published estimates of risk

Outcome	Type of Exposure	Study
Cancer mortality	Atomic bomb explosion	Atom bomb survivors
	Radiation therapy treatment for benign disease	Ankylosing spondylitis; benign gynecologic disorders; peptic ulcer
	Occupational	Radium dial painters; underground miners; nuclear workers
Cancer incidence	Atomic bomb explosion	Atom bomb survivors
	Radiation therapy treatment for malignant disease	Cervical cancers; childhood cancers; Breast disease; endometrial cancer; Hodgkin's disease
	Radiation therapy treatment for benign disease	Breast disease; tinea capitis; thymus; Tonsils; bone disease; hyperthyroidism
	Diagnostic procedure	Tuberculosis; Thorotrast; thyroid

with all ages and both gender covered. The study also includes a wide range of doses estimated on an individual basis. The control group consists of people who were present in Hiroshima or Nagasaki at the time of bombing receiving only small doses of radiation (less than 5 mGy, considered as the background dose level, by being far away from the hypocenters protected by buildings or other structures). The population include about a total of 76,000 people. They have been followed over their entire lifetime (and their offspring) till now and the number of cancers and the related deaths associated with radiation has been derived from these people. The types of cancers examined include: leukemia, stomach, colon, liver, lung, prostate, breast, ovary, bladder, and others.

Other sources of data of epidemiological studies are: (1) patients treated with radiation for various diseases, (2) workers exposed to radiation while working in their job, or (3) patients receiving radiation exposure through medical diagnostic procedures for tuberculosis or thyroid malfunction. Quantitative estimation of dose to specific organs is often available in these studies but quality of the data are not uniform nor controlled. Those diseases treated with radiation include ankylosing spondylitis, benign gynecologic disorders, peptic ulcer, cervical cancers, childhood cancers, breast disease, endometrial cancer, Hodgkin's disease, tinea capitis, thymus, hyperthyroidism, etc. These treatments were made without understanding the hazard of radiation with an expectation that ionizing radiation can cure most of these diseases. Also those workers receiving radiation in their occupation include radium dial painters, underground miners, and workers in national nuclear R&D. While the information covers various studies among diverse groups of people, the data quality was not consistent with lack of records on the dose delivery for individuals. Thus utilizing these studies for risk assessment was problematic. Accordingly these studies were used mainly for comparison purposes with the ABSS but not directly used in developing quantitative estimates of risk.

5.2.2 Calculation of Dose

Before we continue the discussion on risk assessment, the concept of dose needs to be clarified. The term, dose, represents the energy deposited in mass. As the energy of radiation is transferred from the incident radiation and deposited into the interacting medium, the resulting energy deposition would control the extent of the damage in the target materials. Therefore, dose is used as the quantity to represent or limit radiation damage (e.g., cancer risk) associated with radiation exposure.

5.2.2.1 The Absorbed Dose

The first quantity to be defined in the estimation of dose is the absorbed dose. The absorbed dose specifies the amount of energy absorbed per unit mass of material from radiation interaction with the matter. More specifically, it is the average energy imparted to the mass contained in an incremental volume of a specified material. The unit for absorbed dose is gray, written as Gy. 1 gray represents 1 joule of energy deposited in 1 kg of mass. 1 Gy is also equal to 100 rad. Rad (radiation absorbed dose) is another (older) unit of absorbed dose. 1 rad is equal to 100 ergs of energy deposition per gram of mass (1 erg $= 10^{-7}$ J).

5.2.2.2 The Equivalent Dose

The absorbed dose does not take into account the fact that radiation of different types can have different biological effectiveness due to the differences in physical characteristics such as LET. The equivalent dose is a modified dose concept introduced for the consideration of biological effect. The equivalent dose is obtained by multiplying the absorbed dose with the radiation weighting factor (the weighting factors of different radiation received). The radiation weighting factor reflects the differences in biological effectiveness depending on the type and energy of the radiation. As shown in Table 5.6, the value of radiation weighting factor of gamma rays or electrons is 1. For α particles, the value is 20.

The equivalent dose H_T is always defined for a specific organ or tissue type and is given by

$$H_T = w_R \times D_{T,R} \tag{5.2}$$

where w_R is the radiation weighting factor and $D_{T,R}$ is the average absorbed dose in organ (or tissue T) from a given type of radiation R.

When the radiation field is composed of multiple radiations with different types, the equivalent dose is the summation of all the incremental, average doses due to

Table 5.6 Radiation weighting factors (W_R) and quality factor (Q) for calculation of equivalent dose (or dose equivalent)

Radiation type and energy	w_R (ICRP 103) 2007	w_R (ICRP 60) 1990
X-rays, g-rays, electrons, positrons, and muons	1	1
Neutrons, E < 10 keV	Cont. Function[a]	5
Neutrons, 10 keV to 100 keV	Cont. Function[a]	10
Neutrons, 100 keV to 2 MeV	Cont. Function[a]	20
Neutrons, 2 MeV to 20 MeV	Cont. Function[a]	10
Neutrons, E > 20 MeV	Cont. Function[a]	5
Protons, other than recoils, > 2 MeV	2	5
Alpha particles, fission fragments, and relativistic heavy nuclei	20	20

aContinuous function for neutron radiation weighting factor

each of the component radiations. In the summation, the radiation weighting factors, w_R, are again used as weights:

$$H_T = \sum_R w_R D_{T,R} \tag{5.3}$$

The unit for the equivalent dose is Sievert, abbreviated as Sv. 1 Sv is equal to 1 Gy times the radiation weighting factor of one, corresponding to 1 joule energy deposition per 1 kg by a radiation with radiation weighting factor of one. Another (older but still widely used) unit for equivalent dose is rem which is equal to 0.01 Sv.

Calculation of radiation weighting factor is based on determining so-called relative biological effectiveness of the radiation particle as a function of LET. The RBE is the ratio of the amount of energy required to produce the same biological effect (e.g., 10% survival of the irradiated cells) between a reference radiation (200-keV x-rays) and the radiation under consideration. For example, if it takes 1 Gy of neutrons to kill 90% of cells while x-ray produces the same end point with 5 Gy of absorbed dose, the RBE of neutrons with respect to x-rays is 5/1 = 5.0. While using RBE as a basis of comparing biological effectiveness of different types of radiation is problematic as the value of RBE varies with the changes in the endpoint chosen, the value of radiation weighting factor for a given radiation represents the conservative estimate of the RBE by selecting the values to be greater than the bulk of experimental values observed for the most sensitive cell types under irradiation. In reality, RBE is directly controlled by LET which also varies as the energy of the radiation changes. Therefore, the radiation weighting factor is a function of particle energy from a theoretical point of view.

In the past, the concept of dose equivalent was used in place of the equivalent dose. The equivalent dose is based on an average absorbed dose in the tissue or organ (D_T) and weighted by the radiation weighting factor (W_R). The dose equivalent is based on the absorbed dose at a “point” in tissue which is weighted by quality factors (Q). The quality factor and the radiation weighting factor are both to represent biological effectiveness of radiation but quality factor is defined for a point while radiation weighting factor is an organ or tissue averaged quantity. In terms of numerical values, the two are basically the same.

According to the ICRP (ICRP 1991), the relationship between LET (in keV/μm) and the Q values is given as $Q(L) = 1$ (when LET<10); $Q(L) = 0.32*\mathrm{L} - 2.2$ (when $10 \leq \mathrm{L} \leq 100$); $Q(L) = 300*\mathrm{L}^{-0.5}$ (when LET >100). Based on these relationships, the Q value will reach the maximum value of ~30 at around 100 keV/μm.

In the case of electrons, as the LET of electrons ranges between 12 keV/μm (at 1 keV) and 0.2 keV/μm (at 1 MeV), the Q value is generally lower than 1 but becomes higher than 1 at very low energy (e.g., ~ 1 keV). Thus using 1 as the radiation weighting factor of electrons is generally conservative but not for very low energy electrons. Also, the LET of alpha particles varies from 130 keV/μm (at 3 MeV) to 60 keV/μm (at 9 MeV). Therefore, the Q value of alpha particle is usually lower than 20, the assigned value of radiation weighting factor, but can be higher than 20 when the energy of the particle becomes lower (<~6 MeV). These observations indicate that the currently assigned values of the radiation weighting factors are not absolute but adequate approximate values reflecting reasonable degree of conservatism.

The radiation weighting factor for different types of radiation is given in Table 5.6. Also, the weighting factor for neutrons is a continuous function of energy according to the latest estimates (ICRP 103 report) as shown in Fig. 5.18.

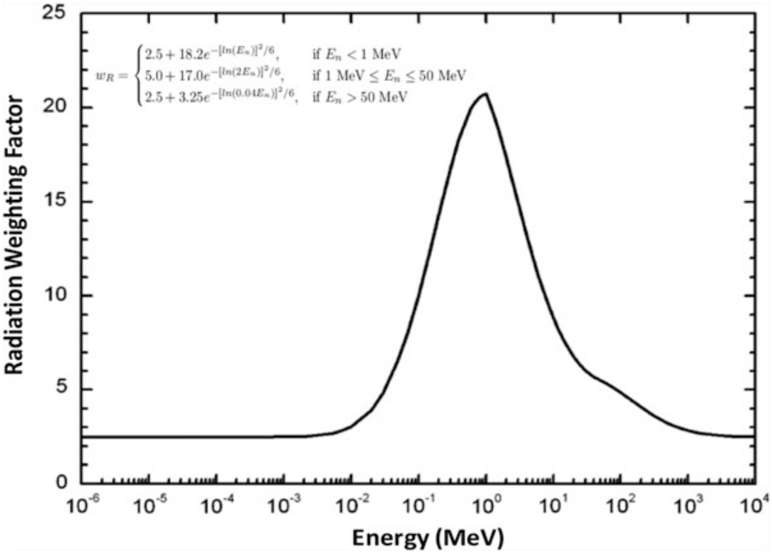


Fig. 5.18 Neutron radiation weighting factor, as a function of energy E_n (source: ICRP 2007)

5.2.2.3 The Effective Dose

Once the dose to various organs of an exposed person is determined as equivalent dose, these equivalent doses are combined to represent the whole body dose.

The whole body dose called the effective dose (H_E) is defined as an aggregated quantity by using tissue weighting factors. The effective dose is the weighted sum of the equivalent doses to various organs as shown below.

$$H_E = \sum_T w_T H_T = \sum_T w_T \cdot \sum_R w_R D_{T,R} \qquad (5.4)$$

The unit for effective dose is the same as the equivalent dose (i.e. *rem* or *Sv*).

In this aggregation process, the differences in the sensitivity of organs to radiation damage are taken into account through the use of so-called tissue (i.e., organ) weighting factor. The tissue weighting factors of various organs are derived from observing how many organ specific cancers appeared per given dose to the organs of the exposed individuals (mostly by using the results from ABSS). Therefore, the tissue weighting factors (w_T) represent the fraction of the cancer risk that is contributed by each of the body organs that are irradiated. The values of tissue weighting factors according to ICRP are given in Table 5.7. Development of the values of tissue weighting factor is explained below.

For the total combined dose of the exposed individuals at 10,000 person-Sv (which is called “collective dose”) to specific organs, incidence of excess cancers of the organs due to radiation can be measured. For incident, such effort led to the observation of an excess of about 90 lung cancers due to the dose of 10,000 person-Sv to the lungs. Considering that about 95% of the lung cancers induced by ionizing radiation are fatal, the risk of lung cancer death due to radiation exposures would be:

$$\begin{aligned}&\left(\frac{90\,excess\ lung\ cancers}{10,000\,person - Sv}\right) \cdot (0.95\,fatal\,fraction)\\ &= 85 \times 10^{-4}\,per\ Sv \text{ for lung cancer}\end{aligned}$$

Similarly, an excess of 50 leukemia and 80 thyroid cancers was observed from 10,000 person-Sv to the bone marrows and the thyroids, respectively. Assuming the fatality fraction of 0.99 and 0.1 for leukemia and thyroid cancer, respectively, the risk of death from leukemia and thyroid cancer can be calculated as,

$$\begin{aligned}&\left(\frac{50\,excess\ lung\ cancers}{10,000\,person - Sv}\right) \cdot (0.99\,fatal\,fraction)\\ &= 50 \times 10^{-4}\,per\ Sv \text{ for leukemia}\end{aligned}$$

Table 5.7 Values of tissue weighting factors and parameters considered based on ICRP recommendations (ICRP 1991; ICRP 2007; Moeller 1997, Table 14.4)

Tissue (organ)	Fatal cancer risk coefficient (10^{-4} Sv^{-1})	Cancer lethality fraction	Years of life lost (relative ratio)	Adjusted cancer risk coefficient (10^{-4} Sv^{-1})	Relative ratio of adjusted cancer risk coefficient	Tissue weighting factor (ICRP 60)	Tissue weighting factor (ICRP 103)
Gonads	–		20.0 (1.33)	133	0.18	0.20	0.08
Ovary	10	0.70	16.8 (1.12)	15	0.02		
Bone marrow	50	0.99	30.9 (2.06)	104	0.14	0.12	0.12
Colon	85	0.55	12.5 (0.83)	103	0.14	0.12	0.12
Lung	85	0.95	13.5 (0.90)	80	0.11	0.12	0.12
Stomach	110	0.90	12.4 (0.83)	100	0.14	0.12	0.12
Bladder	30	0.50	9.8 (0.65)	29	0.04	0.05	0.04
Breast	20	0.50	18.2 (1.21)	36	0.05	0.05	0.12
Liver	15	0.95	15.0 (1.21)	16	0.02	0.05	0.04
Esophagus	30	0.95	11.5 (0.77)	24	0.03	0.05	0.04
Thyroid	8	0.10	15.0 (1.00)	15	0.02	0.05	0.04
Bone surface	5	0.70	15.0 (1.00)	7	0.01	0.01	0.01
Skin	2	0.002	15.0 (1.00)	4	0.01	0.01	0.01
Salivary glands							0.01
Remainder	50	0.80	13.7 (0.91)	59	0.08	0.05	0.12
Total	500			725	1	1	1

$$\left(\frac{80\,excess\ lung\ cancers}{10,000\,person - Sv}\right)\cdot(0.10\,fatal\,fraction)$$
$$= 8\times 10^{-4}\ per\ Sv\ \text{ for thyroid cancer}$$

Similar calculations can be made for other body organs. The results of each fatal cancer risk coefficients can be added to give the total fatal cancer risk coefficients in all body tissues and organs. This is shown in Table 5.7. The calculated value of total fatal cancer risk coefficients was 500×10^{-4} per Sv. This total value can be used as denominator to determine the quotients as relative importance of each organ cancers. The resulting quotient values represent the relative sensitivity of the respective organs with respect to radiation-induced fatal cancer.

Additional consideration is also given to the fact that not only the death itself but also *the years of productive life lost* among the individuals who passed away is to be recognized. In addition, we note that even though a person survives a cancer, he or she has to experience *reduced quality of life*. These points can used to further adjust the values of tissue weighting factors

Consider an example of lung cancer. The cancer lethality fraction of lung cancer is 95%, therefore 5% survives but faces difficult life. This fraction is added back to the calculation to reflect life impairment. Also in comparison to 15 years as the average value of the years of productive life lost due to all cancer deaths, the years of life lost due to lung cancer is 13.5 years on average. The ratio, 13.5/15 = 0.9, is used to represent the relative importance of lung cancer with respect to representing the years of life lost from cancer death. Then, the resulting adjustment to the fatal cancer probability coefficient for lung cancer, 85×10^{-4} per Sv, becomes $\{(85 \times 10^{-4}\ /\mathrm{Sv})(1.0 + 0.05)\}*0.9 = 80 \times 10^{-4}/\mathrm{Sv}$

This adjustment can be made for all other cancer types. The sum of these adjusted values is about $725 \times 10^{-4}/\mathrm{Sv}$. Then the adjusted relative importance of lung cancer becomes, 80/725, or about 12%. This value implies that the risk and related consequences of developing lung cancer is about 12% of the total risk arising through the development of cancers in all of the organs if the entire body were exposed to ionizing radiation. Results of similar adjustments are shown in Table 5.7. Due to the uncertainties involved, the values were categorized into four groups of 0.20, 0.12, 0.05, and 0.01 as the suggested tissue weighting factors by the ICRP in 1991 (ICRP 1991).

In the 2007 ICRP 107 report (ICRP 2007), additional changes were suggested to the tissue weighting factors. The most significant change was with gonads/ovary with the new tissue weighting factor of 0.08, reduced from 0.20. This change reflects the findings that heritable effects of radiation were not evident among the survivors of atomic bombing. Other changes are minor adjustments considering the values of the relative ratio of adjusted cancer risk coefficient as shown in Table 5.7. The organs included as part of the remainder category are adrenals, extrathoracic region, gall bladder, heart, kidneys, lymphatic nodes, muscle, oral mucosa, pancreas, small intestine, spleen, thymus, prostate (male), and uterus/cervix (female). The tissue weighting factor for the remainder (0.12) applies to the arithmetic mean dose of the 13 organs for each gender.

5.2.2.4 The Collective Dose

Note that the concept of collective effective dose was employed in the calculation of fatal cancer risk coefficient in the derivation of tissue weighting factors. Collective dose is a term to express the integrated dose to a given population group. For example, if a population composed of 10,000 people each received an average whole-body dose of 1 Sv, the collective effective dose for the population is 10,000 person-Sv (or one million person-rem). If a population of 20,000 people each received an average dose of 0.5 Sv, the collective effective dose as total population dose would again be 10,000 person-Sv.

While the concept of collective effective dose is useful in evaluating the public health impact of radiation exposures, key assumption in its use is that the doses are sufficiently high to produce statistically significant stochastic effects (cancer) (NCRP 1995). The data used in the calculation mostly come from the ABSS with high enough doses among the exposed population (an exception is the use of miner data for lung cancer calculation with high enough individual doses). Therefore, the collective dose-based calculations performed in the analysis are valid. However, if the dose received by the exposed individuals is very small (<< 1 mGy), occurrence of cancer at the dose level is not expected and use of collective dose and related risk projections are not scientifically valid.

Different Concepts of Dose

The absorbed dose: the amount of energy absorbed per unit mass of material from radiation interaction with the matter.

The dose equivalent: the absorbed dose at a “point” in tissue or organ weighted by the quality factors (Q) for the radiation

The equivalent dose: the average absorbed dose in the tissue or organ weighted by the radiation weighting factor (W_R) for the radiation

The effective dose: the whole body dose as the weighted sum of the equivalent doses to various organs.

The collective dose: the total integrated dose among a given population group as the sum of the individual effective doses.

Example 5.2: Calculation of the Effective Dose

During one year, a worker receives 5.0 mGy from internally deposited alpha particles in the lung, 100 mGy from beta particles in the thyroid, and 10 mGy externally from uniform, whole-body gamma irradiation. What is the effective dose for this worker?

Solution:

First, determine the equivalent dose to each target organ by using, $H_T = \sum_R W_R \, D_{T,R}$

(continued)

- $H_{Lung} = 20 \text{ x } 5.0 \text{ (mGy)} = 100 \text{ mSv}$
- $H_{Thyroid} = 1 \text{ x } 100 \text{ (mGy)} = 180 \text{ mSv}$
- $H_{Whole\ Body} = 1 \text{ x } 10 \text{ (mGy)} = 10 \text{ mSv}$

For whole-body effective dose to the worker,

$$H_E = \Sigma w_T H_T = 0.12 \times 100 + 0.04 \times 180 + 1 \times 10 = 29.2 \text{ mSv}$$

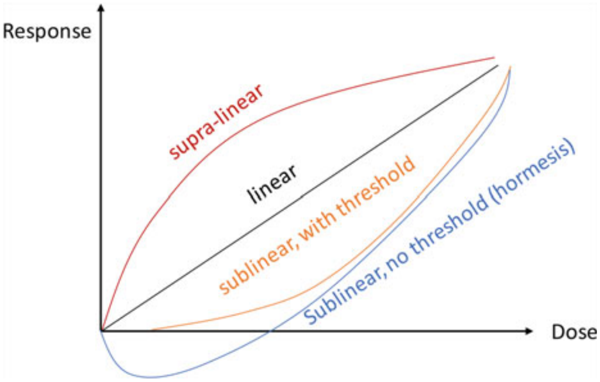
where ICRP 103 tissue weighting factors in Table 5.7 are used (for whole body, $w_T = 1$)

5.2.3 Dose Response Relationships

While radiation-induced cancer in humans is well documented resulting in the estimation of radiation induced cancer risk, the levels of radiation exposure experienced by the subjects in the study are much higher than the levels expected in our daily living or a typical occupational setting. In other words, the levels of radiation exposure under consideration for the protection of workers and the public should be much lower than what was experienced by the ABSS subjects. As the risk of radiation exposure at low doses has to be estimated from the compiled data measured at relatively high doses, extrapolation from high to low doses was necessary.

Such high-to-low dose extrapolation requires understanding of the underlying mechanism of radiation-induced cancer. However, due to the complexity in cancer mechanisms, large uncertainty remains in defining such mechanism. Accordingly, several hypotheses in proposing different types of extrapolation as dose-response curves have been suggested. Here, dose-response curve means graphical representation of the relationship between increase in dose and the response observed. Figure 5.19 shows four different types of such curves as linear, sub-linear, sub-linear with threshold, and supra-linear curves. The number of cancers at zero dose is higher

Fig. 5.19 General types of dose-response relationships for radiation-induced cancer



than zero due to the presence of natural cancer incidence (although this is not depicted in the figure). The linear curve implies that each delivery of particle energy by radiation leads to cancer response. Therefore, the linear curve assumes that every energy delivery creates ionization and subsequent DNA damages disrupting cell growth control towards cancerous development. This is an overly simplistic but a conservative approach. It assumes that the probability of a detrimental response (cancer) is present at any level of exposure with the response linearly proportional to dose.

The sub-linear curve represents the case where energy delivery by radiation is less effective in causing damage at lower dose but becomes more and more effective at higher doses. This may be the case if the DNA repair mechanism is effective at low dose but becomes less effective at high doses (as the system may become overwhelmed).

If such phenomenon occurs only when the dose level is higher than a certain level (i.e., threshold), the curve becomes the sub-linear curve with a threshold. In this case, below a certain level of dose, there is no observed response. This curve may even imply beneficial effect (so-called, the hormesis effect) of radiation at least at some low dose levels. Beneficial effect of radiation may occur if radiation stimulates normal cells with respect to cell proliferation, cellular repair mechanisms, or immune responses. Stimulatory and beneficial effect of toxic chemical agents (e.g., nickel, zinc) have been observed when they are given in small doses. Lower spontaneous cancer incidence rates have also been noticed among the people living in areas of very high natural background radiation. However, such beneficial effect was not observed among the atomic bomb survivors.

The supra-linear curve assumes that the energy delivered by radiation is very effective at low dose but effectiveness of additional dose delivery decreases at higher dose. This implies that at high dose the delivery of additional energy is wasted (leading to an overkill).

Experimental studies using animals have also been used to identify the dose-response relationship for cancer induction from gamma irradiation. The results more or less followed a linear-quadratic, no-threshold function: The response has both a linear and a quadratic proportionality to dose and a detrimental response occurs at any level of exposure. The linear-quadratic model yields slightly lower risk estimates compared with the results from a linear relationship.

In the risk assessment made by the United Nations Scientific Committee on Atomic Radiation (UNSCEAR) in 1988 (UNSCEAR 1988) and the Biological Effects of Ionizing Radiation (BEIR) Committee of the U.S. (NAS 1990; NAS 2006), a linear model was adopted for all radiation-induced cancer deaths, except for leukemia. For leukemia, a linear-quadratic model was assumed. In these models, the dose-response function, f(d) as a function of the dose is represented as $f(d) = \alpha_1 d$ ("linear") or $f(d) = \alpha_2 d + \alpha_3 d^2$ ("linear-quadratic").

5.2.4 Relation Between Cancer by Natural Incidence and Radiation Induced Cancer

One of the key questions to address in radiation risk estimation using epidemiologic data is whether radiation induced cancer has any relationship with the occurrence of cancer from natural causes. Some people have argued that there is no relationship and that cancer incidence due to radiation exposure is independent of cancer incidence from natural causes.

These two different approaches are captured by the use of so-called the absolute (additive) risk model or the relative (multiplicative) risk model. In the absolute risk model, the risk associated with radiation is independent of the background risk, i.e., the spontaneous occurrence of the same disease in the absence of radiation exposure. Thus, the cancer risk from radiation is additional to the natural cancer incidence. This additive model, i.e., the absolute risk model is given as,

$$\gamma(d) = \gamma_0 + f(d)g(\beta) \tag{5.5}$$

where, $\gamma(d)$ is the total cancer risk, not the excess cancer risk, γ_0 denote the risk of natural cancer death for an individual at a given age, $f(d)$ is the dose response function as explained in the previous section (the unit for d is Sv), and $g(\beta)$ captures the effects of other factors (e.g., gender, age at exposure, time since exposure) on radiation-induced cancer death. The excess cancer risk from radiation is obtained by subtracting the natural cancer risk, i.e. $\gamma(d)$ -γ_0,

The relative risk model assumes that the risk of radiation-induced cancer increases with the age-specific baseline rate of cancer. Thus, radiation exposure causes additional damage building on the damage caused by natural causes. This implies that radiation affects biological factors that serve as promoters of cancer in exposed individuals. Accordingly, the risk of radiation-induced cancer increases with the age of an exposed individual in parallel with the risk of cancer death in general. In the relative risk model, the excess risk of cancer due to radiation is proportional to the spontaneous risk. This multiplicative model, i.e., the relative risk model is given as,

$$\gamma(d) = \gamma_0 + \gamma_0 f(d)g(\beta) = \gamma_0 \left[1 + f(d)g(\beta)\right] \tag{5.6}$$

Both the absolute and the relative risk models were separately used in the report published by National Academy of Science of the U.S. in 1972 and 1980, called BEIR I (NAS 1972) and III (NAS 1980) report, respectively, to estimate the lifetime risk of cancer from radiation exposure. In the 1990 BEIR V report, only the relative risk model was adopted (NAS 1990). Then in the 2006 BEIR VII report (NAS 2006), both the absolute and the relative risk models were used together (with the assigned weights of 0.3 and 0.7, respectively) for most cancer types in the logarithmic space. For lung cancer, however, the weights were reversed (0.3 to the relative risk model and 0.7 to the absolute risk model). Also, for thyroid cancer, only the relative risk model was used while for breast cancer, the absolute risk model was preferred. In the UNSCEAR report, the 1977 report (UNSCEAR 1977) used only the absolute risk model and the 1988 report (UNSCEAR 1988) used both the absolute and the relative risk models.

5.2.5 *Cancer Risk Estimation in BEIR V and BEIR VII*

The populations of Hiroshima and Nagasaki who survived through atomic bomb explosion in 1945 were the subjects of the largest epidemiologic study (i.e., ABSS) of radiation health effects. Among about 195,000 survivors residing in two cities, a total of 75,991 people with reconstructed dose information available were included in the study and followed throughout their lifetime. Among them, 34,272 of the survivors who were far away from the hypocenters became a "control" group as their radiation doses were comparable to the natural background level (i.e., less than 0.005 Gy (0.5 rad)). The remaining 41,719 people formed different dose groups for the examination of dose-response relationship. The data from these people were further stratified by gender, city of residence, ten dose groups, and five-year intervals of attained age, age at exposure, time since exposure, and the number of cancers observed for each cancer. Table 5.8 shows the makeup of the cohorts at different dose groups.

The dose to twelve specific organs of the survivors were also estimated. Based on the observed number of cancers in the control group and different dose groups, the number of excess cancers due to radiation was estimated. In this estimation, the probability of observing cancer deaths in each stratified cell was modeled by using the Poisson distribution. The values of the unknowns in the assumed dose-response models were obtained by fitting the models while controlling other risk related factors, such as, gender, attained age, age at exposure and time after exposure was performed by using the technique of the maximum likelihood estimation.

5.2.5.1 BEIR V Results

The result of the fitted models in BEIR V report (NAS 1990) for different cancers are given below. In the equations, d is dose in Sv, T is years after exposure, and E is age at exposure. I(T) and I(S) are called the indicator functions. The indicator function $I(T)$ is defined as 1 if what is inside the parenthesis is true and 0 if not, e.g. $I(T < 10)$ is 1 if T < 10 and 0 otherwise. For the indicator function $I(S)$, it is 1 if female and 0 if male.

Table 5.8 Number of Cohorts in Atomic Bomb Survivors Study at each dose group

Kerma (Gy)	0-0.005	0.006-0.05	0.06-0.09	0.10-0.99	1.00-1.99	2.00+	**Total**
Cohort size	34,272	19,192	4129	15,346	1949	1106	**75,991**

Leukemia:

$$\begin{aligned} \mathrm{f(d)} &= 0.243\ \mathrm{d} + 0.271\ \mathrm{d}^2 \\ g(\beta) &= \begin{cases} exp\,[4.885 \cdot I(T \leq 15) + 2.380 \cdot I(15 < T \leq 25)] if\ E \leq 20 \\ exp\,[2.367 \cdot I(T \leq 25) + 1.638 \cdot I(25 < T \leq 30)] if\ E > 20 \end{cases} \end{aligned} \tag{5.7}$$

Respiratory cancer:

$$\begin{aligned} \mathrm{f(d)} &= 0.636\ \mathrm{d} \\ g(\beta) &= exp\left[-1.437 \cdot ln\left(\frac{T}{20}\right) + 0.711 \cdot I(S)\right] \end{aligned} \tag{5.8}$$

Breast cancer:

$$\begin{aligned} \mathrm{f(d)} &= 1.220\ \mathrm{d} \\ g(\beta) &= \begin{cases} exp\left[1.385 - 0.104 \cdot ln\left(\frac{T}{20}\right) - 2.212 \cdot ln^2\left(\frac{T}{20}\right)\right] & if\ E \leq 15 \\ exp\left[-0.104 \cdot ln\left(\frac{T}{20}\right) - 2.212 \cdot ln^2\left(\frac{T}{20}\right) - 0.0628(E - 15)\right] & if\ E > 15 \end{cases} \end{aligned} \tag{5.9}$$

Digestive cancer:

$$\begin{aligned} \mathrm{f(d)} &= 0.809\ \mathrm{d} \\ g(\beta) &= exp\,[0.553 \cdot I(S) + \sigma_E] \\ \sigma_E &= \begin{cases} 0 & if\ E \leq 25 \\ -0.198\ (E - 25) & if\ 25 < E \leq 35 \\ -1.98 & if\ E > 35 \end{cases} \end{aligned} \tag{5.10}$$

Other cancers:

$$\begin{aligned} \mathrm{f(d)} &= 1.220\ \mathrm{d} \\ g(\beta) &= \begin{cases} 1 & if\ E \leq 10 \\ exp\,[-0.0464(E - 10)] & if\ E > 10 \end{cases} \end{aligned} \tag{5.11}$$

Examining the coefficients of the linear term in these dose response relationships, *f(d)*, can provide approximate insight on the relative likelihood of different organ

Table 5.9 BEIR V estimates of excess cancer mortality for single exposure and continuous exposure, as lifetime risk per 100,000 exposed persons

	Male			Female		
Exposure type	Total	Nonleukemia	Leukemia	Total	Nonleukemia	Leukemia
Single exposure to 0.1 Sv (10 rem)	770	660	110	810	730	80
Continuous lifetime exposure to 1 mSv/y (0.1 rem/y)	520	450	70	600	540	60

cancer occurrences. For example, the largest value of the coefficients was with breast cancer and other cancers (i.e., thyroid cancer) at 1.220 which is about five times higher than the corresponding value for leukemia (0.243). This implies that breast cancer or thyroid cancer are fives time more prevalent than leukemia among the exposed.

To estimate the excess number of cancers among general population, the fitted risk models were applied to a stationary population with known death rates from natural causes. In the BEIR V report, the population used was the U.S. population with death rate data for 1979 through 1981. By using such death rate data as background data, i.e., γ_0, in the equation $\gamma(d) = \gamma_0[1 + f(d)g(\beta)]$ and using the fitted models for $f(d)$ and $g(\beta)$, the expected number of excess cancer death was calculated for various age groups.

The resulting excess cancer death estimates are shown in Table 5.9. The estimates assume an equal dose to all organs. The non-leukemia cancer represents sum of respiratory, breast, digestive, and other cancers.

Application of the above cancer risk estimates to the general population is not straight forward as the general population are exposed to radiation through chronic long-term exposure (i.e., at low dose rate) at low dose levels. Thus extrapolation of the risk estimates from high dose and high dose rate exposures experienced by the ABSS subjects to low dose and low dose rate exposure is needed. It turns out that the risk estimates for leukemia from the ABSS subjects contain an implicit dose rate reduction factor, eliminating the need for the consideration of dose rate issue. However the low dose rate effect needs to be taken into account for solid cancer (non-leukemia) risk estimates.

The BEIR V committee recommended a factor of two as the dose rate effectiveness factor (DREF) for solid (non-leukemia) cancers. Using the data from Table 5.9 for a single exposure, applying the DREF of 2 to solid cancers results in the cancer risk are estimated to be $4.4 \times 10^{-2}\ \mathrm{Sv}^{-1}$ for males and $4.45 \times 10^{-2}\ \mathrm{Sv}^{-1}$ for females, as calculated below.

For male, $\frac{(660/2+110)\ \textit{excess cancer mortality}}{0.1\ Sv \times 100,000\ \textit{persons}} = 0.044\ \textit{cancer risk per Sv}$

For female, $\frac{(730/2+80)\ \textit{excess cancer mortality}}{0.1\ Sv \times 100,000\ \textit{persons}} = 0.0445\ \textit{cancer risk per Sv}$

Then the probability of fatal cancer from a single exposure (as an average between male and female) for the general population is about 4.425×10^{-2} per Sv^{-1}.

Likewise, for a continuous exposure, the probability of fatal cancer from radiation is calculated as follows.

For male, $\frac{(450/2+70)\ excess\ cancer\ mortality}{1mSv/y \times 100,000\ persons} = 0.00295\ cancer\ risk\ per\ mSv/\text{y}$

For female, $\frac{(540/2+60)\ excess\ cancer\ mortality}{1mSv/y \times 100,000\ persons} = 0.0033\ cancer\ risk\ per\ mSv/\text{y}$

Thus the average value of cancer fatality risk for a continuous exposure among the general population would be 3.1×10^{-3} per mSv/yr.

5.2.5.2 BEIR VII Results

The latest results of risk estimation on radiation induced cancer are available from the 2006 BEIR VII report (NAS 2006). In comparison to BEIR V, the risk estimates were based on largely expanded epidemiologic data for the ABSS subjects including cancer incidence data and 15 additional years of follow-up. Due to the availability of data, risk estimates were made for 11 specific cancer sites. Results indicated that the lifetime risk estimate for solid cancer mortality (averaged over two genders) is $5.1 \times 10^{-2}\ \text{Gy}^{-1}$. This result is similar to that of BEIR V. The newly estimated leukemia risk from radiation exposure did not change significantly from BEIR V.

The study found that genetic effects of radiation (low-dose, low-LET radiation) are very small compared to baseline frequencies of genetic disease. The updated molecular and cellular data did not support the hormesis theory according to the BEIR VII study.

Results of the study are summarized for single radiation exposure (Table 5.10 for mortality, Table 5.11 for morbidity) as well as for chronic exposure (Table 5.12).

Example 5.3: Estimating the Risks of Cancer from Chronic Exposure to Radiation

A male is exposed to natural background of 0.004 Gy (4 mGy) per year throughout life. Based on the results of the BEIR VII report, estimate the lifetime risk of solid cancer, leukemia, and death from cancer for the person. Compare the result with the estimates from the BEIR V report.

Solution:

Based on the results given in Table 5.12, the estimated risk of a male being diagnosed with a solid cancer = 4 mGy· (554 per 100,000 per 1 mGy) = 2.216×10^{-2} (~ 1 in 45)

(continued)

Table 5.10 BEIR VII lifetime attributable risk of cancer mortality (from single exposure) for the general population (NAS 2006)

	Age at exposure (years)										
Cancer site	0	5	10	15	20	30	40	50	60	70	80
Males											
Stomach	41	34	30	25	21	16	15	13	11	8	4
Colon	163	139	117	99	84	61	60	57	49	36	21
Liver	44	37	31	27	23	16	16	14	12	8	4
Lung	318	264	219	182	151	107	107	104	93	71	42
Prostate	17	15	12	10	9	7	6	7	7	7	5
Bladder	45	38	32	27	23	17	17	17	17	15	10
Other	400	255	200	162	134	94	88	77	58	36	17
All solid	1028	781	641	533	444	317	310	289	246	181	102
Leukemia	71	71	71	70	67	64	67	71	73	69	51
All cancers	**1099**	**852**	**712**	**603**	**511**	**381**	**377**	**360**	**319**	**250**	**153**
Females											
Stomach	57	48	41	34	29	21	20	19	16	13	8
Colon	102	86	73	62	53	38	37	35	31	25	15
Liver	24	20	17	14	12	9	8	8	7	5	3
Lung	643	534	442	367	305	213	212	204	183	140	81
Breast	274	214	167	130	101	61	35	19	9	5	2
Uterus	11	10	8	7	6	4	4	3	3	2	1
Ovary	55	47	39	34	28	20	20	18	15	10	5
Bladder	59	51	43	36	31	23	23	22	22	19	13
Other	491	287	220	179	147	103	97	86	69	47	24
All solid	1717	1295	1051	862	711	491	455	415	354	265	152
Leukemia	53	52	53	52	51	51	52	54	55	52	38
All cancers	**1770**	**1347**	**1104**	**914**	**762**	**542**	**507**	**469**	**409**	**317**	**190**

Note: Number of deaths per 100,000 persons exposed to a single dose of 0.1 Gy
These estimates are obtained as combined estimates based on relative and absolute risk transport and have been adjusted by a DDREF of 1.5, except for leukemia, which is based on a liner-quadratic model

Example 5.3 (continued)

The lifetime risk of being diagnosed with leukemia = 4 · (67 per 100,000) = 2.68×10^{-3} (~ 1 in 370)

The lifetime risk of dying of cancer = 4 · (332 per 100,000) = 1.328×10^{-2} (~ 1 in 75)

From BEIR V, the average value of cancer fatality risk for a continuous exposure is 3.1×10^{-3} per mSv/yr.

Therefore, the lifetime risk of dying of cancer according to BEIR V = 4 · $3.1 \times 10^{-3} = 1.21 \times 10^{-2}$ (~ 1 in 83)

Table 5.11 BEIR VII lifetime attributable risk of cancer incidence (from single exposure) for the general population (NAS 2006)

	Age at Exposure (years)										
Cancer site	**0**	**5**	**10**	**15**	**20**	**30**	**40**	**50**	**60**	**70**	**80**
Males											
Stomach	76	65	55	46	40	28	27	25	20	14	7
Colon	336	285	241	204	173	125	122	113	94	65	30
Liver	61	50	43	36	30	22	21	19	14	8	3
Lung	314	261	216	180	149	105	104	101	89	65	34
Prostate	93	80	67	57	48	35	35	33	26	14	5
Bladder	209	177	150	127	108	79	79	76	66	47	23
Other	1123	672	503	394	312	198	172	140	98	57	23
Thyroid	115	76	50	33	21	9	3	1	0.3	0.1	0
All solid	2326	1667	1325	1076	881	602	564	507	407	270	126
Leukemia	237	149	120	105	96	84	84	84	82	73	48
All cancers	**2563**	**1816**	**1445**	**1181**	**977**	**686**	**648**	**591**	**489**	**343**	**174**
Females											
Stomach	101	85	72	61	52	36	35	32	27	19	11
Colon	220	187	158	134	114	82	79	73	62	45	23
Liver	28	23	20	16	14	10	10	9	7	5	2
Lung	733	608	504	417	346	242	240	230	201	147	77
Breast	1171	914	712	553	429	253	141	70	31	12	4
Uterus	50	42	36	30	26	18	16	13	9	5	2
Ovary	104	87	73	60	50	34	31	25	18	11	5
Bladder	212	180	152	129	109	79	78	74	64	47	24
Other	1339	719	523	409	323	207	181	148	109	668	30
Thyroid	634	419	275	178	113	41	14	4	1	0.3	0
All solid	4592	3265	2525	1988	1575	1002	824	678	529	358	177
Leukemia	185	112	86	76	71	63	62	62	57	51	37
All cancers	**4777**	**3377**	**2611**	**2064**	**1646**	**1065**	**886**	**740**	**586**	**409**	**214**

Note: Number of deaths per 100,000 persons exposed to a single dose of 0.1 Gy
These estimates are obtained as combined estimates based on relative and absolute risk transport and have been adjusted by a DDREF of 1.5, except for leukemia, which is based on a liner-quadratic model

Example 5.4: Cancer Risk Assessment from Radiation Exposure

Suppose a female subject receives at age 22 an accidental occupational whole-body absorbed dose of 5 rad due to low-LET radiation. At age 32, the subject receives a series of diagnostic x-rays resulting in a dose of 5 rad to the organs of the digestive system. At age 52 the subject is diagnosed to have stomach cancer expected to be fatal.

Using the results from BEIR V report, give your assessment as to the probability of cancer causation, that is, the likelihood that the cancer is caused by occupational radiation exposure, to medical radiation exposure, or to

(continued)

Table 5.12 BEIR VII Lifetime attributable risk of solid cancer incidence and mortality from chronic exposure among the general population (NAS 2006)

	Incidence: Exposure Scenario		Mortality: Exposure Scenario	
Cancer Site	1 mGy/yr. throughout life	10 mGy/yr. from ages 18 ~ 65	1 mGy/yr. throughout life	10 mGy/yr. from ages 18 ~ 65
Males				
Stomach	24	123	13	66
Colon	107	551	53	273
Liver	18	93	14	72
Lung	96	581	99	492
Prostate	32	164	6.3	32
Bladder	69	358	16	80
Other	194	801	85	395
Thyroid	14	28		
All solid	554	2699	285	1410
Leukemia	67	360	47	290
All cancers	**621**	**3059**	**332**	**1700**
Females				
Stomach	32	163	19	94
Colon	72	368	34	174
Liver	8.7	44	8	40
Lung	229	1131	204	1002
Breast	223	795	53	193
Uterus	14	19	3.5	18
Ovary	29	140	18	91
Bladder	71	364	21	108
Other	213	861	98	449
Thyroid	75	139		
All solid	968	4025	459	2169
Leukemia	51	270	38	220
All cancers	**1019**	**4295**	**497**	**2389**

Note: Number of deaths per 100,000 persons exposed to 1 mGy /yr. throughout life or 10 mGy/yr. from ages 18 to 64

These estimates are obtained as combined estimates based on relative and absolute risk transport and have been adjusted by a DDREF of 1.5, except for leukemia, which is based on a liner-quadratic model

Example 5.4 (continued)

natural causes. Assume that the baseline digestive cancer mortality risk from natural causes for a female at age 52 is 35.9 per 100,000 people per year.

Solution:

$$\text{Baseline stomach cancer mortality risk} = \gamma_0 = 35.9/100{,}000 = 3.59 \text{ x } 10^{-4}$$

(continued)

Example 5.4 (continued)

Find the total cancer risk for each event (i.e. from occupational and medical exposures as a function of dose and of age at the exposure.

Using the (Eq. 5.6) for relative risk model and (Eq. 5.10) for digestive cancer,

$\gamma(d) = \gamma_0\,[1 + f(d)g(\beta)]$ (the relative risk model)

Digestive cancer:

$$\mathrm{f(d)} = 0.809\ \mathrm{d}$$

$$g(\beta) = \mathit{exp}\,[0.553 \cdot I(S) + \sigma_E]$$

$$\sigma_E = \begin{cases} 0 & \mathit{if}\ E \le 25 \\ -0.198\,(E - 25) & \mathit{if}\ 25 < E \le 35 \\ -1.98 & \mathit{if}\ E > 35 \end{cases}$$

Because the BEIR V model includes risk from natural causes, the excess risk for each event must be determined before summing them up to find the relative risks:

$$\begin{aligned}\text{Excess risk for the event} = {} & \text{Total cancer risk for the event} \\ & - \text{Baseline cancer risk (natural)}\end{aligned}$$

$$\text{Excess risk} = \gamma(d) - \gamma_0$$

Age at Exposure (y)	*Exposure Event*	*f(d)*	σ_E	*g(β)*	*γ(d)*	*Excess Risk*
22	Occupational	2.427×10^{-2}	0	1.70403	3.738×10^{-4}	1.485×10^{-5}
32	Medical	2.427×10^{-2}	−1.386	0.42613	3.627×10^{-4}	3.713×10^{-6}

Thus, the relative cancer risk can be calculated by dividing the each calculated risk by the total risk

$$\begin{aligned}\text{Relative risk (natural)} &= \frac{3.59 \times 10^{-4}}{3.59 \times 10^{-4} + 1.485 \times 10^{-5} + 3.713 \times 10^{-6}} \\ &\quad \times 100\% \\ &= 95.1\%\end{aligned}$$

(continued)

Example 5.4 (continued)

$$\begin{aligned}\text{Relative risk (occupational)} &= \frac{1.485\times 10^{-5}}{3.59\ \ \times 10^{-4}+1.485\times 10^{-5}+3.713\times 10^{-6}}\\ &\quad\times 100\% \\ &= 3.9\%\end{aligned}$$

$$\text{Relative risk (medical)} = \frac{3.713\times 10^{-6}}{3.59\ \ \times 10^{-4}+1.485\times 10^{-5}+3.713\times 10^{-6}}\times 100\% = 1.0\%$$

5.2.5.3 Comparisons of the Estimated Cancer Risk Coefficients

As mentioned in Sect. 5.2.3, in addition to the efforts of the BEIR Committee of the U.S, the UNSCEAR Committee also performs analysis of the data from the ABSS and provides risk estimates of radiation-induced cancer. In fact, over the years, the UNSCEAR Committee published a series of reports on radiation-induced cancer in 1977 (UNSCEAR 1977), 1988 (UNSCEAR 1988), 2000 (UNSCEAR 2000), and 2006 (UNSCEAR 2006). The BEIR Committee also published the BEIR I report (NAS 1972) and the BEIR III report (NAS 1980) prior to the publication of the BEIR V and BEIR VII reports.

The estimates of cancer risk in these reports varied mostly due to the changes in the estimates of dose received by the subjects and the model employed for statistical data fitting (i.e., relative risk model vs. absolute risk model). In particular, the dose estimates used as the basis of dose-response analysis in the ABSS changed in 1987 and 1988 due to the new dosimetry study by the Radiation Effects Research Foundation. Prior to 1987, the studies were based on a dosimetry system developed in the mid-sixties. The new dose estimates were based on reassessment of A-bomb dosimetry using detailed calculation of neutron and gamma ray transport through weapons materials and the air followed by accurate shielding calculations within Japanese houses and organ dose calculations for the survivors having various shielding circumstances, location, orientation, and physical size. The new dosimetry system gave lower estimates of dose received by the survivors resulting in higher values of cancer risks.

An international advisory body, International Council on Radiation Protection (ICRP) and the U.S. advisory body, National Council on Radiation Protection and Measurements (NCRP) also publish reports on risk estimates of radiation-induced cancer based on independent review of the results from the UNSCEAR and the BEIR Committee reports. The latest work from the ICRP (ICRP 2007) reviewed the results in the BEIR VII report (NAS 2007), the UNSCEAR 2000 report (UNSCEAR 2000), and the NCRP 36 report (NCRP 2001) and recommended the values of cancer risk estimates as shown in Table 5.13. The table also includes the values recommended in the previous version of ICRP review (ICRP 1991) for comparison.

Table 5.13 also shows the risk estimates of heritable effects of radiation. It turns out that there are no statistically demonstrable adverse heritable effects attributable

Table 5.13 Risk estimates of fatal cancer and heritable damage due to non-chronic radiation exposure for working population and the general public

Exposed population	Risk of Fatal Cancer (Sv^{-1})		Risk of Heritable Effects (Sv^{-1})		Total Risk (Sv^{-1})	
	ICRP103	ICRP60	ICRP103	ICRP60	ICRP103	ICRP60
Adult – working population: 20 to 64 y of age	4.1×10^{-2}	4.8×10^{-2}	0.1×10^{-2}	0.8×10^{-2}	4.2×10^{-2}	5.6×10^{-2}
Whole – whole population: 0 to 90 y of age	5.5×10^{-2}	6.0×10^{-2}	0.2×10^{-2}	1.3×10^{-2}	5.7×10^{-2}	7.3×10^{-2}

to radiation exposure among the survivors of the atomic bomb explosions in Japan (NAS 1990; NAS 2006). The estimates given in the table is based on the data obtained from studies using animal (mice).

As shown in the table, in the ICRP 60 (1991) report, the estimate of total detriment from radiation-induced stochastic effects (fatal cancer plus heritable effects) is 5.6×10^{-2} Sv^{-1} for radiation workers and 7.3×10^{-2} Sv^{-1} for the whole population. In the ICRP 103 report, these estimates were revised as 4.2×10^{-2} Sv^{-1} for radiation workers and 5.7×10^{-2} Sv^{-1} for the whole population. These estimates indicate that the risk of fatal cancer from radiation exposure is about 5% per Sv of dose.

The estimated cancer risk coefficient of 0.005% per mSv of radiation dose is often used along with the quantification of collective effective dose among population to project fatalities resulting from the release of radionuclides from nuclear facilities. However, such projection is not scientifically valid and should not be used if the dose received by the individuals is much less than 1 mGy. In such approach, the likelihood of cancer occurrence to an exposed person is extremely low. However, aggregation of individual doses among large number of the population can give a large collective dose value leading to a projection of significant number of fatalities. ICRP forewarned against such misuse of collective dose concept stating "*The aggregation of very low individual doses over extended time periods is inappropriate, and in particular, the calculation of the number of cancer deaths based on collective effective dose from trivial individual doses should be avoided* (ICRP 2007)".

In terms of continuous exposure, the average value of cancer risk for incidence and death is summarized in Table 5.14 based on the BEIR VII report. The fatal cancer risk from a continuous lifetime exposure is 0.33% per mSv/yr. for a male and 0.5% per mSv/yr. for a female.

Table 5.14 Risk estimates of cancer incidence and cancer death due to chronic radiation exposure for working population and the general public

Exposed population	Cancer Incidence (per 1 mGy/year)		Cancer Death (Sv^{-1}) (per 1 mGy/yr)	
	Male	Female	Male	Female
Adult – working population: 18 to 64 y of age	3.059×10^{-3}	4.295×10^{-2}	1.70×10^{-3}	2.389×10^{-3}
Whole – whole population: Throughout life	6.21×10^{-3}	1.019×10^{-2}	3.32×10^{-3}	4.97×10^{-3}

5.3 Development of Standards for Radiation Protection

Cancer risk estimation for radiation exposure provides the basis for regulatory approaches for protection of humans from radiation. With the information on radiation dose-response, the expected incidence of cancer from specific dose levels of radiation can be estimated. Once the target level of protection against cancer incidence is set, the corresponding level of radiation dose can then be derived and used as the limit.

As discussed in Chap. 2, achieving absolute safety is impossibility although that is what the public wants from the exposure to radiation. The critical question is “While cancer occurs naturally from various causes, what level of excess cancer occurrence can be considered acceptable to the society?” This consideration guides the development of standards for radiation protection.

Two principal objectives in developing standards for radiation protection are (NCRP 1993):

1. To prevent clinically significant radiation-induced deterministic non-cancer effects.
2. To minimize the risk of stochastic effects, i.e., cancer and genetic effects.

It is believed that deterministic effects occur only above a threshold dose level and that by controlling the dose below the threshold, the first objective can be achieved. In the case of stochastic effect, i.e., cancer or heritable effect, there is always a finite probability of effect occurrence even at low dose levels. Thus the dose level should be controlled as low as possible under the consideration of resource limitations. This latter approach is terms “as low as reasonable achievable (ALARA)”.

The process of radiation protection standards development goes through four stages. The first stage is the work by independent scientific investigators such as government research groups or academic institutions. Their work is to observe and detect adverse health effects from radiation exposure. Radiation Effects Research Foundation (RERF), an organization established to conduct ABSS, has played a key role in this stage.

The second stage is performing risk estimation of radiation exposure based on the compiled scientific work. Two scientific committees, as mentioned earlier, played

Table 5.15 Recommended dose limits by ICRP-60

Application	Occupational	Public
Whole body	Effective dose averaged over 5 years: 20 mSv (2 rem) / year Maximum annual dose in the above 5 year period is 50 mSv	1 mSv (100 mrem)/ year
Annual equivalent dose to		
Lens of the eye	150 mSv (15 rem)	15 mSv (1.5 rem)
Skin	500 mSv (50 rem)	50 mSv (5 rem)
Hands and feet	500 mSv (50 rem)	

essential role in this stage, i.e., United Nations Scientific Committee on the Effects of Atomic Radiation (UNSCEAR) and National Research Council/National Academy of Science (NRC/NAS) Committee on the Biological Effects of Ionizing Radiation (BEIR). Cancer risk estimates from the BEIR committee are widely used as the basis for radiation protection standards development.

The third stage is the work by advisory bodies such as International Commission on Radiological Protection (ICRP) or National Council on Radiation Protection and Measurements (NCRP) of the U.S. These advisory bodies impose value judgments on the level of acceptable risk and translate health effects knowledge to recommendations on basic radiation protection limits. Please note the discussions in Sect. 2.4.2.1 regarding “what is the acceptable level of risk?”. These limits from ICRP or NCRP are recommended values, not legal limits.

The fourth stage is to develop specific rules based on the recommended values as the basis of legal enforcement for radiation protection. The entities involved in the last stage include national regulatory agencies of each respective government (e.g., US EPA, US Nuclear Regulatory Commission (US NRC), or Korean Nuclear Safety and Security Commission). International Atomic Energy Agency (IAEA), although not a national entity, takes similar responsibility for the international community.

The widely used values of radiation protection standards are based on the recommendations by the ICRP, i.e., the ICRP 60 recommendations (ICRP 1991). The recommendations are shown in Table 5.15. The effective dose limits for whole body are to limit stochastic effects while the annual equivalent dose limits are to prevent non-stochastic (deterministic) effects. The values are also retained in the ICRP 103 report (ICRP 2007).

As an example of regulatory limits, the current U.S. NRC regulations are shown in Table 5.16. The current NRC limit is based on limiting the sum of external (deep dose equivalent) dose and the internal dose (committed effective dose equivalent). This sum is denoted as the “total effective dose equivalent,” (H_{TEDE}). TEDE is the result of adding “deep dose equivalent” which is the dose equivalent from external radiation and “committed effective dose equivalent” for the internal dose. Deep-dose equivalent applies to whole-body exposure and the evaluation depth for deep dose equivalent is at 1 cm from the surface.

Table 5.16 US federal occupational dose limits (in Rems)

10 CFR Part 20 Limits	
Exposed Area	Annual dose limit (rems)
Whole-body[a]	5 TEDE[b]
Gonads	5 TEDE[b]; 20 organ
Lens of the eyes	15
Skin (averaged over 1cm^2)	50[c]
Extremities [d]	50[c]
Minors	10% of above limits
Embryo/fetus [e]	0.5 [f]

a Whole-body = head, trunk, legs above knees, and arms above elbows

b TEDE = Total effective Dose Equivalent = Sum of the deep-dose equivalent (external dose) + the committed effective dose equivalent (internal dose)

c Capping dose to prevent non-stochastic effects

d In the previous Part 20, "extremities" means hands and forearms, feet and ankles. In the revised Part 20, "extremities" mean hand, elbow, arm below the elbow, foot, knee, and leg below the knee

e Of a declared pregnant woman

f Over the period of gestation (9 months)

In terms of practical application of radiation dose limits, any radiation exposure of humans must be justified. This means that, while doses to individuals must not exceed established limits, there should not be any worker exposure to ionizing radiation without the expectation of an overall benefit from the activity. Therefore, the exposure to radiation must have a positive net benefit. Difficulty in applying this guideline lies with the question of how to quantify costs (and risks) and benefits in comparable terms. Such quantitative comparison may impose the use of monetary measures for the risk incurred from radiation exposure.

Furthermore, the exposure of workers to radiation must be optimized. This means that all exposures must be kept as low as reasonably achievable (ALARA), with economic and social factors taken into account (please refer to Sect. 2.4.2.1 for further discussions). In other words, the net benefit of radiation exposure should be maximized while taking economic and social factors and constraints into consideration. Informed value judgments are necessary in applying the dose limits.

5.4 Radiation Safety Applications

To ensure that exposures to radiation are within the prescribed regulatory limits, various radiation safety measures can be used. As mentioned in 2.4.2.4, use of these measures are mainly based on combination of controlling the time of exposure and the distance from the source and use of shielding. This is called the principle of time, distance, and shielding (Fig. 5.20).

The shorter the time one is exposed to a source, the lower will be the number of particles incident on the body and the resulting dose. Thus the time a worker spend

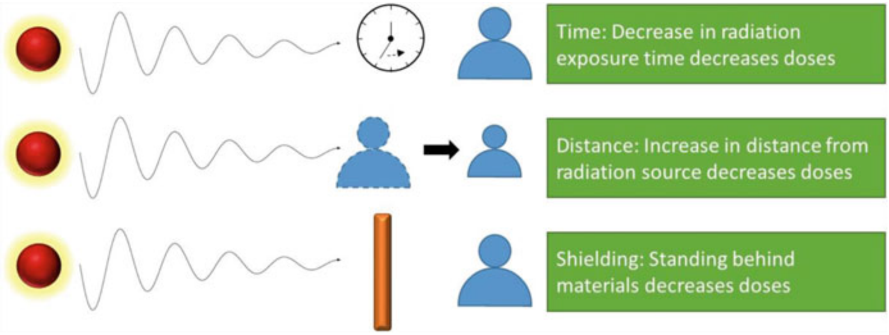


Fig. 5.20 The principle of time, distance, and shielding

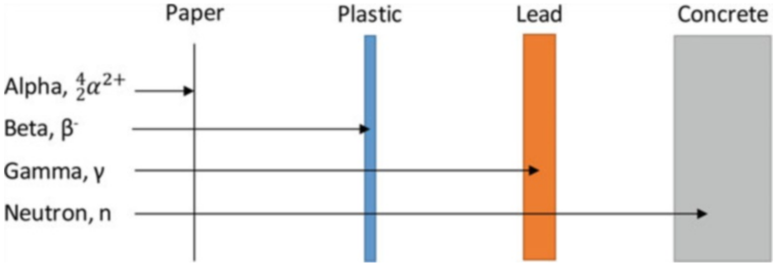


Fig. 5.21 Differences in the necessary shielding material and thickness for different radiation particles

while being exposed to a radiation source should be minimized. This can be achieved through proper advanced planning.

Distance is very effective in reducing the intensity of the radiation particles incident on the body, especially for small sources – It follows the inverse square law. The intensity of radiation particles decreases inversely proportional to the square of the distance from the source. In the planning of activities or design of structures involved in handling nuclear waste, keeping as much distance as possible between the source and the worker is necessary.

The term “shielding“implies deliberate introduction of material between the radiation source and an object to be protected so that the radiation level at the position of that object will be reduced. With proper implementation of shielding, radiation dose to humans can be effectively controlled. Such implementation is exercised in nuclear waste generation through the use of various structures, container, casks, and shield blocks.

These shielding applications are mainly against penetrating radiations such as gamma rays and neutrons. Properly designed shield against these radiations will also effectively block charged particles such as beta and alpha particles as these charged particles are readily stopped within very short distances in typical shield materials. Differences in the necessary shielding material and thickness for different radiation particles are illustrated in Fig. 5.21. However, care must be exercised if high energy beta particles are involved as they can produce x-rays through bremsstrahlung, a penetrating radiation. Also, in the case of charged particles, control of contamination becomes important as ingestion or inhalation of these particles could pose significant hazards through (human body) internal exposure. Further details on shielding against gamma rays and neutrons are discussed in Sect. 7.2

5.5 Conclusion

Understanding biological effects of radiation lays the foundation for safety in nuclear waste management as such understanding enables characterization of risk of radiation and development of regulatory standards. This chapter provided an overview of biological effects of radiation and the discussions of how the concept of "dose" is defined and used to characterize and control the risk of radiation exposure among occupational workers and the public. Latest analysis of the A-bomb survivors data provided the estimate of fatal cancer risk coefficient as 0.0051% per mSv of radiation dose. Studies of occupational radiation exposure also provided the estimate of fatal cancer risk at about 0.3% (male) or 0.5% (female) per annual dose of 1 mSv for chronic radiation exposures. The corresponding annual dose limits for radiation protection recommended by ICRP are 20 mSv and 1 mSv for occupational workers and the members of the public, respectively. More specific national regulatory standards for safety in nuclear waste disposal are derived as the fraction of these numbers (see also Table 2.1 and Example 2.7). Some specifics in the practices of radiation safety are also discussed in the chapter. While the issue of uncertainty in limiting cancer risk from radiation is well recognized, use of conservatism in radiation risk assessment and in establishing regulatory limits provides margin of safety in regulatory control of radiation risk.

Homework

Problem 5.1: Explain how the damages to DNA created by radiation can be repaired.

Problem 5.2: Explain how the damages to DNA created by radiation can lead into cell killing, mutation, and malignant transformation.

Problem 5.3: Explain why most cancers occur not right after the exposure to radiation but rather late (years after the exposure) in a person's life.

Problem 5.4: Under the hypothetical situation of the Yucca Mountain repository in the U.S. being licensed and operational, estimate the maximum possible value of the fatal cancer probability of the following individual.

(a) A worker handling HLW at the waste handling facility at Yucca Mountain for the preparation of waste package for disposal. He started his work at the age of 40 and worked at the site for 5 years.

(b) A person who was born after the closure of the repository and lives near Yucca Mountain throughout life.

Problem 5.5: A male person worked as a maintenance worker in a nuclear power plant from age 18 till his retirement at age 64. Assume his occupational dose from was 0.5 mSv per year. When he became 50 years old, he also worked as part-time industrial radiographer for three years receiving a total of 20 mSv. During his lifetime, he also received 3 mSv of whole body from natural background radiation. If he died at age 70 from leukemia, calculate the probability of this cancer being caused by his normal job, part-time job, or natural background radiation.

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Chapter 6
Generation of Nuclear Waste from Nuclear Power

Abstract Operation of nuclear power plants and the activities related to nuclear fuel preparation and handling of the associated waste represent the most significant source of nuclear waste generation. This chapter describes how nuclear fuels are prepared and utilized for nuclear reactor operation and how nuclear wastes are produced from the related processes. The chapter also describes how various nuclear wastes are classified for subsequent activities to appropriately manage the risk of nuclear waste.

Keywords Nuclear fuel preparation · Nuclear reactor operation · Wastes from fuel cycle · Mass balance · Waste classification

While nuclear wastes are generated from diverse activities or organizations, one of the most significant sources of nuclear waste is nuclear power plant (NPP): Nuclear fuels needed for electricity production by NPP eventually become nuclear waste. The life cycle of nuclear fuel, called nuclear fuel cycle, is also an important source of nuclear waste. Nuclear fuel cycle refers to all of the activities related to preparation, use, and disposal of nuclear fuel. These activities include mining, concentration and purification of uranium, uranium enrichment, fuel fabrication for nuclear reactor operation, and management of used nuclear fuel (and other nuclear wastes and byproducts).

Besides nuclear fuels discharged from nuclear reactors, other nuclear wastes from nuclear power plants come from treatment of contaminated liquids, gases, or solids from NPP operations. Radioactive resin, chemical sludge, and daily refuse such as contaminated paper, gloves, and plastics are among them. The contaminated parts, hardware, and equipment from the nuclear reactor system also become nuclear waste. Nuclear fuel cycle facilities (other than NPP) generate nuclear wastes as residual materials from the processing of feed materials or from the treatment of radioactively contaminated materials. If used nuclear fuels are recycled, high level nuclear waste containing fission products and actinides from reprocessing are produced which require very special handling and treatment.

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https://doi.org/10.1007/978-94-024-2106-4_6

This chapter gives an overview of nuclear waste generation from civilian nuclear fuel cycles and commercial nuclear reactor operation.

6.1 Overview of Nuclear Fuel Cycle

All nuclear reactors use fissile isotope of uranium, ^{235}U, as fuel. Except CANDU and MAGNOX reactors (where natural uranium with ^{235}U at 0.711 wt% is used as fuel), all other types of nuclear reactors require fuel with ^{235}U at higher concentration than the natural level. Therefore, enrichment of uranium, i.e., increasing the ^{235}U isotope content, is a necessary step as part of nuclear fuel development. Nuclear fuel cycle begins with extracting uranium from uranium mine and ends with disposition of the residual uranium and other byproduct materials. These activities are divided into three stages, i.e., the front-end and the back-end of the fuel cycle and reactor operations in between.

The front-end of the fuel cycle includes all activities needed to manufacture nuclear fuel. This includes (1) Mining and milling of uranium ore to produce U_3O_8 (known as yellowcake); (2) Purification of U_3O_8 and conversion to UF_6 as a preparatory step for enrichment; (3) Enrichment of ^{235}U using the chemical form, UF_6, to the required ^{235}U concentration level, and; (4) Converting UF_6 to UO_2 and fabrication of nuclear fuel. The reason for the changes of the chemical forms of uranium is explained below.

Operation of nuclear reactor is to use nuclear fuel for energy generation. The energy from nuclear fission is converted to electric energy in the plant system. Activities such as fresh fuel loading, reload core fuel management, and plant maintenance are performed as part of reactor operation. This stage eventually produces used nuclear fuels along with large volume of various other nuclear wastes. The used nuclear fuels are typically called “spent nuclear fuels”. These spent nuclear fuels still contain fissile materials (e.g., ^{235}U, ^{239}Pu) and could be recycled for further energy generation. If the remaining fissile materials in the spent fuel were to be utilized, activities necessary for recycling of spent fuel become part of the back-end fuel cycle.

The back-end of the fuel cycle is to manage what comes out of nuclear reactor operation, i.e., spent fuels and other radioactive wastes. All of the follow-on activities until these wastes are permanently disposed of belong to the back-end of the fuel cycle. These activities include on-site spent-fuel storage, spent-fuel transport, away-from-reactor storage, and final disposition.

When recycling of fissile material back to reactor operation is employed, the back-end of the fuel cycle includes reprocessing of spent nuclear fuel. Reprocessing includes steps of breaking spent fuel, dissolving fuel materials, separating materials to be reused, and handling of waste. Details of spent fuel reprocessing are discussed in Chap. 8.

The nuclear fuel cycle is classified as once-through/open, modified open, or closed, depending on whether the recycling scheme is utilized or not (Fig. 6.1). In

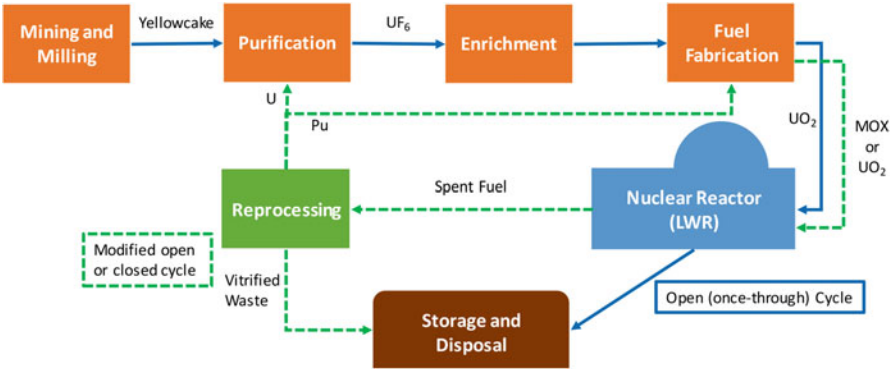

Fig. 6.1 A schematic representation of nuclear fuel cycle

the open/once-through cycles, spent fuel is directly disposed of as waste. The modified open cycle employs at least one re-burn of fissile materials (mainly plutonium) through reprocessing of spent fuel. In the end, spent fuels are disposed of as waste. The closed cycle involves no spent fuel disposal as all useful fissile materials remaining in spent fuel are repeatedly recycled for full resource utilization. Only the process wastes are disposed of in the end in the closed cycle. Further details of these fuel cycle options are described in Chap. 15.

6.2 The Steps in Nuclear Fuel Cycle

6.2.1 Mining

Uranium is a ubiquitous metal that occurs throughout the earth's crust. It is present in most rocks and soils with its concentration varying in different rocks. For example, granite rocks are found to contain uranium at concentration levels of 3–35 parts per million (ppm) whereas sedimentary rocks have lower concentration at ~2 ppm. On average, the natural abundance of uranium in earth's crust is about 2.8 ppm which is at about 500 times higher than gold and about the same as tin.

Some locations have elevated concentration of uranium resulted from the leaching from a rock and subsequent precipitation due to changes in chemical conditions (i.e., becoming reducing environment). These locations with greater than 1000 ppm (0.1%) uranium concentration can serve as uranium mines. Typically, uranium mines have the content of uranium at about 0.15 ~ 0.3%. Thus uranium is mined with an average yield of 1.5 to 3 tons of U recovered from 1000 tons of ore. The mined natural U is composed of two essential isotopes, ^{235}U (0.72 weight %) and ^{238}U (99.27 weight %).

As rocks breakdown through weathering, uranium is also found in soils and waters. Then uranium is subsequently taken up by plants, vegetables, and trees and becomes part of global food chain resulting in human consumption of uranium.

Uranium is mined mainly by three different techniques, i.e., underground mining, surface (open pit) mining, or solution/in-situ leach (ISL) mining. Underground mining is used for uranium deposits deeply located or covered by strata of hard rock and was used mostly in the early years of nuclear technology development. Surface (open pit) mining became more prevalent in later years. It is used when uranium ore is within a few hundred meters from the surface and when the overburden can be removed without excessive use of blasting. The method comes with such benefits as higher productivity, higher recovery, easier dewatering, safer working conditions, and usually lower costs. The disadvantage of the method is greater environmental impacts due to the larger surface area disturbed by the mining activity. The quantity of material to be removed to access the ore is considerably less with underground mining than surface mining.

Solution (ISL) mining is the latest method based on solubilizing the uranium in an underground ore body by injecting leaching solutions. The dissolved uranium solution is then pumped out followed by uranium recovery operations. This technique has the benefits of yielding very little tailings compared to the other two extraction techniques but involves higher cost. Applicability of the solution mining approach is limited to uranium ore body in a generally horizontal bed underlain by a relatively impermeable stratum. The ore deposits must also be sufficiently extensive to offset the higher cost of operation. Solution mining was first introduced in 1975 but became a preferred method since then. Use of a particular method depends on the nature of the orebody, safety and economic considerations.

While uranium mines operate in about 20 countries in the world, six countries (Kazakhstan, Canada, Australia, Namibia, Niger, and Russia) dominate the production of uranium producing about 87% of global supply in 2018. Table 6.1 shows the top 10 largest uranium mines in the world with the method of mining used.

6.2.2 *Milling*

As the level of uranium concentration in mined uranium ore is low at around 0.15 ~ 0.3%, it is necessary to raise the uranium content for fuel preparation. Increasing uranium concentration first requires removal of other constituents by using a combination of physical and chemical processes. These processes to obtain a highly concentrated uranium mass (typically up to 85–90 weight %) are called milling. The product of uranium milling, U_3O_8, is called "yellow cake" (U_3O_8). U_3O_8 is the most stable form of uranium oxide.

In milling, first, crushing and grinding of uranium to a suitable (i.e., small) size is done to increase the surface area. Then uranium is extracted through leaching from the crushed particles by using dissolving acid. The uranium in the acid solution is then separated into organic phase through solvent extraction (as organic phase and

Table 6.1 The Top 10 largest-producing uranium mines in the world in 2018

Mine	Country	Main owner	Type	Production (tonnes U)	% of the World
Cigar Lake	Canada	Cameco/Orano	Underground	6924	13
Olympic Dam	Australia	BHP Billiton	By-product/ underground	3159	6
Husab	Namibia	Swakop Uranium (CGN)	Open pit	3028	6
Inkai, sites 1–3	Kazakhstan	Kazaktomprom/ Cameco	ISL	2643	5
Rössing	Namibia	Rio Tinto	Open pit	2102	4
Budenovskoye 2	Kazakhstan	Uranium One/Kazatomprom	ISL	2081	4
Tortkuduk	Kazakhstan	Orano/ Kazatomprom	ISL	1900	4
SOMAIR	Niger	Orano	Open pit	1783	3
Ranger	Australia	Rio Tinto/ERA	Open pit	1695	3
Kharasan 2	Kazakhstan	Kazatomprom	ISL	1631	3
Top 10 total				26,946	51%

Data source: WNA (2019)

aqueous phase separate naturally). The organic solvent used is TBP (Tri-Butyl Phosphate or Tri-n-Butyl Phosphate, $(C_4H_9)_3PO_4$). TBP is a neutral extractant and forms bonds by using the electron from the phosphoryl oxygen atom in the structure. Note that TBP is the solvent used not only in milling but also in uranium refining and reprocessing (see Sect. 8.1.1). Uranium in the organic phase is then stripped back to aqueous phase followed by chemical precipitation, calcination, pulverization, and packaging to produce the product, yellow cask.

Example 6.1: Conversion Between Atomic Percent (at%) and Weight Percent (wt%)

Some problems in nuclear waste management require conversion between atomic percent and weight percent; at% signifies atomic percentage, i.e., the number of atoms of different atomic species present as percentage among the total, and wt% signifies weight percentage of different atomic species among the total.

Convert 0.72 wt% of ^{235}U to at%.

Solution:

Natural Uranium (NU) contains 0.72 wt% ^{235}U and 99.28 wt% ^{238}U. Consider 100 grams of natural uranium: 100 gram of natural uranium contains 0.72 gm of ^{235}U:

(continued)

Example 6.1 (continued)

# of atoms in 0.72 grams of ^{235}U = 0.72 × Avogadro's Constant/atomic weight of U

$= 0.72 \times 6.022 \times 10^{23}/235$

$= 1.845 \times 10^{21}$ atoms of ^{235}U

Similarly, number of atoms in 99.28 grams of Uranium-238

= 99.28 × Avogadro's Constant/atomic weight of U

$= 99.28 \times 6.022 \times 10^{23}/238$

$= 2.512 \times 10^{23}$ atoms of ^{238}U

Therefore, at% of ^{235}U is:

= [# of ^{235}U atoms]/[total # of ^{235}U and ^{238}U atoms] × 100%

$= [1.845 \times 10^{21}]/[(0.01845 + 2.512) \times 10^{23}] \times 100\%$

= 2.53%

So, 0.72 wt% ^{235}U is equivalent to 2.53 at% ^{235}U

6.2.3 Conversion

The yellowcake produced from uranium milling contains impurities. These impurities need to be removed before the use of uranium as fuel. The impurities include materials like boron, cadmium, and rare-earth elements. These materials possess sizable neutron-absorption cross-section even at small amounts thus work as neutron poison (absorber) in the nuclear reactor. The purification process of yellowcake is called refining. The "refining" process is also based on the solvent extraction technique. The yellowcake is dissolved in nitric acid and uranium is extracted by using the organic solvent, TBP, in the organic phase. The extracted organic complex is then treated to back extract uranium as uranyl nitrate solution in the aqueous phase.

The purified uranyl nitrate solution is then evaporated to a uranyl nitrate molten salt which is subsequently denitrified and further reduced to form UO_2 which is the stable ceramic form of uranium.

Except for CANDU or Magnox reactors, UO_2 needs to go through the process of enrichment to increase the natural isotopic content of ^{235}U to the level required by reactor operation. Typical level of isotopic content of ^{235}U required is between 3.5 percent and 5 percent.

Uranium hexafluoride [UF_6] is the compound of choice for the gas-based enrichment processes. Fluorine as a stable element has only one isotope ^{19}F, so that the difference in molecular weights of UF_6 molecule is due only to the difference in weights of the uranium isotopes. UF_6 is also the only compound of uranium sufficiently volatile at low temperature (solid at room temperature but easily vaporized).

For conversion to UF_6, UO_2 is first converted into UF_4 through reaction with hydrofluoric acid ($UO_2 + 4HF \rightarrow UF_4 + 2H_2O$) and then to UF_6 through reaction with elemental fluorine ($UF_4 + F_2 \rightarrow UF_6$). The purified product UF_6 is shipped as solid to an enrichment facility in metal containers.

CANDU reactors do not require enrichment of uranium, the product, UO_2, can be used as the fuel. In the case of the Magnox reactor, natural uranium is used in metallic form with manganese alloy called "Magnox". In both cases, the uranium purification process could take place at the uranium mill or at the fuel fabrication plant.

6.2.4 Enrichment

Most common techniques of uranium enrichment are gaseous diffusion and gas centrifuge.

In gaseous diffusion, gas will move through a porous wall from a region of higher pressure to a lower pressure region. In this movement, the kinetic energies of molecules as gas are the same (due to thermal equilibrium). As the kinetic energy of a gas is given by $\frac{1}{2}mv^2$, lighter molecules will move with higher speed than the heavier ones. The ratio of velocities between $^{238}UF_6$ molecules and $^{235}UF_6$ is 1 to 1.00429, as shown by

$$\alpha_0 = \sqrt{\frac{mass\ of\ ^{238}UF_6}{mass\ of\ ^{235}UF_6}} = \sqrt{\frac{352}{349}} = 1.00429 \qquad (6.1)$$

This ratio is called the separation factor, representing the degree of separation from a single stage operation of gaseous diffusion. In order to get the needed level of separation for fuel use, the process must be repeated many times. For example, enrichment of natural U to 3% ^{235}U level requires 1272 stages of gaseous diffusion (in the case of the US experience). For comparison, to produce 97% enrichment, 4080 stages of gaseous diffusion are needed.

In gas centrifuge, the UF_6 gas rotates inside cylinders subject to centrifugal acceleration. While the high speed of rotation drives the molecules at the same speed, the energy of heavier $^{238}UF_6$ molecules is higher thereby forcing them to move toward the outside of the cylinder. The lighter $^{235}UF_6$ molecules tend to collect closer to the center. In terms of the number of separation stages, a gas centrifuge is over ten times more efficient than a gas diffusion.

Other techniques of uranium enrichment also include aerodynamic separation, electromagnetic separation, laser separation, and chemical separation. In aerodynamic separation, a carrier gas, such as H_2, is mixed with UF_6, at a carrier-to-UF_6 ratio of about 20 to 1 (e.g., 96% H_2 and 4% UF_6). The carrier gas moves at a very high speed within a tube or nozzle to produce shock waves without becoming supersonic. The UF_6 is dragged along with it. Due to the difference in mass, the

^{235}U compound (the lighter compound) is separated from the rest in this dragging process. Two different methods of aerodynamic separation have been developed on a large scale, including Vortex tube and jet nozzle technique.

Electromagnetic separation was used for uranium enrichment during the U.S. Manhattan project (in the 1940s). The approach is based on the fact that when passing through a magnetic field, particles with different masses will have different momentum and different radii of curvature. In electromagnetic separation, uranium tetrachloride (UCl_4), a gas volatilized at about 600 °C, is used and ionized to U^+ ions by running an electric discharge through it. The ions are then accelerated and subsequently deflected by magnetic fields onto their respective collection targets.

Laser separation, the newest technique, is based on utilizing excited energy levels of uranium. Laser beam can be used with a specific frequency to excite atoms of only single isotope (^{235}U) to a higher state. Further energy can be delivered for the excited ^{235}U isotope to be ionized. The ionized ^{235}U isotope can then be collected by an electric field for separation from the gas mixture. Currently available laser separation techniques include ALVIS (atomic vapor laser isotope separation) and MLIS (molecular laser isotope separation). In the ALVIS process, a vapor of natural uranium, containing a mixture of ^{235}U and ^{238}U, is produced by heating a uranium ingot to above 2000 °C in a crucible using an electron beam. The laser beams are passed through the vapor and selectively excite and ionize only ^{235}U. The resulting positively charged ^{235}U is attracted to the negatively charged collection plates being separated from ^{238}U. In MLIS, an infra-red laser is used to excite the $^{235}UF_6$ molecule. Then an ultra-violet laser is used to dissociate $^{235}UF_6$ molecule into fluorine gas and $^{235}UF_5$. The resulting $^{235}UF_5$ is collected as the product.

Chemical separation of ^{235}U is based on using the differences in adsorption coefficients between ^{235}U and ^{238}U to ion-exchange resins or in ^{235}U and ^{238}U propensity to change valence in oxidation/reduction.

All components of a gaseous enrichment plant using UF_6 (i.e., gaseous diffusion/centrifuge or MLIS) need to be maintained at an appropriately elevated temperature to assure that the UF_6 remains in gaseous form. As fluorine is highly reactive with water and corrosive to most common metals, the internals of gaseous enrichment process must be leak tight and fabricated with corrosion resistant metal (i.e., nickel or austenitic stainless steel).

Large scale commercial enrichment plants based on the gaseous diffusion technique have dominated the world's enrichment service market for the last half century but all have been closed since 2015. The world's current uranium enrichment is all based on gaseous centrifuge since 2015 with the running capacity of 57,350 kgSWU (or 57.35 MSWU) with planned addition of laser enrichment plants. A special unit called the separative work unit, abbreviated as SWU, is used to define the required effort for the uranium enrichment as explained in the next subsection.

Basic Material Balance Relationship Underlying Enrichment Process

The amount of feed material needed to obtain a given amount of enriched uranium product depends on three parameters: The original enrichment of the feed, the desired enrichment of the product, and the level of residual enrichment of the depleted uranium, DU (this is also called the tails). The relationship among the parameters are described by considering mass balance in the enrichment process.

Assume that by using F kg of feed with the ^{235}U enrichment level of x_F, (weight fraction), production of P kg of uranium at the enrichment level of x_p is needed, leaving behind T kg of tails at enrichment level x_T. Assuming no loss of uranium in the process, the mass balance of both the total materials processed and the total amount of ^{235}U in the process is described by the following two equations:

$$F = P + T \tag{6.2}$$

$$x_F F = x_P P + x_T T \tag{6.3}$$

Eliminating T from these two equations gives

$$F = (x_{P-}\, x_T)/(x_{F-}\, x_T) * P \tag{6.4}$$

Or, eliminating F gives the expression

$$T = (x_{P-}\, x_F)/(x_{F-}\, x_T) * P \tag{6.5}$$

Now, let's define the separative work unit (SWU) as a way of comparing various ways of enriching ^{235}U. The SWU necessary to separate a mass F of feed of assay x_f into a mass P of product assay x_p, and tails of mass T and assay x_t is given by the expression,

$$SWU = P \cdot V(x_P) + T \cdot V(x_T) - F \cdot V(x_F), \tag{6.6}$$

where x_p, x_T, *and* x_F are the respective enrichment levels, and V(x) is the value function at a given, defined as

$$V(x) = (1 - 2x)\, ln \left[\frac{1 - x}{x}\right] \tag{6.7}$$

where x is in weight fraction.

The concept of SWU is based on the observation that isotope separation is equivalent to the unmixing of gases that had previously been irreversibly mixed. Since separated isotopes represent a more ordered situation than unseparated isotopes, the entropy (i.e., disorderliness) of the separated state is clearly smaller than that of the unseparated state. Therefore work must be performed on a system in order to decrease its entropy in an isothermal process. This is reflected in the definition of SWU. The SWU is proportional to the amount of material processed and the energy needed to operate the machine but is not a measure of the amount of work required to separate isotopes. We notice that SWU is a function of the concentrations of the

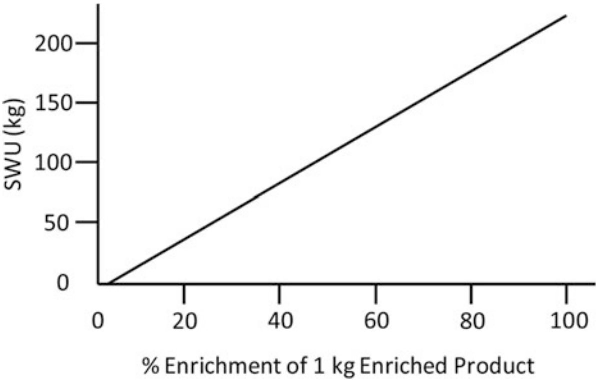

Fig. 6.2 Required amount of SWU to produce 1 kg of product at different enrichment level (with natural U feed and 0.25% tail) (figure reproduced from Mozley 1998)

feedstock, the enriched output, and the depleted tailings. The number of SWU provided by an enrichment facility is directly related to the amount of energy that the facility consumes as the standard measure of the exerted efforted.

The unit for SWU is kg SW or tSW (i.e., 1 SWU = 1 kg SW; 1 kSWU = 1tSW), and the capacities of enrichment facilities are rated in tSWU/yr. The total cost divided by the capacity provides the basis for the charge of enrichment service as $/tSWU. The same amount of SWU requires different amount of energy depending on the efficiency of the separation technique, i.e., depending on the efficiency of the separation technology, the same capacity of SWU will consume different amounts of energy. For example, gas centrifuge plants require 50 to 60 kW·h (180–220 MJ) of electricity per SWU while modern gaseous diffusion plants typically require 2400 to 2500 kWh, or 8.6–9 GJ of electricity per SWU. In contrast, ALVIS, the laser-based technology, requires 100–150 kWh/SWU.

By definition, 1 SWU is equivalent to 1 kg of separative work. As an example, to produce 10 kg of 4.5 wt% enriched uranium starting with 100 kg of natural uranium and leaving 0.3% enriched tails (DU), it takes about 61 SWU (61 kg SW). To support the operation of a typical 1000 MWe reactor, 100,000 to 140,000 kg SW is required annually (100,000 to 140,000 SWU/yr) depending on the enrichment level of the fuel (NRC 2008).

For fixed values of x_F and x_T, the separative work necessary to produce a given amount of product increases monotonically with enrichment. This can be seen in Fig. 6.2 where the required amount of SWU to produce 1 kg of enriched product at different enrichment levels is shown (starting with natural uranium, assuming the tail has 0.25% ^{235}U). According to the figure, about 3.9 kg SWU is needed to produce 1 kg of 3%-enriched uranium while about 200 kg SWU is needed to produce 90%-enriched uranium.

In addition to the separative work units (SWU) provided by an enrichment facility, another parameter of importance is the mass of feed needed to produce a desired mass of enriched uranium. As with the number of SWUs, the amount of feed material required will depend on the target enrichment level and the remaining amount of ^{235}U in tails. Fig. 6.3 shows the changes in the required SWU to produce 1 kg of 90% enriched product (i.e., weapons grade uranium) with the use of different enrichment levels of feed. The figure indicates that amount of SWU is much reduced

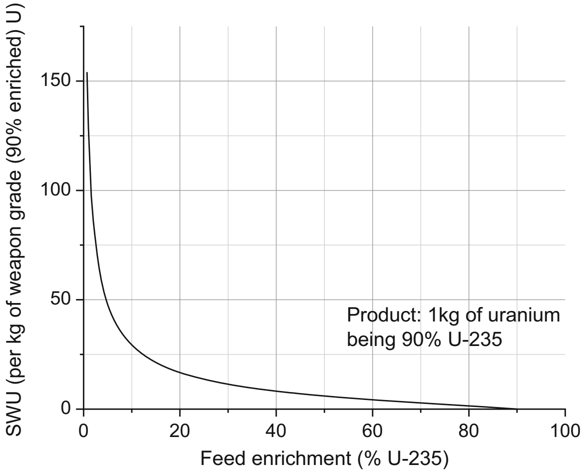

Fig. 6.3 Required amount of SWU to produce 1 kg of 90%-enriched product with the use of different enrichment levels of feed

if the feed is not the natural uranium but an already enriched uranium at certain level. For example, when the feed is natural uranium or 4% enriched one, the SWU needed to produce 1 kg of 90% enriched ^{235}U is ~154 SWU or ~ 55 SWU, respectively.

Example 6.2: How Much Natural Uranium Is Required for a Nuclear Power Plant?

Determine the amount of natural uranium required to operate 1000 MWe nuclear power plant (NPP) for a year if the reactor uses 3.75% enriched fuel.

Assumptions:

wt% of DU: 0.3%,
wt% of NU: 0.7%
Assume 100,000 SWU per year is needed to operate a 1000 MWe NPP

Solution:

$F = P + T,$ (from Eq. 6.2)

$x_F F = x_P P + x_T T,$ (from Eq. 6.3)

$SWU = P \cdot V(x_P) + T \cdot V(x_T) - F \cdot V(x_F),$ (from Eq. 6.6)

where, $V(x) = (1 - 2x)\, ln \left[\frac{1-x}{x}\right]$ (from Eq. 6.7)

Thus,

$x_F = 0.007,\ x_P = 0.0375,\ x_T = 0.003,\ SWU = 100{,}000$

$V(x_F) = (1 - 2 \times 0.007)\, ln\left(\frac{1-0.007}{0.007}\right) = 4.885$

(continued)

Example 6.2 (continued)

$V(x_P) = (1 - 2 \times 0.0375)\, ln \left(\frac{1-0.0375}{0.0.375}\right) = 3.002$

$V(x_T) = (1 - 2 \times 0.003)\, ln \left(\frac{1-0.003}{0.003}\right) = 5.771$

Plugging in the numbers, we get 3 equations with 3 unknowns:

$$F = P + T \tag{1}$$

$$0.007F = 0.0375P + 0.003T, \tag{2}$$

$$100000 = 3.002P + 5.771T - 4.885F, \tag{3}$$

Substituting (1) into (2), we get

$$0.007(P + T) = 0.0375P + 0.003T \rightarrow 0.004T = 0.0305P$$

$$\rightarrow T = 7.625P \tag{4}$$

Substituting (4) into (1), we get

$$F = P + T = P + 7.625P = 8.625P$$

$$\rightarrow F = 8.625P \tag{5}$$

Substituting (4) and (5) into (3) to solve for P,

$$100000 = 3.002P + 5.771 \cdot 7.625P - 4.885 \cdot 8.625P = 4.87275P$$

$$\rightarrow P = 20522 \tag{6}$$

Substituting the value of P to (4) and (1),

$$\rightarrow T = 7.625 \cdot 20522 = 156482 \tag{7}$$

$$\rightarrow F = 20522 + 156482 = 177004 \tag{8}$$

Thus, the solution is:

P = 20,522 kg,
T = 156,482 kg,
F = 177,004 kg.

∴1.770 × 10^5kg U_{nat} is required per year to produce 20,522 kg of 3.75 wt% U as product.

6.2.5 *Fuel Fabrication*

Fuel fabrication is the process of converting the enriched UF_6 gas into the fuel form of choice and manufacturing nuclear fuels in the form of fuel rods and assembly. This is the final step of nuclear fuel preparation for reactor irradiation. It includes three main stages, i.e., 1) producing uranium in the fuel form of choice, 2) fuel rod fabrication, and 3) fuel assembly fabrication.

The most widely used fuel form in commercial nuclear reactors (e.g., LWR, HWR, and FBR) is UO_2. In this case, the enriched UF_6 gas is converted to UO_2 for UO_2 pellet production. UO_2 is a stable ceramic that can be heated almost to its melting point (around 2865 °C) without serious mechanical deterioration. As it does not react with water, the properties of UO_2 are not affected by leakage in cladding in water-cooled reactors. Compared to other alternative fuel form (e.g., Magnox), however, UO_2 has the disadvantage of low theoretical density (uranium atom density) and low thermal conductivity.

In earlier nuclear reactors, metallic uranium was also used as the fuel. Use of metals is to maximize the number of uranium atoms per unit volume in the fuel. The reactors with metallic uranium fuel include the military reactors for plutonium production and the older gas-cooled reactors in the U.K. and France. Uranium metal is highly reactive and reacts with water, air, hydrogen, and most nonmetallic elements. Therefore, when metallic uranium is used as fuel, cladding failure would have serious consequences. This is the main reason why metal fuels were not chosen for commercial nuclear reactors. Metallic uranium also has a lower melting point of 1132 °C compared to 2865 °C of UO_2.

In the case of sodium cooled reactors, metal alloys, such as uranium carbide and uranium nitride, are used as fuel. These alloys do not react with sodium, while oxide fuels have the concern of chemical reaction with sodium. Uranium carbide provides much higher thermal conductivity (by almost a factor of ten compared to UO_2) and higher theoretical density (13.6 g/cm^3) – thus more favorable neutron usage for breeding and lower thermal gradients in the fuel is attained. As a disadvantage, uranium carbide is quite reactive with water, less capable of retaining gaseous fission products, and is more prone to irradiation induced swelling. Uranium nitride has an advantage of high uranium atom density (a theoretical density of 14.3 g/cm^3, higher than uranium carbide) and high thermal conductivity. Uranium nitride is also thermally stable and features low swelling and low fission gas release rates under irradiation. Disadvantages of uranium nitride include: high neutron absorption cross section of ^{14}N or ^{16}N; possible dissociation of nitride compound at different temperatures depending on the nitrogen over-pressure, and; generation of large inventory of ^{14}C from neutron activation which raises a concern in spent fuel management.

The UF_6 shipped in a solid form is sublimed to its gaseous form for UO_2 fuel preparation by heating. The gaseous UF_6 is reacted with superheated steam using a rotary kiln for conversion to UO_2 powder. The UO_2 powder is milled and pressed into pellets. The pressed pellets are sintered to be converted into a high density ceramic by reducing porosity to about 5 percent (i.e., up to 95% of theoretical density). The sintered pellets are outgassed and ground for finishing to meet the specification of fuel pellet geometry.

In fuel rod fabrication, the finished pellets are stacked up inside cylindrical zircaloy tube. This zircaloy tube is an encasement structure to contain uranium pellets, and is called cladding. Zircaloy, either zircaloy-2 or − 4, is used by most of the LWR fuel as the material for cladding. Zircaloy has good compatibility with the fuel and coolant. Zircaloy is predominantly zirconium containing small amounts of tin, nickel, chromium, and iron. Presence of tin and oxygen helps with improving the mechanical strength while iron, chromium, or nickel improves corrosion resistance in water. Nickel is used in zircaloy-2 (used for BWR) whereas zircaloy-4 (used for PWR) does not contain nickel. Stainless steel or zircaloy is also used as cladding for metallic fuel. The cladding is sealed up through welding after filling the voids with inert (e.g., helium) gas for a final product, fuel rod. The finished rods are inspected for any fabrication defects.

In fuel assembly fabrication, the fabricated fuel rods are placed in a cage assembly to form an assembly. Use of cladding supports the assembly to withstand the loads during normal operation. An example showing a PWR and a BWR fuel assembly is given in Fig. 6.4. The position of each fuel rod is fixed in the prefabricated framework structures that hold the rods in a precisely defined grid arrangement. Fuel assemblies of western PWRs and BWRs feature square lattice arrangement while the fuel assemblies of Russian VVER reactor have hexagonal arrangement. PWRs use typically 17×17 or 14×14 fuel pin arrays while BWRs use pin geometries ranging from 6×6 to 10×10 arrays. The assembly also has vacant rod positions. These vacant positions are for control rod insertion or as a "guide thimble" for placement of a neutron source rod (for reactor startup), burnable poison rods (as neutron poisons), neutron level instrumentations, or a test fuel segment. These assemblies are about 4–4.5 m long.

In a BWR or PWR, one nuclear fuel assembly contains uranium weighing about 180 or 460 kg, respectively. After adding the weight of oxygen in uranium oxide, zircaloy cladding, and other metallic hardware, the total weight of a finished fuel assembly is about 280–320 or 590–660 kg for a BWR or PWR assembly, respectively. The weight of a PWR assembly is about 70% uranium, 9% oxygen, and 21% metal. More specific information about the fuel assemblies is also given in Table 7.1.

In the case of recycling spent fuel, MOX (mixed uranium oxide and plutonium oxide) fuel is used. The process of MOX fuel fabrication is similar to that of UO_2 fuel fabrication except UO_2 powder is mixed with PuO_2 powder through vigorous blending. Usually depleted uranium as tails from the enrichment process is used for MOX fuel. In this case, using about 7% of plutonium from LWR spent fuel for a MOX fuel is equivalent to a typical enriched UO_2 fuel. The mixed $(U,Pu)O_2$ powder then goes through pressing and sintering process. Installation of shielding is necessary to protect the workers from spontaneous neutrons emitted from plutonium (e.g., ^{238}Pu, ^{240}Pu). There are about thirty and ten LWRs using MOX fuel in Europe and in Japan, respectively (WNA 2020).

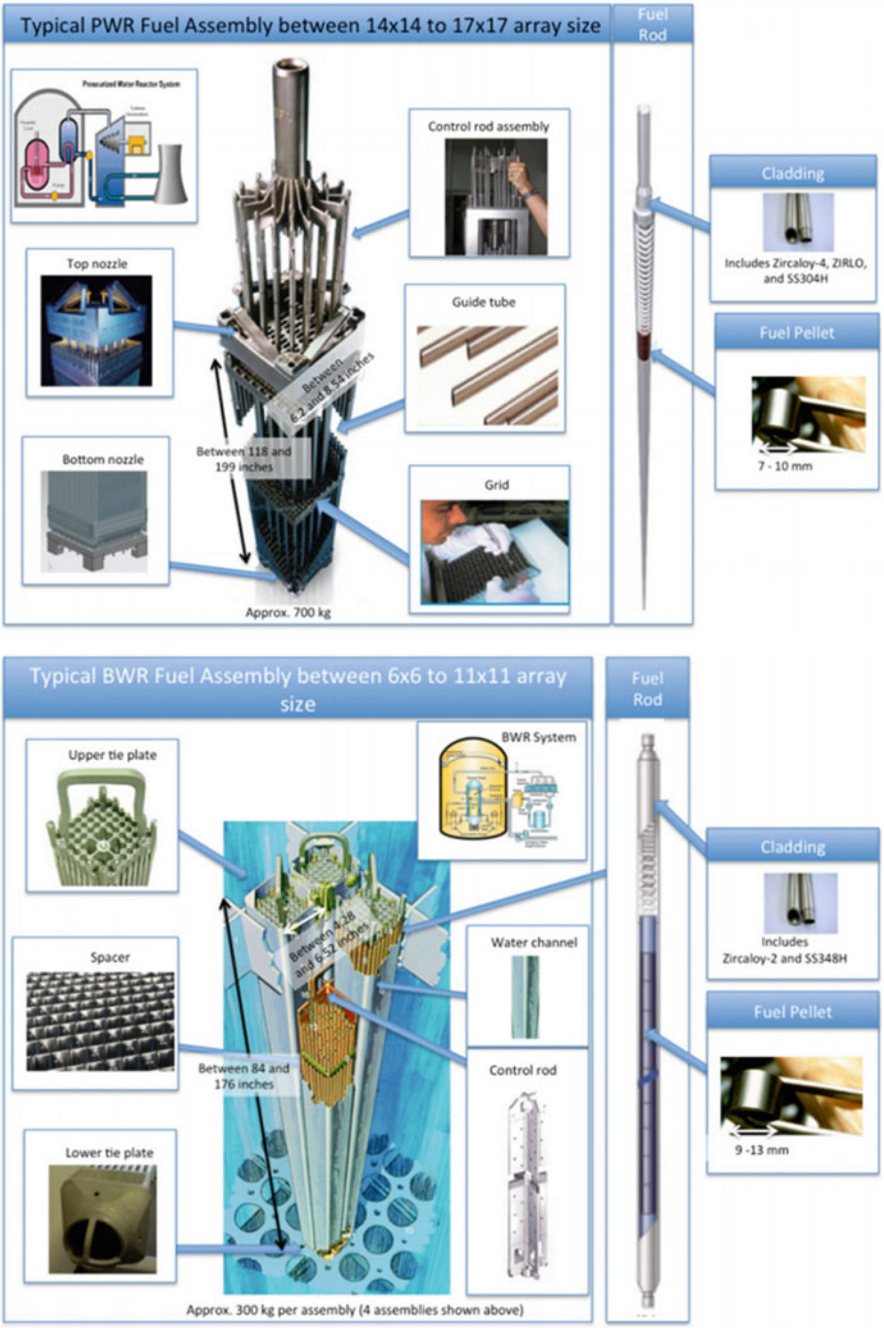

Fig. 6.4 PWR and BWR fuel assemblies and the related components with their processing steps. (Source: Wagner 2012)

6.2.6 Nuclear Reactor Operations

The fabricated fuel assemblies are loaded into the nuclear reactor. Nuclear reactor is where the processes of fission energy generation take place. Nuclear fuels are irradiated by neutrons to induce fission and the resulting energy is utilized for steam production and electricity generation.

6.2.6.1 In-Core Fuel Management

One of the most important considerations for reactor operation is to provide enough fissile material to the reactor to maintain criticality throughout the designed operating period. Given the continuous depletion of fissile during operation, the reactor must contain excess fissile material at the beginning of reactor operation. This means that a control mechanism must be present to maintain the reactor critical while the amount of excess fissile material existing in the reactor decreases due to fuel depletion during the entire operating life. This is done by adjusting the level of neutron poisons in the core (the region in the reactor containing nuclear fuel). The neutron poisons are the materials intentionally introduced into the reactor with very high neutron absorption cross sections to compensate for the presence of excess fissile materials. To keep the reactor exactly at the critical condition, the level of neutron poisons is gradually reduced as the fissile materials are being depleted. Neutron poisons are provided in the form of burnable poison rods, soluble boron, or control rods.

Burnable poison rods are the rods containing materials with high neutron absorption cross-section (compound of boron or gadolinium) that slowly lose the neutron poison characteristics by absorbing neutrons (by being converted into materials of relatively low absorption cross section). Soluble boron refers to boric acid added to the coolant. The concentration of boric acid is at the highest level at the beginning of core life and gradually decreases following the steady depletion of fissile. Control rods are the rods containing neutron poisons in the form of solid assemblies and can be removed or inserted quickly depending upon the degree of neutron poisons needed. Control rods are used only when necessary such as during startup and shutdown as they involve large perturbations in spatial distributions of neutrons in the core.

When the reactor reaches a point where maintaining criticality is no longer feasible even with complete removal of neutron poisons (due to significant levels of fissile material depletion), the fuel needs to be replaced with fresh fuel. At this point the reactor reached so called the cycle length. The cycle length is the designed irradiation period of the fuel. The cycle length typically ranges between 3 and 5 years. Therefore, the fuel, once loaded, stays in the core for 3–5 years depending on the design of the operating cycle. The higher the initial content of the fissile ^{235}U in the fuel, the longer the cycle length is. Then the fuel is discharged from the reactor and becomes used nuclear fuel (which is most often called spent nuclear fuel).

Loading of fresh fuels, while reshuffling or removing old fuels, is called refueling. During refueling, some of the old fuels, usually one third or one quarter of the reactor fuels, are removed from the core and moved to on-site storage. The remainder is rearranged to locations in the core better suited to energy generation with its remaining level of fissile content. Refueling is done every 12–18 months. All of the fuel assemblies available in the core including new fuel assemblies must be arranged in such a way that the total energy yield is maximized and the spatial distributions of power are maintained as uniform/flat as possible not to create local power peaking and thus to meet the safety limitations and operational constraints.

Management of such fuel loading/irradiation during nuclear reactor operation is called in-core fuel management. In core fuel management requires that the design limits for safety are not violated during the entire reactor operation. These limits are enforced to minimize local power peaking while maintaining exact balance between the power produced and the power extracted through cooling. These limits are in the form of temperature limit, power limit, the peak to average power ratio, limit on excess in neutron multiplication capability, and maintaining effectiveness in heat transfer. If any of the safety limits are violated, the reactor is automatically shut down.

Keeping the spatial power distributions within the reactor core as uniform as possible is important to prevent the occurrence of local hot spots and also for effective utilization of fuel. In this regard, in-core fuel management is an optimization problem. The problem requires optimizing the rearrangements of all fuel assemblies including the old and fresh ones, while maximizing fuel burn-up, minimizing fuel-cycle costs, and satisfying all the safety constraints.

6.2.6.2 Production of Radioactivity from Reactor Operation

The fresh nuclear fuel is entirely made of uranium as a mixture of ^{235}U and ^{238}U (besides the oxygen as UO_2 and the mechanical or structural components). As the uranium in nuclear fuel is irradiated in a nuclear reactor, it undergoes nuclear transformation and its composition is drastically changed. The end result of nuclear transformations is the production of a very diverse mixture of nuclides in the fuel.

This transformation can be summarized as: 1) consumption of fissionable material, 2) production of fission products, 3) production of heavy nuclides such as isotopes of uranium, plutonium and minor actinides, 4) production of activation products from the materials in the mechanical components of fuel rods and assemblies. The transuranic elements such as neptunium ($Z = 93$), americium ($Z = 95$), and curium ($Z = 96$) in spent fuel are also called minor actinides. Actinides refer to elements that range in atomic numbers (Z) from 89 to 103.

A typical light water reactor (LWR) spent nuclear fuel contains ^{238}U (at 93–94%), ^{235}U (at ~1%), plutonium isotopes (at around 1%), fission products (3–5%), and other actinides (0.1%). Two of the plutonium isotopes, ^{239}Pu and ^{241}Pu, are fissile materials that can also be recycled for further energy generation. The changes in the composition of fuel depend on the number of nuclear reactions including fission taken place in the fuel, which is in turn related to the amount of energy produced by fission. The amount of energy extracted from nuclear fuel per unit mass is represented by using a special term, called "burnup", which is a term as a measure of fuel utilization.

The burnup is determined by the level of power produced and the length of fuel irradiation (reactor operation) and is represented by megawatt days (MWD) per unit mass of fuel used (e.g., ton). Therefore, burnup is calculated by multiplying the thermal power of the nuclear reactor by the duration of operation and dividing it by the mass of the initial fuel loading. The unit of burnup is then MWD/MTHM (see Example 6.4). The most common term used for the mass of fuel is MTHM (metric ton of heavy metal), but sometimes MTIHM (metric ton of initial heavy metal) is also used. Here the mass of heavy metal refers to only uranium in the original fuel although the actual nuclear fuel is in the form of uranium oxide (UO_2). Therefore the mass of oxygen in the fuel (which takes up about 12% of the fuel mass in UO_2) is not included in the calculation of MTHM.

Example 6.3: Simple Burnup Calculation

If a 3000 MW thermal plant (equivalent to about 1000 MW electric, assuming 33% of thermal efficiency) uses 24 ton of enriched uranium (tU) and operates at full power for 1 year, what is the average burnup of the fuel?

Solution:

Burnup
= [Total thermal energy produced]/[mass of fuel used]
= [3000 (MWt) × 365.25 (d)]/[24 MTHM]
= 45,656 MWD/MTHM (or ~45.7 GWD/MTHM)

Typical burnup of spent fuel from a LWR is at the level of about 30,000 MWD/MTHM but can reach up to over 60,000 MWD/MTHM. In the case of PHWR, the spent fuel reaches the burnup of about 7500 MWD/MTHM. The low burnup (thus low amount of energy generation) is due to the use of natural (unenriched) uranium fuel.

From a typical nuclear power plant, the amount of spent fuel discharged per 1 GW (1000 MWe) electrical output ranges between 20 and 30 MTHM per year depending on the burnup.

The amount of spent fuel generated from reactor operation is represented by the following equation.

$$\text{Annual spent fuel production rate (tU/y)} \\ = \frac{\text{Electric power produced (MWe/y)} \cdot 365.25 \cdot \text{Capacity factor (\%)}}{\text{Thermal efficiency (\%)} \cdot \text{Burnup (MWD/MTU)}} \tag{6.8}$$

For example, at the burnup level of 45,000 MWD/MTHM and 65,000 MWD/MTHM, the amount of spent fuel discharged from a 1000 MWe nuclear reactor per year is 27.6 ton and 19.1 tons, respectively. The breakdown of the mass of fission products and actinides in the corresponding mass of spent fuel is as follows.

For 27.6 tonnes of spent fuel at 45,000 MWD/MT: 26 t uranium oxide (<1.0% U-235), 280 kg transuranics (mainly plutonium), 1 tonne fission products.

For 19.1 tonnes of spent fuel at 65,000 MWD/MT: 18.3 t uranium oxide (<1.0% U-235), 200 kg transuranics (mainly plutonium), <0.6 t fission products.

The total combined presence of fissile (such as ^{235}U, ^{239}Pu, and ^{242}Pu) in the spent fuel is at about 1.4 ~ 1.5% which can be utilized in the case of employing spent fuel recycling.

The irradiated fuel rods contains the inventory of radioactivity produced during reactor operation from the fission products, actinides, and other activation products (from the activation of materials by neutron irradiation).

Most of the fission product (see Sect. 3.3.4) are very short-lived. For instance, the number of radionuclides with greater than 10 years of half-life among fission products is thirteen. However, some of these nuclides (^{99}Tc, ^{129}I, ^{135}Cs) are very long-lived.

Actinides are the man-made radionuclides occurring after uranium and the actinide elements capture neutrons along with their subsequent radioactive decays. They become important part of radioactive inventory in spent nuclear fuel.

Activation products are the products of neutron capture by reactor structural materials, coolant, moderator, and fuel impurities, resulting in gamma-ray emissions. Common examples of them are from neutron activation of reactor structure such as steel (e.g., ^{58}Ni(n,γ)^{59}Ni, ^{62}Ni(n,γ)^{63}Ni, ^{54}Fe(n,γ)^{55}Fe), of cladding (e.g., ^{92}Zr(n,γ)^{93}Zr, ^{94}Zr(n,γ)^{95}Zr), of coolant and moderator (e.g., hydrogen, oxygen, ^{2}H (n,γ)^{3}H, ^{14}N(n,p)^{14}C, ^{17}O(n,α)^{14}C), of the chemicals added to coolant (e.g., lithium hydroxide or boric acids; ^{10}B(n, ^{8}Be)^{3}H, 6Li(n,α)^{3}H, ^{7}Li(n,nα)^{3}H), of fuel impurities (e.g., ^{14}N(n,p)^{14}C), and corrosion products (e.g., ^{59}Co(n,γ)^{60}Co).

If any of the fuel rods has breach in the cladding, radioactive materials can escape from the fuel to become entrained in the primary coolant and be distributed throughout the reactor coolant system through coolant circulation. Most of these species stay in structural materials and piping. Some of them in liquid or gaseous form (i.e., noble gases, iodine, particulate matter, CO_2, CH_4, etc.) become part of effluents from the nuclear power plant. Some of them also become part of solid waste, called low level or intermediate level waste. These gases, liquids, and solids are collected, treated, and stored by radioactive waste treatment systems before they are released to the environment or shipped to a waste disposal facility.

Example 6.4: Sources of Radioactivity During the NPP Operation

List major sources of radioactivity during nuclear reactor operation (according to their source categories).

(continued)

Example 6.4 (continued)

Solution:

The sources of radioactivity in the operation of nuclear reactor can be categorized into 3 general categories.

(1) Irradiated fuel rods:

- Fission products: strontium-90 (^{90}Sr), cesium-137 (^{137}Cs), tritium (^{3}H), iodine-129 (^{129}I), technetium-99 (^{99}Tc), niobium-95 (^{95}Nb), xenon-133 (^{133}Xe), cerium-144 (^{144}Ce), cesium-135 (^{135}Cs), etc.
- Actinides: neptunium-237 (^{237}Np), plutonium-239 (^{239}Pu), plutonium-240 (^{240}Pu), plutonium-241 (^{241}Pu), plutonium-242 (^{242}Pu), americium-241 (^{241}Am), americium-243 (^{243}Am), curium-242 (^{242}Cm), curium-244 (^{244}Cm), etc. Most of these are also activation products.
- Activation products: carbon-14 (^{14}C)

(A small fraction of the radioactive materials may escape from the fuel through perforations in the cladding into the primary coolant; in-core fuel failures may also lead to the release of radioactivity)

(2) Neutron activation of

- Structure (steel; nickel-59 (^{59}Ni), nickel-63 (^{63}Ni), iron-55 (^{55}Fe))
- Cladding (Zircaloy; zirconium-93 (^{93}Zr), zirconium-95 (^{95}Zr))
- Chemicals added to coolant (tritium (^{3}H))
- Coolant & moderator (tritium (^{3}H), carbon-14 (^{14}C))
- Corrosion products (cobalt-60 (^{60}Co))

(3) Fissioning of “tramp” uranium (traces of uranium dioxide remaining as a contaminant on the exterior surface of fuel cladding after fabrication)

6.2.7 Reprocessing

Recycling of fissile materials in spent fuel requires reprocessing. Reprocessing consists of a series of physical and chemical operations to separate nuclear materials of interest from spent fuel. Pu isotopes, once separated, can be recycled into nuclear reactors through MOX fuel fabrication for energy generation. The separated uranium can also be recycled or be disposed of as low level waste. Reprocessing also offers opportunity for the separation and removal of selected nuclides for specific waste management purposes, e.g., to significantly reduce the decay heat inventory (e.g., ^{137}Cs, ^{90}Sr, ^{241}Am, ^{242}Cm, ^{244}Cm, and plutonium isotopes) cutting down the amount of heat to be managed in the final disposal stage. More detailed discussions on reprocessing are in Chap. 8.

6.3 Material Balance in the Nuclear Fuel Cycle

From the perspective of material balance of uranium, nuclear fuel cycle can be summarized as follows:

- Uranium ore containing less than 0.5% of uranium is mined.
- Milling process extracts some 85–95% of the uranium present in the ore with about 0.5% uranium loss.
- Conversion achieves more than 99.95% purity of uranium.
- Enrichment increases the concentration of ^{235}U from 0.7% to around 3 ~ 5%.
- Reactor operation results in spent fuel with the content of uranium at about 94–95% (with ^{235}U at less than 1%).

An approximate mass flow of uranium without fuel recycling in a typical light water reactor (LWR) is given below as an example. This example is for 3% enriced fuel to support 1.0 GWe-year power generation:

Uranium mining and milling - > (as U_3O_8, 211,246 kg total uranium)- > ***Conversion to UF$_6$*** - > (as UF_6, 210,246 total uranium, after the loss of 1000 kg uranium) - > ***Enrichment*** - > (as ~3w/o UF_6, 32,078 kg total uranium (966 kg of ^{235}U, with the loss of uranium in the 0.3w/o tails) - > ***Fuel fabrication*** - > (as UO_2, 31,758 kg of total uranium at the BOC, after the loss of 320 kg uranium (9.6 kg of ^{235}U)) - > ***Reactor*** - > ***Spent fuel assemblies*** - > ***Spent fuel storages*** - > ***Waste disposal***.

Please note that the amount of uranium going into the reactor as fuel at the BOC can be reduced significantly with increase in the U enrichment. For example, at 3.75% enrichment, the amount of U in the fuel at BOC is ~20,500 kg in comparison to 31,758 kg at 3% enrichment. This difference also leads to the difference in the amount of spent fuel produced. This increase in uranium enrichment level also results in higher burnup of spent fuel. Therefore, increasing fuel burnup reduces the initial mass of uranium needed and the amount of spent fuel produced. For example, increasing the burnup from 33,000 to 50,000 MWD/MTHM in PWRs reduces the required mass of yellow cake (U_3O_8) by about 15%, the separative work units (SWUs) by 1%, and the spent fuel generation by about 40%, annually.

6.4 Waste Generation and Release of Radioactivity from the Nuclear Fuel Cycle

This section addresses the specifics of nuclear waste generation from the front-end and the back-end of nuclear fuel cycle as well as from nuclear reactor operations.

6.4.1 Wastes from the Front-End of Nuclear Fuel Cycle

Uranium mining produces wastes as waste rocks or effluents from the mining operation. The effluents include uranium-bearing dusts, radon gas, and mine drainage liquids containing uranium and its daughter products. Mine drainage is typically stored in settling ponds. Solids wastes of overburden are also produced as barren rock containing naturally radioactive materials.

Uranium mill tailings are the main waste from the milling process representing the largest volume of radioactive waste generated in the nuclear fuel cycle. Uranium mill tailings are the undissolved, nonmetallic constituents remaining after the extraction of uranium by leaching. Thus the tailings are part of the liquid waste behind the leaching process. They are stored in settling ponds, and after being dried off, become solid and are stored in large piles, known as uranium mill tailing piles. These piles need to be covered with clay and soil to prevent runoff and to reduce radon emissions and dispersion of fine particles. Other effluents from uranium milling include gaseous wastes of uranium and its daughter products dusts and liquid wastes of spent chemicals from the grinding and leaching processes.

Wastes from the conversion process are mostly gases and liquids effluents. The gaseous effluents are the particulates produced from the processing of feed uranium materials and the liquid effluents are from the process of uranium recovery and using liquids for waste treatment through wet scrubbing. Similar radioactive wastes are also produced from the operation of uranium enrichment and fuel fabrication.

In terms of radioactivity release, mining and milling contribute to the largest amount of release mostly as ^{222}Rn gas. Uranium and its decay products (^{238}U, ^{234}U, ^{234}Th, ^{232}Th, ^{230}Th, ^{226}Ra, ^{210}Po, ^{210}Pb), released as dust particles make up for most of other radioactivity release. Similarly, from conversion, enrichment, and fuel fabrication, uranium and its decay products are released (except ^{232}Th, ^{230}Th, ^{226}Ra, ^{210}Po, ^{210}Pb that are separated out from the uranium purification processes). Compared to uranium mining and milling, the level of radioactivity release is much lower from conversion, enrichment, and fuel fabrication.

Table 6.2 shows the levels of activity release from the front-end of nuclear fuel cycle for various nuclides. The amount of release in the table is normalized based on providing nuclear fuel for annual production of 1 GWe electricity. In terms of waste classification, the wastes produced in the front-end nuclear fuel cycle are low level waste or exempt waste (with very low activity contents).

6.4.2 Wastes from Reactor Operation

Besides spent nuclear fuel, various nuclear wastes are generated from nuclear reactor operation as gaseous, liquid, and solid wastes. We will discuss mainly the cases of PWRs and BWRs.

Table 6.2 Release of radionuclides from the front-end of nuclear fuel cycle

		Normalized annual release of radioactivity (GBq per GWe per year)							
	Mining	Milling		Conversion		Enrichment		Fuel fabrication	
	Total (mostly airborne)	Airborne	Liquid	Airborne	Liquid	Airborne	Liquid	Airborne	Liquid
^{210}Pb		0.02	0.01						
^{210}Po		0.02							
^{222}Rn	75,000	3000							
^{226}Ra		0.02	0.02		1.1×10^{-4}				
^{228}Th				2.2×10^{-5}					
^{230}Th		0.02	0.01	4.0×10^{-4}					
^{232}Th				2.2×10^{-5}					
^{234}Th		0.4		0.13		1.3×10^{-3}		3.4×10^{-4}	0.17
^{234}U				0.13	0.094	1.3×10^{-3}	0.01	3.4×10^{-4}	0.17
^{235}U				6.1×10^{-3}	4.3×10^{-3}	6×10^{-5}	5×10^{-4}	1.4×10^{-6}	1.4×10^{-3}
^{238}U		0.4	0.3	0.13	0.094	1.3×10^{-3}	0.01	3.4×10^{-4}	0.17

Data source: UNSCEAR (1993)

In PWRs, gaseous wastes are produced from the primary system, the secondary system, and the plant building ventilation system. The gases from the primary system are from the chemical and volume control system (CVCS), pressurizer relief tank (of the reactor cooling system), and the liquid waste holdup tanks. The CVCS is a system used to remove undesirable radioactive materials (i.e., fission products and corrosion products) from the reactor coolant system, thus to reduce radiation level of the coolant. The system is also used to add appropriate chemicals for the control of criticality or materials corrosion. One of the functions of CVCS is also to control the pressure in the primary system through the addition of both hydrogen and nitrogen to the volume control tank of CVCS. So the primary system gases include fission product gases, and hydrogen and nitrogen gases. These gases are collected in a storage tank, passed through a catalytic recombiner (where hydrogen and oxygen are combined to form water to prevent hydrogen levels from building up to flammable or explosive concentration levels). The collected gases are stored in 60 day holdup tanks for the decay of the short-lived noble gases, treated with filters (e.g., HEPA filters or charcoal filters) for particulate matter removal, and are discharged. The HEPA (high efficiency particulate air) filter is expected to provide greater than 99.97% efficiency for the capture of particles of various sizes. Through the treatments, the amount of radioactivity release should be controlled within the regulatory safety limit.

The gases from the secondary system comes from the release during steam generator blowdown (removal of water, suspended solids, and bottom sludge from the steam generator to avoid concentration of impurities), or as ejected gases by a steam jet from the low pressure side of the turbine, or as effluent from the turbine gland seal. Steam generator blowdown refers to forced replacement of used water in the secondary side of the steam generators with fresh water for impurities removal. Radioactivity contained in the secondary system gases is typically much lower than the primary gases and may come from the presence of holes in steam generator tubes allowing movement of radioactive gases from the primary system. Turbine gland seal refers to the use of auxiliary steam supply or inlet steam to prevent or reduce steam leakage between the rotating and stationary components of the *turbines*. Air from building ventilation has large volume but contains a low level radioactivity. These gaseous wastes are treated with filtration for activity removal (e.g., iodine) before discharge.

In the case of BWRs, the coolant is directly converted into steam within the reactor. Therefore, the turbine and generator are directly contaminated. The gaseous waste from BWRs arises from steam jet air ejector in turbine, containment/drywell, or turbine gland seal. Steam jet air ejector is to produce vacuum in the turbine through high pressure steam expansion. The collected gaseous wastes are stored in hold-up tanks for the decay of short-lived fission product noble gases and treated for iodine removal with charcoal bed and for the removal of particulate matters with high efficiency particulate air (HEPA) filters. Hydrogen recombiners are also used to reduce hydrogen concentration levels.

Liquid wastes in LWRs represent water and other liquids that have leaked or been released from the primary coolant system, the CVCS, pump shafts, valve seals, or

Table 6.3 Annual release of radionuclides (per GWe of electricity generation) from nuclear reactors (based on the data reported for the 1990–1994 period) (data source: UNSCEAR 2000)

	Normalized release per GWe of electricity generation (TBq per GWe per year)						
	Airborne Release					Liquid Release	
	Noble gas (^{133}Xe, ^{135}Xe, ^{85}Kr)	^{3}H	^{14}C	^{131}I	Particulates (mostly as ^{88}Rb, ^{89}Rb, ^{139}Ba)	^{3}H	Other (^{90}Sr, ^{137}Cs, ^{51}Cr, ^{54}Mn, ^{58}Co, ^{60}Co, and ^{95}Zr)
PWR	27	2.3	0.22	0.0003	0.0002	22	0.019
BWR	350	0.94	0.51	0.0008	0.18	0.94	0.043
GCR	1600	4.7	1.4	0.0014	0.0003	220	0.51
HWR	2100	650	1.6	0.0004	0.00005	490	0.13
LWGR	1700	26	1.3	0.007	0.014	11	0.005
FBR	380	49	0.12	0.0003	0.012	1.8	0.049
Total	330	36	0.44	0.0007	0.040	48	0.047
	Total combined release: 415 (TBq)						

from steam generator blowdown. The wastes also include waste oil, liquids from various floor drains and sumps, laboratory and laundry drains.

Typically, the liquid wastes are collected in large storage tanks for holdup (up to 30 days). The liquids are then processed using filters (to remove particulate matter) and ion exchange resins (to remove dissolved radioactive and chemical contaminants). The liquid wastes must meet regulatory criteria before discharge to the environment. To meet the criteria, liquid waste may be further treated through additional ion exchangers or reverse osmosis units (see Sect. 13.5.1). Evaporators also used to purify/process water and to minimize the waste volume.

Release of radioactivity from nuclear power plant operation takes place mostly through liquids and gaseous effluents. The largest amount of activity release is from fission product noble gases (mostly as xenon (^{133}Xe, ^{135}Xe) and krypton (^{85}Kr) in gaseous effluents. The total value of activity release as noble gases from nuclear reactors is 330 TBq (per GWe of electricity generated) as an annual average. The next largest contributor to activity release from nuclear reactors is tritium in both gaseous (36 TBq per GWe) and liquid effluents (48 TBq per GWe). Other important radionuclides released include ^{14}C, ^{131}I, and various fission products or activated corrosion products (e.g., ^{88}Rb, ^{89}Rb, ^{139}Ba in gaseous release and ^{90}Sr, ^{137}Cs, ^{51}Cr, ^{54}Mn, ^{58}Co, ^{60}Co, and ^{95}Zr in liquid release). These values are based on the observations during the period between 1990 and 1994 as global average for all types of nuclear power plants. These numbers are summarized in Table 6.3 (for different types of nuclear reactors).

As radioactive species are distributed throughout the nuclear power plant, various types of solid radioactive waste are also produced from nuclear reactor operations. These wastes are low level waste (LLW) (in the U.S.) or low and intermediate level waste (LILW) (typically outside the U.S.). A large portion of these wastes is generated from the treatment of radioactive liquid and gaseous effluents as spent ion-exchange resins, discarded filter material, or evaporator residue. Spent ion exchange resins are placed into a shipping container where excess water is removed prior to transfer to a disposal site. Sometimes, resins are solidified. Concentrated liquids of evaporator residue are produced by the evaporation of a wide variety of reactor liquid streams. These concentrated liquids are solidified in various materials such as cement. Filter sludges are the waste produced by precoat filters and consist of both the filter (granular/powdered/fiber) material and the thin layer of dissolved radioactive solids retained by the filter. More details of these treatment processes are given in Sect. 13.5.

Other low level wastes include various types of activated reactor hardware, discarded equipment & tools, or contaminated trash (called dry active waste) from day-to-day operations, maintenance, or modification activities. Contaminated trash is often referred to as compactible dry active waste (DAW). This is bulk material or solids that become contaminated due to daily operations and maintenance activities. Anti-contamination clothing that is no longer usable or cannot be laundered (many plants clean and reuse this clothing until no longer re-useable) is part of this waste along with rags or other materials used in cleaning. Paper, plastic, rubber, metal cans, etc. are also part of this group.

Discarded equipment and tools as noncompactible trash also become LLW/LILW. These include bulk materials or solids that are typically composed of miscellaneous piping, small valves, conduit, fitting tools, concrete, woods, dirt, glass, leaded shielding materials, filter frames or canisters, and other scrap material discarded following maintenance or modification activities. Activated reactor hardware refers to nonfuel irradiated reactor components such as fuel channels, control rods, and in-core instrumentation that has been exposed to neutron flux.

The activity in these LLW/LILWs are mostly from activation products (e.g., ^{60}Co, ^{63}Ni, ^{55}Fe) and fission products (e.g., ^{137}Cs and ^{90}Sr). These materials are normally stored onsite and periodically shipped to authorized waste storage facilities and disposed of for long-term isolation in an engineered disposal facility. Further details of low level wastes and their management are discussed in Chap. 13.

6.4.3 Wastes from Reprocessing

After going through reprocessing of spent nuclear fuel, the activity in spent fuel will be concentrated in the liquid high level waste. This liquid waste is solidified through vitrification resulting in immobilized glass. Solid wastes from reprocessing also include fuel-cladding hulls, particulate filters, discarded equipment tools, and contaminated trash. Unlike the nuclear waste from nuclear power plants, these solid wastes are contaminated with TRUs along with fission products and activation products and are disposed of after compaction. Most of the shorter-lived fission products (e.g., ^{131}I, ^{133}Xe) will have decayed away in these waste.

Any gaseous nuclides released from reprocessing operation must be collected from the off-gas streams and be treated. Certain fractions of radionuclides after treatment are still released to the environment. As shown in Table 6.4, krypton-85 (^{85}Kr) represents the largest inventory of the release (6300 TBq per GWe through air) followed by tritium (24 TBq through air and 270 TBq in the liquids per GWe).

Table 6.4 Annual average release of radioactivity from reprocessing (as release in TBq per GWe of electricity generation during the period 1990–1994) (data source: UNSCEAR 2000)

	Airborne release	Liquid release
^{3}H	24	270
^{14}C	0.4	0.8
^{85}Kr	6300	–
^{90}Sr	–	2.0
^{106}Ru	–	2.1
^{129}I	0.001	0.03
^{131}I	9×10^{-5}	–
^{137}Cs	8×10^{-5}	1.03

Other major nuclides released include carbon-14 (both gaseous and liquid release) and strontium-90, ruthenium-106 (^{106}Ru), iodine-129, and cesium-137 in the liquid effluents. The details of the release are summarized in Table 6.4. Release of ^{85}Kr is expected to be significantly reduced due to recent regulatory policy development requiring the removal of ^{85}Kr from effluents. Except ^{85}Kr and ^{106}Ru, most other radionuclides are included in solid wastes from reprocessing.

6.4.4 *Classification of Radioactive Waste*

As wide varieties of materials in different physical and chemical forms are generated as nuclear waste from nuclear power plants or nuclear fuel cycle facilities, classifying these wastes is desirable for management efficiency. Classification, i.e., categorization of wastes, allows the necessary handling of the wastes to be commensurate to the hazard contained in the waste and also helps to identify suitable disposal options. In terms of the criteria for classification, the level of potential hazard to humans (including both the occupational workers and the public) is considered such that the effort made to handle each class of waste matches the level of effort needed to cope with the potential hazard. Potential hazards of nuclear waste material depend on: (1) the amount and type of radionuclides contained, (2) the activity and half-life of the radionuclides contained, (3) the environmental behavior of the contained radionuclides, and (4) the chemical/physical forms of the radionuclides.

International Atomic Energy Agency (IAEA) provides a comprehensive framework of waste classification covering all types of nuclear waste including exempt waste, very short-lived waste, very low level waste, low level waste, intermediate level waste, and high level waste (Fig. 6.5). The IAEA framework directly connects the classes of nuclear waste with disposal options. While the approach is focused on solid radioactive waste, IAEA indicates that the classification is also applicable to the management of liquid and gaseous waste, with appropriate consideration of necessary waste processing to produce a solid waste form that is suitable for disposal. Below are the suggestions from IAEA (2009).

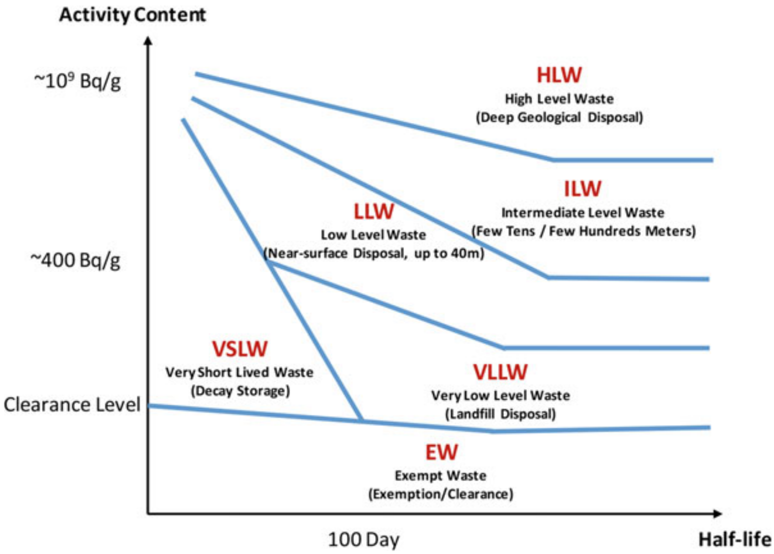

Fig. 6.5 A conceptual illustration of the waste classification scheme suggested by IAEA (The horizontal and vertical axis represents the half-lives of the radionuclides contained in the waste and the activity content of the waste, respectively. For short-lived radionuclides, a period of institutional control (i.e., providing security and surveillance over a given period, ~ 300 years) is imposed along with the classification system to provide a reasonable degree of assurance of safety.)

Exempt Waste (EW) This is a type of nuclear waste with the level of contamination at or below so-called the clearance level. An example of such clearance level is that radiation dose caused by the contamination is less than 0.01 mSv per yr. For the disposal of this waste, no radiological restriction deems necessary. The waste could be exempt or excluded from regulatory control for radiation protection purposes (IAEA 2004).

Very Short Lived Waste (VSLW) This class includes waste containing very short-lived radionuclides while allowing the presence of longer-lived radionuclides at the clearance level. The level of longer-lived radionuclides content was suggested at 4000 Bq/g for alpha emitters and at 400 Bq/g as overall average per waste package. This waste type often arises from research and medical institutions. This waste is to be stored (~a few years) until the short-lived radionuclides decay away. Thus the waste can eventually be disposed of without radiological restriction.

Very Low Level Waste (VLLW) The level of contamination in this waste is very low such that the waste can be disposed of without requiring high level of containment and isolation from the accessible environment. So disposal in near surface landfill type facilities with limited regulatory control for radiation protection and safety can be acceptable. A large portion of radioactive waste arising from uranium mining and milling or nuclear power plant decommissioning or environmental restoration of contaminated sites would fall into this category.

Low Level Waste (LLW) This waste contains long lived radionuclides at relatively low level concentration but may include higher levels of short-lived radionuclides. Typically a radionuclide with a half-life of less than about 30 years is considered short lived. It is necessary to isolate and contain this waste from the accessible environment in a robust engineered disposal system. The disposal system must remain intact for periods of up to a few hundred years and can be in the form of near surface facilities. Several hundreds or up to tens of kBq per gram have been considered as the limit for the long-lived radionuclides (such as ^{14}C, ^{36}Cl, ^{63}Ni, ^{93}Zr, ^{94}Nb, ^{99}Tc and ^{129}I) for this class of waste depending upon the characteristics of the disposal site and facility.

Intermediate Level Waste (ILW) This type of waste contains higher concentration of long lived radionuclides compared to LLW. The waste may also contain alpha emitting long lived radionuclides. Therefore, compared to LLW, a greater degree of containment and isolation from the accessible environment is needed for the disposal of the waste. Instead of near surface facilities, a disposal system at deeper depth in the order of tens or hundreds of meters is suggested for the waste. In the past, need for shielding to limit radiation dose during handling and transport was also considered to distinguish ILW from LLW. In the past, the LLW waste with a contact dose rate greater than 2 mSv/h was considered requiring shielding and became ILW. However, such consideration is no longer recommended by IAEA as contact dose is not necessarily a determining factor for the long term safety of a disposal facility.

High Level Waste (HLW) Compared to ILW, HLW contains large concentrations of both short and long lived radionuclides. Typically, the levels of activity concentration in HLW are in the range of 10^4–10^6 TBq/m^3. The waste also contains significant quantities of decay heat over the periods of several centuries. A deep geological disposal system with a greater degree of containment and isolation from the accessible environment is used for HLW disposal. The disposal system is usually several hundred meters below the surface using stable geological formations and engineered barriers. Consideration of heat dissipation through the application of temperature limits in the system is an important part of the facility design.

Example 6.5: Estimation of Nuclear Waste Generation

1. Calculate the amount of natural uranium that would have to be mined to result in an inventory of spent fuel of 100,000 MTIHM (metric tons initial heavy metal). Assume a once-through LWR fuel cycle, using fresh fuel enriched to 3% in ^{235}U. Assume an enrichment plant tails assay of 0.2% of ^{235}U, and neglect any material losses at other front-end stages of the fuel cycle.
2. Estimate the volume of uranium mill tailings produced from uranium mining/milling operation in Part 1). Also, estimate the amount of radioactivity in the produced uranium mill tailings (at the time of its production).

(continued)

Example 6.5 (continued)

Uranium ore milling typically results in the recovery of 90% of the uranium originally in the ore. The remainder is discharged in the mill tailings, along with the radioactive daughter products of the uranium decay chain originally present in the ore.

3. The recovered uranium from Part 1) is refined, converted to UF_6 and then enriched. Estimate the amount the radioactivity of the enrichment plant tails (at the time of its production). Suppose that 80% of the uranium feed to the enrichment plant is rejected as tails.

   Note: In all calculations, ignore the contribution of the ^{235}U decay chain to the total radioactivity. Use the information of 4n + 2 (^{238}U) decay chain given Sect. 3.1.4.
4. Estimate the total volume of spent fuel and LLW after generating 100,000 MTIHM of spent fuel. Note that 31,758 kgU is needed to produce 1 GWe year. of electricity (from Sect. 6.3).
5. Estimate the total area required to dispose of the wastes (from Part a), separately for each type of waste. State any assumptions made. Compare the resulting estimate with a number you are familiar with.

Given:

<table>
<tr><td rowspan="6"><i>LLW generation from fuel cycle</i>
Conversion 20 m^3 /GWe
Enrichment 45 m^3 /GWe
Fuel fabrication 85 m^3/GWe
Reactor operation 56 m^3/GWe (average of 107 and 30 m^3/GWe for PWR and BWR, repectively)</td><td colspan="2"><i>Mass density</i> (ton/m^3)</td></tr>
<tr><td>Uranium ore</td><td>5.145</td></tr>
<tr><td>^{238}U</td><td>19.1</td></tr>
<tr><td>U_3O_8</td><td>8.3</td></tr>
<tr><td>UO_2</td><td>10.97</td></tr>
<tr><td>U mill tailing</td><td>1.58</td></tr>
<tr><td><i>Spent fuel generation</i>
10 m^3/GWe</td><td colspan="2" rowspan="2"><i>We ignore the contribution of ^{235}U decay chain to the total radio-activity calculation.</i></td></tr>
<tr><td><i>Area required for disposal</i>
Spent fuel: ~2.8 MTHM/100m^2
LLW: Depth of trenches ~5 m
Mill tailings: Height of U mill tailing pile ~10 m</td></tr>
</table>

Solutions:

1. We neglect any material losses at the front-end stages in the fuel cycle. Then the amount of natural uranium to be mined is equal to the amount of feed uranium needed (F) for enrichment.

   Material balance involved:

(continued)

Example 6.5 (continued)

$$F = P + T$$

$$x_F F = x_p P + x_T T$$

where, F is feed, P is product, T is tail, x_F is feed of ^{235}U concentration, x_P is product of ^{235}U concentration and x_T is tail of ^{235}U concentration.

Amount natural uranium have to be mined = F = $(X_P - X_T) / (X_F - X_T) \cdot P$

With P = amount of enriched product = 100,000 MTIHM,
X_P = enrichment of product = 3%,
X_F = enrichment of feed = 0.72% and
X_T = enrichment plant tails = 0.2% then,

The amount of natural uranium to be mined = (3%–0.2%)/(0.73%–0.2%) ·100,000 MTIHM
= **528,302 MTIHM**

2. To have 528,302 MTIHM of natural uranium as feed for enrichment, the amount of U_3O_8 needed is

528,302 ·(842/714) = 623,012 metric tons U_3O_8.

Assume that average grade of U ore is 0.25% in U_3O_8 (while ranging between 0.15 and 0.3%). Because uranium milling typically results in the recovery of 90% of the uranium originally present in the ore, the percent of U_3O_8 from the ore is 90% · 0.25% = 0.225%.

→ Total amount of ore needed is 623,012/0.00225 = 2.769×10^8 tons
→ Amount of uranium mill tailings produced is 2.769×10^8 ·0.99775 = 276,276,975 tons ≈ 276 million tons

Mass density of mill tailing is around 1.58 tons/m^3

→ Total volume of uranium mill tailings produced is around 276,276,975/ 1.58 ≈ **1.748×10^8 m^3**

With the recovery of 90% of the uranium with milling, 10% of uranium is discharged from uranium mill tailing. Assume all the radioactivity in uranium comes from the ^{238}U and its daughter products. Since the half-lives of the daughter products are much smaller than the half-life of uranium, secular equilibrium is assumed between ^{238}U and its decay products (see the discussions on secular equilibrium in Sect. 3.1.7). Therefore, all the daughter products have the same activity level as that of the parent uranium. Radioactivity from each radionuclide in the U-238 decay chain are shown in the table below.

Radioactivity of each radionuclide in the U-238 decay chain:

(continued)

Example 6.5 (continued)

Number	Nuclide	Half-life	Activity after milling
1	U-238	4.51E9 y	$0.1A_0$
2	Th-234	24.1 d	A_0
3	Pa-234	1.17 m	A_0
4	U-234	2.47E5 y	$0.1A_0$
5	Th-230	8.0E4 y	A_0
6	Ra-226	1602 y	A_0
7	Rn-222	3.82 d	A_0
8	Po-218	3.05 m	A_0
9	Pb-214	26.8 m	A_0
10	Bi-214	164 ms	A_0
11	Po-214	21 y	A_0
12	Pb-210	5.01 d	A_0
13	Bi-210	138.4 d	A_0
14	Pb-206	Stable	A_0
Total			$12.2\ A_0$

Specific activity of ^{238}U = Activity/ atomic mass in gram
$= (6.023 \times 10^{23}/238) \cdot (0.693/(4.5 \times 10^9 \cdot 365 \cdot 24 \cdot 3600)$
$= 12358.05205$ Bq/g
$= 3.34 \times 10^{-7}$ Ci/g

→ Activity of ^{238}U in the ore is $A_0 = 528{,}302 \times 10^6 \cdot 99.28\% \cdot 3.34 \times 10^{-7}$ Ci = 175,182 Ci

→ Activity in the uranium mill tailing is 12.2 times this activity (as shown in the table) = 12.2 · 175,182 Ci

= **2.137×10^6 Ci**

3. The amount of uranium remaining after enrichment (Part 1) is 528,302–100,000 (enriched U portion) = 428,302 MTIHM. As 80% of the uranium feed to the enrichment plant is rejected as tails, the mass of uranium in the tails is: 80% · 428,302 = 342,642 MTIHM.

   Activity of ^{238}U in 342,642 MTIHM is around $342{,}642 \times 10^6$ (g) $\cdot 3.34 \times 10^{-7}$ (Ci/g) ≈ 114,442 Ci

   The activity of enrichment tails is around **114,442 Ci.**

4. From the mass balance in Sect. 6.3, we have:

31,758 kg of total uranium at BOC to produce 1 GWe-year → 100,000 MTIHM will make $100000 \times 10^3/(31{,}758)$ GWe-year = 3149 GWe-year.

(continued)

Example 6.5 (continued)

From given information from the problem, 10 m^3 spent fuel is generated per GWe-year

→ Total volume of spent fuel is 10 × 2780 = **31,490 m^3 LLW**

Each GWe-year corresponds to the generation of LLW at 20 m^3 from conversion, 45 m^3 from enrichment, and 85 m^3 from fuel fabrication.

→ Total LLW after generating 100,000 MTIHM is 2780 · (20 + 45 + 85 + 56) **= 648,620 m^3 LLW**.

5. Assume that mass of uranium in the spent fuel is the same as the mass of uranium at BOC and the area required for disposal spent fuel is ≈ 2.8 MTHM/100 m^2

→ The area required for spent fuel is 100,000/(2.8/100) = 3.57×10^6 m^2 = 3.57 km^2 ≈ **1.8 times the total area of Monaco (≈ 2 km^2)**.

Assume that LLW will be disposal in trenches with ~5 m depth.

→ The area required for LLW is 648,620/5 m^2 = 129,723 m^2 ≈ **20 (12 ~ 32) times the area of a FIFA-sanctioned soccer field (4050–10,800 m^2)**.

Assume that mill tailings is disposal in U mill tailing pile with height ≈ 10 m

→ The area required for mill tailings (from Part 2) is 1.748×10^8/10 m^2 = 1.748×10^7 m^2 ≈ **1/5 times the area of Manhattan (≈ 87 km^2)**

6.4.5 Overall Radiation Exposure from Nuclear Fuel Cycles

Presence of various radionuclides in the nuclear fuel cycle causes radiation exposure to the workers and the members of the public. In terms of occupational radiation exposure, the current average effective dose among the workers is 1 mSv per year per person (UNSCEAR 2008) in the nuclear fuel cycle. This number represents a fourfold reduction from the dose level in the 1970s (i.e., 4.4 mSv per worker) and reflects steady decrease in the annual effective dose to the monitored workers over the years.

The collective dose to local and regional populations from these radionuclides has been estimated by the United Nations. The results for the period of 1998–2002 are summarized in Table 6.5. The annual collective dose to the local and regional population due to the sources of radiation in the nuclear fuel cycle was 0.72 person-Sv per GWe. The largest contribution was from nuclear reactor operation with 38% share followed by uranium mining responsible for 26% of the total. The main radionuclides contributing to the local and regional population from nuclear power operation were ^{14}C and tritium. In uranium mining, radon gas is the key dose

Table 6.5 Annual Collective Dose to the Public (to the local and regional populations) from the Nuclear Fuel Cycle to Support the Generation of 1 GWe Electricity (data source: UNSCEAR 2008)

Source	Annual collective effective dose per GWe (person-Sv)	Annual per caput dose (μSv) to the local population	Major radionuclides
Mining	0.19	25	Rn
Milling	0.008		Rn (86%), U, Th, Ra
Tailings	0.04		Rn
Fabrication	0.003	0.2	U
Reactor operation (airborne effluents)	0.22	0.1	^{14}C, ^{3}H, noble gas, particulates, iodine
Reactor operation (liquid effluents)	0.05		^{3}H, particulates
Reprocessing (airborne effluents)	0.028	2	^{14}C, particulates, ^{3}H, ^{85}Kr
Reprocessing (liquid effluents)	0.081		^{14}C, ^{134}Cs, ^{137}Cs, ^{106}Ru, ^{90}Sr, particulates
Transportation	<0.1	<0.1	^{137}Cs, ^{60}Co, ^{244}Cm
Disposal of radioactive waste		<0.001	
Total	0.72	27.4	

contributor. In the case of reprocessing operation, fission products such as cesium (^{134}Cs and ^{137}Cs) and ruthenium (^{106}Ru) are the main dose contributors. Important radionuclides in the nuclear fuel cycle as the main source of radiation exposure to the public and workers are summarized in Fig. 6.6. The total collective dose among the global population from the release of gaseous and liquid effluents from the nuclear fuel cycle is about 200 person-Sv per year.

The dose to the public has also steadily decreased over the years. The normalized collective effective dose to the public was 12 person-Sv per GWe per year in the 1970s in comparison to 0.92 person-Sv per GWe in 1990–1994 and 0.72 person-Sv per GWe in 1998–2002. The recent dose impact of the nuclear fuel cycle on the public is more than a factor of 16 lower compared to the dose impact in the 1970s.

In addition to the nuclear fuel cycle and nuclear power plants, there are a variety of other (man-made or natural) sources of radiation exposure (UNSCEAR 2008). Contributions of these sources to public radiation exposure are summarized in Table 6.6. About 80% of public radiation dose comes from natural sources with radon gas in the soil being the biggest contributor. The remaining 20% comes mostly from medical diagnostic imaging, e.g., x-rays and CT scanning. For an average member of the public, radiation exposure caused by the nuclear fuel cycle is about 0.2 μSv per person per year. In comparison to other sources of radiation exposure, the average individual radiation dose from the nuclear fuel cycle is less than 0.007%

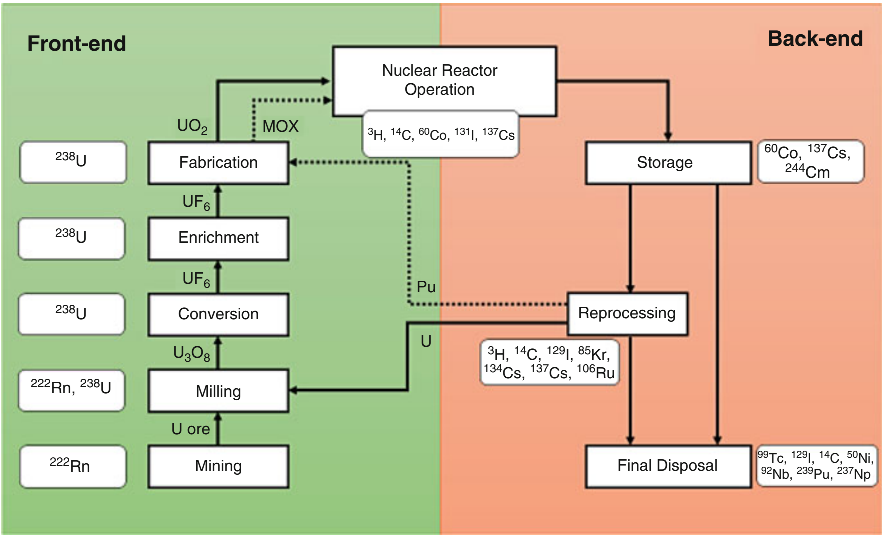

Fig. 6.6 Key radionuclides of concern from the nuclear fuel cycle as source of radiation exposure to the public and workers

Table 6.6 Annual dose to human from various sources of ionizing radiation as world average (data source: UNSCEAR 2008)

Source		Annual dose as world average (mSv/yr)	Typical range of individual doses
Natural	Radon gas (inhalation)	1.26	0.2–10
	External terrestrial	0.48	0.3–1
	Ingestion of food	0.29	0.2–1
	Cosmic radiation	0.39	0.3–1
	Total natural	2.4	1–13
Man-made	Medical diagnosis	0.6	0–2.0
	Atmospheric nuclear testing	0.005	Declining due to decay from 0.11 in 1963
	Occupational exposure	0.005	0–20 (highest dose is from radon among miners)
	Chernobyl accident	0.002	Decreasing from a maximum of 0.04 in 1986
	Nuclear fuel cycle (public exposure)	0.0002	Could go up to 0.02 mSv/y for some critical groups
	Total man-made	0.6	0 – Several tens
Total		3.0	

of the total. If the dose from the Chernobyl accident is added to the dose from the nuclear fuel cycle, the contribution becomes 0.073%. This indicates that radiation impact from nuclear power or nuclear fuel cycle on humans, on average, are not significant in comparison to the contributions from other natural or man-made sources of radiation exposure.

6.5 Conclusion

Understanding the processes of physical and chemical treatment of nuclear materials in both the front-end and back-end of nuclear fuel cycle is fundamental to understand the generation of nuclear waste from the use of nuclear energy. This chapter provided an overview of nuclear fuel cycle for such understanding. All of the steps in the front-end nuclear fuel cycle are to manufacture fuel for nuclear reactor operation. The fuel is initially only made of uranium but, after reactor irradiations, becomes a mixture of uranium, plutonium, the minor actinides, fission products, and other activation products. The variations in the composition with byproducts production depend mainly on the total fission energy extracted from the fuel. Further discussions of the spent fuel composition are provided in Chap. 7. Handling of the discharged nuclear fuels through the stage of final disposal is an important part of the back-end of nuclear fuel cycle. For the purpose of resource utilization or waste transmutation, recycling of spent fuel with reprocessing can be considered. Details of the required steps and materials processing are discussed in Chap. 8. Analysis of system-wide cross cutting issues such as economics and nuclear security and nonproliferation are described in Chap. 15. As observed, the radiological impact on humans from the release of radioactive materials from nuclear power or nuclear fuel cycle, in comparison to the contributions from other natural or man-made sources of radiation exposure, is not significant, on average. This also implies that, with proper emplacement of regulatory control, the supporting nuclear waste management activities can be conducted without undue burden on the society.

Homework

Problem 6.1 List two or three key radionuclides of concern generated in nuclear power plant in nuclear waste management for each of the following category. Explain why they are important.

(a) Fission products in spent fuel
(b) Activation products in spent fuel
(c) Activation products in steel structure
(d) Activation products in the coolant

Problem 6.2 The purpose of this problem is to identify relative importance of different radionuclides in nuclear waste management. For this purpose, you are to use different evaluation methods to identify relative importance of different radionuclides.

Assume that there are 13 different types of nuclear waste, each containing 1 Ci of each of these 13 radionuclides (as shown in Table 6.7). Determine the relative importance of each of the waste by using the following three evaluation methods:

Table 6.7 Supporting information for the radionuclides of concern "(for Problem 6.2)"

Nuclide	Half-Life	MPC (Bq/m3)	DPUI (Sv/Bq)	Solubility (mol/L)	K_d (ml/g)
H-3	12.3 year	5E+4	1.73E-11	10	0
C-14	5730 year	1E+4	5.64E-10	0.1	5
Co-60	5.26 year	1E+3	7.28E-9	0.1	60
Ni-63	100 year	2E+3	1.56E-10	1E-3	400
Sr-90	29.1 year	4E+0	3.23E-9	1E-3	15
Tc-99	2.12 x 10^5 year	1E+5	3.95E-10	1E-3	0.1
I-129	1.6 x 10^7 year	5E+2	7.46E-8	10	1
I-131	8.05 days	2E+3	1.44E-8	10	1
Cs-137	30.17 year	2E+2	1.35E-8	3E-3	280
Np-237	2.14 x 10^6 year	1E+2	1.20E-6	3E-5	5
U-238	4.5 billion yrs	1E+4	6.42E-9	4E-5	35
Pu-239	24,400 year	3E+2	9.56E-7	2E-08	550
Am-241	432 year	1E+2	9.84E-7	1E-09	550

K_d values are for the case of sand from Health Physics, Vol. 59, No. 4, 1990
Solubility values are from EPRI, Report (Table 6.6) 1000802, November 2000; POSIVA Report TILA-96 (Table 11.2), 1996; SKB Report TR-06-32 (Table 8.1), 2006
$R_d = 1 + K_d$*[density of soil]/[porosity of soil]
Use, [density of soil] = 1.8 g/cm^3 and [porosity of soil] = 0.4
DPUI values are from the US Federal Guidance Report No. 11 (EPA-520/1-88-020, 1988)

a) Toxicity Index; b) Integrated Radiological Toxic Potential (IRTP), and; c) Health Risk Index (HRI).

a) Toxicity Index (TI): The TI for water ingestion is the volume of water (in m^3) in which wastes would have to be diluted to meet standards for radionuclide concentrations in drinking water. The TI is defined as,

$$\mathrm{TI}_i = (\mathrm{A_i}/\mathrm{MPC_{iw}})$$

where, A_i is the activity of ith element (e.g., Bq or Ci); MPC_{iw} is the maximum permissible concentration of ith element in water (e.g., Bq/m^3).

b) Integrated Radiological Toxic Potential (IRTP): IRTP is the time integral of the volume of water required to dilute a unit volume (1 m^3) of waste to a level at which it could be drunk by an individual member of the public without exceeding a current dose limit. The IRTP expresses toxic potential of nuclear wastes based on the TI concept but can take account of the time delays in groundwater transport. The IRTP is calculated as,

$$\mathrm{IRTP}_i = C.A_i.DPUI_i.\exp\left(-\lambda_i\, \mathrm{t_{gw}}\right)/\lambda_i\ \left(\mathrm{m^3.year}\right)$$

where, C is a constant (0.712 m^{3}Sv^{-1}); A_i is the activity of the radionuclide i at time t (Bq); $DPUI_i$ is the dose per unit intake of the radionuclide i by ingestion (Sv/Bq); λ_i is the decay constant (1/y), and; t_{gw} is the groundwater travel time, as 5000 year.

c) Health Risk Index $= RS_i * A_i * DPUI_i * \exp\left(-\lambda_i t_{gw}\right)\frac{1}{SQRT(Rd_i).}$

where, RS_i is the relative solubility, A_i is the activity of waste, $DPUI_i$ is the dose per unit intake of the radionuclide by ingestion, t_{gw} is the groundwater travel time as 5000 years, and Rd_i is the retardation factor of radionuclide *i*.

d) Compare and discuss the results from a), b), and c) by explaining the relative importance of radionuclides and why they are different with different evaluation methods.

Problem 6.3 Using the concept of toxicity index given in Problem 6.3, calculate the toxicity index of a waste containing 1 gram of each of the following radionuclides. Plot the changes in toxicity index in approximate scale with time and comment on the plot based on your observation (MPCs are given in μCi/ml).

- Cs-137 ($t_{1/2} = 30$ year.; $MPC_{water} = 2 \times 10^{-5}$)
- Ra-226 ($t_{1/2} = 1620$ year.; $MPC_{water} = 1 \times 10^{-8}$)
- I-129 ($t_{1/2} = 1.7 \times 10^{7}$ year.; $MPC_{water} = 4 \times 10^{-7}$)

Problem 6.4 The current regulatory definition for transuranic (TRU) waste encompasses any high level waste with total long-lived alpha activity greater than 10^{-7} Ci/g of waste. According to this definition, long-lived radionuclides are those with half-lives greater than 20 years. An alternative standard that has been proposed is natural ore containing 10% by weight of uranium, i.e., any material with specific long-lived alpha activity greater than the specific long-lived alpha activity greater than the specific long-lived activity of such ore would be considered to be TRU waste.

(a) Which of these is the stricter definition?
(b) The solid waste stream from mixed uranium-plutonium oxide fuel fabrication plants contains 2% by weight of plutonium isotopes. Assume that the isotopic composition of the plutonium is 60% by weight ^{239}Pu, 30% ^{240}Pu, and 10% ^{241}Pu. Would this waste stream be classified as TRU waste?

Problem 6.5

(a) In 1993, the U.S. Government agreed to begin purchasing highly enriched uranium (93%wt ^{235}U) from Russia in 1993 at a rate of 10 MTU/yr. for the first 5 years and 50 MT/yr. thereafter. This is part of the Megatons to Megawatts program (also called the U.S.-Russia Highly Enriched Uranium Purchase Agreement). The program was successfully completed in December 2013.

If the estimated inventory of highly enriched material at that time was 700 MTU, in what year was the last uranium purchased from Russia?

(b) The value of 10 MT of 93% enriched uranium when diluted with natural uranium (0.71% ^{235}U) to form fuel at 3.5% enrichment in ^{235}U is approximately

$200 million. How many metric tons of reactor fuel (in terms of uranium) was produced from 10 MT of the enriched uranium?

(c) If the 10 MT of highly enriched material was originally enriched from natural uranium how many metric tons of uranium tails were produced and disposed of in Russia (assume tails have a ^{235}U content of 0.2%)?

(d) How many metric tons of tails would be produced by the enrichment of natural uranium to produce the amount of 3.5% enriched ^{235}U reactor fuel in part (b)?

(e) The dilution to reactor fuel is effectively a loss of separative work, how much separative work was utilized in enriching 10 MT of 93% enriched uranium from natural uranium?

(f) What would be the separative work involved in enriching the number of metric tons of reactor fuel in part (b) from natural uranium to 3.5% enriched?

(g) Comment on the relative amounts of separative work in part (e) vs. that in part (d).

(h) Assuming the going rate for separative work is $70/kgSWU, how much money is lost in dilution? Is this loss justifiable relative to the value of the fuel in part (b)? Are there any other justifications for or consequences of this purchase?

Further Reading

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Chapter 7
Characteristics of Spent Fuel and Its Storage and Transportation

Abstract As the most significant type of nuclear waste in terms of risk to humans, the irradiated nuclear fuels discharged from nuclear reactor demand utmost care in their handling. This chapter describes the composition, radionuclide inventory, decay heat, and radiation dose of spent fuel as a function of burnup and cooling time to provide the basis of its safe handling. Radiation shielding, cooling, and criticality control during storage and transportation of spent fuel are also described.

Keywords Key radionuclides · Activity and decay heat · Radiation shielding · Criticality control · Storage and transportation

When the irradiated nuclear fuel is discharged from the reactor, it becomes a used fuel (called spent fuel by convention). Due to the presence of very high levels of radiation field and heat produced by these byproducts, spent fuel presents extreme levels of hazard demanding utmost care in handling. In addition, presence of fissile in spent fuel demands physical protection against theft or misuse of nuclear weapons usable materials for malicious purposes during storage, transportation, and disposal. Special precautions are also needed to prevent any criticality accidents during the handling in the back-end nuclear fuel cycle. Accordingly, various safety and security measures are in place throughout the entire phase of spent fuel management. This chapter covers basic issues of spent fuel management under the open/once-through fuel cycle. The recycling related issues such as reprocessing and transmutation are discussed in Chap. 8. Final disposal of spent fuel is covered in Chaps. 9, 10, 11, and 12. Nuclear security and nonproliferation issues are discussed in Chap. 15.

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7.1 Characteristics of Spent Fuel

7.1.1 General Characteristics of Spent Fuel

When spent fuel comes out of a nuclear reactor, it is in the form of a fuel assembly. One spent fuel assembly is the basic unit in all stages of spent fuel management. A single PWR or BWR spent fuel assembly weighs about 0.59 ~ 0.67 ton or 0.28 ~ 0.32 ton, respectively (as mentioned in Sect. 6.2.5). Its length is about 4 meters for a PWR and 4.5 meters for a BWR. The total radioactivity contained in a single freshly discharged PWR and BWR spent fuel assembly is about 3.7×10^3 pBq (1.0×10^8 Ci Ci) and 1.03×10^3 pBq (2.8×10^7 Ci), respectively (a low burnup LWR fuel according to the example given in Table 7.1). To give a perspective, note that these values exceed the total radioactivity released from the 2011 Fukushima accidents (~1000 pBq (penta Becquerel), 2.7×10^7 Ci) in Japan but is slightly lower than the total radioactivity released from the 1986 Chernobyl accident (5200 pBq, 1.4×10^8 Ci) in Ukraine.

Table 7.1 An example of physical characteristics of spent fuel from LWRs (GE BWR and Westinghouse PWR)

	BWR	PWR
Overall assembly length (m)	4.470	4.059
Cross section (cm)	13.9 × 13.9	21.4 × 21.4
Fuel pin array	8 × 8	17 × 17
Fuel pins/assembly	63	264
Nominal volume/ assembly (m^2)	0.0864	0.186
Assembly total weight (kg)	275.7	657.9
Uranium/assembly (kg)		
Initial	183.3	461.4
Discharge	176.5	441.2
Enrichment(wt% ^{235}U)		
Initial	2.75	3.20
Discharge	0.69	0.84
Plutonium/assembly at discharge (kg))	1.54	4.18
Other TRU elements/assembly at discharge (kg)	0.10	0.43
Fission products/assembly at discharge (kg)	5.2	15.7
Average discharge bumup (MWd/MTHM)	27,500	33,000
Average thermal power (kW/assembly)		
Discharge	278	1017
1 year after discharge	1.3	4.7
10 years after discharge	0.2	0.5
Average radioactivity (megacuries/assembly)		
Discharge	28.3	102.0
1 year after discharge	0.35	1.16
10 years after discharge	0.06	0.18

Derived from data source (Croff and Alexander 1980)

Table 7.2 Changes in total activity in pBq, i.e., 10^{15} Bq (Ci in paranthesis) of PWR and BWR spent fuel in a single assembly at different burnup and cooling time

	Spent fuel cooling time (years)						
Burnup (GWD/MTHM)	At discharge	0.5	1.0	3.0	5.0	10.0	30.0
BWR							
30	1060 (2.87E+07)	20.9 (5.66E+05)	12.5 (3.38E+05)	5.18 (1.40E+05)	3.47 (9.38E+04)	2.44 (6.60E+04)	1.31 (3.55E+04)
50	1110 (2.99E+07)	26.0 (7.04E+05)	16.7 (4.52E+05)	7.62 (2.06E+05)	5.32 (1.44E+05)	3.09 (1.03E+05)	2.26 (5.61E+04)
PWR							
35	4810 (1.30E+08)	84.7 (2.29E+06)	47.4 (1.28E+06)	22.2 (4.60E+05)	10.5 (2.85E+05)	7.14 (1.93E+05)	3.85 (1.04E+05)
60	3960 (1.07E+08)	86.6 (2.34E+06)	54.4 (1.47E+06)	23.5 (6.34E+05)	16.0 (4.32E+05)	11.3 (3.05E+05)	6.22 (1.68E+05)

Source: NRC (2000)

Basic physical characteristics of a typical spent fuel assembly are shown in Table 7.1 (as an example for a low burnup fuel from LWRs). As seen in the table, each fresh nuclear fuel assembly contains about 0.45 ton or 0.18 ton of UO_2 in PWR or BWR, respectively. The schematic of a fuel assembly is shown in Fig. 6.4. As spent fuel is heavy, long, and highly radioactive, its handling requires special procedures and precautions.

As a result of neutron irradiation, a host of irradiation byproducts are present in used fuel. These byproducts include the products of fission (called fission products) as well as uranium activation products (called activation products).

The inventory of radioactivity in spent fuel varies with burnup and the cooling time (time since reactor discharge). The activity inventory increases with burnup and decreases with cooling time as shown in Table 7.2. Details of how the activity of spent fuel varies as a function of burnup or cooling time are discussed in the next section. For example, at 1 year after discharge, the level of spent fuel activity is reduced to about 1% of the freshly discharged spent fuel. The activity is further reduced to ~0.2% at 10 years of cooling post discharge. These changes are shown in Table 7.2 for both PWRs and BWRs.

7.1.2 Nuclide Compositions of Spent Fuel

Between the fresh fuel and spent fuel, the content of ^{235}U is reduced from about 3–4% (weight percent) to less than 1% remaining in spent fuel. The content of ^{238}U is also reduced from about 96–97% to about 93–94% due to neutron capture. Fission products are produced, as a replacement of the lost mass of ^{235}U (and ^{238}U due to fast fission), in the amount of about 3–5%. Plutonium and other minor actinides are also produced from the activation of uranium, plutonium and the minor actinides followed by subsequent radioactive decays. The plutonium takes up about 1% of mass in spent fuel. An approximate composition of a 50,000 MWD/MT burnup case spent fuel is graphically depicted in Fig. 7.1.

As an example, differences in the composition of nuclear fuel between before and after reactor irradiation are also shown in Table 7.3 for two cases of burnup (35 and

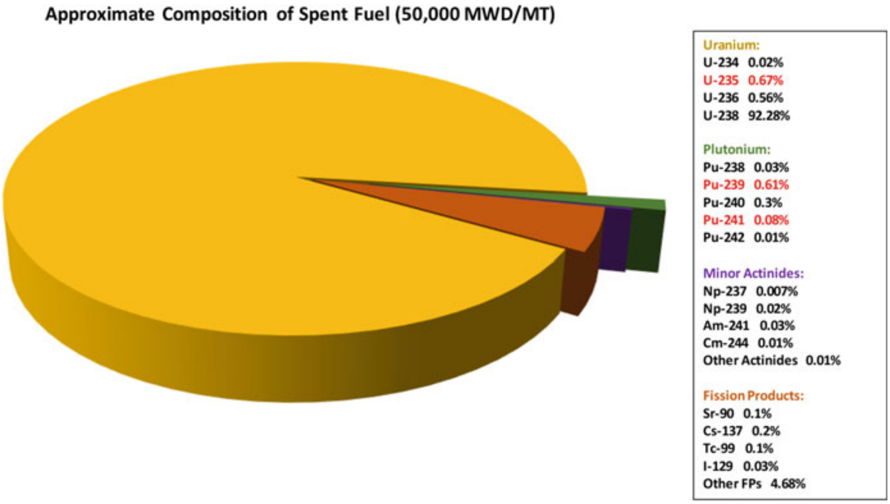


Fig. 7.1 Constituents of a typical nuclear spent fuel: an example of 50,000 MWd/MT burnup case

Table 7.3 An example of nuclear fuel composition change (in weight percent) between fresh and spent fuel

	3.3 wt.% enriched fuel, 35 GWD/MTHM			4 wt.% enriched fuel, 50 GWD/MTHM		
Nuclide	Before irradiation	After irradiation	Difference	Before irradiation	After irradiation	Difference
U-235	3.3	0.79	−2.51	4.0	0.67	−3.33
U-236	0	0.4	+0.4	0	0.56	+0.56
U-238	96.7	94.29	−2.41	96	92.28	−3.72
Fission products	0	3.51	+3.51	0	5.11	+5.11
Plutonium	0	0.905	+0.905	0	1.22	+1.22
Other minor actinides (Np+Am+Cm)	0	0.095	+0.095	0	0.14	+0.14
Total	**100**	**99.99**	**0.01** (mass of ^{234}U)	**100**	**99.98**	**0.02** (mass of ^{234}U)

50 GWD/MTHM). It can be noted that production of fission products, plutonium, and minor actinides is increased with burnup with higher uranium loss. In the case of nuclear reactor operation using natural (unenriched) uranium fuel (i.e., PHWR), the spent fuel (typically at 7.5 GWD/MTHM burnup) contains much smaller content of ^{235}U (0.23%), Pu (0.38%) and fission products (0.6%) with the ^{238}U content at about 98.8% due to low burnup.

The key radionuclides present in LWR spent fuel (at the time of reactor discharge) are listed in Table 7.4 with their half-lives, kinetic energy of the released particles, and radioactivity inventories at the time of reactor discharge.

Table 7.4 Type, half-life and initial activity of major nuclides in spent fuel

Nuclide	Type (production mechanism)	Half-life	Radiation (particle energy in MeV; averaged[a] if % is not given)	Activity (MBq) per assembly at discharge (33,000 MWD/MTU)
H-3	Fission/activation product	12.3 yr	β (0.0186)	1.40E+07
C-14	Activation product	5730 yr	β (0.157)	2.54E+04
Mn-54	Activation product	312.2 days	γ (0.835)	1.68E+06
Fe-55	Activation product	2.73 yr	β (0.231); beta disintegration energy	9.52E+07
Ni-59	Activation product	7.6×10^4 yr	β (1.073); beta disintegration energy	7.84E+04
Co-58	Activation product	70.88 days	γ (0.811), β(0.474)	1.17E+08
Co-60	Activation product	5.271 yr	γ (1.17 & 1.33), β (0.318)	1.34E+08
Ni-63	Activation product	100 yr	β (0.0669)	1.13E+07
Se-79	Fission product	$\leq 6.5 \times 10^4$ yr	β (0.16)	6.99E+03
Kr-85	Fission product	10.73 yr	γ (0.514), β (0.687)	1.62E+08
Sr-90	Fission product	29.1 yr	β (0.546)	1.29E+09
Zr-95	Fission/activation product	64.02 days	β (0.366, 0.400) γ (0.7567, 0.7242)	2.63E+10
Nb-95	Fission/activation product	34.97 days	γ (0.766), β (0.160)	2.67E+10
Tc-99	Fission/activation product	2.13×10^5 yr	β (0.293), γ (0.0897)	2.26E+05
Sn-119m	Activation product	293 days	γ (0.024)	1.14E+08
Sb-125	Activation product	2.76 yr	β (0.302, 0.13), γ (0.4279, 0.5005, 0.6359, 0.4634)	2.91E+07
Sn-126	Fission product	1.0×10^5 yr	β (0.25), γ (0.0876)	1.32E+04
I-129	Fission product	1.57×10^7 yr	β (0.15) γ (0.0398)	5.26E+02

(continued)

Table 7.4 (continued)

Nuclide	Type (production mechanism)	Half-life	Radiation (particle energy in MeV; averaged[a] if % is not given)	Activity (MBq) per assembly at discharge (33,000 MWD/MTU)
I-131	Fission product	8.04 days	β (0.606, . . .), γ (0.3645, . . .)	1.56E+10
Xe-133	Fission product	5.243 days	β (0.346, . . .), γ (0.081,. . .)	3.12E+10
Cs-137	Fission product	30.17 yr	β (0.514, . . .), γ (0.6617)	1.77E+09
Ce-144	Fission product	284.6 days	β (0.318, 0.185,..), γ (0.1335, 0.08, . . .)	2.06E+10
Eu-154	Fission product	8.59 yr	β (0.58, 0.27,..), γ (0.1231, 1.2745, . . .)	1.81E+08
Eu-155	Fission product	4.71 yr	β (0.15,. . .), γ (0.0865, 0.1053, . . .)	1.19E+08
Np-237	β^- decay of ^{237}U or α decay of ^{241}Am	2.14×10^6 yr	α (4.788, 4.771, ...) γ (0.0294, 0.0865, ...)	5.19E+03
Pu-238	β^- decay of ^{238}Np or α decay of ^{244}Cm	87.7 yr	α (5.4992, 5.4565, ...) γ (0.0435, 0.0999, . . .)	3.64E+07
Pu-239	^{238}Pu neutron capture or β^- decay of ^{239}Np or α decay of ^{243}Cm	2.41×10^4 yr	α (5.156, 5.143, 5.105, ...) γ (0.0516, 0.0301–1.057)	5.18E+06
Pu-240	^{239}Pu neutron capture or α decay of ^{244}Cm	6560 yr	α (5.1683, 5.1237, ...) γ (0.0452, 0.1042, ...)	8.72E+06
Pu-241	^{240}Pu neutron capture	14.4 yr	β (0.0208), α (4.897, 4.853, ...) γ (0.1486, 0.1037, ...)	2.12E+09
Pu-242	Electron capture by ^{242}Am or ^{241}Pu neutron capture	3.75×10^5 yr	α (4.901, 4.856, ...) γ (0.0449, . . .)	3.07E+04
Am-241	β^- decay of ^{241}Pu	432.7 yr	α (5.4857, 5.4430, ...) γ (0.0595, 0.0263−955)	1.86E+06
Am-243	β^- decay of ^{243}Pu or ^{242m}Am neutron capture	7370 yr	α (5.276, 5.234, ...) γ (0.0747, 0.0311–0.6622)	2.82E+05
Cm-242	β^- decay of ^{242}Am	162.8 days	α (6.1127, 6.0694, ...) γ (0.0441, . . .)	6.45E+08
Cm-244	β^- decay of ^{244}Am or ^{243}Cm neutron capture	18.1 yr	α (5.8048, 5.7627, ...) γ (0.0428, . . .)	2.62E+07

Source of the activity data: Alexander et al. (1977)

[a] The fuel is from a PWR with fuel burnup of 33,000 MWD/MTU. The fuel assembly has 461.4 kg of U as specified in Table 7.1

As to the presence of fission products in spent fuel, they take up approximately 5% of the mass inventory. Nevertheless, they are the dominant source of radioactivity in spent fuel. Also an interesting feature of fission products is that their half-lives vary widely. While about one hundred of them are non-radioactive, most of fission products have very short half-lives: More than 430 nuclides have half-life less than 24 hours. Iodine-131, one of the most important nuclides in terms of the potential public radiation dose in severe nuclear reactor accident, has the half-life of 8.04 days. Xenon-133, one of the most abundant nuclides among fission products, has a half-life of 5.25 days. Forty two of the fission products have half-life of longer than 1 day but shorter than a year. The total number of fission products with longer than 1 year half-life is 16. Only 13 of them have half-life longer than 10 years. Among them, strontium-90 ($t_{1/2} = 29.1$ years) and cesium-137 ($t_{1/2} = 30$ years) are important contributors to the heat and radiation dose from spent fuel, each with about 30 years of half-life. There are also six fission products with half-life longer than 100,000 years. Such long lived fission products include technetium-99 ($t_{1/2} = 2.1 \times 10^5$ years) and iodine-129 ($t_{1/2} = 15.7$ million years) along with ^{126}Sn, ^{79}Se, ^{93}Zr, ^{135}Cs, and ^{107}Pd as shown in Table 7.5. The variations of half-life among the fission products are summarized in Table 7.5.

Production of various isotopes of uranium, plutonium, and minor actinides in spent fuel comes from various nuclear reactions in the fuel. This is captured in Fig. 7.2. For example, when uranium-235 absorbs one neutron without being fissioned, it becomes ^{236}U. If ^{236}U subsequently absorbs another neutron, it produces ^{237}U ($t_{1/2} = 6.75$ days). Uranium-237 then decays to neptunium-237 through a beta minus decay ($t_{1/2} = 2.14 \times 10^6$ years). Production of ^{237}U and ^{237}Np dominates the activity buildup of actinides, immediately after discharge of spent fuel from the reactor.

Table 7.5 Examples of fission products at different half-life groups

Half-lives	Number	Examples
<24 h	438+	^{102}Mo (11.6 m), ^{139}Ba (83.2 m)
1 day to 1 yr	42	^{131}I (8.04 d), ^{133}Xe (5.25 d), ^{95}Nb (3.5 d), ^{95}Zr (64 d), ^{144}Ce (284.4 d)
>1 yr. to 10 yrs	4	^{106}Ru (368 d), ^{155}Eu (4.76 y)
>10 yrs	13	^{85}Kr (10.76y), ^{113m}Cd (14.1 y), ^{90}Sr (29.1 y), ^{137}Cs (30.2 y), ^{121m}Sn (43.9 y), ^{151}Sm (96.6 y), ^{99}Tc (2.12×10^5 y), ^{126}Sn (2.3×10^5 y), ^{79}Se (3.27×10^5 y), ^{93}Zr (1.53×10^6 y), ^{135}Cs (2.3×10^6), ^{107}Pd (6.5×10^6), ^{129}I (1.57×10^7 y)
Stable	101	^{72}Ge, ^{73}Ge, ^{74}Ge, ^{75}As, ^{77}Se, ^{78}Se, ^{80}Se, ^{82}Se, ^{83}Kr, ^{84}Kr, ^{86}Kr, ^{85}Rb, ^{88}Sr, ^{91}Zr, ^{92}Zr, ^{94}Zr, ^{95}Mo, ^{97}Mo, ^{98}Mo, ^{101}Ru, ^{102}Ru, ^{104}Ru, ^{105}Pd, ^{106}Pd, ^{108}Pd, ^{110}Pd, ^{112}Cd, ^{114}Cd, ^{118}Sn, ^{120}Sn, ^{122}Sn, ^{124}Sn, ^{123}Sb, ^{126}Te, ^{127}I, ^{132}Xe, ^{134}Xe, ^{133}Cs, ^{138}Ba, ^{139}La, ^{140}Ce, ^{142}Ce, ^{143}Nd, ^{145}Nd, ^{146}Nd, ^{148}Nd, ^{149}Sm, ^{152}Sm, ^{154}Sm, ^{153}Eu, ^{155}Gd, ^{156}Gd, ^{157}Gd, ^{158}Gd, ^{160}Gd, ^{159}Tb, ^{161}Dy
Total	615	

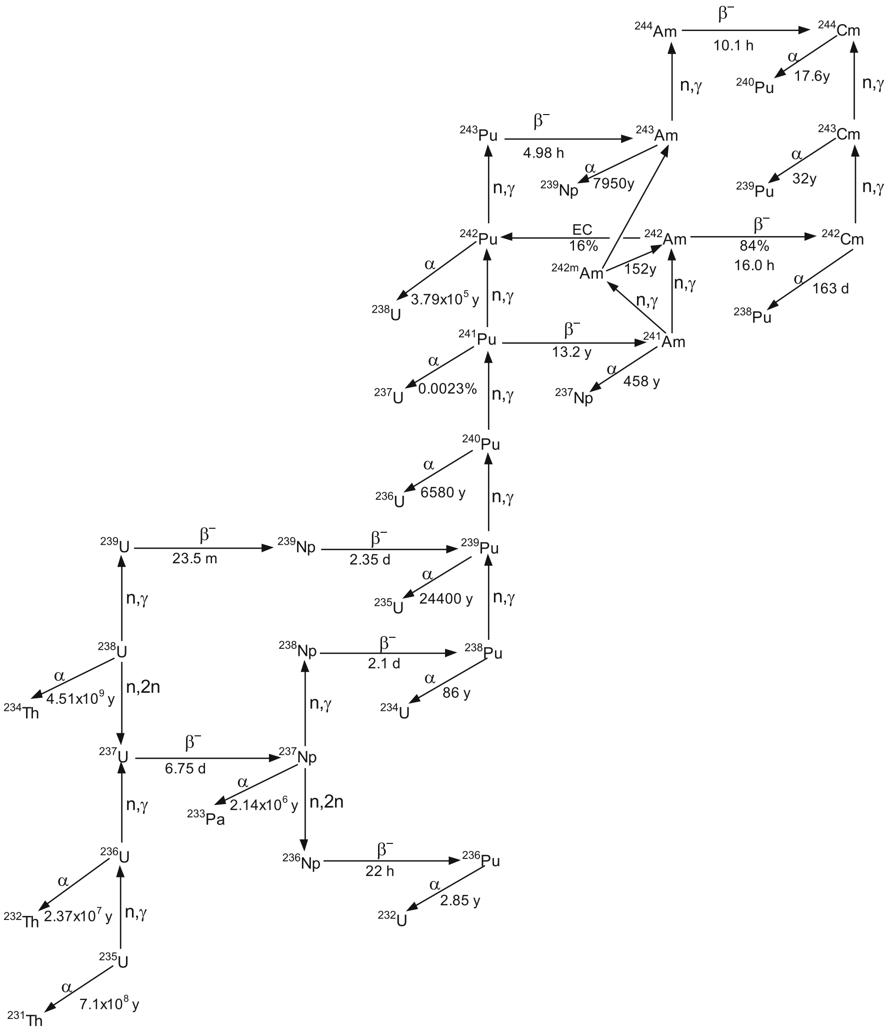


Fig. 7.2 Nuclide reactions and decay chains producing plutonium, neptunium, americium and curium in nuclear fuel. (Reproduced from Benedict et al. 1981)

Uranium-238, which makes up bulk of nuclear fuel mass, becomes ^{239}U after capturing one neutron. U-239 then undergoes beta minus decay with a half-life of 24 min producing ^{239}Np. Np-239 (beta minus) decays with a half-life of 2.3 days into ^{239}Pu. Then, by successive neutron absorptions, ^{239}Pu can become ^{240}Pu, ^{241}Pu, ^{242}Pu, and also ^{243}Pu ($t_{1/2} = 4.96$ h). These isotopes of plutonium can also produce americium and curium. For example, beta minus decay of ^{241}Pu and ^{243}Pu forms ^{241}Am ($t_{1/2} = 432$ years) and ^{243}Am ($t_{1/2} = 7380$ years), respectively. Neutron capture by ^{241}Am and the subsequent beta minus decay result in ^{242}Cm ($t_{1/2} = 163$ days). Successive neutron captures lead to ^{243}Cm ($t_{1/2} = 28.5$ years), ^{244}Cm ($t_{1/}$

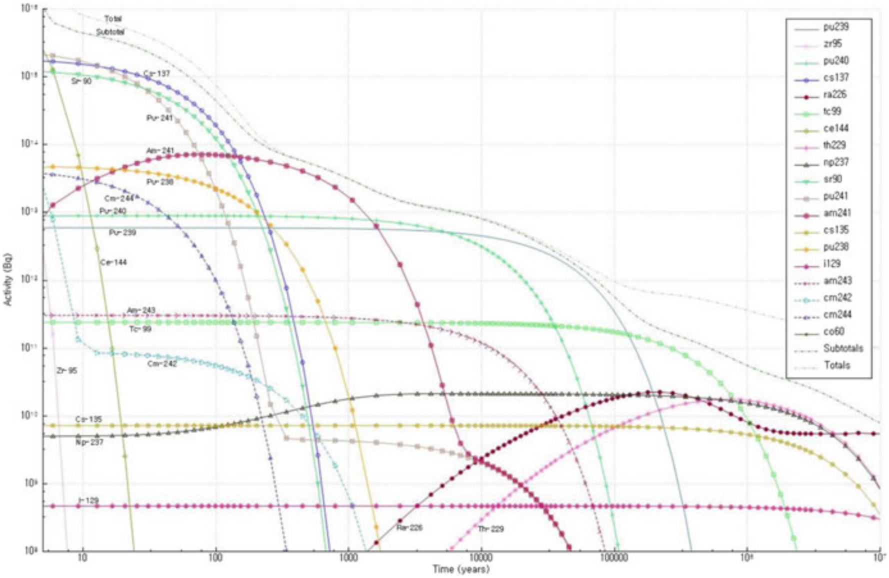


Fig. 7.3 The changes of activity of spent fuel in a LWR per single assembly (on average) as a function of time since discharge (33,000 MWD/MTU, 3.2% initial enrichment)

$_2$ = 18.11 years), and ^{245}Cm ($t_{1/2} = 8.5 \times 10^3$ years). Through alpha decays, ^{242}Cm produces ^{238}Pu, ^{243}Cm produces ^{239}Pu, ^{244}Cm produces ^{240}Pu, and ^{245}Cm produces ^{241}Pu. ^{243}Am also decays to ^{239}Np after emitting α particle which quickly beta decays to ^{239}Pu.

The actinides and their decay daughters are the principal source of long-term radioactivity in spent fuel.

Figure 7.3 gives an example of the time-dependent changes of activity of various radionuclides in LWR spent fuel. It can be seen that total activity in spent fuel is dominated by fission products up to several hundred years after reactor discharge. For example, at the time of discharge, the activity is largely contributed by short-lived fission products such as ^{95}Nb, ^{95}Zr, ^{103m}Rh, and ^{103}Ru. After 10 years, the important activity contributors are ^{137}Cs, ^{137m}Ba, ^{90}Sr, and ^{90}Y. These same radionuclides remain important as key activity contributors after 100 years, After about 1000 years, the activity becomes dominated by the α-emitting actinides such as ^{241}Am, ^{239}Pu and ^{240}Pu. At 10,000 years, ^{239}Pu remains the important activity contributor followed by ^{240}Pu. Even at 100,000 years, ^{239}Pu remains the top activity contributor followed by ^{99}Tc.

Somewhat more details of the changes in the activity of key radionuclides over time in spent fuel (per assembly) are also shown in Table 7.6 (covering 10,000 years of cooling time). The spent fuel, in this example, is a PWR spent fuel with initial enrichment of 3.2% and the discharge burnup of 33 GWD/MT. Again, during the first several hundred years of cooling, the dominant contributors to the total activity are ^{90}Sr and ^{137}Cs and their respective decay products, ^{90}Y and ^{137m}Ba. At later time

Table 7.6 Changes in the activity of selected radionuclides in spent fuels at different times since discharge (for PWR, at 45 GWD/MTHM, 4.5% ^{235}U enrichment, 32% of thermal efficiency)

	Nuclide	Half-life	Activity (Ci/GWyear)[a]				
			0 y (MCi/GWyr)	10y (MCi/GWyr)	100y (kCi/GWyr)	1000y (kCi/GWyr)	10,000y (Ci/GWyr)
Fission products	^{85}Kr	10.8 y	0.108	0.057	0.168	0.000	0.000
	^{90}Sr	29.1 y	0.844	0.660	71.92	0.000	0.000
	^{99}Tc	211 ky	0.000	0.000	0.147	0.146	142.1
	^{129}I	15.7My	0.000	0.000	0.000	0.000	0.292
	^{137}Cs	30.1y	1.169	0.928	115.9	0.000	0.000
Actinides	^{230}Th	75.4ky	0.000	0.000	0.000	0.000	2.166
	^{233}U	159ky	0.000	0.000	0.000	0.000	0.546
	^{237}Np	2.14My	0.000	0.000	0.005	0.011	13.40
	^{238}Pu	87.7y	0.037	0.037	18.13	0.016	0.000
	^{239}Pu	24.1ky	0.003	0.003	3.122	3.049	2393
	^{240}Pu	6.56ky	0.005	0.005	4.646	4.227	1634
	^{241}Pu	14.3y	1.365	0.842	10.89	0.003	4.003
	^{241}Am	432y	0.002	0.019	40.90	9.760	1.684
	^{244}Cm	18.1y	0.039	0.027	0.851	0.000	0.000
Sum[b]			3.571	2.576	266.7	17.21	4191
Total[b]			359.2	0.933	79.39	17.67	4362

Activities are based on ORIGEN calculation (45 GWd/t, 4.5 wt.% enrichment, 32% thermal efficiency)

The symbols k and M denote 10^3 and 10^6, respectively

[a] To convert to activity per MTHM, use the conversion factor: 28.5 MTHM/GWyr

[b] The “sum” is the sum for radionuclides listed in the column (before rounding off); the: “total” is the sum for all radionuclides, including those not listed in this table

periods, ^{241}Am (a decay product of ^{241}Pu), ^{240}Pu, ^{239}Pu, ^{99}Tc, and ^{237}Np are important contributors. The aged spent fuel will allow the buildup of ^{241}Am from ^{241}Pu decay and the buildup of ^{239}Pu from the decay of ^{239}Np. The activity of long-lived fission products, such as ^{99}Tc and ^{129}I, although at very low level, persists through very long time periods.

A different display of the changes in total activity inventory is also shown in Fig. 7.4 where the activity of spent fuel at different burnups with different cooling

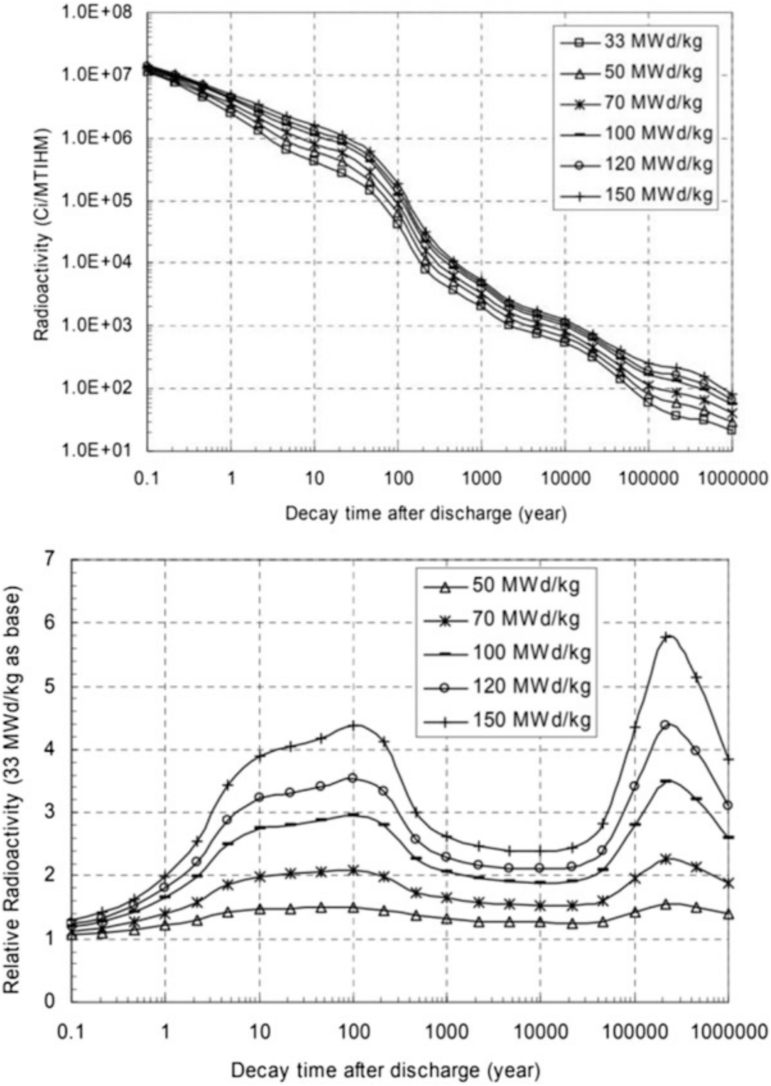


Fig. 7.4 Comparison of spent fuel radioactivity per MTIHM with different burnups as a function of cooling periods (i.e., time after discharge). (Source: Xu 2003)

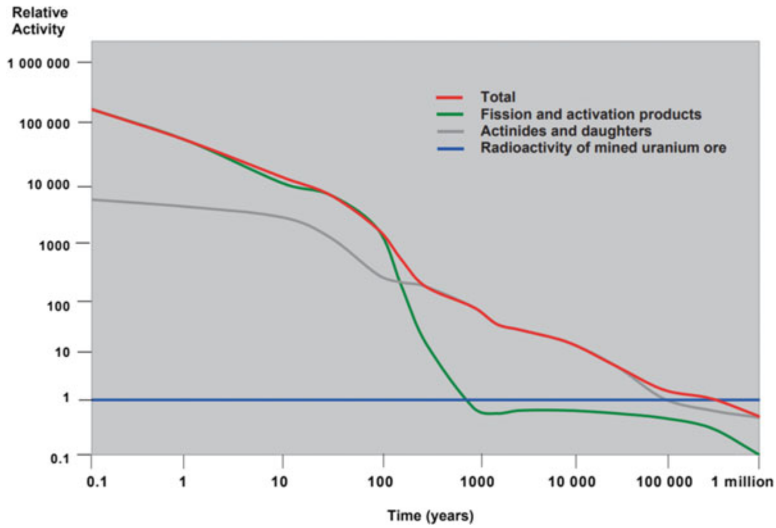


Fig. 7.5 Time dependent changes of activity of spent fuel relative to the activity of the uranium ore (IAEA 2013)

periods is given (the results are in activity per MTIHM). Figure 7.4a shows that the overall activity decreases with radioactive decay and that the activity increases with burnup. Figure 7.4b also shows that the relative degree of increase with burnup becomes less at beyond 100 years of cooling due to the buildup of TRUs.

Figure 7.5 shows the comparison of spent fuel activity (for the same mass) in relation to that of natural uranium. It can be noted that the activity of spent fuel is more than five orders of magnitude higher than that of natural uranium at the time of discharge. The majority of the activity is from fission products. Then the activity falls rather quickly after 100 years. The activity of actinides and their daughters is much less at the beginning but becomes higher than that of fission products after a few hundred years. After this point, the activity of spent fuel is maintained by actinides and their daughter products as shown in the figure. At about 100,000 years after discharge, the total activity of spent fuel becomes close to the level of natural uranium.

While the detailed composition of spent fuel varies depending on how long the fuel is irradiated in the reactor (i.e., as a function of burnup) or how long it has been cooled since discharged from the reactor, presence of plutonium in spent fuel is an important feature drawing special attention. Plutonium can be used for nuclear fuel as well as a material for nuclear explosives. One PWR spent fuel assembly contains about 4.2 kg of plutonium which is about the half of so-called “significant quantity” of plutonium (i.e., 8 kg, the amount needed to make one nuclear explosive device). Theoretically speaking, one nuclear weapon can be made with the plutonium available in two of the PWR spent fuel assemblies.

Along with plutonium, presence of minor actinides in spent fuel must be noted. Basically, all of the minor actinides are fissionable and some are fissile (see the

Table 7.7 Nuclear properties of various fissile (fissile in the table is marked with *) and fertile nuclides

Isotope	Half-life (y)	Neutrons/sec-kg	Watts/kg	Critical mass (kg) (bare sphere)	Comment
Pa-231	32.8×10^3	Nil	1.3	162	
Th-232	14.1×10^9	Nil	Nil	Infinite	
U-233*	159×10^3	1.23	0.281	16.4	
U-235*	700×10^6	0.364	6×10^{-3}	47.9	
U-238	4.5×10^9	0.11	8×10^{-6}	Infinite	
Np-237*	2.1×10^6	0.139	0.021	59	
Pu-238	88	2.67×10^6	560	10	Heat and neutrons
Pu-239*	24×10^3	21.8	2.0	10.2	
Pu-240	6.54×10^3	1.03×10^6	7.0	36.8	Neutrons
Pu-241*	14.7	49.3	6.4	12.9	
Pu-242	376×10^3	1.73×10^6	0.12	89	Neutrons
Am-241*	433	1540	115	57	Heat and neutrons
Am-243	7.38×10^3	900	6.4	155	
Cm-244	18.1	11×10^9	2.8×10^3	28	Heat and neutrons
Cm-245*	8.5×10^3	147×10^3	5.7	13	
Bk-247*	1.4×10^3	Nil	36	10	
Cf-251*	898	Nil	56	9	

definition in Sect. 3.3.2) and can possibly be used for fission energy generation. The nuclear properties of several actinides are listed in Table 7.7. The listed information includes the mass needed to build a nuclear explosive device (assuming bare spherical geometry), the spontaneous neutron generation rate, and the heat generation rate. The fissile such as ^{233}U, ^{235}U, and ^{239}Pu has small critical mass, small neutron generation rate and low level heat. Other fissionable nuclides have a large critical mass, a large spontaneous neutron generation rate, or large heat generation rate. Nevertheless, except for ^{232}Th and ^{238}U, all of the nuclides listed in the table can be potentially used as part of nuclear explosive materials although the design and detonation of nuclear explosives using them will be complicated due to the presence of large number of spontaneous neutrons and heat. Due to this concern, diversion or theft of spent fuel should be carefully guarded to prevent the use for non-civilian purposes.

The related work performed to address this concern belongs to the activities of nuclear safeguards and/or nuclear security. Nuclear safeguards are the measures to verify that nuclear materials are used only for peaceful purposes. Nuclear security refers to activities to protect nuclear materials against terrorism or sabotage. These are discussed further in Sect. 15.4.

Other materials used in nuclear fuel to build fuel rods and assemblies also become the source of radioactivity through neutron activation. These materials include iron, zirconium, and other normally inert materials, including trace impurities such as nitrogen and oxygen. For example, nitrogen exists in the fuel as impurity from fuel manufacturing. Nitrogen produces carbon-14 through neutron capture and release of proton, i.e., $^{14}N(n,p)^{14}C$. Oxygen, present as part of UO_2 also captures neutron and produces carbon-14 through $^{17}O(n,\alpha)^{14}C$ reaction. Other nuclides are formed by neutron activation in the zircaloy cladding, stainless steel end fittings, and Inconel spacers. The big contributors to the activity in these materials include zirconium-95 (^{95}Zr, $t_{1/2} = 65$ days), cobalt-60 (^{60}Co, $t_{1/2} = 5.26$ years), chromium-51 (^{51}Cr, $t_{1/2} = 27.8$ days), iron-55 (^{55}Fe, $t_{1/2} = 2.6$ years), cobalt-58 (^{58}Co, $t_{1/2} = 71.3$ days), nickel-63 (^{63}Ni, $t_{1/2} = 92$ years), niobium-95 (^{95}Nb, $t_{1/2} = 35$ days), tin-119m (^{119m}Sn, $t_{1/2} = 250$ days), and antimony-125 (^{125}Sb, $t_{1/2} = 2.7$ years). Very long term behavior of activation products activity is dominated by ^{59}Ni ($t_{1/2} = 8 \times 10^4$ years).

7.1.3 Determining Nuclide Concentrations in Spent Fuel

Understanding the constituents of spent fuel including their isotopic distributions helps to plan activities in spent fuel management. The isotopic distributions of radionuclides in spent fuel is the end result of the competing actions between the production mechanisms and the loss mechanisms and also depend on the irradiation history during the reactor operation and the length of cooling periods after discharge. The production mechanisms in nuclear fuel include fission, neutron capture, and the (alpha or beta) decay of precursors. The loss mechanisms include radioactive decay of the produced nuclide, neutron capture by the produced nuclide, and removal from the system due to release or migration.

The concentrations of radionuclides present in spent fuel can then be calculated by considering time-dependent changes in the production and loss of each and every nuclide present as represented by the following equation:

$$\begin{aligned}\frac{dN_i}{dt} &= [\text{rate of production of radionuclide } i] - [\text{rate of loss of radionuclide } i] \\ &= [\text{rate of production by fission}] + [\text{rate of production by neutron capture}] \\ &\quad +[\text{rate of production by precursor decay}] - [\text{rate of loss by decay}] \\ &\quad -[\text{rate of loss by neutron capture}] - [\text{rate of loss by removal}]\end{aligned} \tag{7.1}$$

where, N_i is atomic density of nuclide i.

Each of the production and loss term is explained below.

(a) [Rate of production by fission] = *fission rate* × *fission yield*

$$
\begin{aligned}
&= N_{235}\sigma_f^{235}\phi \cdot \gamma^{235} + N_{239}\sigma_f^{239}\phi \cdot \gamma^{239} \\
&= \Sigma_f^{235}\phi \cdot \gamma^{235} + \Sigma_f^{239}\phi \cdot \gamma^{239}
\end{aligned}
\tag{7.2}
$$

where N_{235} *and* N_{239} is the atomic number density of ^{235}U and ^{239}Pu, σ_f^{235} and σ_f^{235} are the microscopic cross section for fission from the fissile, ^{235}U and ^{239}Pu, and γ^{235} and γ^{239} are the fission yield of ^{235}U and ^{239}Pu, respectively. Neutron flux, ϕ, is the number of neutrons in the system per unit area per second. The product of N_F and σ_f^F is called macroscopic cross section of fissile F for fission and is denoted as Σ_f^F (see Sect. 3.2.3). Product of Σ_f^F and ϕ gives fission rate density from the fissile F.

(b) [Rate of production by neutron capture] = rate of all neutron capture reactions leading to the production of nuclide i
$= N_1\sigma_1^c\phi \cdot f_{i1} + N_2\sigma_2^c\phi \cdot f_{i2} + \ldots + N_L\sigma_L^c\phi \cdot f_{iL}$

$$
= \phi \sum_{k=1}^{L} f_{ik} \cdot \sigma_k^c N_k \tag{7.3}
$$

where, f_{ik} is fraction of neutron capture reaction by nuclide k leading to formation of nuclide i, σ_k^c is average neutron capture cross section of nuclide k, and L is the total number of such nuclides leading to the production of nuclide i.

(c) [Rate of production by precursor decay] = Rate of precursor decay × yield

$$
\begin{aligned}
&= \lambda_1 N_1 d_{i1} + \lambda_2 N_2 d_{i2} + \ldots + \lambda_M N_M d_{iM} \\
&= \sum_{j=1}^{M} d_{ij} \cdot \lambda_j N_j
\end{aligned}
\tag{7.4}
$$

where d_{ij} is the fraction of decays of precursor nuclide j ($j = 1,\ldots, M$) leading to the formation of nuclide i and M is the total number of precursor nuclides leading to the production of i.

(d)

$$
[\text{Rate of loss by decay}] = -\lambda_i N_i \tag{7.5}
$$

where, λ_i is decay constant of nuclide i.

(e)

$$[\text{Rate of loss by neutron capture}] = N_i \sigma_i \phi \tag{7.6}$$

where, σ_i is average neutron capture cross section of nuclide *i*.

(f)

$$[\text{Rate of loss by removal}] = -r_i N_i \tag{7.7}$$

where r_i is continuous removal rate of nuclide *i* from the system.

Combining all of the terms gives the concentration of nuclide *i* as time-dependent quantity as,

$$\frac{dN_i}{dt} = N_{235}\sigma_f^{235}\phi \cdot \gamma_i^{235} + N_{239}\sigma_f^{239}\phi \cdot \gamma_i^{239} + \phi\sum_{k=1}^{L} f_{ik} \cdot \sigma_k^c N_k + \sum_{j=1}^{M} d_{ij} \cdot \lambda_j N_j - (\lambda_i + \sigma_i\phi + r_i)N_i \tag{7.8}$$

As the equation is an ordinary differential equation, an analytical solution may be obtained. However, obtaining the solution is not straightforward as many nuclides are involved in a time-dependent fashion in the decay chain. If *N* is the total number of nuclides to be analyzed, there are *N* equations to be solved. Solving these equations requires time-dependent values of the parameters, including the values of neutron flux. This complicates the process of solving the equations.

Solving these equations (as a set of simultaneous differential equations) is usually done through computer code-based fuel depletion analysis. Typically, the calculation in fuel depletion analysis assumes that the neutron flux remains constant during a small but finite time increment allowing the calculation of the amount of nuclide given in Eq. 7.8. The results of nuclide concentrations are then used to recalculate the neutron flux. This allows to calculate the amount of nuclides in the next time step. By repeating these steps through subsequent time steps, a set of simultaneous differential equations can be solved.

Equation 7.8 can also be solved by using the analytical solutions approach with simplifying assumptions. Assume that the production of a fission product is only from ^{235}U fission and that the loss is only from the decay of the fission products. Then the equation becomes,

$$\frac{dN_i}{dt} = -\lambda_i N_i + F \tag{7.9}$$

where

$$F = \Sigma_f^{235}\phi \cdot \gamma_i^{235} \equiv \gamma_i \Sigma_f \phi \text{ (i.e.the rate of production by fission)} \tag{7.10}$$

This equation implies that the fission occurs at a constant rate and any neutron-absorption related production or loss of the fission products is ignored.

Then the equation can be solved as,

$$N_i = \frac{F}{\lambda_i}\left(1 - e^{-\lambda_i T_R}\right)e^{-\lambda_i T_C} \tag{7.11}$$

where, T_R is the irradiation time and T_C is the cooling time.

If the half-life of the fission product is short compared to the time of reactor irradiation ($T_{1/2} \ll T_R$), it can be further assumed that the fission product reaches saturation prior to the end of the irradiation. In this case, the effect of decay during the irradiation period is negligible, i.e. $\left(1 - e^{-\lambda_i T_R}\right) \approx 1$. Then the activity of the fission product becomes,

$$\lambda_i N_i = F e^{-\lambda_i T_C}\left(\text{when } T_{1/2} \ll TR\right) \tag{7.12}$$

Then, the saturation activity per unit reactor power can be expressed as (using the unit of curies per watt of thermal power),

$$\frac{Activity\,(Curies)}{Thermal\,power\,(Watt)} = \gamma_i e^{-\lambda_i T_c} \cdot \left(\frac{\text{disintegration/s}}{\text{fission/s}}\right)\left(\frac{\text{fission}}{200\,\text{MeV}}\right)\left(\frac{1\,\text{MeV}}{1.6 \times 10^{-13}\text{W}\cdot\text{s}}\right)$$
$$\left(\frac{1\,\text{Ci}}{3.7 \times 10^{10}\,\text{disintegrations/s}}\right)\frac{Activity\,(Curies)}{Thermal\,power\,(Watt)} = 0.845\,\gamma_i e^{-\lambda T_c} \quad \left(\text{when } T_{1/2} \ll T_R\right) \tag{7.13}$$

On the other hand, in the case of long-lived fission products, their half-lives are much longer than the irradiation time (i.e., $T_{1/2} \gg T_R$). In this case, $\left(1 - e^{-\lambda_i T_R}\right) \approx \lambda_i T_R$ and Eq.7.11 is simplified as

$$\lambda_i N_i = F \lambda_i T_R e^{-\lambda_i T_C} = F\frac{0.693}{T_{1/2}} T_R e^{-\lambda_i T_C} \tag{7.14}$$

Again, similar to Eq. 7.13, the activity of fission products per unit reactor power can be expressed as (using the unit of curies per watt of thermal power),

$$\frac{Activity\ (Curies)}{Thermal\ power\ (Watt)} = 0.586\frac{T_R}{T_{1/2}}\gamma_i e^{-\lambda T_c}\left(\text{when } T_{1/2} \gg TR\right) \tag{7.15}$$

Example 7.1: Activity Calculation of Radionuclides in the Spent Fuel
A nuclear reactor operated for 2 years at a steady power level of 1000 MWe after entering the service and was shutdown. Assume that all of the spent fuels were removed and kept in storage for 30 years. What would be the activity of ^{137}Cs and ^{90}Sr in the spent fuel?

Solutions:

The half-lives of ^{137}Cs and ^{90}Sr are both about 30 years which is much longer than the irradiation period. In this case, Eq. 7.15 can be used.

Given: $T_R = 2$ years, $T_C = 30$ years, $P = 1000$ (MWe) ≈ 3300 (MWt) assuming 33% thermal efficiency.

For ^{137}Cs

$T_{1/2} = 30.2$ years, $\gamma_i = 6.22\% = 0.0622$

$$\frac{\text{Activity (Curies)}}{\text{Thermal power (Watt)}} = 0.586\frac{T_R}{T_{1/2}}\gamma_i e^{-\lambda T_c}$$

$\rightarrow$ Activity (Curies) = Thermal power (Watt) $\cdot 0.586\frac{T_R}{T_{1/2}}\gamma_i e^{-\lambda T_c}$

$\rightarrow$ Activity (Curies) $= 3300 \times 10^6(\text{W}) \times 0.586 \times \frac{2}{30.2} \times 0.0622 \times e^{-\frac{0.693}{30.2}\times 30} = 4.0 \times 10^6$(Curies)

For ^{90}Sr, similarly as above

$T_{1/2} = 29.1$ years, $\gamma_i = 5.73\% = 0.0573$

$\rightarrow$ Activity (Curies) $= 3300 \times 10^6(\text{W}) \times 0.586 \times \frac{2}{29.1} \times 0.0573 \times e^{-\frac{0.693}{29.1}\times 30} = 3.78 \times 10^6$(Curies)

7.1.4 Decay Heat Production in Spent Fuel

The energy produced from fission in nuclear reactors is thermal energy. This thermal energy is released as the kinetic energy of the radioactive particles in two steps, i.e., the initial release and the delayed release. The initial release is carried by the particles directly released from the process of fission. These particles include fission products (carrying about 168.2 MeV), neutrons promptly produced at the time of fission (carrying about 4.8 MeV), and gamma rays produced at the time of fission (carrying about 7.5 MeV). Together, this initially released energy is about 180.5 MeV per fission.

The second stage of energy release (delayed energy release) is through the decay of fission products emitting gamma rays, beta particles, and neutrinos. The total of this delayed energy is about 26.6 MeV (~6.8 MeV from gamma rays, ~7.8 MeV from beta rays, and ~ 12 MeV from the neutrinos, emitted as part of beta decays). As neutrinos very rarely interact with matter, the energy of neutrinos is not recoverable.

Table 7.8 Energy release from ^{235}U fission and the resulting heat generation

Energy source	Emitted energy	Recoverable energy (heat)
Fission fragments	168.2	168.2
Fission neutrons	4.8	4.8
Prompt γ-rays	7.5	7.5
β-rays from fission product decays (delayed)	7.8	7.8
Neutrinos from fission product decays (delayed)	12	
γ-rays from fission product decays (delayed)	6.8	6.8
Capture γ-rays		3–12
Total	207.1	198.1–207.1

Therefore, the recoverable energy from delayed energy release is about 14.6 MeV (6.8 + 7.8 MeV) per fission. There is also the energy associated with the release of delayed neutrons from fission but this delayed neutron energy is negligible as the fraction of delayed neutrons is less than 1% of all fission induced neutrons. Therefore, it can be said that total recoverable amount of energy release from fission is about 195 MeV (180.5 + 14.6 MeV) per fission event for ^{235}U. In the case of fission with ^{239}Pu, this value is about 202 MeV.

There are also neutrons in the reactor that are not causing fission but captured by the uranium atom (i.e., ^{235}U and ^{238}U) as well as in the structural components of nuclear fuel. Capture of neutrons in these materials leads to gamma ray emissions (through (n,γ) reactions). The energy carried by such capture-induced gamma rays is about 1.4–5.8% (3–12 MeV) of the total energy release from the fission itself, depending upon the materials used in the reactor. Adding this, the total average energy release from a ^{235}U fission event is about 200 MeV. This ~200 MeV is deposited in the surrounding materials in the form of heat. Table 7.8 shows the summary of energy release from ^{235}U fission.

Note that the delayed energy release from fission becomes the source of heat in nuclear reactor after reactor shutdown (initially 14.6 MeV which is about 7% of total fission energy). This delayed energy release needs to be appropriately dissipated to prevent over-heating of nuclear fuel after reactor shutdown. This is one of the key features of the safety system designs in a nuclear reactor. Without proper cooling, this delayed energy release can cause excessive thermal expansion and eventual failure of nuclear fuel leading to the release of radioactivity (note: the melting temperature of UO_2 fuel is around 2800 °C). The decay heat in spent fuel has to be also managed after the discharge from nuclear reactor and during various stages of spent fuel management.

To minimize the concern of failure of spent fuel, the cladding surface temperature must be maintained to be lower than 340 ~ 420 °C (depending on the type of fuel) by cooling during long-term storage. For a short term situation, up to 570 °C is allowed. Typically cooling of spent fuel is provided through convection by air or water.

The rate of heat release from the fuel depends on the rate of fission in the fuel during reactor irradiation, the length of irradiation time, and the length of time the fuel was allowed to “cool” before shipping or processing.

A simple statistical correlation by Way and Wigner (1948) allows approximate calculations of the decay heat and fission product activity. At a time t (in days) after fission, the rate of beta decay from a single fission product is given by the following equation (the equation is valid when $t > 1$ day),

$$\text{Beta decay rate} = \beta(t) = 5.2 \times 10^{-6} t^{-1.2} \ \text{particles}/\sec/\text{fission} \tag{7.16}$$

Then by considering all of the fission events happening during reactor irradiation, the total amount of beta decay activity in spent fuel can be calculated. The rate of disintegration of fission products in the spent fuel (in Ci/Watts of power) when the fuel went through fissions for T days and then was cooled for t days (thus, time since fission is t for minimum and $T + t$ for maximum) is given as,

$$= \int_{t}^{T+t} \left[\beta(t)\left(\frac{dis}{s - fission}\right)\left(\frac{86400s}{d}\right)\right] dt \cdot \left(\frac{\text{fission}}{200\ \text{MeV}}\right) \times \left(\frac{1\ \text{MeV}}{1.6 \times 10^{-13}\text{W} \cdot \text{s}}\right)\left(\frac{1\ \text{Ci}}{3.7 \times 10^{10}\ \text{disintegrations/s}}\right) \tag{7.17}$$

Result of this integration gives the rate of disintegration of fission products in the fuel (in curies per watt of reactor power),

$$\frac{Ci}{W} = 1.9\left[t^{-0.2} - (T + t)^{-0.2}\right] \tag{7.18}$$

A similar relationship can also be used to represent gamma decays from a single fission product as follows.

$$\text{Gamma decay rate} = \gamma(t) = 1.9 \times 10^{-6} t^{-1.2} \text{photons}/\sec/\text{fission} \tag{7.19}$$

Example 7.2: Total Radioactivity in the Spent Fuel

A nuclear reactor operating at a steady power level of 1000 MWe was shutdown 2 years after entering service. Calculate the total fission product activity in the spent fuels in the reactor (a) 1 day and (2) 30 years after the shutdown.

Solution:

From the given condition,

T (the operating period) = 730 days, t (the cooling period) = 1 day or 30 years (10,950 days)

(continued)

Example 7.2 (continued)

P (power level) = 1000 MWe = 3300 MWt (assuming 33% thermal efficiency)

Activity t days after shutdown (in Ci) = $1.9 \times 3300 \times 10^6 \times [t^{-0.2} - (t + T)^{-0.2}]$

(a) Activity 1 day after shutdown = $1.9 \times 3300 \times 10^6 \times [(1)^{-0.2} - (731)^{-0.2}]$ (in Ci)

$$= 1.9 \times 3300 \times 10^6 \times 0.732$$
$$= 4.56 \times 10^9 \text{ (Ci)}$$
$$= 1.69 \times 10^{20} \text{ (Bq)}$$

(b) Activity 30 years after shutdown (in Ci) = $1.9 \times 3300 \times 10^6 \times [(10{,}950)^{-0.2} - (11{,}680)^{-0.2}]$

$$= 1.9 \times 3300 \times 10^6 \times 0.001996$$
$$= 1.25 \times 10^7 \text{ (Ci)}$$
$$= 4.83 \times 10^{17} \text{ (Bq)}$$

Based on results of Examples 7.1 and 7.2, the total activity of ^{137}Cs (4.0×10^6 Ci) and ^{90}Sr (3.78×10^6 Ci) after 30 years of cooling is about 7.8×10^6 Ci. This corresponds to about 62% of the total fission product activity in the spent fuel at that time.

7.1.4.1 Decay Heat Calculation

In association with Eq. 7.16, a similar statistical approach is also available to represent the rate of energy release in the form of beta particles, gamma rays, and neutrinos as shown below (Way and Wigner 1948).

$$\begin{aligned} &\text{Energy release rate at time } t(\text{days}, > 1 \text{ min}) = E(t) \\ &= 3.9\, t^{-1.2} + 11.7\, t^{-1.4} (\text{eV/s/fission}) \end{aligned} \tag{7.20}$$

(this equation is valid when $t > 1$ day).

Assuming that about half of the energy represented by the equation is recoverable (equally divided by gamma and beta rays), an approximate calculation of decay heat can be made as a function of the reactor irradiation period and the cooling time. Consider a nuclear reactor that has been operating at some constant power level, P, for T days (as depicted in Fig. 7.6). The decay heat rate at $T_R + T_c$ days (T_c days after shutdown) from power generation at P at specific time t' is determined as follows.

Consider a fission event that occurred at time t'. By using Eq. 7.20, the subsequent release of energy from this single fission event as decay heat at T_c days after reactor shutdown is,

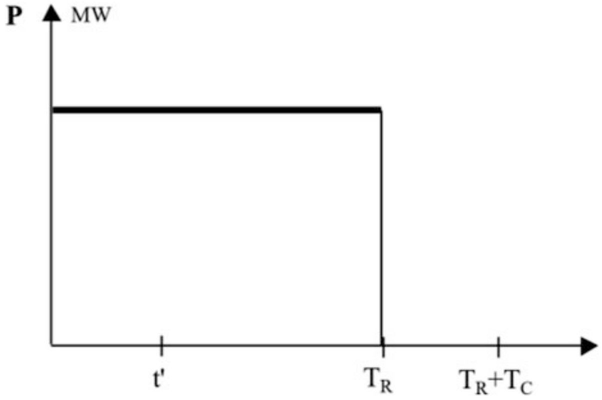


Fig. 7.6 Assumed power generation history for decay heat calculation

$$\begin{aligned}&[\text{Energy release rate at } T_R + T_c \text{ from a fission at } t'] \\ &\quad = E\,(T_R + T_c - t')\ (eV/s)\end{aligned} \tag{7.21}$$

The decay heat rate at T_R + T_c from power generation level of $P(MW)$ at time t' can be obtained by considering the total number of fission events at time t' in the given power generation level as,

$$\begin{aligned}&= Pdt'(MWD) \times F(Fissions/MWD) \times Recoverable\ energy\ release\ (eV/s) \\ &= Pdt' \cdot F \cdot \left(\frac{1}{2}\right) \cdot \left[3.9(T_R + T_c - t')^{-1.2} + 11.7(T_R + T_c - t')^{-1.4}\right]\end{aligned} \tag{7.22}$$

where, F is the total number of fissions from power generation at level P(MW) for a day.

By taking integration of the rate over the period of T_R irradiation days, the total decay heat at T_c days after shutdown from the power generation at the power level P for T_R days of operation becomes,

$$P_d(T_R, T_c) = \int_0^{T_R} dt'\ P \cdot F \cdot \left(\frac{1}{2}\right) \cdot \left[3.9(T_R + T_c - t')^{-1.2} + 11.7(T_R + T_c - t')^{-1.4}\right] \tag{7.23}$$

where $P_d(T_R, T_c)$ is decay heat T_C days after shutdown from the power generation for T_R days.

Note the following conversions are used in the calculation:

$$\begin{aligned}1\ \text{Fission} &= 200\ \text{MeV} \\ &= 200\ (\text{MeV}) \times 1.6 \times 10^{-13}\ (\text{J/MeV}) \\ &= 3.2 \times 10^{-11}\ \text{J}\end{aligned}$$

1 J $=1 / (3.2 \times 10^{-11})$ fissions
$\approx 3 \times 10^{10}$ fissions
1MW $=3 \times 10^{16}$ (fissions/s)

Then, the total decay heat becomes,

$$P_d(T,t) = \int_0^{T_R} dt'\, P(MWD) \cdot 3 \times 10^{16} \left(\frac{\frac{fission}{s}}{\mathrm{MW}}\right) \times \left(\frac{86400\,\mathrm{s}}{1\,\mathrm{d}}\right) \cdot \left(\frac{1}{2}\right) \cdot \left[3.9\left(T_R + T_c - t'\right)^{-1.2} + 11.7\left(T_R + T_c - t'\right)^{-1.4}\right] \cdot \left(\frac{eV/s}{fission}\right) \cdot 1.6 \times 10^{-25} \left(\frac{MW}{eV/s}\right) \tag{7.24}$$

This produces the following equation,

$$\frac{P_d(T_R, T_c)}{P} = 0.0042\left[T_C{}^{-0.2} - (T_R + T_c)^{-0.2}\right] + 0.0063\left[T_C{}^{-0.4} - (T_R + T_c)^{-0.4}\right] \tag{7.25}$$

Equation 7.25 can also be written as

$$\frac{P_d(T_R, T_c)}{P} = \frac{P_d(\infty, T_C)}{P} - \frac{P_d(\infty, T_R + T_c)}{P} \tag{7.26}$$

where, the two quantities on the right-hand-side are calculated for infinite irradiation time and for cooling times of T_c and $T_R + T_c$, respectively.

Example 7.3: Decay Heat and Activity Calculation in the Spent Fuel
A nuclear reactor operating at a steady power level of 1000 MWe was shutdown 2 years after entering service. Assume the thermal efficiency of the plant is 33%.

1. Using Eq. 7.24, calculate the total amount of decay heat produced in the reactor (a) 1 day after shutdown and (b) 30 years after the shutdown.
2. Assuming that all of the spent fuels were removed and kept in storage for 30 years, calculate the total amount of decay heat from ^{137}Cs and ^{90}Sr in the spent fuel at that time. Use the activity of ^{137}Cs and ^{90}Sr estimated in Example 7.1. Compare these results from the results from part 1).

(continued)

Example 7.3 (continued)

Solution: (1) with 33% thermal efficiency, the total thermal power output from 1000 MWe is 3300 MWth. From (Eq. 7.25)

$P_d(730d, t) = P \times 0.0042[t^{-0.2} - (730 + t)^{-0.2}] + 0.0063[t^{-0.4} - (730 + t)^{-0.4}]$

At t = 1 day

$P_d(730d, 1) = 3300 \times (0.0042[1^{-0.2} - (730 + 1)^{-0.2}] + 0.0063[t^{-0.4} - (730 + 1)^{-0.4}])$

$P_d(730d, 1) = 3300 \times 0.00893 = 29.5\ MW$ (this is about 0.9% of the reactor full power)

At t = 30 years = 10,950 days

$P_d(730d, 10950) = 3300 \times (0.0042[10950^{-0.2} - (730 + 10950)^{-0.2}] + 0.0063[t^{-0.4} - (730 + 10950)^{-0.4}])$

$P_d(730d, 10950) = 3300 \times 1.227 \times 10^{-5} = 40.5\ kW$ (this is about 0.012% of the reactor full power)

We can see that the decay heat decreases significantly after 30 years of cooling (from 29.5 MW after 1 day after shutdown to 40.5 kW after 30 years of cooling).

(2) From Example 7.1, total activity of ^{137}Cs and ^{90}Sr in the spent fuel after 30 years of cooling was estimated at 4.0×10^6 Ci and 3.78×10^6 Ci, respectively.

Disintegration of ^{137}Cs and ^{90}Sr produces the following particles.

^{137}Cs: (decay mode: β-)

Beta decay frequency (%)	E_{avg} (keV)	E_{max} (keV)
94.43	173.4	511.5
5.57	424.6	1173.2
Energy release on average	187.4 (keV/decay)	

^{90}Sr (decay mode: Beta minus)

Beta decay frequency (%)	E_{avg} (keV)	E_{max} (keV)
100	195.7	546.0
Energy release on average	195.7 (keV/decay)	

On average, ^{137}Cs releases about 0.1874 MeV of energy per decay and ^{90}Sr releases about 0.1957 MeV of energy (as beta particles) per decay.

^{137m}Ba ($t_{1/2} = 2.552$ min) and ^{90}Y ($t_{1/2} = 2.67$ d), the decay products of ^{137}Cs and ^{90}Sr, respectively, disintegrate very quickly with the release of radiation particles. Thus the energy released from the decay of ^{137m}Ba and ^{90}Y can also be included as part of the total energy release of ^{137}Cs and ^{90}Sr,

(continued)

Example 7.3 (continued)
respectively (assuming secular equilibrium). Energy release from ^{137m}Ba and ^{90}Y is as follows.

^{137m}Ba: (decay mode: Gamma decay and internal conversion)

Auger electron release frequency (%)	E (keV)
8.32	624.2
1.19	655.7
Energy release on average	65.1 (keV/decay)

Gamma and x-rays frequency (%)	E (keV)
2.13	31.8
3.92	32.2
89.78	661.6
Energy release on average	596.5 (keV/decay)

^{90}Y (decay mode: Beta minus and internal conversion)

Beta decay frequency (%)	Eavg (keV)	Emax (keV)
99.98	934.8	2284.0
Energy release on average	934.6 (keV/decay)	

Auger electrons	E (keV)
Energy release on average	0.280 (keV/decay)

Gamma and x rays	E (keV)
Energy release on average	0.002 (keV/decay)

The total amount of decay heat carried by radiation particles from ^{137}Cs decay after 30 years of cooling by using the results from Example 7.1 $= 4.0 \times 10^{6}$ (Ci) $\times\ 3.7 \times 10^{10}$ (dps/Ci) $\times$ (0.1874 + 0.0651 + 0.5965) (MeV/dps) $= 1.256 \times 10^{18}$ (MeV/s) $\times\ 10^{6}$ (eV/MeV) $\times\ 1.6 \times 10^{-19}$ (J/eV) $= 2.01 \times 10^{4}$ (J/s) $= 2.01 \times 10^{4}$ (W) = 20.1 kW

Total amount of decay heat carried by radiation particles from ^{90}Sr decay after 30 years of cooling $= 3.78 \times 10^{6}$ (Ci) $\times\ 3.7 \times 10^{10}$ (dps/Ci) $\times$ (0.1957 + 0.9346 + 0.00028 + 0.000002) (MeV/dps) $= 1.58 \times 10^{17}$ (MeV/s) $\times\ 10^{6}$ (eV/MeV) $\times\ 1.6 \times 10^{-19}$ (J/eV) $= 2.53 \times 10^{4}$ (J/s) $= 2.53 \times 10^{4}$ (W) = 25.3 kW

Adding the two values gives 45.4 kW. This value is bigger than the estimated total decay heat calculated from Part (1) (40.5 kW). The difference is expected to be due to the uncertainty in the use of the simplified correlation.

Example 7.3 illustrates a point that decay heat decreases rather rapidly as a function of time. Also, the level of decay heat remains an important source of heat for long periods of time. The changes in the amount of decay heat as a function of the

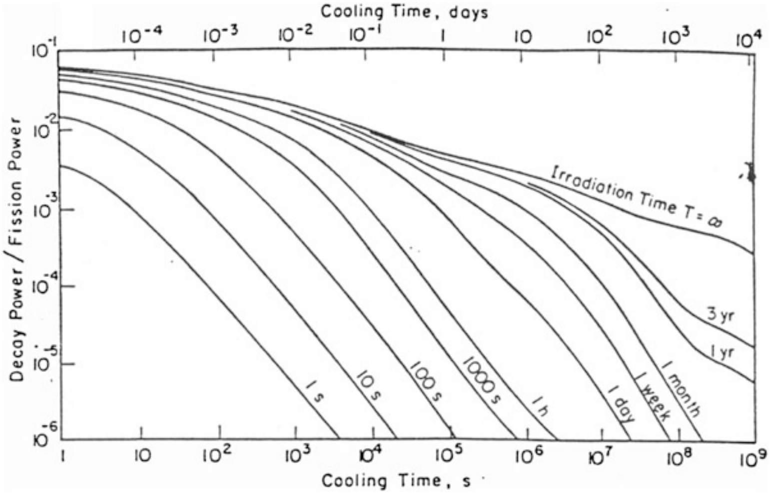


Fig. 7.7 Changes in decay power of LWR as a function of cooling time (at different irradiation periods)

reactor irradiation periods and the cooling time is captured in Fig. 7.7 for the case of ^{235}U fission.

Decay heat becomes higher with increase in fuel burnup. This is due to the increase in the production of fission products. Comparison of decay heat from spent fuel per unit mass basis at different burnups is also shown in Fig. 7.8. The values in Fig. 7.8 are relative to that of the reference burnup fuel at 33,000 MWD/MTHM. As indicated in the figure, decay heat increases with burnup but the rate of increase varies as a function of decay time after discharge. This is due to the variations in the half-life of the fission products as well as the changes in the contributions of various isotopes of uranium, plutonium, and minor actinides. For example, after around several hundred years, the contributions from ^{137}Cs and ^{90}Sr go away and the effect of burnup on decay heat becomes small. Then with the buildup of plutonium and minor actinides, the effect of burnup becomes pronounced again at around 30,000 years until they decay away.

In terms of which nuclides contribute the most to the decay heat production, Fig. 7.9 shows an example case of a standard PWR. We can see that, during the short term period after reactor shutdown, ^{137}Cs and ^{90}Sr dominate the decay heat production. They both decay with approximately 30 years of half-life and produce ^{90}Y and ^{137m}Ba, respectively. As both of these daughter products have very short half-life (64 hour for ^{90}Y and 2.552 min for ^{137m}Ba). The decay heat generated by the daughter products ^{90}Y and ^{137m}Ba can be grouped together with their parent nuclides assuming secular equilibrium (as explained in Chap. 3).

After ~100 years of cooling, the decay heat in spent fuel is dominated by ^{241}Am ($t_{1/2} = 432$ y) which is produced by beta decay of ^{241}Pu ($t_{1/2} = 14.7$y). Then beyond a thousand years of cooling, plutonium isotopes (Pu-240, etc.) start to become

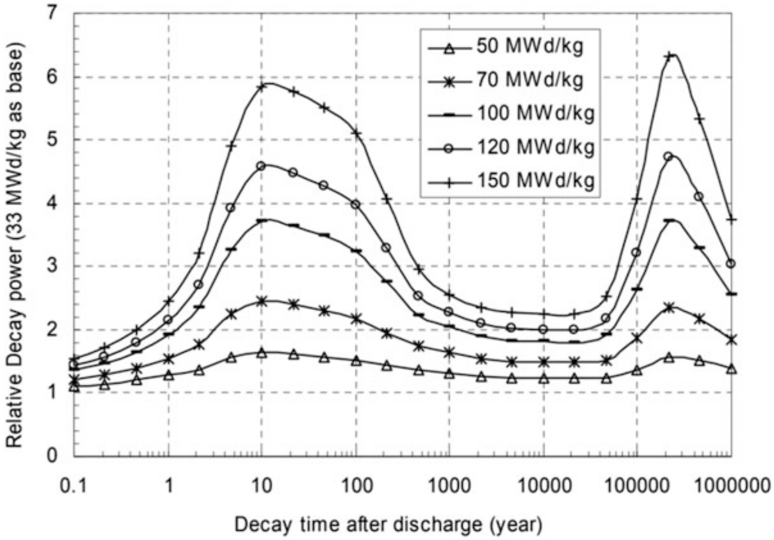


Fig. 7.8 Changes in decay power per MTIHM after discharge at different fuel burnup levels (PWR spent fuel). (Source: Xu 2003)

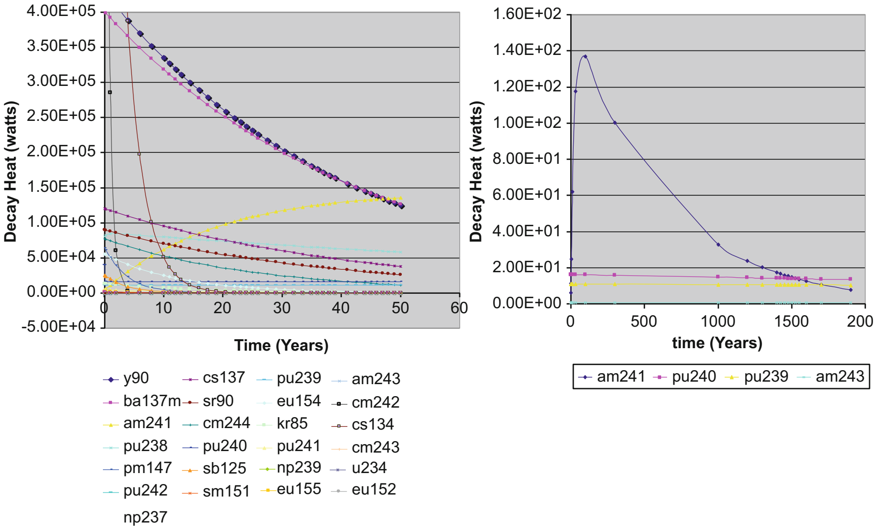


Fig. 7.9 Projected decay heat production for a typical PWR spent fuel (discharged @44 GWD/MTU) (Stahala 2006)

important. Importance of radionuclides in decay heat production also varies with the changes in burnup, cooling time, and initial enrichment of the fuel.

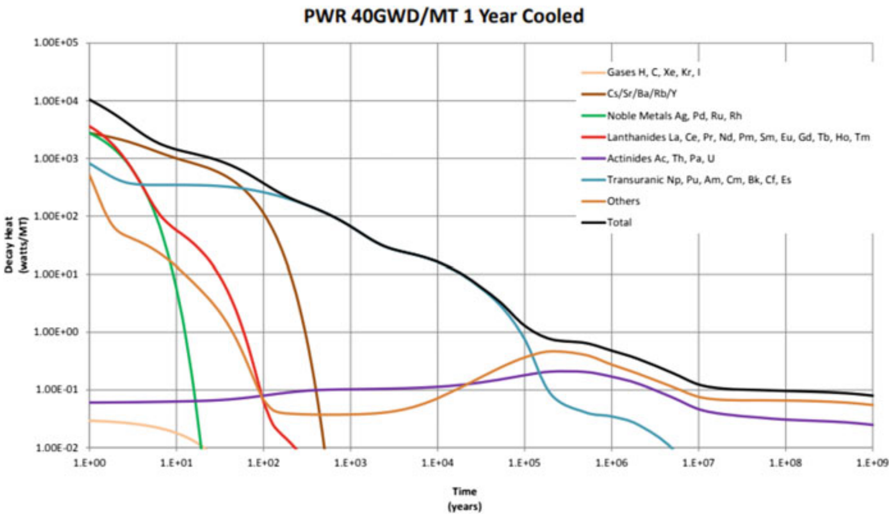


Fig. 7.10 Decay heat from spent nuclear fuel discharged at 40,000 MWD/MTU with 1 year cooling (Hardin et al. 2012, Table E-1, p. 259)

The overall contributions from different radionuclides to spent fuel decay heat for a 40,000 MWD/MTU, 1 year cooled case are captured in Fig. 7.10. Besides the initial contributions from Cs/Sr/Y/Ba, the transuranic elements dominate decay heat production in spent fuel through up to 100,000 years.

Table 7.9 shows more specifics on the contributions of different nuclides to decay heat production in PWRs at different burnups and cooling times. Other than ^{90}Y, ^{137m}Ba, ^{90}Sr, ^{137}Cs, and ^{241}Am, a few other nuclides such as ^{244}Cm, ^{106}Rh, and ^{238}Pu are found as important sources of decay heat. For a short cooled spent fuel, ^{134}Cs ($t_{1/2} = 2.065$ y) makes significant contribution.

7.1.4.2 Correlation Models for Decay Heat Calculation

Use of a correlation model has been suggested to facilitate accurate determination of decay heat of LWR spent fuel. Two of such models are given below.

The first model was developed for spent fuels at burnup less than 37,000 MWd/MTU (Malbrain et al. 1982):

For cooling periods, $1 < t < 30$ years

$$Q(t) = C_1 \cdot e^{(C_2 + C_3 t)^{-1}} \cdot \left(\frac{B}{33000}\right) \left[\frac{watts}{MTU}\right] \tag{7.27}$$

where,
$C_1 = 550$, $C_2 = 0.223$, $C_3 = 0.177$, and B is in burnup in [MWd/MTU].

Table 7.9 Radionuclides decay-heat rankings for PWR fuel

Burnup/enrichment												
	20 GWd/t 3 wt%		50 GWd/t 3 wt%		30 GWd/t 4 wt%		60 GWd/t 4 wt%		40 GWd/t 5 wt%		70 GWd/t 5 wt%	
Nuclide[a]	Percent	Rank	Percent	Rank	Percent	Rank	Percent	Rank	Percent	Rank	Percent	Rank
5-year cooling												
Am-241	1.96	10	1.54	12	1.96	10	1.44	12	1.90	12	1.36	12
Ba-137m	20.10	2	16.81	2	20.01	2	16.68	2	19.75	2	16.51	2
Ce-144	0.75	15	0.29	18	0.58	17	0.25	18	0.46	17	0.21	18
Cm-244	0.69	16	13.25	4	1.66	12	14.10	4	2.84	10	14.91	4
Co-60	4.68	8	3.80	8	3.81	9	3.18	9	3.23	9	2.74	9
Cs-134	10.88	3	17.45	1	13.43	3	17.70	1	15.01	3	17.74	1
Cs-137	6.02	6	5.03	7	5.99	6	4.99	7	5.92	5	4.95	7
Eu-154	1.41	11	2.26	11	1.84	11	2.32	11	2.15	11	2.38	11
Kr-85	0.6	17	0.39	15	0.59	16	0.40	15	0.57	15	0.40	15
Pm-147	1.36	12	0.53	14	1.11	13	0.47	14	0.93	13	0.43	14
Pr-144	8.42	5	3.24	9	6.49	5	2.77	10	5.15	7	2.40	10
Pu-238	2.38	9	6.43	6	3.83	8	7.59	5	5.24	6	8.66	5
Pu-239	0.87	14	0.34	16	0.68	15	0.31	16	0.57	16	0.28	16
Pu-240	0.94	13	0.68	13	0.82	14	0.60	13	0.72	14	0.54	13
Pu-241	0.19	19	0.14	19	0.18	19	0.13	19	0.17	19	0.12	19
Rh-106	10.08	4	8.57	5	8.74	4	7.19	6	7.55	4	6.17	6
Sb-125	0.41	18	0.34	17	0.38	18	0.30	17	0.34	18	0.27	17
Sr-90	4.84	7	3.20	10	4.78	7	3.32	8	4.72	8	3.39	8
Y-90	23.08	1	15.27	3	22.82	1	15.83	3	22.48	1	16.15	3
Total (W/t)	1090		3170		1630		3790		2180		4430	
100-year cooling												

(continued)

Table 7.9 (continued)

Burnup/enrichment												
	20 GWd/t 3 wt%		50 GWd/t 3 wt%		30 GWd/t 4 wt%		60 GWd/t 4 wt%		40 GWd/t 5 wt%		70 GWd/t 5 wt%	
Nuclide[a]	Percent	Rank	Percent	Rank	Percent	Rank	Percent	Rank	Percent	Rank	Percent	Rank
Am-241	46.58	1	39.22	1	44.98	1	35.98	1	42.48	1	33.32	1
Am-243	0.08	10	0.50	10	0.13	10	0.49	10	0.17	10	0.47	10
Ba-137m	13.93	2	12.99	3	13.69	2	12.84	3	13.47	3	12.62	3
Cm-244	0.11	9	2.42	7	0.27	9	2.56	7	0.46	9	2.69	7
Cs-137	4.17	7	3.89	6	4.10	7	3.85	6	4.04	6	3.78	6
Pu-238	7.00	4	21.09	2	11.13	4	24.80	2	15.20	2	28.09	2
Pu-239	5.43	6	2.39	8	4.20	6	2.13	9	3.49	7	1.92	9
Pu-240	5.79	5	4.90	5	4.99	5	4.35	5	4.43	5	3.92	5
Sr-90	2.90	8	2.14	9	2.83	8	2.21	8	2.78	8	2.24	8
Y-90	13.84	3	10.22	4	13.52	3	10.55	4	13.28	4	10.68	4
Total (W/t)	175		456		265		549		356		645	

Data source: NRC (2000)

[a] Greater than 0.1% of total decay-heat-generation rate for any burnup/enrichment combination

For cooling periods, 30 < t < 10^6 years

$$Q(t) = D_1 \cdot t^{-\beta} \cdot \left(\frac{B}{33000}\right) \left[\frac{watts}{MTU}\right] \tag{7.28}$$

where, $D_1 = 9410$ and $\beta = 0.749$. The quoted error in using the model for LWR spent fuel is generally less than 10% and not exceeding 35%.

The second decay heat model is a more detailed model for the consideration of a wider range of burnup given as (Stahala 2006):

$$Q(t) = e^{D_1} \cdot t^{-\beta} \cdot \left(\frac{B}{33000}\right) \left[\frac{watts}{MTU}\right] \tag{7.29}$$

where

$$\begin{aligned} D_1 = \alpha_1 + \ln(B)\{\alpha_2 + \alpha_5 T + \alpha_6 x_F + \alpha_8 \ln(B)\} + T(\alpha_3 + \alpha_7 x_F + \alpha_9 T) \\ + x_F(\alpha_4 + \alpha_{10} x_F) \end{aligned} \tag{7.30}$$

$$\begin{aligned} \beta = \gamma_1 + \ln(B)\{\gamma_2 + \gamma_5 T + \gamma_6 x_F + \gamma_8 \ln(B)\} + T(\gamma_3 + \gamma_7 x_F + \gamma_9 T) \\ + x_F(\gamma_4 + \gamma_{10} x_F) \end{aligned} \tag{7.31}$$

B is in burnup in [MWd/MTU], T is irradiation days, and x_F is enrichment (wt%). The applicable ranges of the equation are: burnup from 1000 to 58,500 MWd/MTU; irradiation days between 150 and 1350 days, and; initial ^{235}U enrichment from 1.5 to 5 wt%.

The data for each coefficients used in the equation are given as a function of burnup and cooling periods in Tables 7.10 and 7.11 for PWRs and Tables 7.12 and 7.13 for BWRs. The maximum error in the results from the use of this model was estimated at 8.5%, with a mean error of 2.9%.

In the case of recycled mixed oxide (MOX) spent fuel, the decay heat inventory in spent fuel becomes much higher than the once-burned spent fuel even at the same burnup (by more than a factor of 2). This is depicted in Fig. 7.11. The difference is mainly due to higher buildup of actinides in the MOX spent fuel. For MOX spent fuel, ^{244}Cm ($t_{1/2} = 18.1$ y), along with ^{238}Pu ($t_{1/2} = 87.7$ y) and ^{241}Am ($t_{1/2} = 432.7$ y), is estimated to contribute the most to the heat load as represented in Table 7.14. Table 7.15 shows a typical inventory of radioactivity and decay heat in a UO_2 and MOX fuel including the contributions of actinides at different burnups and cooling periods.

7.2 Shielding for Spent Nuclear Fuel

As the constituents of spent fuel include sources of penetrating radiation such as gamma rays and neutrons, proper means of shielding must be in place to protect the worker and the public during handling, shipping and storage of spent fuel.

Table 7.10 PWR spent fuel decay heat model coefficients (BU > 10,000 MWd/MTU)

1 to 8		8 to 49		49 to 74		74 to 150	
Cooling period (years)							
α_1	1.025×10^{1}	α_1	8.355	α_1	9.678	α_1	9.541
α_2	1.698×10^{-1}	α_2	8.403×10^{-1}	α_2	1.464	α_2	1.619
α_3	-1.030×10^{-3}	α_3	-8.600×10^{-5}	α_3	-5.700×10^{-5}	α_3	3.300×10^{-5}
α_4	-8.375×10^{-2}	α_4	-1.178×10^{-1}	α_4	-8.200×10^{-2}	α_4	1.107×10^{-1}
α_5	9.490×10^{-5}	α_5	-4.300×10^{-6}	α_5	-2.000×10^{-6}	α_5	3.390×10^{-5}
α_6	-4.580×10^{-3}	α_6	-1.159×10^{-1}	α_6	-3.071×10^{-1}	α_6	-3.751×10^{-1}
α_7	-1.400×10^{-5}	α_7	2.380×10^{-6}	α_7	1.690×10^{-7}	α_7	-9.860×10^{-6}
α_8	3.856×10^{-2}	α_8	3.238×10^{-1}	α_8	7.118×10^{-1}	α_8	8.897×10^{-1}
α_9	2.520×10^{-7}	α_9	-2.400×10^{-9}	α_9	-7.600×10^{-9}	α_9	-3.810×10^{-8}
α_{10}	6.319×10^{-3}	α_{10}	1.078×10^{-2}	α_{10}	1.517×10^{-2}	α_{10}	4.219×10^{-3}
γ_1	1.418	γ_1	4.894×10^{-1}	γ_1	8.466×10^{-1}	γ_1	8.185×10^{-1}
γ_2	-1.433×10^{-1}	γ_2	2.014×10^{-1}	γ_2	3.719×10^{-1}	γ_2	4.087×10^{-1}
γ_3	-4.700×10^{-4}	γ_3	-2.800×10^{-5}	γ_3	-1.800×10^{-5}	γ_3	2.730×10^{-6}
γ_4	1.729×10^{-3}	γ_4	-2.555×10^{-2}	γ_4	-1.658×10^{-2}	γ_4	$2.763.\mathrm{E} \times 10^{-2}$
γ_5	4.740×10^{-5}	γ_5	-5.400×10^{-6}	γ_5	-4.500×10^{-6}	γ_5	3.590×10^{-6}
γ_6	1.728×10^{-2}	γ_6	-3.259×10^{-2}	γ_6	-8.504×10^{-2}	γ_6	-1.011×10^{-1}
γ_7	-9.800×10^{-6}	γ_7	1.360×10^{-6}	γ_7	6.870×10^{-7}	γ_7	-1.590×10^{-6}
γ_8	-5.534×10^{-2}	γ_8	7.826×10^{-2}	γ_8	1.847×10^{-1}	γ_8	2.262×10^{-1}
γ_9	1.250×10^{-7}	γ_9	7.610×10^{-10}	γ_9	-8.000×10^{-10}	γ_9	-7.740×10^{-9}
γ_{10}	3.430×10^{-5}	γ_{10}	2.538×10^{-3}	γ_{10}	3.817×10^{-3}	γ_{10}	1.317×10^{-3}

150 to 300		300 to 1500		1500 to 10,000	
Cooling period (years)					
α_1	7.938	α_1	9.338	α_1	7.989
α_2	6.803×10^{-1}	α_2	-8.019×10^{-1}	α_2	-6.536×10^{-1}
α_3	3.010×10^{-4}	α_3	1.530×10^{-4}	α_3	7.680×10^{-5}
α_4	1.622×10^{-1}	α_4	3.957×10^{-2}	α_4	-9.485×10^{-2}
α_5	1.120×10^{-4}	α_5	4.340×10^{-5}	α_5	1.460×10^{-5}
α_6	-8.820×10^{-3}	α_6	3.891×10^{-1}	α_6	3.298×10^{-1}
α_7	-1.860×10^{-5}	α_7	-5.700×10^{-6}	α_7	-3.600×10^{-6}
α_8	3.258×10^{-1}	α_8	-8.680×10^{-1}	α_8	-7.193×10^{-1}
α_9	-9.350×10^{-8}	α_9	-4.500×10^{-8}	α_9	-2.800×10^{-8}
α_{10}	-1.376×10^{-2}	α_{10}	-1.638×10^{-2}	α_{10}	-5.920×10^{-3}
γ_1	4.983×10^{-1}	γ_1	7.275×10^{-1}	γ_1	5.560×10^{-1}
γ_2	2.235×10^{-1}	γ_2	-3.259×10^{-2}	γ_2	-1.655×10^{-2}
γ_3	5.640×10^{-5}	γ_3	3.180×10^{-5}	γ_3	2.120×10^{-5}
γ_4	3.855×10^{-2}	γ_4	1.710×10^{-2}	γ_4	-9.300×10^{-4}
γ_5	1.920×10^{-5}	γ_5	7.810×10^{-6}	γ_5	3.680×10^{-6}
γ_6	-2.882×10^{-2}	γ_6	4.034×10^{-2}	γ_6	3.385×10^{-2}
γ_7	-3.390×10^{-6}	γ_7	-1.200×10^{-6}	γ_7	-8.600×10^{-7}
γ_8	1.154×10^{-1}	γ_8	-8.934×10^{-2}	γ_8	-7.437×10^{-2}
γ_9	-1.890×10^{-8}	γ_9	-1.100×10^{-8}	γ_9	-8.300×10^{-9}
γ_{10}	-2.280×10^{-3}	γ_{10}	-2.760×10^{-3}	γ_{10}	-1.390×10^{-3}

Table 7.11 PWR spent fuel decay heat model coefficients (BU $\leq$ 10,000 MWd/MTU)

1 to 8		8 to 49		49 to 74		74 to 150	
Cooling period (years)							
α_1	1.022 × 101	α_1	7.408	α_1	6.859	α_1	4.681
α_2	1.250×10^{-1}	α_2	-2.876×10^{-1}	α_2	−1.526	α_2	−2.848
α_3	-1.150×10^{-3}	α_3	-2.700×10^{-5}	α_3	-2.900×10^{-5}	α_3	-4.970×10^{-5}
α_4	-9.312×10^{-2}	α_4	1.630×10^{-1}	α_4	7.870×10^{-1}	α_4	1.405
α_5	5.390×10^{-5}	α_5	2.640×10^{-5}	α_5	2.230×10^{-5}	α_5	2.840×10^{-5}
α_6	-1.397×10^{-2}	α_6	2.092×10^{-2}	α_6	8.826×10^{-2}	α_6	7.764×10^{-2}
α_7	-7.320×10^{-6}	α_7	1.390×10^{-6}	α_7	1.630×10^{-6}	α_7	-5.590×10^{-6}
α_8	1.070×10^{-2}	α_8	-3.356×10^{-2}	α_8	-1.778×10^{-1}	α_8	-3.378×10^{-1}
α_9	2.910×10^{-7}	α_9	-4.600×10^{-9}	α_9	-5.600×10^{-9}	α_9	3.110×10^{-9}
α_{10}	7.205×10^{-3}	α_{10}	-1.248×10^{-2}	α_{10}	-5.802×10^{-2}	α_{10}	-1.022×10^{-1}
γ_1	1.561	γ_1	2.046×10^{-1}	γ_1	5.878×10^{-2}	γ_1	-4.409×10^{-1}
γ_2	3.370×10^{-2}	γ_2	-1.142×10^{-1}	γ_2	-4.472×10^{-1}	γ_2	-7.530×10^{-1}
γ_3	-5.500×10^{-4}	γ_3	-2.400×10^{-5}	γ_3	-2.300×10^{-5}	γ_3	-2.750×10^{-5}
γ_4	-3.569×10^{-2}	γ_4	6.469×10^{-2}	γ_4	2.322×10^{-1}	γ_4	3.754×10^{-1}
γ_5	1.500×10^{-5}	γ_5	-2.400×10^{-6}	γ_5	-3.400×10^{-6}	γ_5	$-2.050 \times 10^{–6}$
γ_6	-5.340×10^{-3}	γ_6	8.642×10^{-3}	γ_6	2.685×10^{-2}	γ_6	2.477×10^{-2}
γ_7	-5.740×10^{-6}	γ_7	8.940×10^{-7}	γ_7	9.260×10^{-7}	γ_7	-6.980×10^{-7}
γ_8	2.836×10^{-3}	γ_8	-1.312×10^{-2}	γ_8	-5.188×10^{-2}	γ_8	-8.887×10^{-2}
γ_9	1.450×10^{-7}	γ_9	3.480×10^{-9}	γ_9	2.980×10^{-9}	γ_9	4.970×10^{-9}
γ_{10}	3.114×10^{-3}	γ_{10}	-4.820×10^{-3}	γ_{10}	-1.704×10^{-2}	γ_{10}	-2.729×10^{-2}

150 to 300		300 to 1500		1500 to 10,000	
Cooling period (years)					
α_1	7.391	α_1	1.489 × 101	α_1	1.067×10^{1}
α_2	6.295×10^{-2}	α_2	4.487	α_2	2.348
α_3	1.650×10^{-4}	α_3	1.890×10^{-4}	α_3	1.240×10^{-4}
α_4	8.354×10^{-2}	α_4	−1.311	α_4	−1.012
α_5	7.320×10^{-5}	α_5	5.400×10^{-5}	α_5	4.180×10^{-5}
α_6	-1.245×10^{-1}	α_6	-1.452×10^{-1}	α_6	-9.638×10^{-2}
α_7	-1.840×10^{-5}	α_7	-1.280×10^{-6}	α_7	-1.100×10^{-6}
α_8	-1.932×10^{-2}	α_8	5.295×10^{-1}	α_8	2.757×10^{-1}
α_9	-2.740×10^{-8}	α_9	-6.150×10^{-8}	α_9	-3.800×10^{-8}
α_{10}	-7.050×10^{-3}	α_{10}	8.211×10^{-2}	α_{10}	6.904×10^{-2}
γ_1	8.248×10^{-2}	γ_1	1.375	γ_1	8.254×10^{-1}
γ_2	-1.829×10^{-1}	γ_2	5.842×10^{-1}	γ_2	3.066×10^{-1}
γ_3	1.510×10^{-5}	γ_3	1.970×10^{-5}	γ_3	1.090×10^{-5}
γ_4	1.158×10^{-1}	γ_4	-1.275×10^{-1}	γ_4	-9.025×10^{-2}
γ_5	6.970×10^{-6}	γ_5	3.640×10^{-6}	γ_5	1.990×10^{-6}
γ_6	-1.586×10^{-2}	γ_6	-1.868×10^{-2}	γ_6	-1.279×10^{-2}
γ_7	-3.330×10^{-6}	γ_7	-3.130×10^{-7}	γ_7	-2.800×10^{-7}

(continued)

Table 7.11 (continued)

150 to 300		300 to 1500		1500 to 10,000	
γ_8	-2.659×10^{-2}	γ_8	6.833×10^{-2}	γ_8	3.572×10^{-2}
γ_9	-9.790×10^{-10}	γ_9	-6.980×10^{-9}	γ_9	-3.900×10^{-9}
γ_{10}	-8.600×10^{-3}	γ_{10}	7.033×10^{-3}	γ_{10}	5.435×10^{-3}

Table 7.12 BWR spent fuel decay heat model coefficients (BU > 10,000 MWd/MTU)

1 to 8		8 to 49		49 to 74		74 to 150	
Cooling period (years)							
α_1	1.025×10^{1}	α_1	8.400	α_1	9.817	α_1	9.709
α_2	1.692×10^{-1}	α_2	8.652×10^{-1}	α_2	1.557	α_2	1.731
α_3	-1.030×10^{-3}	α_3	-8.640×10^{-5}	α_3	-6.200×10^{-5}	α_3	2.750×10^{-5}
α_4	-8.444×10^{-2}	α_4	-1.278×10^{-1}	α_4	-1.091×10^{-1}	α_4	8.942×10^{-2}
α_5	9.410×10^{-5}	α_5	-4.120×10^{-6}	α_5	-4.900×10^{-6}	α_5	2.730×10^{-5}
α_6	-5.240×10^{-3}	α_6	-1.186×10^{-1}	α_6	-3.125×10^{-1}	α_6	-3.758×10^{-1}
α_7	-1.400×10^{-5}	α_7	2.310×10^{-6}	α_7	4.900×10^{-7}	α_7	-9.550×10^{-6}
α_8	3.999×10^{-2}	α_8	3.194×10^{-1}	α_8	6.995×10^{-1}	α_8	8.637×10^{-1}
α_9	2.520×10^{-7}	α_9	-2.270×10^{-9}	α_9	-6.400×10^{-9}	α_9	-3.820×10^{-8}
α_{10}	6.340×10^{-3}	α_{10}	1.159×10^{-2}	α_{10}	1.760×10^{-2}	α_{10}	6.272×10^{-3}
γ_1	1.412	γ_1	5.031×10^{-1}	γ_1	8.860×10^{-1}	γ_1	8.649×10^{-1}
γ_2	-1.491×10^{-1}	γ_2	2.065×10^{-1}	γ_2	3.956×10^{-1}	γ_2	4.370×10^{-1}
γ_3	-4.700×10^{-4}	γ_3	-2.760×10^{-5}	γ_3	-1.900×10^{-5}	γ_3	1.320×10^{-6}
γ_4	3.684×10^{-3}	γ_4	-2.781×10^{-2}	γ_4	-2.352×10^{-2}	γ_4	2.195×10^{-2}
γ_5	4.650×10^{-5}	γ_5	-4.980×10^{-6}	γ_5	-5.000×10^{-6}	γ_5	2.310×10^{-6}
γ_6	1.833×10^{-2}	γ_6	-3.258×10^{-2}	γ_6	-8.577×10^{-2}	γ_6	-1.007×10^{-1}
γ_7	-9.900×10^{-6}	γ_7	1.310×10^{-6}	γ_7	7.490×10^{-7}	γ_7	-1.530×10^{-6}
γ_8	-5.581×10^{-2}	γ_8	7.537×10^{-2}	γ_8	1.796×10^{-1}	γ_8	2.180×10^{-1}
γ_9	1.250×10^{-7}	γ_9	8.770×10^{-10}	γ_9	-3.900×10^{-10}	γ_9	-7.620×10^{-9}
γ_{10}	-1.300×10^{-4}	γ_{10}	2.718×10^{-3}	γ_{10}	4.441×10^{-3}	γ_{10}	1.861×10^{-3}

150 to 300		300 to 1500		1500 to 10,000	
Cooling period (years)					
α_1	7.941	α_1	9.127	α_1	7.877
α_2	7.129×10^{-1}	α_2	-9.109×10^{-1}	α_2	-7.718×10^{-1}
α_3	3.180×10^{-4}	α_3	1.600×10^{-4}	α_3	7.590×10^{-5}
α_4	1.805×10^{-1}	α_4	6.910×10^{-2}	α_4	-8.144×10^{-2}
α_5	1.100×10^{-4}	α_5	4.650×10^{-5}	α_5	1.550×10^{-5}
α_6	-6.020×10^{-3}	α_6	3.811×10^{-1}	α_6	3.287×10^{-1}
α_7	-1.930×10^{-5}	α_7	-5.900×10^{-6}	α_7	-3.560×10^{-6}
α_8	3.390×10^{-1}	α_8	-8.281×10^{-1}	α_8	-7.213×10^{-1}

(continued)

Table 7.12 (continued)

150 to 300		300 to 1500		1500 to 10,000	
α_9	-1.010×10^{-7}	α_9	-4.670×10^{-8}	α_9	-2.740×10^{-8}
α_{10}	-1.575×10^{-2}	α_{10}	-1.837×10^{-2}	α_{10}	-6.720×10^{-3}
γ_1	5.120×10^{-1}	γ_1	7.042×10^{-1}	γ_1	5.450×10^{-1}
γ_2	2.360×10^{-1}	γ_2	-4.424×10^{-2}	γ_2	-3.035×10^{-2}
γ_3	5.940×10^{-5}	γ_3	3.320×10^{-5}	γ_3	2.150×10^{-5}
γ_4	4.077×10^{-2}	γ_4	2.130×10^{-2}	γ_4	1.146×10^{-3}
γ_5	1.890×10^{-5}	γ_5	8.350×10^{-6}	γ_5	3.910×10^{-6}
γ_6	-2.775×10^{-2}	γ_6	3.954×10^{-2}	γ_6	3.394×10^{-2}
γ_7	-3.520×10^{-6}	γ_7	-1.210×10^{-6}	γ_7	-8.860×10^{-7}
γ_8	1.148×10^{-1}	γ_8	-8.502×10^{-2}	γ_8	-7.581×10^{-2}
γ_9	-2.030×10^{-8}	γ_9	-1.110×10^{-8}	γ_9	-8.420×10^{-9}
γ_{10}	-2.540×10^{-3}	γ_{10}	-3.040×10^{-3}	γ_{10}	-1.500×10^{-3}

Table 7.13 PWR spent fuel decay heat model coefficients (BU $\leq$ 10,000 MWd/MTU)

1 to 8		8 to 49		49 to 74		74 to 150	
Cooling period (years)							
α_1	1.023×10^{1}	α_1	7.442	α_1	6.926	α_1	5.087
α_2	1.225×10^{-1}	α_2	-2.290×10^{-1}	α_2	-1.385	α_2	-2.422
α_3	-1.140×10^{-3}	α_3	-3.670×10^{-5}	α_3	-2.900×10^{-5}	α_3	-4.400×10^{-5}
α_4	-9.590×10^{-2}	α_4	1.496×10^{-1}	α_4	7.873×10^{-1}	α_4	1.328
α_5	5.440×10^{-5}	α_5	2.600×10^{-5}	α_5	2.230×10^{-5}	α_5	2.840×10^{-5}
α_6	-1.406×10^{-2}	α_6	1.736×10^{-2}	α_6	7.773×10^{-2}	α_6	4.169×10^{-2}
α_7	-7.640×10^{-6}	α_7	1.740×10^{-6}	α_7	1.560×10^{-6}	α_7	-6.400×10^{-6}
α_8	9.890×10^{-3}	α_8	-2.565×10^{-2}	α_8	-1.544×10^{-1}	α_8	-2.760×10^{-1}
α_9	2.890×10^{-7}	α_9	-3.060×10^{-9}	α_9	-5.700×10^{-9}	α_9	2.570×10^{-9}
α_{10}	7.419×10^{-3}	α_{10}	-1.177×10^{-2}	α_{10}	-5.871×10^{-2}	α_{10}	-9.705×10^{-2}
γ_1	1.561	γ_1	2.153×10^{-1}	γ_1	7.530×10^{-2}	γ_1	-3.469×10^{-1}
γ_2	3.222×10^{-2}	γ_2	-9.287×10^{-2}	γ_2	-4.067×10^{-1}	γ_2	-6.473×10^{-1}
γ_3	-5.500×10^{-4}	γ_3	-2.750×10^{-5}	γ_3	-2.300×10^{-5}	γ_3	-2.700×10^{-5}
γ_4	-3.693×10^{-2}	γ_4	6.049×10^{-2}	γ_4	2.332×10^{-1}	γ_4	3.589×10^{-1}
γ_5	1.520×10^{-5}	γ_5	-2.630×10^{-6}	γ_5	-3.400×10^{-6}	γ_5	-2.100×10^{-6}
γ_6	-5.310×10^{-3}	γ_6	7.445×10^{-3}	γ_6	2.399×10^{-2}	γ_6	1.609×10^{-2}
γ_7	-6.040×10^{-6}	γ_7	1.080×10^{-6}	γ_7	9.650×10^{-7}	γ_7	-8.200×10^{-7}
γ_8	2.620×10^{-3}	γ_8	-1.000×10^{-2}	γ_8	-4.495×10^{-2}	γ_8	-7.312×10^{-2}
γ_9	1.440×10^{-7}	γ_9	4.090×10^{-9}	γ_9	2.960×10^{-9}	γ_9	4.890×10^{-9}
γ_{10}	3.264×10^{-3}	γ_{10}	-4.590×10^{-3}	γ_{10}	-1.730×10^{-2}	γ_{10}	-2.622×10^{-2}
150 to 300		300 to 1500		1500 to 10,000			

(continued)

Table 7.13 (continued)

150 to 300		300 to 1500		1500 to 10,000	
Cooling period (years)					
α_1	7.699	α_1	1.455×10^{1}	α_1	1.067×10^{1}
α_2	2.394×10^{-1}	α_2	4.071	α_2	2.248
α_3	1.850×10^{-4}	α_3	1.940×10^{-4}	α_3	1.240×10^{-4}
α_4	-1.600×10^{-4}	α_4	-1.249	α_4	-9.989×10^{-1}
α_5	7.320×10^{-5}	α_5	5.610×10^{-5}	α_5	4.190×10^{-5}
α_6	-1.422×10^{-1}	α_6	-1.168×10^{-1}	α_6	-8.634×10^{-2}
α_7	-2.100×10^{-5}	α_7	-1.360×10^{-6}	α_7	-9.490×10^{-7}
α_8	-3.860×10^{-3}	α_8	4.567×10^{-1}	α_8	2.586×10^{-1}
α_9	-3.200×10^{-8}	α_9	-6.240×10^{-8}	α_9	-3.840×10^{-8}
α_{10}	7.040×10^{-4}	α_{10}	7.803×10^{-2}	α_{10}	6.710×10^{-2}
γ_1	1.596×10^{-1}	γ_1	1.338	γ_1	8.340×10^{-1}
γ_2	-1.246×10^{-1}	γ_2	5.378×10^{-1}	γ_2	3.032×10^{-1}
γ_3	1.870×10^{-5}	γ_3	2.040×10^{-5}	γ_3	1.100×10^{-5}
γ_4	9.725×10^{-2}	γ_4	-1.203×10^{-1}	γ_4	-8.984×10^{-2}
γ_5	6.910×10^{-6}	γ_5	3.950×10^{-6}	γ_5	2.020×10^{-6}
γ_6	-2.110×10^{-2}	γ_6	-1.577×10^{-2}	γ_6	-1.238×10^{-2}
γ_7	-3.800×10^{-6}	γ_7	-3.250×10^{-7}	γ_7	-2.630×10^{-7}
γ_8	-1.975×10^{-2}	γ_8	5.962×10^{-2}	γ_8	3.460×10^{-2}
γ_9	-1.800×10^{-9}	γ_9	-7.150×10^{-9}	γ_9	-3.950×10^{-9}
γ_{10}	-6.950×10^{-3}	γ_{10}	6.591×10^{-3}	γ_{10}	5.284×10^{-3}

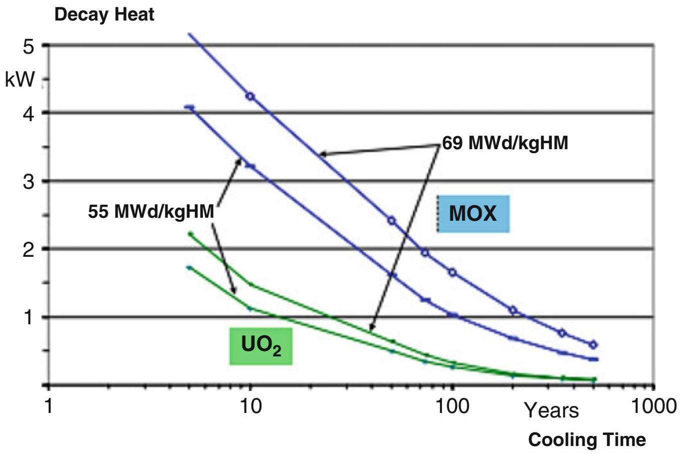


Fig. 7.11 Comparison of decay heat between UO2 and MOX spent fuel at different burnup and cooling time (IAEA 2003, Fig. 2 p. 34)

Table 7.14 Examples of heat load (in W) of various isotopes in MOX and UO_2 spent fuel

	Spent MOX fuel (50 GWD/MTHM; after 7 years cooling)	Spent UO_2 fuel (60 GWD/MTHM; 4.2 w/o; after 3 years of cooling)
Pu-238	6390	905
Pu-239	188	39.1
Pu-240	591	72.8
Pu-241	103	18.2
Pu-242	4.39	0.454
Am-241	2000	137
Am-242m	0.334	0.0324
AM-243	57.9	7.88
Cm-242	27.1	128
Cm-243	34.2	5.23
Cm-244	11,100	1710
Cm-245	3.27	0.275
Cm-246	0.386	0.0819

Source: Bathke et al. (2002)

Table 7.15 Changes in activity and heat load of different spent fuels as a function of cooling time

	Standard Burnup UO_2 Fuel (33 GWD/THM)		Extended Burnup UO_2 Fuel (60 GWD/THM)		MOX Fuel (50 GWD/THM)	
Cooling periods-yr	Actinides	Total	Actinides	Total	Actinides	Total
	Activity (ci)					
1	5.75×10^5	8.72×10^6	8.06×10^5	9.73×10^6	4.11×10^6	1.24×10^7
3	4.85×10^5	3.35×10^6	6.66×10^5	4.42×10^6	3.47×10^6	6.52×10^6
5	4.42×10^5	2.13×10^6	6.07×10^5	3.10×10^6	3.18×10^6	4.96×10^6
7	4.04×10^5	1.73×10^6	5.56×10^5	2.61×10^6	2.91×10^6	4.30×10^6
10	3.54×10^5	1.46×10^6	4.89×10^5	2.24×10^6	1.92×10^6	2.83×10^6
	Heat load (W)					
1	2.84×10^3	3.75×10^4	6.04×10^3	4.51×10^4	2.92×10^4	6.48×10^4
3	1.20×10^3	1.24×10^4	3.01×10^3	1.80×10^4	1.66×10^4	2.86×10^4
5	1.13×10^3	6.92×10^3	2.80×10^3	1.16×10^4	1.56×10^4	2.18×10^4
7	1.12×10^3	5.25×10^3	2.71×10^3	9.28×10^3	1.51×10^4	1.94×10^4
10	1.11×10^3	4.36×10^3	2.60×10^3	7.84×10^3	1.32×10^4	1.57×10^4

Source: Bathke et al. (2002)

Table 7.16 Important radionuclides in shielding for spent fuel

	Low-burnup fuel (20 GWD/t, 3.0 w/o initial enrichment)		High-burnup fuel (70 GWD/t, 5.0 w/o initial enrichment)	
	Activation/fission products	Actinides	Activation/fission products	Actinides
High importance	^{60}Co ^{137m}Ba		^{60}Co	^{244}Cm
Medium importance	^{90}Y ^{154}Eu		^{90}Y ^{137m}Ba	^{246}Cm
Low importance	^{134}Cs	^{238}Pu ^{240}Pu ^{242}Pu ^{241}Am ^{244}Cm	^{134}Cs ^{154}Eu	^{238}Pu ^{240}Pu ^{242}Pu ^{241}Am

The important radionuclides in contributing to radiation dose are:

- Neutron source:
  - ^{244}Cm, ^{241}Am, ^{246}Cm, ^{240}Pu, ^{242}Pu, ^{238}Pu, etc.
- Gamma source:
  - ^{60}Co, ^{90}Sr, ^{134}Cs, ^{137}Cs, ^{144}Pr, ^{154}Eu, ^{244}Cm, etc.

These important nuclides are also listed in Table 7.16 with the indication of their relative importance. Regardless of the burnup, ^{60}Co, ^{90}Y (the decay product of ^{90}Sr), and ^{137m}Ba (the decay product of ^{137}Cs) are the biggest sources of gamma irradiation from spent fuel (followed by ^{154}Eu and ^{134}Cs with much less importance). The importance of actinides as the source of neutron dose depends on the level of burnup and becomes prominent at high burnup. Key contributors to neutron dose at high burnup are ^{244}Cm and ^{246}Cm.

At the time of discharge, the dose rate level at the surface of an unshielded spent fuel assembly can reach ~260 Gy/h (between 200 and 300 Gy/hr). Therefore, delivery of lethal dose (4 ~ 5 Gy) is quite possible if any individual gets close to spent fuel. The largest source of radiation dose from spent fuel is ^{60}Co along with ^{137}Cs (and its decay product ^{137m}Ba) and ^{244}Cm. This dose level decreases to ~130 Gy/h (between 100 and 200 Gy/h) after 30 years. At 1 meter from the center of an assembly (on a line perpendicular to its axis), the dose from a 30 year cooled spent fuel would be about ~8 Gy/h. Thus one-hour exposure to the spent fuel at 1 meter could still give a dose in excess of the lethal dose. Accordingly, any handling of spent fuel must always take place remotely with a shield in place (i.e., under water) until spent fuel is enclosed in a heavily shielded system, eliminating the possibility of human access to spent fuel.

Shielding for spent fuel is achieved using the combinations of shield materials against neutrons and gamma rays. As the radiation interaction mechanisms involved are distinctively different between neutrons and gamma rays, the materials and approaches to shielding against neutrons and gamma rays are different.

In the case of gamma rays, the radiation loses energy through chance encounters. Therefore, to increase the degree of attenuation and energy loss in the shield, the shield material should be dense and of high-atomic-number. Commonly used gamma shield material includes steel, lead, concrete, depleted uranium, or tungsten. While steel or lead are very popular, depleted uranium or tungsten is used at higher cost if a high performance minimum thickness shield is required. Depleted uranium is a by-product from enriching natural uranium with residual ^{235}U content of 0.2–0.3% with ^{238}U comprising the remainder. Concrete is also used if large thickness and mass can be allowed in the application. Iron ore and iron scrap may be used together with concrete as an aggregate for heavy or high-density concrete, achieving better attenuation than the ordinary concrete.

Light elements like hydrogen or carbon are effective neutron shield materials but using them as shield requires the energy levels of neutrons to be low to have sizable interaction cross sections. As the neutrons produced by fission reactions are fast (energy of several hundred keV or above), it is necessary to slow them down, as the capture cross section of neutrons increases with decreasing energy and is only large enough in the thermal range. Accordingly, neutron shield is designed first to slow down the neutrons (e.g., by using iron through inelastic scattering) and then to absorb them as thermal neutrons. Secondary gamma rays are often produced by the slowing down of fast neutrons, either from inelastic scattering or neutron capture in the shield. Thus the shield has to attenuate both gamma rays and neutrons. Common neutron-shielding materials include water, polyethylene, and concrete. Lithium hydride, though very expensive, has been used when use of small amount of shield mass is necessary. In addition, for neutron capture at thermal energy, a material with high neutron cross-section (such as boron or lithium) is used. In any case, good neutron shielding material must contain high concentration of hydrogen as hydrogen effectively slows down neutrons.

Comparison of various shield materials with respect to density, atomic number, hydrogen content, and other relevant information is given in Table 7.17. Implementation of different materials for spent fuel shielding will depend upon the purpose. For transportation casks, lead, steel, and depleted uranium is mostly commonly used for gamma shielding and plastic resins are often used for neutron shielding. For storage systems, concrete is widely used as shielding material as it can shield both gamma rays and neutrons.

7.2.1 Analysis for Gamma Ray Shielding

In the implementation of radiation shielding, the degree of radiation attenuation with respect to the specifics of shield design is considered. Commonly, the shielding analysis is done through attenuation calculation by using a simplified modeling approach. The following equation is an example of the simplified modeling

Table 7.17 Comparison of major shielding materials and their characteristics

	Lead	Iron	Tungsten	Depleted U	Concrete ordinary	Concrete heavy	Water	Polyethy-lene	LiH
Density (g/cm^3)	11.35	7.87	19.3	18.9	2.2 ~ 2.4	3.7 ~ 4.8	1	0.95	0.78
Atomic number	82	26	74	92	11 (effective)	~26 (effective)	7.4	5.5	1.5
Melting point °C	327	1535	3410	1132			100 (boiling)	40 (softening)	686
Hydrogen concentration					0.8 to 2.4 × 10^{22} atoms/cm^3		6.7 × 10^{22} atom/cm^3	8.0 × 10^{22} atom/cm^3	5.9 × 10^{22} atom/cm^3
Radiation damage	Nil	Small	Small	High	Negligible		Small	Significant	Small
Thermal neutron activation	Impurities	Moderate	High	High	Low	High	Impurities	Nil	Negligible
Approximate cost ($/kg)	2	0.2	~30		0.09	0.52	Container cost	2	30

approach, called the point-kernel technique, to represent the changes in the strength of radiation with a shield in place for the case of radiation source as a point.

$$I(x) \equiv \phi(x) = \frac{S}{4\pi r^2} \cdot B(x) \cdot e^{-\mu x} \tag{7.32}$$

where, S is the source strength (i.e., number of the gamma rays emitted per unit time), r is the distance between the source and the location of shield, x is the thickness of the shield, and μ is the attenuation coefficient as defined in Sect. 3.2.3.

The point kernel represents the response to the unit point-source (i.e., $S = 1$),

$$K(x) = \frac{1}{4\pi r^2} \cdot B(x) \cdot e^{-\mu x} \tag{7.33}$$

The fundamental assumption in the point kernel technique is that extended/distributed sources of radiation can be regarded as consisting of many isotropic point sources. Thus the solution for a distributed source can be obtained by taking the integration of the solution for a simple unit point source over the geometry of the distributed source. The term $4\pi r^2$ represents the surface area of a sphere surrounding the source with radius r. For an isotropic source, each portion of the sphere will receive the same number of photons and the term $1/(4\pi r^2)$ becomes the degree of "spread out" (i.e., following "inverse square" attenuation) in the source strength as photons travel away from the source.

The buildup factor, $B(x)$, is used as the correction factor to consider the buildup of the intensity of radiation if the geometry of beam is broad and backscatter of the beam within the shield takes place to increase the intensity. The buildup factor also includes the contributions from low intensity, subsidiary radiation released locally in the intervening medium, such as annihilation gamma rays or bremsstrahlung photons due to the interactions between the source photons and the medium. Therefore, the buildup factor effectively works as a correction factor to adjust the result to include the contributions from the scattered and subsidiary radiation based on the calculation using the uncollided photons.

The most widely used form of the buildup factor is Taylor's formula, as given below. The parameters A, α_1, and α_2 are defined for different shield materials at different photon energy. Table 7.18 show the values of the parameters for water, ordinary concrete, aluminum, lead, and tungsten.

$$B(\mu r) = Ae^{-\alpha_1 \mu r} + (1 - A)e^{-\alpha_2 \mu r} \tag{7.34}$$

Example 7.4: Calculation of Required Thickness for Shielding (with Dose Limit)

Consider a 50-GBq point isotropic source emitting one 1-MeV photon per disintegration. The source is to be stored in a spherical lead cask (density 11.5

(continued)

Table 7.18 Parameters for Taylor form of the exposure build-up factor for point isotropic source for different shield media

Energy (MeV)	A	$-\alpha_1$	α_2
Water			
0.5	100.845	0.12687	−0.10925
1.0	19.601	0.09037	−0.02522
2.0	12.612	0.05320	0.01932
3.0	11.110	0.03550	0.03206
4.0	11.163	0.02543	0.03025
6.0	8.385	0.01820	0.04164
8.0	4.635	0.02633	0.07097
10.0	3.545	0.02991	0.08717
Ordinary concrete			
0.5	38.225	0.14824	−0.10579
1.0	25.507	0.07230	−0.01843
2.0	18.089	0.04250	0.00849
3.0	13.640	0.03200	0.02022
4.0	11.460	0.02600	0.02450
6.0	10.781	0.01520	0.02925
8.0	8.972	0.01300	0.02979
10.0	4.015	0.02880	0.06844
Aluminum			
0.5	38.911	0.10015	−0.06312
1.0	28.782	0.06820	−0.02973
2.0	16.981	0.04588	0.00271
3.0	10.583	0.04066	0.02514
4.0	7.526	0.03973	0.03860
6.0	5.713	0.03934	0.04347
8.0	4.716	0.03837	0.04431
10.0	3.999	0.03900	0.04130
Lead			
0.5	1.677	0.03084	0.30941
1.0	2.984	0.03503	0.13486
2.0	5.421	0.03482	0.04379
3.0	5.580	0.05422	0.00611
4.0	3.897	0.08468	−0.02383
6.0	0.926	0.17860	−0.04635
8.0	0.368	0.23691	−0.05684
10.0	0.311	0.24024	−0.02783
Tungsten			
0.5	2.655	0.01740	0.11340
1.0	3.234	0.04754	0.13058
2.0	3.504	0.06053	0.08862
3.0	4.722	0.06468	0.01404
4.0	5.520	0.08857	−0.04570
6.0	1.273	0.17257	−0.12178
8.0	0.664	0.20710	0.04692
10.0	0.509	0.21743	0.05025

Source: Trubey (1966)

Example 7.4 (continued)

g/cm^3). The distance between the point source and the inner cask surface is 50 cm. What minimum cask-wall thickness is required to assure that the dose rate to the person standing right next to the outer surface of the cask is limited to 2.3 μSv/h, neglecting the buildup of scattered photons?

Solution:

Assumption:

1. Energy loss of photon through the air is negligible due to close distance
2. The person is standing looking at the cask, and the surface area of the person is approximately 1 m^2 (assume 2 m tall and 0.5 m wide)
3. The person weighs 70 kg, and the radiation going through the person is completely absorbed (as a conservative assumption)

$I(x) = \frac{S}{4\pi r^2} \cdot B(x) \cdot e^{-\mu x}$ from (Eq. 5.12), and neglecting the buildup of scattered photons

Also, $1\ Gy = 1\ J/kg = 6.24 \times 10^9 [MeV/g]$

For $E = 1\ MeV$ arriving at the surface of the lead cask (assuming negligible loss of energy),

$S = 50 \times 10^{-9} [s^{-1}]$ (given)

$E_0 = 1\ [MeV]$ (given)

$\rho_{Pb} = 11.5 \left[\frac{g}{cm^3}\right]$ (given)

$\left(\frac{\mu_{en}}{\rho}\right)_{Pb} = 0.0379 \left[\frac{cm^2}{g}\right]$ (Table 3.10, Sect. 3.2.3)

$\rightarrow \mu_{Pb} = 0.43585\ [cm^{-1}]$

Neglecting buildup of scattered photons

$\rightarrow I_u(x) = \frac{S}{4\pi r^2} e^{-\mu_{en} x}$ since we are interested in how much of "photon energy" is transmitted through the cask thickness to calculate the dose rate outside the cask

$\rightarrow$ *Energy absorbed by person* $= \frac{50 \times 10^9\ \left[\frac{MeV}{s}\right]}{4\pi(50)^2\ [cm^2]} e \qquad \cdot 10000[cm^2] =$ $\left(1.592 \times 10^{10}\right) \cdot e^{-0.43585x}\ \left[\frac{MeV}{s}\right]$

$$\textit{The given dose limit} = 2.3 \left[\frac{\mu Sv}{h}\right] = 2.3 \times 10^{-6} \left[\frac{Gy}{h}\right] = 4 \left[\frac{MeV}{g - s}\right]$$

$\rightarrow$ *Dose limit* $= 4 \left[\frac{MeV}{g-s}\right] \cdot 70000[g] = 2.8 \times 10^5\ \left[\frac{MeV}{s}\right]$

Setting above two equal,

$(1.592 \times 10^{10}) \cdot e^{-0.43585x} = 2.8 \times 10^5$

$\boldsymbol{x = 25.1cm}$

(continued)

Example 7.4 (continued)

So with very conservative assumptions, the required thickness would be around 25cm.

Note: People are normally exposed to natural radiation of 1 ~ 5 mSv/y, with some areas receiving over 10 mSv/y. an average individual dose is approximately 2.5 mSv/y, corresponding to about 2.85 × *10*$^{-4}$ *mSv/h in the world.*

For a mono-directional gamma ray beam, the exposure rate through a shield thickness *b* can be calculated as

$$\dot{X} = 0.0659\phi_b E_0 \left(\frac{\mu_a}{\rho}\right)_{air} = 0.0659\frac{S}{4\pi R^2}\cdot e^{-\mu R}\cdot B_p(\mu R)\cdot E_0\left(\frac{\mu_a}{\rho}\right)_{air} \tag{7.35}$$

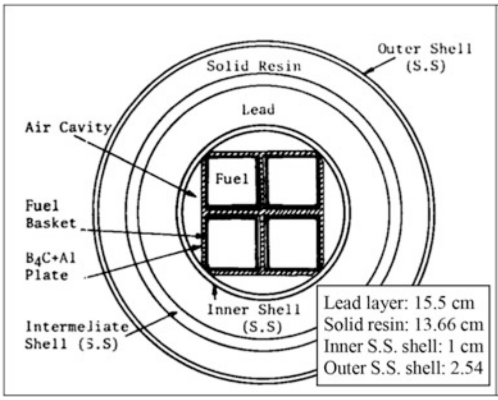
where $\dot{X}$ is exposure rate at the surface of the shielding material [mR/hr], ϕ_b is the intensity (or flux) with buildup in [γ-rays/cm^2-s], E_0 is incident energy [MeV], $B_p(\mu R)$ is the buildup factor, and $\left(\frac{\mu_a}{\rho}\right)_{air}$ is mass absorption coefficient of air for corresponding gamma ray energy [cm^2/g].

7.2.2 Analysis for Neutron Shielding

The attenuation of neutrons in a shield can be described in a similar way, i.e., by using the point kernel technique, as in the case of gamma rays. However, neutron interactions with matter are much more complex as different forms of scattering (i.e., elastic and inelastic) as well as absorption take place while the cross section for such interactions varies widely with energy. Also, complete analysis of neutron shielding requires a knowledge of the spatial distributions of neutrons of all energies. Such distributions can be provided by the so-called removal-diffusion method but using simple analytical approaches is not possible in neutron shielding analysis. A combination of a removal-attenuation calculation (for fast neutrons) and a multigroup diffusion method (for the description of slowing down of neutrons and the behaviors of thermal neutrons) involving the use of computer codes are typically exercised in neutron shielding analysis.

7.2.3 An Example of Spent Fuel Shielding

The material for neutron shielding for spent fuel is typically solid organics in the form of plastics or resins. For gamma ray shielding, most common material used is



Summary of dose rates under the normal and accident conditions with the shielding (mSv/hr)

	Normal		Accident
	Surface	At 1 m	At 1 m
Primary gamma	0.176	0.065	0.288
Secondary gamma	0.037	0.012	0.001
Neutron	0.048	0.017	1.519
Total	0.261	0.094	18.08

Fig. 7.12 Example of Shielding for PWR Spent Fuel (4 assemblies) Shipping Cask. (Source: Kang et al. 1988)

lead or depleted uranium in combination with stainless steel. Use of stainless steel is to allow inelastic scattering of fast neutrons for slowing down to thermal or epithermal neutrons. Examples of shielding materials in the currently used spent fuels casks are shown in Table 7.27.

A detailed example of shield for spent fuels against gamma rays and neutrons is shown Fig. 7.12 (Kang et al. 1988). The shield is for a shipping cask to transport four assemblies of PWR spent fuel. A combination of lead (as gamma shielding) and solid resins (as neutron shielding) was used. Stainless steel shell layers were also used as gamma and neutron shields. As shown, the design employs gamma ray shielding first followed by neutron shielding. This is because gamma shield can serve the purpose of slowing down fast neutrons thus increase the effectiveness of neutron shielding. The thickness of the lead and solid resin was 15.5 and 13.66 cm, respectively. The stainless steel shells were 1 cm thick for the inner and outer shell and 2.54 cm for the intermediate shell. Boron carbide combined with aluminum was used for criticality control. Based on the use of these shields, radiation dose rate at the surface and 1m from the surface of the cask was found to be 0.261 mSv/h. (26.1 mrem/h) and 0.094 mSv/h. (9.4 mrem/h), respectively. The total weight of the cask system with loaded spent fuel was 38 tons in this example case.

7.2.4 Major Radionuclides of Concern in Spent Fuel Shielding

In this section, our interest is to understand the relative contributions of individual radionuclides to radiation dose from a transport/storage cask when different shielding materials are utilized. Three different casks considered include a carbon steel transport cask, a lead transfer cask, and a concrete storage cask. Table 7.19 shows the summary of the findings in terms of the ranks of nuclides and their

Table 7.19 Major gamma and neutron dose contributors from a PWR and BWR spent fuel at different burnup and cooling time with different shield materials [Shielding rankings for dominant radionuclides in PWR fuel]

	20 GWd/t 3.0 wt%		70 GWd/t 5.0 wt%		20 GWd/t 3.0 wt%		70 GWd/t 5.0 wt%		20 GWd/t 3.0 wt%		70 GWd/t 5.0 wt%	
Nuclides[a]	Percent[b]	Rank	Percent	Rank	Percent	Rank	Percent	Rank	Percent	Rank	Percent	Rank
PWR	**Carbon steel transport cask**				**Concrete storage cask**				**Lead transfer cask**			
5-year cooling												
Ba-137m	3.0	6	2.3	7	9.2	4	8.9	4	0.2	8	0.1	8
Cm-244	1.8	7	36.1	1	0.3	8	8.3	5	1.9	6	36.6	1
Co-60	49.8	1	26.4	2	50.3	1	34.6	1	56.4	1	29.9	2
Cs-134	10.7	3	15.8	3	14.0	2	26.7	2	10.2	3	15.0	3
Eu-154	4.6	5	7.0	4	5.5	6	10.9	3	4.5	5	6.9	4
Pr-144	19.6	2	5.0	5	12.4	3	4.2	7	17.3	2	4.4	6
Rh-106	9.0	4	5.0	6	6.9	5	5.0	6	8.2	4	4.5	5
Y-90	1.0	8	0.6	8	1.1	7	0.9	8	1.0	7	0.6	7
100-year cooling												
Am-241	18.6	2	4.3	4	2.2	3	1.3	5	2.9	1	5.4	3
Ba-137m	42.0	1	12.4	3	85.6	1	66.9	1	3.6	6	0.7	8
Cm-244	6.0	5	46.6	1	0.7	5	14.8	2	9.5	4	52.3	1
Cm-246	0.3	9	23.7	2	0.0	9	7.5	3	0.5	9	26.6	2
Pu-238	3.2	6	4.2	5	0.4	6	1.3	6	5.6	5	5.2	4
Pu-239	1.8	8	0.2	9	0.2	8	0.1	9	3.2	8	0.3	9
Pu-240	13.3	3	2.9	7	1.6	4	0.9	7	21.5	2	3.3	6
Pu-242	2.2	7	1.7	8	0.3	7	0.5	8	3.5	7	1.9	7
Y-90	11.9	4	3.0	6	8.7	2	5.8	4	18.9	3	3.4	5

(continued)

Table 7.19 (continued)

	20 GWd/t 3.0 wt%		70 GWd/t 5.0 wt%		20 GWd/t 3.0 wt%		70 GWd/t 5.0 wt%		20 GWd/t 3.0 wt%		70 GWd/t 5.0 wt%	
Nuclides[a]	Percent[b]	Rank	Percent	Rank	Percent	Rank	Percent	Rank	Percent	Rank	Percent	Rank
BWR	**Carbon steel transport cask**				**Concrete storage cask**				**Lead transfer cask**			
5-year cooling												
Ba-137m	4.5	6	2.7	7	13.9	4	11.6	4	0.3	8	0.2	9
Cf-252	0.0	9	1.3	8	0.0	9	0.3	9	0.0	9	1.4	7
Cm-244	3.0	7	43.9	1	0.5	8	10.9	5	3.3	6	45.8	1
Co-60	24.5	2	10.7	3	24.9	1	15.1	2	29.7	1	12.5	3
Cs-134	16.2	3	19.1	2	21.3	2	34.9	1	16.5	3	18.7	2
Eu-154	7.0	5	8.1	4	8.4	6	13.5	3	7.4	5	8.2	4
Pr-144	29.3	1	6.1	5	18.7	3	5.4	7	27.6	2	5.5	6
Rh-106	13.4	4	6.0	6	10.3	5	6.4	6	13.0	4	5.6	5
Y-90	1.5	8	0.8	9	1.6	7	1.2	8	1.6	7	0.8	8
100-year cooling												
Am-241	17.8	2	3.8	5	2.1	3	1.2	6	31.5	1	4.8	4
Ba-137m	42.0	1	12.2	3	85.6	1	66.7	1	3.6	6	0.7	8
Cm-244	6.6	5	46.2	1	0.8	5	14.8	2	10.4	4	51.7	1
Cm-246	0.4	9	25.0	2	0.0	9	8.1	3	0.6	9	28.0	2
Pu-238	3.4	6	4.0	4	0.4	6	1.3	5	5.8	5	4.9	3
Pu-239	1.8	8	0.2	9	0.2	8	0.1	9	3.2	8	0.2	9
Pu-240	13.4	3	3.0	6	1.6	4	0.9	7	21.6	2	3.4	5
Pu-242	2.2	7	1.6	8	0.3	7	0.5	8	3.4	7	1.8	7
Y-90	11.9	4	3.0	7	8.7	2	5.8	4	19.0	3	3.3	6

Source: NRC (2000)

[a] Dominant nuclides that contribute more than 1% to the total dose rate

[b] Indicates percentage contribution of the nuclide to the total dose rate for the cask configuration

contributions to the estimated dose. The spent fuel as the source of radiation included both low and extended-burnup LWR fuels with two different cooling periods (5 and 100 years). The results are shown in Table 7.19.

In general, the most important nuclide for a short cooled (at 5 years) spent fuel was ^{60}Co (a gamma source with the emission of 1.17 MeV and 1.33 MeV photons, $t_{1/2} = 5.27$ years). The most important neutron source was ^{244}Cm with a half-life of 18.1 years. The importance of these two nuclides didn't change much between low and high burnup spent fuels although the neutron dose from ^{244}Cm can be more significant than the gamma dose from ^{60}Co in high burnup spent fuel. Other important nuclides include ^{144}Pr, ^{134}Cs, ^{106}Rh, ^{134m}Ba, ^{90}Y, and ^{154}Eu. For a longer cooled spent fuel (at 100 years), various actinides such as ^{244}Cm, ^{246}Cm, ^{241}Am, ^{238}Pu, ^{239}Pu, ^{240}Pu, and ^{242}Pu, became important. For gamma radiation, only ^{137m}Ba and ^{90}Y, the decay products of ^{137}Cs and ^{90}Sr, respectively, remained important.

With the employment of different shielding materials, the major contributors to the total dose did not change although the relative ranking changed with different burnups and cooling times. With the use of concrete, the importance of neutron source actinides got slightly reduced, perhaps due to better neutron shielding capability of concrete over steel or lead.

7.3 Criticality Control in Spent Fuel Management

Discharge of spent fuel from a nuclear reactor occurs when the fuel does not contain enough fissile materials to sustain the fission reaction for reactor operation. However, fissile materials are still present in spent fuel. Thus care needs to be exercised to prevent spent fuel from reaching the condition of criticality during all phases of spent fuel management.

Criticality in a given spent fuel system can be calculated by using the following equation.

$$k_{eff} = \frac{\boldsymbol{N_F}}{\boldsymbol{N_A} + \boldsymbol{N_L}} \tag{7.36}$$

where, $\boldsymbol{N_F}$ represents the number of neutrons that result from fission, and $\boldsymbol{N_A}$ represents the number of neutrons that are absorbed and vanish, and $\boldsymbol{N_L}$ that represents the number of neutrons lost as they leak from the system.

As mentioned in Sect. 3.3.5, criticality control is based on controlling the factors that influence the neutron balance in a fissile system. Neutrons produced by fission in the spent fuel system collide with other atomic particles in the system and are either absorbed or scattered (i.e. they change direction) or move out of the system (i.e., leaked out). Whether a neutron is absorbed, scatted, or leaked out depends on the energy of the neutron as the probability (i.e., cross section) of different interactions depends on the energy of neutrons.

Key nuclides of importance in criticality control of spent fuel are ^{238}U, ^{239}Pu and ^{235}U, regardless of the burnup. For other nuclides, relative importance among

Table 7.20 Important radionuclides in criticality control for LWR spent fuel

	Low burnup fuel (20 GWD/t, 3.0 w/o initial enrichment)		High burnup fuel (70 GWD/t, 5.0 w/o initial enrichment)	
	Activation/fission products	Actinides	Activation/fission products	Actinides
High importance		^{238}U ^{239}Pu ^{235}U		^{238}U ^{239}Pu ^{235}U
Medium importance	^{149}Sm	^{240}Pu	^{103}Rh ^{133}Cs ^{143}Nd ^{149}Sm ^{155}Gd	^{236}U ^{237}Np ^{240}Pu ^{241}Pu ^{242}Pu ^{242}Pu ^{241}Am
Low importance	^{95}Mo ^{99}Tc ^{103}Rh ^{131}Xe ^{133}Cs ^{143}Nd ^{145}Nd ^{147}Sm ^{150}Sm ^{151}Sm ^{152}Sm ^{153}Eu ^{155}Gd	^{236}Pu ^{237}Np ^{238}Pu ^{241}Pu ^{241}Am	^{95}Mo ^{99}Tc ^{101}Ru ^{105}Pd ^{108}Pd ^{109}Ag ^{131}Xe ^{141}Pr ^{145}Nd ^{147}Sm ^{150}Sm ^{151}Sm ^{152}Sm ^{153}Eu	^{238}Pu

Source: NRC (2000)

actinides and fission products in low and high burnup spent fuel vary as summarized in Table 7.20.

Criticality control of spent fuel is implemented by requiring spent fuel to be subcritical all the time typically requiring the k_{eff} of spent fuel to be less than 0.95. Thus, the number of neutrons produced in spent fuel (from fission in the remaining fissile or fissionable material) should be less than the number of neutrons absorbed or leaked from spent fuel by more than 5%. Analysis of a spent fuel system for criticality safety requires the use of computer codes with the description of the geometry of the system and the materials used in the system.

The k_{eff} value of the fresh fuel is much less than 0.95 (typically below 0.85 in the transportation cask). Then, at the time of discharge, the k_{eff} values of the fuel may drop below 0.7 (depending upon the burnup). The reduction is due to the depletion of fissile nuclides while producing neutron-absorbing nuclides (e.g., non-fissile actinides and fission products) and new fissile actinides (e.g., ^{239}Pu and ^{241}Pu). k_{eff} further decreases as it continues to cool during storage. It can be said that under the normal operation conditions, there exists enough safety margin for criticality control in spent fuel management. However, in the case of spent fuel storage in a densely packed system, criticality could become an issue. In this case, to ensure presence of safety margin for criticality control, design considerations are made such that absorption and leakage of neutrons in a spent fuel system (e.g., in the storage pool) always exceed the level of neutron production. Under the hypothetical situations of the most undesirable configuration (i.e., accidents with favorable moderation and reflection characteristics for neutron production), the k_{eff} of the system may exceed 0.95 but should be maintained to be less 1.

Most important aspect in criticality control of spent fuel is the use of neutron poisons. Whenever any concern with criticality control of spent fuels arises, such as from high density storage using re-racking in a storage pool, neutron poisons (e.g., boron) are used. Boron can be in the form of boric acid (H_3BO_3) as aqueous solution or solid structure as boron carbide (B_4C) within a metal matrix or polymer matrix.

7.4 Storage of Spent Fuel

The storage of spent fuel is initially at a water pool inside the fuel building next to the nuclear reactor building for the purpose of cooling. Spent fuel discharged from the reactor is moved to the fuel building through a transfer tube as shown in Fig. 7.15. If the storage space in the reactor pool of the fuel building runs out, use of separate structure/space is needed.

In a typical light water reactor, about a quarter to a third of the total fuel load is removed from the core through refueling every 12 to 24 months. Thus certain portion of the loaded fuels is always replaced with fresh fuels and becomes spent fuel. These spent fuels are stored on site until they are shipped away from the reactor for storage in other facility, reprocessing, or permanent disposal.

During the early phase of nuclear reactor development in countries like the U.S., France, and the U.K., the spent fuels from nuclear power plants were either reprocessed or expected to be reprocessed within a few months after the discharge. Therefore, the spent fuel pools in the reactors were built with limited storage capacity (e.g., to hold two or three core). In the case of the U.S., reprocessing was exercised only from 1966 till 1972. During that period, reprocessing of commercial spent fuel was done at the West Valley Fuel Processing Plant which stopped the operation in 1972 due to concerns over environmental release of radioactivity and the difficulty in necessary back-fitting of the facility. Then came the executive order by President Carter in 1977 which banned spent fuel reprocessing. That ban was the U.S. policy decision against the 1974 Indian nuclear explosions which was based on taking advantage of the civilian reprocessing capability. Reprocessing of spent fuel from commercial nuclear power plant discontinued in the U.S. since then while reprocessing continued in Europe. From that point on, the electric utilities in the U.S. had to keep the spent fuel at their reactor sites until a centralized interim storage facility or the final disposal facility becomes available. Thus the originally planned short term storage of spent fuel at a reactor site turned into an extended storage facility until the option for permanent disposal of spent fuel becomes available. Nuclear reactors built after these periods still followed the similar design of the limited spent fuel storage capacity. As of 2020, there is no operating permanent disposal facility for spent fuel in the world.

Due to lack of final disposal capability, extended storage of spent fuel becomes a necessity. The pros and cons of extending the storage period is summarized in Table 7.21.

Spent fuel storage can be classified into two categories: water pool (wet) and dry storage. Most of the nuclear industry's spent fuels are stored in water pools at the reactor sites (AR). Spent fuel storage facility can also be built away from the reactor site (AFR) to meet the additional storage space need as a wet or dry facility. Currently, there are three general options of spent fuel storage including AR storage, AR storage extension, and AFR storage as shown in Fig. 7.13.

Table 7.21 Pros and cons of extended storage of spent fuel

Comparisons of pros and cons of extended storage of spent fuel	
Pros	• Decay of radionuclides and reduction in decay heat • Earn time before decision on national strategy of spent fuel management • Give time to develop information needed to support new and efficient regulation • Leave the possibility of reusing fissile materials
Cons	• Increase in the inventory of actinides • Major hazard still left on the surface at the storage facility (possibility of accident or misuse or degradation of the storage system remains until final disposal is implemented) • Government's responsibility in taking title of spent fuel is not exercised • At the end of storage period, spent fuels need to be packaged and shipped for final disposal (unless multi-purpose canisters for shipping and disposal are implemented)

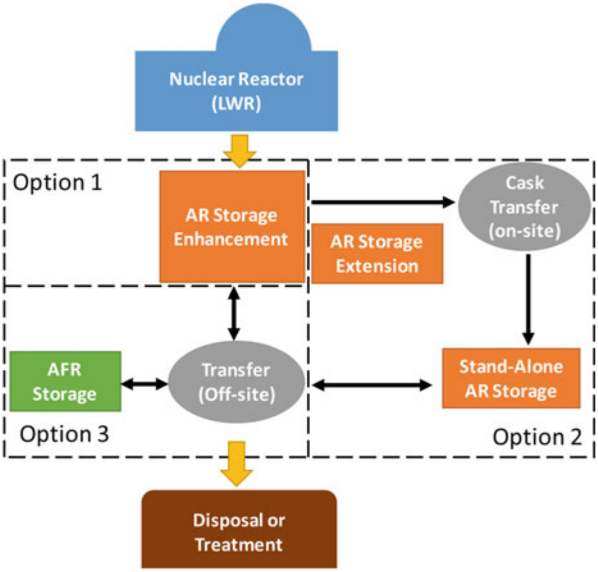


Fig. 7.13 Three options for the storage of spent fuel before permanent disposal or treatment (arrows are possible transport operations) (AR: At-reactor; AFR: Away-from-reactor)

In both wet and dry storage technologies, spent fuel must be appropriately protected through implementation of shielding, cooling, and criticality control. The followings are typical design requirements for a spent fuel storage facility (IAEA 1999):

1. Fuel cladding integrity should be maintained.
2. Fuel degradation during storage should be prevented.
3. Subcriticality of spent fuel is to be maintained under normal and accidental conditions.
4. Adequate shielding of spent fuel should be provided to protect plant operators, the public and the environment from receiving radiation doses in excess of regulatory limits.
5. Environmental protection should be assured by minimizing the release of radioisotopes.
6. Easy retrievability must be provided requiring continued care, maintenance, and monitoring of the storage facility.

As many of nuclear power plants in the world are built with limited capacity for spent fuel storage, utilities have come up with strategies to deal with the shortage in storage capacity. These strategies include: (1) increasing energy extraction per unit mass of spent fuel through efficient in-core fuel management and use of high-burnups; (2) expanding the physical size of the storage pool; (3) shipping the spent fuel out to a storage pool at another reactor; (4) increasing the efficiency of existing pool capacity by implementing closer spacings between spent fuels through reracking or fuel consolidation, and; (5) building a new and independent storage facility on-site for interim storage.

In-core fuel management for enhancing energy extraction efficiency involves special fuel shuffling schemes. The shuffling results in more uniform burnup and temperature distributions across the entire core, minimizing the presence of local hot spots and maximizing the energy production during the lifetime of the fuel. Because of the even core power distributions, more power can be extracted from the fuel. Therefore, for the same amount of energy production, the amount of spent fuel to be discharged is reduced.

Extending the burnup of fuel to increase fuel utilization has been widely exercised in the nuclear industry. However, as burnup increases, production of fission products increases resulting in higher radiation dose and decay heat. Handling of such high burnup fuels requires additional care in spent fuel transportation and disposal.

Expansion of pool capacity by building another pool or expanding an existing one has also been done. However, this is an expensive option. Some utilities have moved or plan to move spent fuels between reactor pools at different sites to delay the need for additional storage space. Yet, this option requires availability of additional storage space at a different plant site within the utility company.

Increasing the efficiency of spent fuel storage through reracking for closer spacings is the most widely implemented option. Spent fuel was originally stored on 53-cm centers in the pools but can be changed to closer spacing, such as 23-cm centers, with the use of so-called high density racks by incorporating additional

Table 7.22 Experiences of fuel rod consolidation in the U.S.

Reactor	Date	Consolidation ratio	Companies
Oconee	1982	2:1 (for NFBC[a] 6:1)	Duke Energy, Westinghouse Electric
Maine Yankee	1981–1984	1.6:1	Maine Yankee, Proto-Power Corp.
West Valley	1985–1986	1.8:1	Rochester Gas & Electric, NAC Int'l, EPRI, NYSERDA
Battelle Columbus	1986	1.85:1 ~ 2:1	Rochester Gas & Electric, US Tools & Die, EPRI, NYSERDA
Millstone 2	1983–1988; 1991	2:1	Northeast Utilities, Baltimore Gas & Electric, CE, EPRI
Prairie Island	1987	2:1 (for NFBC 6:1)	Northern States Power, Westinghouse

[a]NFBC: Non-fuel bearing components (spacer, end-fitting, guide tube)

neutron poisons. Such high density racks still meet the criticality requirement, seismic requirement, and the additional floor loadings. This can be combined with high-burnup fuels that reduce the amount of fissile material and neutron absorbers. To incorporate additional neutron poisons, a neutron absorber can be introduced to the rack by sandwiching boron containing B_4C between stainless steel plates to make up the walls of each storage cell. Many reactor basins were designed originally to hold up to 4 to 5 annual spent fuel discharges. By reracking, utilities can increase the capacity up to 10 additional annual discharges.

An alternative way, called fuel consolidation, was also considered. Under the option, the fuel assembly is dismantled and the spent fuel rods are rearranged into a close-packed geometry in a storage canister. This approach notes that fuel rods occupy about 40% of the space in typical LWR fuel assemblies. By removing the mechanical components of the assembly (the so called non-fuel bearing components (NFBC) including spacer, end-fitting, guide tube) are compacted, an increase by a factor of 1.7 to 2.0 in the volumetric efficiency of fuel storage can be achieved. A successful demonstration was performed by Duke Power Company at its Oconee Station in the U.S. in 1982 by including four PWR assemblies in two storage canisters. Activities of spent fuel rod consolidation in the U.S. are summarized in Table 7.22. Although fuel consolidation has not been licensed by the NRC, it remains as option for dry storage to increase the storage capacity.

Building a separate storage facility, called "Independent spent fuel storage installations (ISFSI)" has been widely exercised by the U.S. utilities in recent years out of necessity for interim storage of spent nuclear fuel while waiting for a centralized storage or geological repository. ISFSI typically uses dry storage concept.

Example 7.5: Dry Storage vs Wet Storage

Assume that we have spent fuels at following two burnup levels to be emplaced into a storage cask:

(1) 25,000 MWD/MTIHM; (2) 50,000 MWD/MTIHM

The cask is 2.3 m in diameter (D) and 4.6 m long (H) and can store 24 PWR assemblies (~ 10 MTIHM). To meet the following temperature limits for spent fuel, determine the cooling period required before the fuel can be placed in the cask for either dry or wet storage.

Given:

The maximum surface temp in air:	250 °C
The maximum surface temp in water:	80 °C
The ambient air/water temp:	18 °C

Use (Eq. 7.27) for the calculation of decay heat from the spent fuel:

$$Q(t) = C_1 \cdot e^{(C_2 + C_3 t)^{-1}} \cdot \left(\frac{B}{33000}\right)\left[\frac{watts}{MTU}\right] \text{ where } C_1 = 550, C_2 = 0.223, C_3 = 0.177$$

Also, use natural convection heat transfer coefficient: Air 1.8 W/m^2-°C; water 60 W/ m^2-°C for the equation,

$$Q = hA\left(T_{surface} - T_{ambient}\right) \tag{7.37}$$

where h is convective heat transfer coefficient.

Solution:

$h_{air} = 1.8\ W/\,\mathrm{m}^{2-}\ ^\circ\mathrm{C}$, $h_{water} = 60\ W/\,\mathrm{m}^{2-}\ ^\circ\mathrm{C}$

$T_{max,\ air} = 250\ ^\circ\mathrm{C}$, $T_{max,\ water} = 80\ ^\circ\mathrm{C}$, $T_{ambient\ air/water} = 18\ ^\circ\mathrm{C}$

The allowable heat (Q) in the storage condition can be represented by, $Q = hA(T_{surface} - T_{ambient})$.

If we assume that natural convection occurs mostly along the length of the container, the surface area for heat transfer is,

$$A = \pi DH = \pi(2.3m)(4.6m) = 33.24m^2$$

For dry storage, $T_{surface} \le 250^\circ\ \mathrm{C}$

$\rightarrow Q = hA \quad \left(T_{surface} - T_{ambient}\right) = 1.8\frac{\mathrm{W}}{\mathrm{m}^2 - ^\circ C} \cdot 33.24m^2 \cdot (250 - 18)^\circ\ \mathrm{C} = 13,880\ \ \mathrm{W}$

(continued)

Example 7.5 (continued)

For wet storage, $T_{surface} \leq 80^{\circ}$ C

$\rightarrow Q = hA\left(T_{surface} - T_{ambient}\right) = 60\,\frac{\mathrm{W}}{\mathrm{m}^2-{}^{\circ}C} \cdot 33.24m^2 \cdot (80 - 18)^{\circ}$ C $=$ $123,646$ W

(1) For 25,000 MWD/MT burnup fuel in air:

$$Q(t) = 550 \cdot e^{(0.223+0.177t)^{-1}} \cdot \left(\frac{25000}{33000}\right)\frac{W}{MTU} \cdot 10\,MTIHM$$

$\left(\text{since 24 PWR assemblies, }\tilde{1}0\text{MTIHM}\right) = 416.667e^{(0.223+0.177t)^{-1}} \cdot 10 =$

$13,880\ \ W$

$$e^{(0.223+0.177t)^{-1}} = 3.33 \rightarrow (0.223 + 0.177t)^{-1} = \ln(3.33) \rightarrow 0.223 + 0.177t = \frac{1}{\ln(3.33)}$$

$\rightarrow t = 3.44$ *years*

The fuel needs to be cooled for 3.44 years before dry storage.

For 25,000 MWD/MT burnup fuel in water:

$$Q(t) = 550 \cdot e^{(0.223+0.177t)^{-1}} \cdot \left(\frac{25,000}{33,000}\right)\frac{W}{MTU} \cdot 10\,MTIHM$$

$\left(\text{since 24 PWR assemblies, }\tilde{1}0\text{ MTIHM}\right) \quad = 416.667e^{(0.223+0.177t)^{-1}} \cdot 10 =$

$123,646\ \ W$

Similarly, by solving for *t* for wet storage, we get

$\rightarrow t = 0.406$ *years*

The equation is not exactly accurate for t < 1 y. however, from the result for wet storage, we can reasonably see that about half a year cooling is sufficient for wet storage while 3.44 years cooling is necessary for dry storage for the same spent fuel.

(2) For 50,000 MWD/MT burnup fuel in air, solving in a similar manner as above examples:

$$550 \cdot e^{(0.223+0.177t)^{-1}} \cdot \left(\frac{55000}{33000}\right) \cdot 10 = 13880$$

$$\rightarrow t = 12.37\ years$$

(continued)

Example 7.5 (continued)

The fuel needs to be cooled for ~13 years for dry storage. This is longer than what is needed (3.4 years) for lower burnup fuel.

For 50,000 MWD/MT burnup fuel in water:

$$550 \cdot e^{(0.223+0.177t)^{-1}} \cdot \left(\frac{55000}{33000}\right) \cdot 10 = 123646$$

$$\rightarrow t = 0.91\ years$$

For wet cooling, 0.91 years of cooling is needed while 13 years of cooling is needed for dry storage for the same spent fuel.

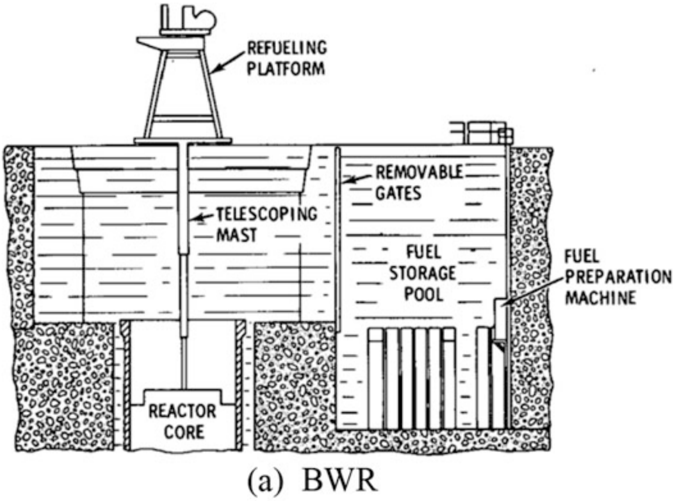
7.4.1 Wet Storage

Water is a convenient, inexpensive storage medium and provides excellent cooling capacity through natural circulation. All nuclear power reactors have at-reactor water-filled pool for the storage of spent fuel. Figure 7.14 shows the location of the storage pool at a BWR and PWR. Most LWR fuel storage pools are of similar design.

As depicted in the figure, discharged nuclear fuel from nuclear reactor is moved to the storage pool by using automated handling system while being always remained underwater. Water also provides effective shielding against neutrons and allows clear visibility for inspection and handling. The pools are rectangular in horizontal cross-section and 12–13 m deep. The pool is typically surrounded by steel lined concrete wall (more than 1 m thick) as a leak-tight structure. Any leak must be detected and contained in the plant system. The storage zone in the pool is about 4 m high, located near the bottom of the pool. This leaves more than 7 m of water above the spent fuel to provide shielding.

In the storage zone, the spent fuel assemblies are stored within the storage racks at the bottom of the pool in a lattice array to provide spacing for coolant flow. The storage racks are made of aluminum steel and are to keep the fuel in controlled positions for physical protection and for ease of tracking and rearrangement. The spacing (or 'pitch') of the spent fuel assemblies in the storage racks is adjusted for criticality control. Boric acid is also added to the pool water as neutron poisons for criticality control. Typically a boron concentration of 2000 ppm is maintained. The spacing (or 'pitch') of the spent fuel assemblies in the storage racks is determined to keep k_{eff} below 0.95 even if the pool is filled with unborated demineralized water.

The cooling system is provided to adequately remove the heat likely to be generated by the maximum inventory of spent fuel anticipated during operation. The water temperature in the pool is maintained normally below 50 °C (120 °F) while the system design requirement is to keep the pool temperature below 71 °C



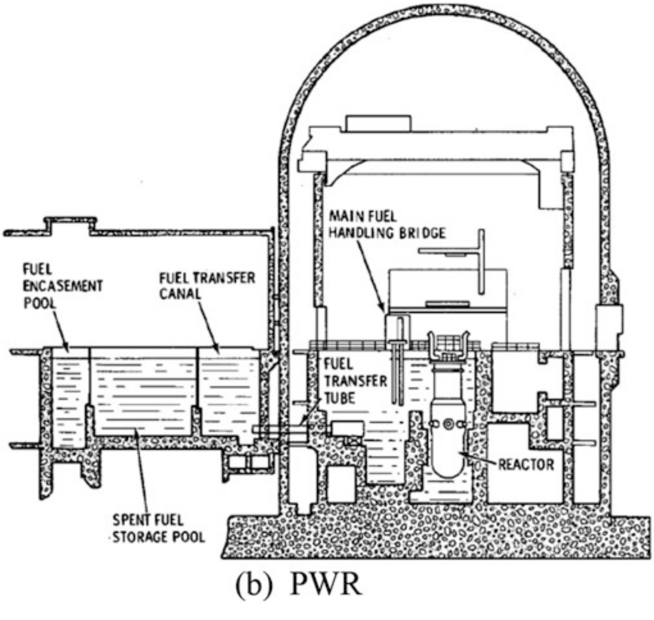


Fig. 7.14 Location of spent fuel pool in a LWR. (Source: IAEA 1982)

(160 °F). The cooling system for a wet storage pool has two separate trains for redundancy, with each train consisting of a pump, a heat exchanger, valves, piping, and instrumentation.

Figure 7.15 shows the location of spent fuel pool inside a fuel handling building next to the containment building. The figure also shows the fuel transfer canal, next

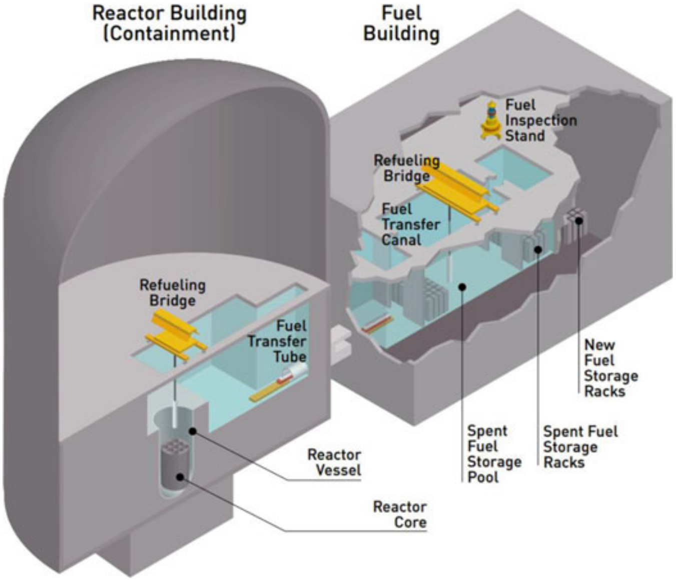


Fig. 7.15 Spent fuel pool, nuclear power plant containment, and fuel handling building in a PWR. (Source: NRC 2018)

to the spent fuel pool, loading pool for putting spent-fuel in shipping cask, and wash-down pit. The fuel transfer canal is connected to the refueling canal by the fuel transfer tube, and the new fuel assemblies are transferred from the new fuel storage facility in the fuel building to the fuel transfer canal (which connects the reactor building and the fuel building) by the fuel handling machine. The wash-down pit is used for decontamination of shipping casks after cask loading.

Radioactive contamination in the pool can take place during transfer of spent fuel due to the mixing of reactor coolant with the storage pool water. Release of fission products from defective fuel rods or the activated corrosion products deposited on the fuel assembly surfaces as a “crud” layer during operation is the source of contamination. Release of radiation from spent fuel can also lead to hydrogen gas production in the pool from radiolysis of water. Accumulation of hydrogen gas in the air in the spent fuel building should be continually monitored for possible treatment to minimize the risk of hydrogen explosion.

Water quality in the pool is controlled to maintain optical clarity of water and to minimize fuel degradation and radiation dose in the building. The fuel pool cleanup system using a mixed-bed demineralizer is used for the purpose. The radiation dose rate to workers in the spent fuel building should be less than 0.025 mSv/h (2.5 mrem/h) during normal operations and 0.1 mSv/h (10 mrem/h) during fuel handling operations.

Potential accidents in the spent fuel pool must also be taken into account in its design. These potential accidents include seismic events causing deformation of the racks, handling equipment failures in which fuel assemblies are dropped onto the racks, or any event that leads to the loss of cooling capability. One of the earliest studies on the issues was the Reactor Safety Study in 1975 which concluded that the risk from any accidents in spent fuel pool was very low. The U.S. Nuclear Regulatory Commission (NRC) has published a series of reports on the safety of spent fuel storage and indicated that the probability of a loss-of-pool coolant event that would lead to zirconium cladding fire resulting in the release of radioactive materials is very small, requiring no specific action.

The US NRC also published a report analyzing spent fuel pool accident risk during plant decommissioning (NRC 2001). The report indicated that large earthquakes and drops of fuel casks from an overhead crane during transfer operations were two event initiators leading to a loss-of-pool-coolant accident. The report also noted that operators have about 100 hours to act before the fuel was uncovered sufficiently to allow the fuel rods to start a fire. Still, the overall conclusion of the report was that the risk is low as the probability of such events was very low. The US NRC also published a report discussing the beyond design-basis accidents (BDA) in spent fuel pool (NRC 2003). The report noted that in a dense-packed pool, a zirconium cladding fire due to lack of coolant is a possibility. However, the report also noted the probability of such events to be insignificant (no greater than that of reactor core damage accidents from seismic events in the scale of beyond the design basis (safe shutdown) earthquake).

Recently, after the Fukushima accidents in Japan, risk from a potential attack by terrorism on spent fuel pool received attention. Whether such attack would lead to propagating zirconium cladding fire and subsequently the release of large amounts of radioactive has not been clearly elaborated. In any case, nuclear power plants must be protected with high assurance against specific threats that are relevant to spent fuel pool safety.

7.4.2 Dry Storage

Storage of spent fuel in a dry environment is being widely pursued. Dry storage is appropriate for spent fuels with low heat loads, e.g., for spent fuels cooled at least 1 year in the spent fuel pool. Given the absence of water for cooling, dry storage is not appropriate for spent fuel with high heat loads such as freshly discharged spent fuel. In dry storage, the spent fuel is stored in a shielded container outside the reactor containment building. In dry storage, potential concerns with the use of water such as corrosion in water and stress on the structures from hydrostatic pressure are eliminated. In case dry storage of high burnup spent fuels is necessary, the spent fuels need to be cooled for longer periods before dry storage is implemented.

7.4.2.1 Options in Dry Storage

Dry storage of spent fuel can be implemented in a variety of ways as long as appropriate safety requirements, such as shielding, cooling, structural safety, are met. The storage can be in casks, small modular structure, or a large building. It can be a surface facility or an underground facility. Therefore, dry storage offers flexibility and can be tailored to meet the needs of a specific site. In the case of using casks or small modular structure, expansion of storage capacity can be readily achieved in very small increments. The heavily shielded containers used in most dry technologies provide a massive physical barrier against accidents (e.g., airplane crashes) or sabotage and would limit the effects of such events to a lower significance. While continuing surveillance and maintenance of the facilities are needed, maintenance requirement for dry storage is lower compared to that of wet storage.

The concepts proposed for dry storage of spent fuel are listed below. In any of these arrangements, spent fuel elements are typically sealed in steal canisters prior to their emplacement in the storage system:

- Metal casks – Large metals casks can be designed for the purpose of storage or as dual-purpose containers for transportation as well. These casks are designed to meet shielding, containment, and heat dissipation requirements. The casks are filled with inert gas and sealed after loading the spent fuel. The cask structural material (e.g., forged steel, modular cast iron, or composite materials) provides shielding and double-welded closures provide for confinement of radionuclides. Heat is removed by conduction through the structural material. Metal casks are placed vertically in the open on a concrete pad or in storage buildings.
- Concrete cask – A concrete cask is generally similar to the metal cask in shape but includes a steel liner in the inner cavity of the concrete casks. The steel liner works as the main physical containment barrier while the concrete cask mainly provides shielding. The steel liner after loading the spent fuel is filled with helium and sealed with a welded lid. The casks are cooled naturally or through forced ventilation between inlet and outlet airflow ducts. These casks can be stored vertically in the open space or horizontally or vertically inside a building. Concrete casks are generally not transported as loaded with spent fuel and thus require facilities for loading/unloading of spent fuel.
- Concrete module – The concrete module is a large monolithic structure to provide horizontal placement of sealed steel canisters with shielding of spent fuels. The module structure is typically anchored to the storage pad on the ground and thus not portable and require facilities for loading/unloading of spent fuel. Use of internal cooling through natural convection in the module is effective and allows for loading of hot fuel in large quantities.
- Vault – A vault is a large reinforced concrete structure containing an array of spent fuel storage cells. Each storage cell can contain one or more spent fuel assemblies stored in metal storage tubes or cylinders. Typically the vault system is above ground but can also be below ground. Cooling is provided by natural air convection over the surface of the cells (or through forced air circulation, if

necessary). Separate facilities for receipt, unloading, and perhaps packaging of spent fuel are needed.

- Drywell – A dry well is a stationary, individual cavity/hole located below ground. Each drywell cavity is lined with either steel or concrete and accommodates one or several spent fuel assemblies. The system is shielded by the surrounding earth and a closure plug above, and cooled by conduction through the surroundings. These drywell holes can be prepared near-surface or in the floor of subterranean tunnels.
- Other underground storage concept – Other proposed system includes an underground ventilated modular dry storage system or tunnel rack storage system with movable racks placed in tunnels inside a mountain.

Key characteristics of these storage concepts are summarized in Table 7.23.

Table 7.23 Comparisons of key characteristics of dry storage technologies

	Heat transfer	Containment (medium)	Shielding	Advantage	Disadvantage
Metal cask	Conduction through cask wall (passive cooling)	Double lid metal gasket (inert gas)	Metallic wall	Proven; good expandability	Cask is expensive; high fuel surface temperature
Concrete cask	Air convection around canister (passive cooling or forced ventilation)	Cavity lining/seal welding (inert gas)	Concrete and steel overpack	Good expandability; low cost	Low thermal conductivity of concrete – Limiting inner concrete surface temperature; concern over long-term stability of concrete
Concrete module	Air convection around canister (passive cooling)	Canister sealing (inert gas)	Concrete wall	Proven/commercial experiences; good expandability	Low storage density; high fuel surface temperature; concern over long-term stability of concrete; potential public acceptance concern
Vault	Air convection around thimble tube (passive cooling or forced ventilation)	Thimble tube	Concrete wall	Low worker dose; proven/ commercial experiences	Low storage density; potential pubic acceptance concern
Dry well	Heat conduction through earth (passive cooling)	Canister (inert gas)	Earth	Reasonably good expandability; below ground	Large area requirement; high fuel surface temperature

Source: IAEA (2007)

The dry storage concept in general provides good expandability, except the case of using vault. It is, however, difficult to perform fuel inspections during the storage. Large structures such as vault and concrete module may have more concern with public acceptance.

7.4.2.2 Cost of Dry Storage Options

In a report published in 1985, U.S. DOE estimated the total undiscounted costs of constructing and operating a 1000-tonne spent fuel storage facility at a reactor site (OTA 1985). The results ranged from \$82 million to \$260 million per facility, thus giving an estimates of storage cost at 82 ~ 260 thousand US dollars per ton of spent fuel. According to an IAEA estimate, the undiscounted total cost of a 48,000-tonne capacity AFR centralized dry-storage facility ranged from \$2.4 billion to \$5.3 billion (IAEA 2007). This corresponds to 50 ~ 110 thousand US dollars per ton of spent fuel. From these estimates, it can be assumed that the construction cost of dry storage facility is approximately one or two hundred thousand dollar per ton of spent fuel (in 1985 US dollars). Table 7.24 shows the details of these estimates. Although the numbers are outdated, results provide a baseline for comparison of different approaches.

In general, metal cask is found an expensive option while dry well appears the least expensive one. In the case of a fixed structure facility, the cost varies as a function of facility capacity per economy of scale. The larger the capacity, the lower the cost per ton of spent fuel. Per ton basis, the cost of fixed structure facility is lower than using modular casks. However, these cost estimates ignore time value of money and do not take into account the total discounted cash flow.

The case of considering time value of money in the estimates of spent fuel storage is captured in Fig. 7.16. In the figure, the cost of modular casks (the CASTOR metal cask) is compared with the cost of a fixed structure facility (a 2000 MtHM storage pool) while taking into account time value of money (assuming 7% discount rate). A large difference in the cost between the two approaches is noted, although the actual difference will vary depending upon the value of the actual discount rate (see also the explanations of discount rate in Sect. 2.3.1).

Table 7.24 Comparison of capital and annual operating costs at reactor storage options (\$/ton of uranium in 1985 US\$, operating cost in parenthesis)

Storage option	Facility capacity (tons) 500	1000	2000
Pool	(4900)	59,000 (3000)	42,000 (2000)
Concrete module/silo	(4900)	59,000 (3000)	42,000 (2000)
Vault (fuel canned)	100,000 (1900)	87,000 (1600)	81,000 (1500)
Drywell		41,000 (700)	35,000 (600)
Metal cask (5-tons capacity)	118,000 (1300)	109,000 (700)	103,000 (400)
Metal cask (10-ton capacity)		75,000 (200)	73,000 (200)

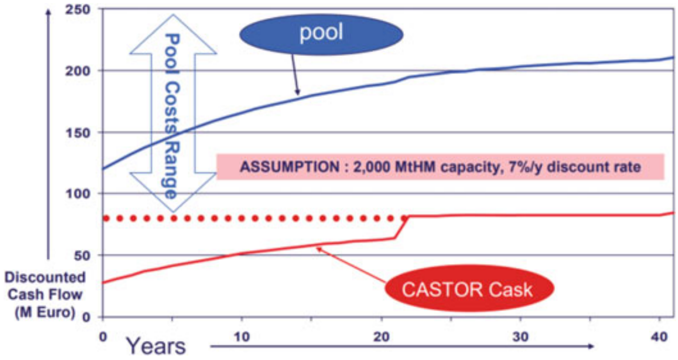


Fig. 7.16 Discounted cash flow for wet and dry storage. (Source: IAEA 2009) (The term, discounted cash flow, in the figure, refers to valuation of an investment based on its expected future cash flows)

Using a fixed structure facility like a storage pool or a vault requires the special effort of facility design and construction. Therefore, there is a time delay between initial investments and starting the actual spent fuel storage service. During this delay, the initial investment, most likely through a loan, incurs interest payments. In contrast, using metal casks does not require development cost, as the casks are commercially available. This implies that the modular dry technologies have a lower initial capital cost, and their remaining costs can be spread out over time as additional containers are needed. Deferring much of the total costs in this way reduces the discounted cost of storage. Therefore, those technologies providing relatively large, fixed capacities (water basins or dry vaults) are more expensive per ton of storage than the dry technologies which readily allow expansion in annual modules (dry well, silos, and casks).

Another way of comparing the cost between a fixed structure and modular units is also shown in Fig. 7.17. In the figure, the area enclosed by the lines represents the magnitude of cost. The open area between the curves is proportional to the carrying cost of the capital expensed before beginning actual storage. The area surrounded by the two dotted lines and the one black solid line reflecting spent fuel generation represents the cost for the pool storage. This area for the pool option is much bigger than the areas covered by the multiple triangles for the cases of MVDS (multiple vault dry storage) or the cask option. This indicates that spreading the fixed cost portion of the project over a large number of storage units lowers the cost of a unit. If the need for total amount of spent fuel storage is uncertain, deferring total cost through deploying modular units is a way to lower the financial risk of investment in comparison to the option of building a fixed storage capacity.

In the case of pursuing the option of building a fixed structure facility (e.g., pools and vaults), consideration of economy of scale should be made. This is because the fixed capital costs required can be significant as seen in Fig. 7.18. This effect is also shown in Table 7.24. For the vault structure, the cost of building a facility as the net

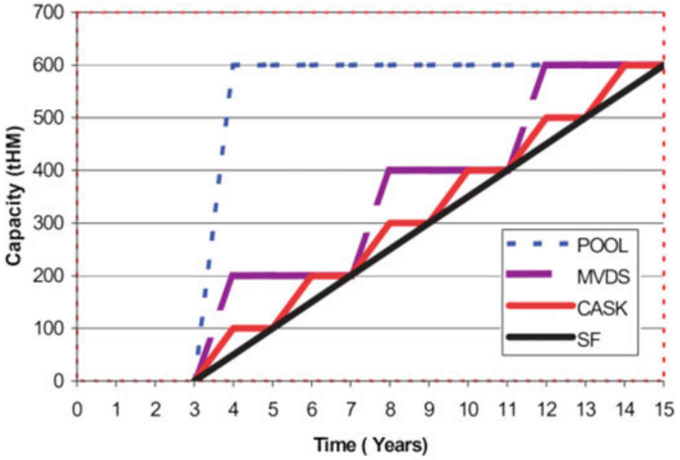


Fig. 7.17 Effect of modularity on costs of spent fuel storage options (IAEA 2009) (MVDS: modular vault dry storage)

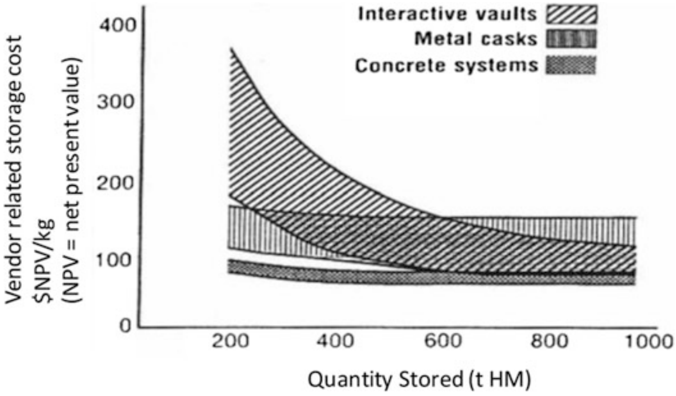


Fig. 7.18 Effect of economy of scale on costs of spent fuel storage options (IAEA 2009)

present value is shown to decrease as the capacity of the facility increases. Therefore, if building a fixed structure facility is necessary, a large capacity storage facility (e.g. >600 t HM) is preferred.

Needless to say, this economy of scale effect is minimal in the case of modular facilities (e.g. metal casks, concrete casks) as the necessary infrastructure for a facility is simple and the level of capital investment is low. In the case of a modular storage facility, a major part of the capital cost is related to the equipment and facilities for the handling and packaging of spent fuel. This portion of the cost (several hundred million dollars) can be minimized by locating the storage facility in a place where existing staff and equipment can be shared (e.g., at a geologic repository or reprocessing plant).

7.4.2.3 The Issue of Storage Periods

The US NRC in 1984 indicated that spent fuel can be stored safely and without significant environmental impacts at an independent spent fuel storage (either wet or dry) installation for at least 30 years beyond the licensed reactor life (NRC 1984). That 30 years period was extended to 60 years under the Waste Confidence Rule of the US. NRC in 1990 (NRC 2010). Considering the licensed reactor life of 60 years in the U.S., this implies that spent fuel can be stored safely at least for 120 years, including the reactor pool storage, contingent upon successful inspection and adequate aging management.

Studies are underway to better understand the implications of long term storage on the integrity of spent fuel. In particular, with the trend in the increase in fuel burnup, potential safety issues related to high burnup and high heat spent fuels need attention. In general, increase in fuel burnup increases the thickness of oxides buildup and hydrogen uptake in spent fuel. This leads to reduction in heat removal capability and cladding embrittlement. Hydride reorientation can also take place under sufficient stress during the storage which may lead to a severe embrittlement of the cladding material. Increased hydrogen uptake can also facilitate propagation of cracks through the cladding thickness.

Increased burnup also leads into the increase in thermal loads and inventory of radionuclides. Increased thermal loads will increase the temperature of spent fuel during storage while higher radionuclide inventory increases the radiation field causing higher level hydrogen gas generation through radiolysis. Both of these phenomena may have implications on possible creep failure of the cladding. Creep (i.e., change of shape of a material) is caused by plastic deformation (e.g., by embrittlement) under the influence of higher temperature. Therefore setting up a limit on maximum allowable temperature for dry storage may be necessary.

There were a few incidents that have highlighted the fact that dry-storage system need to be carefully licensed and closely monitored. On May 28, 1996 at Wisconsin Electric's Point Beach plant in the U.S., an unanticipated hydrogen gas ignition occurred inside Sierra Nuclear Cooperation's VSC-24 storage cask during welding of the shield lid. The hydrogen gas was found to be produced through an electrochemical reaction between zinc in the zinc coating of the cask and the borated water in the spent fuel pool. Similar incidents also took place at Michigan's Palisades unit and at Arkansas Nuclear One. Whenever the procedures of canister welding is performed, possible production of combustible gases during welding operations must be monitored. Once the canister cavity is drained, dried, and backfilled with helium, the source of the hydrogen gas is eliminated.

7.4.3 Monitored Retrievable Storage (MRS)

Due to the difficulty in siting and construction of permanent disposal facility for spent fuel, interim retrievable storage of spent fuel is often necessary. Interim retrievable storage of spent fuel is sometimes called MRS (monitored retrievable storage). The MRS facility is practically no different from the 'away-from-reactor' (AFR) storage facility. The MRS is a stop in between spent fuel storage at nuclear power plants and the permanent disposal. The term 'monitored retrievable storage' (MRS) was introduced in the U.S. in the context of a debate about whether the U.S. Federal Government provide a centralized storage facilities as an interim step until a geological repository becomes available (OTA 1985).

Having a MRS for spent fuel storage for an extended period (e.g., 100 years or longer) provides the benefit of increasing flexibility in the overall scheme of spent fuel management. If construction of a permanent disposal facility is delayed, an MRS can accommodate the increased demand for the storage. In the case of U.S., construction of a dedicated train line between the MRS and the permanent disposal facility reduces the overall shipment distances for spent fuel transportation. The time spent at MRS reduces the decay heat inventory which leads to reduction in disposal costs as more spent fuels can be included in a package or emplaced in the disposal facility.

At an MRS, spent fuel assemblies are handled in the same manner employed in at-reactor spent fuel storage. Decay heat from spent fuels is removed by active/passive means of cooling. Criticality control is achieved by spacing and/or placing neutron absorbers. The facility is under constant monitoring and surveillance. Any operating wastes generated by leakage or corrosion are collected and treated. While the technologies for MRS are dry or wet, sometimes geological formations are also considered. The Climax mine in the U.S. and the Swedish CLAB facility with underground pool are the examples of such facilities.

7.5 Spent Fuel Transportation

Spent fuel transportation takes place by using trucks, trains, or ships between NPPs, storage facilities, and disposal facility. Safety and security must be ensured during transportation against potential radiation accidents and sabotage.

Since 1970s, transportation of spent fuel has been routinely practiced in Europe. These activities are mostly related to commercial reprocessing services in France and the U.K. Similar activities were performed in Germany before their WAK reprocessing plant was closed in 1990. In the U.S., transportation of spent fuel took place over four distinct periods in connection with commercial reprocessing or interim storage of spent fuel. These include: (1) the startup of Nuclear Fuel Services (NFS) – West Valley reprocessing facility (1964–1966) in New York, (2) the additional commercial reprocessing of spent fuel at NFS – West Valley (1971–1974), (3) interim storage at GE-Morris, Illinois (mid 1970s), (4) the decommissioning of NFS at West Valley (1984–1986) and GE-Morris. Spent fuel shipments also took place in this period at the Cooper Nuclear Power Station (1984–1989) of the Nebraska Public Power District to increase spent fuel storage capacity. The failed fuel from the TMI-2 plant were also shipped to Idaho National

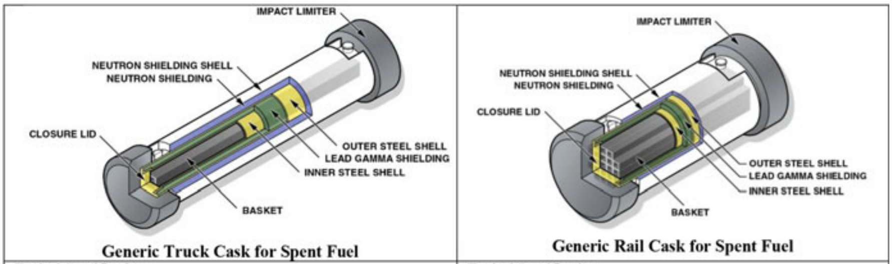
Laboratory between 1986 and 1989. There were expectations for large number of spent fuel shipment starting in mid-1990s with the anticipated opening of Yucca Mountain repository. This did not happen.

7.5.1 Shipping Casks

Transport of spent fuel is typically done by rail or truck or even by the sea when land-based transportation is problematic or not available. In all modes of transport, special shipping casks are used, with their design and size being different depending upon the mode of transportation. Shipping casks are massive and heavy reusable vessels, designed to provide isolation for safety in spent fuel transport. The shipping casks provide, (1) physical containment, (2) radiation shielding, (3) heat removal, (4) criticality protection, and (5) theft protection.

The casks consist of a large cylindrical steel vessel, shielding layers, fuel baskets, a closure lid, and impact-limiters. Typical schematic of spent fuel casks is given in Fig. 7.19. Fuel assemblies are placed inside fuel support baskets within the inner cavity which are designed to accept different types of spent fuel assemblies. The baskets provide mechanical support structure and criticality control by including boron inside the structural material. Impact limiters are made of energy absorbing material (e.g., high density polyurethane foam) surrounded by stainless steel. To load fuel into the cask, the cask containing the fuel basket is placed into the spent fuel pool, and spent fuel assemblies are lowered into the basket. Therefore, loading of spent fuel to the shipping cask takes place underwater.

The fuel baskets are surrounded by two layers of shielding inside the vessel. The first shield layer surrounding the fuel cavity is the gamma shield, made of lead, depleted uranium, or steel. The second layer is the neutron radiation shield (sometimes with added feature of cooling) and consists usually of boron in a mixture of water and ethylene glycol jacket or a specially prepared polyester resin compound.



Generic Truck Cask for Spent Fuel | **Generic Rail Cask for Spent Fuel**

Typical Specifications	Typical Specifications
Gross Weight (including fuel): 50,000 pounds (25 tons)	Gross Weight (including fuel): 250,000 pounds (125 tons)
Cask Diameter: 4 feet	Cask Diameter: 8 feet
Overall Diameter (including Impact Limiters): 6 feet	Overall Diameter (including Impact Limiters): 11 feet
Overall Length (including Impact Limiters): 20 feet	Overall Length (including Impact Limiters): 25 feet
Capacity: Up to 4 PWR or 9 BWR fuel assemblies	Capacity: Up to 26 PWR or 61 BWR fuel assemblies

Fig. 7.19 Schematics of typical spent fuel transportation casks. (Source: NRC 2019)

Table 7.25 Spent fuel transportation casks in the U.S. and their characteristics

Model	Weight (tons)		Capacity		Cavity coolant	Shielding material		Transport mode
	Empty	Loaded	PWR	BWR		Gamma	Neutron	
NLI-1/2	22	23	1	2	He	SS-B-DU	Water	LWT
TN-8	37	39	3	–	Air	SS-Pb	Resin	OWT
TN-9	37	39	–	7	Air	SS-Pb	Resin	OWT
NAC-1/NFS-4	24	25	1	2	Water	SS-Pb	Water	LWT
FSV-1	22–23	23.5–25.5	1	3	Air	SS-DU	Resin	LWT
IF-300	63	68	7	–	Water	SS-DU	Water	Rail
	65	70	–	18	Water	SS-DU	Water	
NLI-10/24	90	97.5	10	24	He	SS-Pb-DU	Water	Rail
TN-12	87	97	12	32	Air	Steel	Resin	Rail

Use of water for cooling and neutron shielding is being discouraged to eliminate the possibility of losing water under accident conditions. The two layers are sandwiched between steel shells. The outer side of the cask is solid steel. The casks are closed by a massive bolt-down closure lid. Generally, the radiation dose rate emitted from the ends of the fuel assembly is less than that emitted from the sides. To limit the impact of collision accidents, impact limiters are placed at each end of the vessel as an impact attenuating medium.

Cooling of spent fuel cask primarily relies on convectional heat transfer (passive heat transfer) through the cask walls. On some casks, cooling fins are provided to increase the heat transfer surface area. Separate auxiliary cooling systems can be used to facilitate handling during loading and unloading.

Table 7.25 gives the list of spent fuel transportation casks with the specification of neutron and gamma shield materials. These casks are for either trucks or trains. Transportation by trucks can be faster but has lower load limits. The casks for shipment by regular trucks (these are called legal-weight truck (LWT) casks) weigh about 25 tons when loaded. Typically, they contain one PWR or two BWR fuel assemblies. The casks designed for special over-weight truck (called, over-weight truck (OWT) casks) allow the loading of multiple PWR or BWR fuel assemblies, with weights up to about 40 tons.

Rail casks come with higher loading capabilities. Light rail casks carry about 7 PWR or 18 BWR fuel assemblies (with a total payload of about 75 tons), and a heavy rail cask can typically hold 12 PWR or 32 BWR assemblies (with a total payload of about 100 tons). Although rail casks can accommodate larger amount of spent fuel per shipment, not all of the nuclear reactor sites have access to the rail lines. For example, about 50% of U.S. reactors do not have rail spurs.

Regulations are in place to ensure the integrity of transportation casks through design, construction, operation, and maintenance. The regulations specify standards for leak-tightness, ability to withstand major accidents, and external radiation

The impact (free drop and puncture), fire, and water immersion tests are considered in sequence to determine their cumulative effects on a given package.

Tests for Spent Fuel Cask Design Certification

Impact:

(a) Free drop - The cask drops 9 m (30 feet) on to a flat, horizontal, unyielding surface so that it strikes at its weakest point.
(b) Puncture - The cask drops 1 m (40 inches) onto a 15 cm (6-inch)-diameter steel bar at least 8 inches long; the bar strikes the cask at its most vulnerable spot.

Fire:
After the impact tests, the cask is totally engulfed in a 800°C (1475 °F) thermal environment for 30 minutes.

Water Immersion:
The cask is completely submerged under at least 3 feet of water for 8 hours. A separate cask is completely immersed under 50 feet of water for 8 hours.

Fig. 7.20 Test requirements for spent fuel shipping casks by the U.S. NRC. (Source: NRC 2015)

exposure limits. For example, the U.S. government regulation (NRC regulations, 10CFR71 and 10CFR73) for spent fuel transportation requires that shipping casks be designed to limit radiation dose to bystanders during normal operations to be less than 0.1 mSv/h (10 mrem/h) at 1.8 m (6 ft) from the cask.

Shipping casks must be designed in such a way that release of radioactive materials from the cask is prevented even in severe accidents. Such required cask performance must be demonstrated through validation of cask designs using model or full-scale tests.

These tests encompass a range of very severe accident conditions including sequential exposure to: impact by drops, fire, and water immersion (as shown in Fig. 7.20). These conditions are for the confirmation of cask performance under simulated severe accident conditions experienced by spent fuel casks.

The regulatory test conditions are engineering criteria that provide a well-defined basis for designing and analyzing casks. They are intended to create stresses on the cask at least as great as those produced by a wide range of extreme accident conditions that could actually be encountered. Thus, while an individual aspect of a specific test (e.g., drop height, or temperature) might be exceeded in real accidents; other test aspects are more severe than what could be encountered in the real world. It has been estimated that the regulatory 30-ft drop onto an unyielding surface would be more severe than about 99.9% of all accidents. The 30 min fire requirement is expected to be longer in duration than 99.8% of actual fires involved in rail or truck accidents (OTA 1985).

A related but different test can also be performed for additional safety demonstration. These include a deep water-immersion test or a crash test. One of the crash tests performed in the U.S. is to crash a 100-ton locomotive at 160 km/h (100 mph) into a shipping cask. The result from an actual crash test showed that the cask maintained its integrity satisfactorily and that the impact was less severe than the regulatory 9-m drop test (NRC 2017).

Proposal for the use of so-called multi-purpose canister (MPC) has also been made for the purpose of streamlining spent fuel management (IAEA 2000). These multi-purpose canisters are not only for transportation but also for storage and disposal, thus, help to minimize handling and any complications in spent fuel shipping or disposal after long-term storage. Using MPC also minimizes secondary waste generation and worker radiation exposure. These benefits, however, may not be realized, if repacking is required due to the changes in licensing requirements. Therefore, use of MPC is most beneficial with the standardization of packaging in spent fuel storage, transport, and disposal, allowing integration of all of spent fuel handling operations. The down side of using MPC is increase in the cost of canister and regulatory burden for licensing.

U.S. DOE, for example, developed plans to implement a multi-purpose canister approach, called Transportation, Aging and Disposal (TAD) canister system in 2007. The followings are the specifications of DOE's TAD cask (US DOE 2008):

- Capacity – 21 PWR or 44 BWR fuel assemblies
- Canister length (including lifting features) – no less than 186.0 and no greater than 212.0 inches
- Maximum weight – 54.25 tons
- Maximum average dose rate from top – 800 mR/h
- Borated stainless steel is the required neutron absorber for disposal.
- TAD canisters to be seal welded.
- TAD canisters, transportation overpack lid, and aging overpack lid will have a common lifting fixture.
- Handling and aging at repository in vertical orientation
- Organic, pyrophoric, and RCRA materials are prohibited

7.5.2 Safety in Spent Fuel Shipment

Regulation of the shipment of radioactive material in the U.S. started in 1939 under the regulations of Postal Service (39 CFR 111.1). In 1954, regulation of spent fuel shipment was provided under the Atomic Energy Act. A dedicated rule for spent fuel shipment was then provided with the passage of U.S. NRC regulation 10 CFR Part 71 (1966, later revised in 1983).

In the U.S., safety record of spent fuel shipment has been good although there were a total of eight accidents between 1960 and early 1990s. These accidents led to accidental radioactive material contamination (see Table 7.26). Among the eight

Table 7.26 Spent fuel transportation accidents in the U.S. involving radioactive material contamination

Date	Mode	Accident description	Contamination beyond the transportation vehicle?
June 2, 1960	Rail	Leak from cask, small area at three rail yards contaminated, no run-off or aerial dispersion	Yes
Nov. 20, 1960	Truck	Small leak from cask onto trailer floor, result of shifting cask, contamination confined to vehicle	No
Sept. 22, 1961	Truck	Leak from cask onto trailer floor, result of shifting, contamination confined to vehicle	No
Aug. 21, 1962	Truck	Cask leakage, trailer and small portion of road contaminated	Yes
Dec. 10, 1963	Rail	Cask leakage, cask contaminated, contamination confined to trailer	No
Nov. 11, 1964	Truck	Cask leakage, trailer, package and terminal contaminated	Yes
July 4, 1976	Truck	Pinhole leak of coolant/moderator on outside jacket of cask, shipment continued without risk to public	No
Jan. 27, 1984	Truck	Slow drip from bottom front end of empty cask while stored in transportation terminal	Yes

Source: State of Nevada (1996)

accidents, six of them took place between 1960 and 1964 when the awareness level of the environmental protection issue was very low and regulatory culture was not well established. Since the promulgation of 10CFR71 in 1966, number of incidents involving leakage of radioactive materials was significantly reduced. This indicates the importance of rules and regulations in ensuring safety in spent fuel transportation. In four of the accidents, contamination was confined to the transportation vehicle, mostly trailer surface contamination. Other four resulted in contamination beyond the transportation vehicle including the truck terminal, some portion of road, or rail yards. The leakages were due to mishandling during loading or unloading of spent fuels. No accidents resulted in the breach of the shipment cask. There was no aerial dispersion of radioactive material.

7.5.2.1 Risk from Incident-Free Shipment

The radiation dose impact on the workers and the public from the incident-free transportation of spent fuel has been estimated (Sprung 2000), examining the dose rate at 1 meter from the surface of truck cask or rail cask containing PWR and BWR spent fuels. The results are summarized in Table 7.27. The dose rate is lower with rail casks than truck casks due to heavier shielding in place. If the spent fuel was cooled for longer than 10 years, the dose rate is less than 0.07 mSv/h (7 mrem/h) at 1 m for all cask types. Actual dose to the public or the members of the shipment crew will

Table 7.27 Estimated dose rate at 1 m for truck and rail cask with spent fuel

Cooling time (yr)	Truck cask dose rate at 1 m	Rail cask dose rate at 1 m
5	0.13 mSv/h (13 mrem/h)	0.067 mSv/h (6.7 mrem/h)
10	0.064 mSv/h (6.4 mrem/h)	0.04 mSv/h (4.0 mrem/h)
15	0.046 mSv/h (4.6 mrem/h)	0.03 mSv/h (3.0 mrem/h)
20	0.035 mSv/h (3.5 mrem/h)	0.024 mSv/h (2.4 mrem/h)
25	0.028 mSv/h (2.8 mrem/h)	0.02 mSv/h (2.0 mrem/h)

Source data: Sprung (2000)

depend on the duration of exposure, the distance from the spent fuel, and placement of shielding.

According to the 10CFR71 regulations, external radiation to the public must be limited to be less than 2 mSv/h at the surface of the casks, or 0.1 mSv/h at a distance of 2 m from it.

Based on various scenarios of radiation exposure during incident-free spent fuel transportation, individual dose from the truck cask and rail cask was projected in a US DOE sponsored study (Sandquist et al. 1985). The results are shown in Table 7.28 (for truck cask) and Table 7.29 (rail cask). As seen in these tables, the maximum expected dose to a member of the public from the assumed exposure scenarios is less than 0.05 mSv which is less than 5% of the recommended dose limit for the public (1 mSv/h) by the IAEA.

7.5.2.2 Risk from Accident During Shipment

To estimate the potential radiation dose received by the public in the hypothetical case of transportation accident involving spent fuel, three sequential studies have been conducted in the U.S. The first one was by the U.S. NRC under the title of "Final Environmental Statement on the Transportation of Radioactive Materials by Air and Other Modes (NRC 1977)". The study was to provide technical basis for 10CFR 71 regulation. Due to lack of experimental data, expert judgements were largely relied upon to determine the response of spent fuel packages to accident environments in the study. Two conceptual models (Model 1 and Model 2) were employed in the study. Model I assumes that the packaging fails catastrophically when spent fuel package was subjected to mechanical or thermal loads in excess of the mechanical and thermal loads encountered during package certification tests. In this case, it was assumed that all inventory of radionuclides are released. This is an unrealistic assumption. Model II assumed gradual release of radionuclides under the accident conditions that exceed the regulatory test level.

The second one was a study by Lawrence Livermore National Laboratory, entitled, "Shipping Container Response to Severe Highway and Railway Accident Conditions" (NRC 1987). This study is also called the Modal Study. In this study, finite element method-based impact and thermal heat transport calculations were

Table 7.28 Projected maximum individual exposures from normal spent fuel transport by truck cask

Activity	Distance to the center of cask (m)	Exposure time	Maximum dose rate and total dose
Caravan – passengers in vehicles traveling in adjacent lanes in the same direction as cask vehicle	10	30 min	40 μrem/min, 1 mrem (0.01 mSv)
Traffic obstruction – passengers in stopped vehicles in lanes adjacent to the cask vehicle which has stopped due to traffic obstruction	5	30 min	100 μrem/min, 3 mrem (0.03 mSv)
Residents and pedestrians – slow transit (due to traffic control devices through area with pedestrians)	6	6 min	70 μrem/min, 0.4 mrem (0.004 mSv)
Residents and pedestrians – truck stop for driver's rest; exposures to residents and passers-by	40	8 h	6 μrem/min, 3 mrem (0.03 mSv)
Residents and pedestrians – slow transit through area with residents	15	6 min	20 μrem/min, 0.1 mrem (0.001 mSv)
Truck servicing – refueling (100-gal capacity)	7	40 min	60 μrem/min, 2 mrem (0.02 mSv)
Truck servicing – load inspections/ enforcement	3	12 min	160 μrem/min, 2 mrem (0.02 mSv)
Truck servicing – tire change or repair to cask trailer	5	50 min	100 μrem/min, 5 mrem (0.05 mSv)
Truck servicing – state weight scales	5	2 min	80 μrem/min, 0.2 mrem (0.002 mSv)

Source: Sandquist et al. (1985)

made to determine the response of spent fuel casks under the collision and fire accident conditions.

The third one was a study by Sandia National Laboratory (SNL), entitled, "Reexamination of Spent Fuel Shipment Risk Estimates (Sprung, 2000)." This study used the findings from the experiments performed at Oak Ridge National Laboratory regarding fission product release from irradiated nuclear fuel under accident. Very detailed and comprehensive characterization of truck and train accidents and their consequences were made available and utilized in the study. A comprehensive uncertainty and sensitivity analysis was also conducted as part of accident consequence quantification. This study represents the most comprehensive and scientifically defensible one.

Results from these three studies are summarized in Table 7.30. As the total population dose from accident for a single shipment involving one shipping cask, the numbers represent the mean estimates of the total combined radiation dose

Table 7.29 Projected maximum individual exposures from normal spent fuel transport by rail cask

Activity	Distance to the center of cask (m)	Exposure time	Maximum dose rate and total dose
Caravan – passengers inrail cars or highway, vehicles traveling in adjacent lanes in the same direction as cask vehicle	20	10 min	30 mrem/min, 0.3 mrem (0.003 mSv)
Traffic obstruction – exposures to persons in vicinity of stopped/slowed cask vehicle due to rail traffic obstruction	6	25 min	100 mrem/min, 2 mrem (0.02 mSv)
Residents and pedestrians – slow transit (through station or due to traffic control devices) through area with pedestrians	8	10 min	70 mrem/min, 0.7 mrem (0.007 mSv)
Residents and pedestrians – train stop for crew's personal needs (e.g., food, crew change, first aid). Exposures to residents and passers-by	50	2 h	50 mrem/min, 0.6 mrem (0.006 mSv)
Residents and pedestrians – slow transit through area with residents (homes, businesses, etc.)	20	10 min	30 mrem/min, 0.3 mrem (0.003 mSv)
Train servicing – engine refueling, car changes, train maintenance, etc.	10	2 h	50 mrem/min, 6 mrem (0.06 mSv)
Train servicing – cask inspections/enforcement	3	10 min	200 mrem/min, 2 mrem (0.02 mSv)
Train servicing – Cask car coupler inspection/ maintenance	9	20 min	70 mrem/min, 1 mrem (0.01 mSv)
Truck servicing – Axle, wheel, or brake inspection/lubrication/maintenance on cask car	7	30 min	90 mrem/min, 3 mrem (0.03 mSv)

Source: Sandquist et al. (1985)

Table 7.30 Estimated mean population dose (person-Sv) from spent fuel transportation accidents per single shipment

Study	Truck accidents	Train accidents
NUREG-0170 Model I (NRC 1977)	1.3×10^{-4}	1.9×10^{-4}
NUREG-0170 Model II (NRC 1977)	7.7×10^{-6}	4.9×10^{-6}
NUREG/CR-4929 (Modal Study) (NRC 1987)	1.3×10^{-6}	1.9×10^{-5}
NUREG/CR-6672 (Sprung 2000)	8.0×10^{-9}	9.4×10^{-8}

among all exposed people during the accident. The human exposure pathways considered as part of dose quantification include the direct exposure to the passing radioactive airborne plume, the exposures caused by inhalation of radioactive materials in the passing airborne plume, the exposures to radioactivity deposited onto the ground from the passing airborne plume, and the exposures caused by inhalation of radioactive materials that are resuspended from contaminated ground into the air. The estimation assumes the transport of PWR spent fuel in a steel-lead-steel spent fuel cask. The shipment routes considered for the calculation include representative set of 200 truck or rail routes in the U.S. involving 79 spent fuel storage locations.

The estimated collective dose to the public from the accidents involving truck or train casks was 8×10^{-9} person-Sv and 9.4×10^{-8} person-Sv, respectively, based on the latest study (the 2000 SNL study). The results from the two previous studies gave higher estimates of dose as the methodologies used were more conservative. The largest dose estimates were from the 1977 NRC study with the total population dose estimates in the order of 10^{-4} person-Sv. While the results vary depending upon the models and assumptions used, the risk associated with spent fuel transportation accidents is found to be non-dramatic. The low level risk estimates from these studies imply that transportation of spent fuel is not a significant concern from the public health perspectives.

Example 7.6 Risk of Spent Fuel Transportation

You are to compare the risk of spent fuel transportation between the rail and truck shipments. Consider the transportation of spent fuels from the Sharon Harris nuclear power plant in New Hill, North Carolina in the U.S to the Yucca Mountain repository. The total shipment mile is 48,908 km (3038.6 miles) for train and 4172.2 km (2607 miles) for truck.

(a) Determine the total number of spent fuel assemblies to be transported from the plant assuming 40 years of plant operation.
(b) Determine the accident rates and fatality rates per shipment mile for both transportation modes.
(c) Determine and compare the total expected accident fatalities between the two transportation modes.
(d) Using the data in Tables 7.28 and 7.29, estimate the total public radiation exposures and the resulting risks for both transportation modes. Make relevant assumptions as necessary.
(e) Assume that less than two accident occurrences in one million rail transport accidents involve the release of radioactivity from the waste packages. The total radiation exposure from these accidents was conservatively estimated at 112,000 person-rem in an urban area or 174 person-rem in a rural area with no cleanup of deposited nuclides (Sandquist et al. 1985). Estimate the total risk associated with rail transport of Sharon Harris spent fuels based on these numbers.

    Assumed plant data: Rating at 3000 MWth; Capacity factor at 75%; Discharge Burnup at 33,000 MWD/MTU

(f) Discuss the overall risk of spent fuel transportation based on your calculation results.

(continued)

Example 7.6 (continued)

Solution:

(a) Thermal energy produced per year = 3000 × 365 × 0.75 = 821,250 MWD

One fuel assembly contains 0.45 MTU.

Therefore, the number of fuel assemblies burnt in 1 year of operation = 821,250/(33,000 × 0.45) = 55.3 → rounded to 56

Number of fuel assemblies in 40 years of operation = 56 × 40 = 2240

(b)

	Accidents[a]	Fatalities[a]
Truck	3.21×10^{-7} per truck-km	1.42×10^{-8} per truck-km
Rail	5.39×10^{-8} per car-km	2.08×10^{-8} per car-km

[a] Data from Saricks, C.L., and M. M. Tompkins, "State-level accidents rates of surface freight transportation – A reexamination", ANL/ESD/TM-150, Argonne National Lab, 1999.

(c)

Train travel distance = 3038.6 miles = 4890 km

Truck travel distance = 2607 miles = 4196 km

Assume cask payload of 1 PWR fuel assembly per cask (light weight truck cask) per one truck shipment and 7 PWR fuel assemblies per one cask (light weight cask). One train can carry 8 casks in one shipment.

Total number of shipment for truck = [2240 assemblies]/[1 per shipment] = 2240

Total number of rail shipment = 2240/(7 × 8) = 40

Total number of accidents for truck shipment = (2240) × (4196km) × (3.21×10^{-7}) = 3.02

Total number of accidents for rail shipment = (40) × (4890km) × (5.39×10^{-8}) = 0.0105

Fatalities for truck shipment = (2240) × (4196) × (1.42×10^{-8}) = 0.133

Fatalities for rail shipment = (40) × (4890) × (2.08×10^{-8}) = 0.00407

(d)

Based on assumed number of people exposed and the time spent for each exposure event, the following collective dose values are calculated.

Dose from one truck shipment

Description	# of people exposed	Exposure duration	Collective dose (person-mrem)
Caravan	20	60 min	40 (0.4 mSv)
Traffic obstruction	20	30 min	60 (0.6 mSv)
Pedestrians	20	12 min	16 (0.16 mSv)

(continued)

Residential area transit	100	24 min	240 (2.4 mSv)
Rest stop	30	24 h	270 (2.7 mSv)
Truck servicing			88 (0.88 mSv)
Total			714 (7.14 mSv) per shipment

Total collective dose for the entire shipments = (Total number of shipment) *(dose per shipment) = 2240 × 7.14 (mSv) = 16 person-Sv

Dose from one rail shipment

Description	# of people exposed	Exposure duration	Collective dose (person-mrem)
Caravan	100	10 min	30 (0.3 mSv)
Traffic obstruction	20	25 min	40 (0.4 mSv)
Transit thru station	100	10 min	70 (0.7 mSv)
Residential area transit	500	10 min	150 (1.5 mSv)
Train stops	100	2 h	60 (0.6 mSv)
Train servicing			240 (2.4 mSv)
Total			590 (5.9 mSv) per shipment

Total collective dose for the entire shipments = (Total number of shipment) × (dose per shipment) = 40 × 5.9 (mSv) = 0.236 person-Sv.

Now, the following calculation assumes that fatal cancer risk among the public can be estimated as the product of the collective dose and cancer potency factor (5.5×10^{-2} per Sv, from Table 5.9). This approach has weak technical basis as the ICRP does not endorse this type of cancer risk calculation when the collective dose represents the sum of very level doses among the public. The following calculation is very conservative and for illustration purpose only.

Fatal cancer risk from truck shipment = 16 (person-Sv) × 5.5×10^{-2} (excess fatal cancer/rem) = 0.88

Fatal cancer risk from rail shipment = 0.236 (person-Sv) × 5.5×10^{-2} (excess fatal cancer/rem) = 0.013

(e)

Total number of accidents for rail shipment = (40) × (4890.2) × (5.39×10^{-8}) = 0.0105

Assume that the train passes through rural areas for 80% of the route and urban areas for the other 20%.

Excess fatal cancer risk in urban area = 0.0105 × (0.2 urban fraction) × (2×10^{-6} probability of radioactivity release per accident) × (112,000 person-rem) × (5×10^{-4} fatal cancer per rem) = 2.35×10^{-7}

(continued)

Example 7.6 (continued)

Excess fatal cancer risk in rural area = 0.0105 × (0.8 rural fraction) × (2×10^{-6} probability of radioactivity release per accident) × (174 person-rem) × (5×10^{-4} fatal cancer per rem) = 1.46×10^{-9}

Total excess fatal cancer risk for the entire rail shipment = 2.36×10^{-7}

(f)

Total fatality risk from rail shipment = fatalities from nonradiological accidents + fatalities from radiation exposure during normal shipment + fatalities from radioactivity release from accidents = $0.00407 + 0.013 + 2.36 \times 10^{-7} = 0.017$

In the case of truck shipments, there were no accidents in the history involving radioactivity release. If we assume that the estimates from the rail transportation accidents can be applied to the accidents for the truck shipment, the following calculation can proceed.

Total fatality risk from truck shipment (assuming the same fatalities from the release of radioactivity due to accidents) = $0.133 + 0.8 + 2.36 \times 10^{-7} = 1.013$

The results indicate that the risk of spent fuel transportation is largely contributed by the risk from incident free shipments.

Risk of shipping all of spent fuels from Harris nuclear power plant is about 1 fatality or much less. Please note that the estimated fatality from the shipment activities by using collective doses among the public is a hypothetical number as ICRP does not endorse such calculation due to lack of scientific plausibility. Rail transport poses lower risk than truck shipment.

7.6 Conclusion

Understanding the physical and nuclear characteristics of spent fuel is essential to grasp the extent of activities required for safe and secure handling of spent fuel. This chapter described the basic characteristics of spent fuel including its composition, radionuclide inventory, decay heat, and radiation dose as a function of burnup and time since reactor discharge. The current state of scientific understanding of spent fuel enables the deployment of necessary technologies to control the risk of spent fuel to be acceptably low. These technologies are in the form of engineered shielding, heat transfer and removal, criticality control, and integrated system operations. Uncertainty in the performance of these technologies are low as they can be readily demonstrated through field tests and experiences. Chapters 8, 9, and 10 will further discuss the treatment and long-term isolation and containment aspects of spent fuel management.

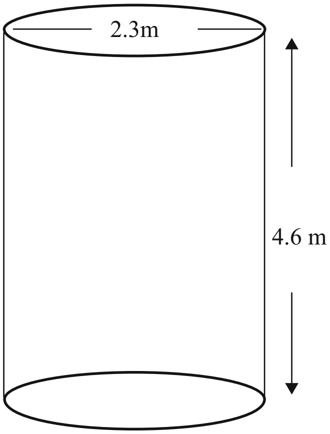
Homework

Problem 7.1: The accident at Three Mile Island Unit 2 in March 1979 occurred approximately 4 months after the reactor entered service. Make a rough estimate of the total fission product activity contained in the reactor core at the time of the accident. You may assume that the reactor had operated continuously at full power (2800 MWt) during the preceding 4 months.

Problem 7.2: Repeat the decay heat calculation given in Example 7.3 for the cooling periods of 30 years and 100 years by using the correlation models of Eqs. (7.27). (7.28), and (7.29) and compare the results.

Problem 7.3: Spent fuel, after being discharged from the reactor, has been cooled in a water-filled pool at the reactor site. Due to the delayed schedule for permanent disposal, spent fuel is accumulating in the reactor pools, which in some cases will soon be full. To overcome this problem, several dry storage concepts including metal cask storage are being developed.

In the case of metal cask storage, the decay heat can be removed only by natural air convection and consequently the spent fuel must be aged for some time before being placed in the metal casks. You are asked to derive a rough estimate of the cooling time required before the fuel can be placed in the cask.



metal cask dimension

Maximum cask surface temperature limit: 250 °C
Ambient air temperature: 18 °C
Natural convection heat transfer coefficient: $h = 1.783\ \mathrm{W/m^2\text{-}^\circ C}$

Assume that the original fuel assemblies, which now have become spent fuels and are to be stored in this single metal cask, contained 1000 kg ^{235}U as fresh fuel (ignore ^{238}U for fission contribution) and were exposed to a thermal neutron flux of 10^{13} $\mathrm{n/cm^2/sec}$ for 3 years. Assume that 200 MeV is the value of recoverable energy per fission and that the thermal fission cross section of ^{235}U is 585 barn. (1 eV = 1.6×10^{-19} J).

(Calculate only the approximate cooling time in years.)

Problem 7.4: List six different ways nuclear electrical generating utilities can use to alleviate the problem of storage of spent fuel prior to disposal in a geologic disposal facility.

Problem 7.5: Assume that a spherical fuel with 2.5 cm diameter is used in a gas cooled reactor containing 3 g of ^{235}U. The ^{235}U is uniformly distributed within the fuel. As a test, the fuel is placed in a reactor for irradiation under uniform thermal neutron flux of 1×10^{12} neutrons/cm^2-sec for 2 months. After the irradiation, the fuel is removed from the reactor and moved to a hot lab in a shield box. Determine the thickness of the wall of the box to reduce the exposure rate at its surface to no more than 1 mR/h, if the box is made of (1) lead, (2) tungsten, or (3) aluminum.

Make all calculations for 1 min after removal from the reactor. You can assume that the sphere is a point source and that the exposure is dominated by gamma rays with negligible contributions from neutrons. Use the assumptions given below as well.

Assume:

E_{avg} (the average energy of gamma rays) = 0.7 MeV

$Y\gamma$ (gamma yield per fission) = $1.9 \times 10^{-6}\, t^{-1.2}$ gammas/sec (t is the time since fission in [days]) (Eq.7.19)

ϕ_n (*neutron flux*)= 1×10^{12} neutrons/cm^2-sec

ρ_{235}(*density of uranium*) = 19.1 g/cm^3

σ_f(*fission cross section*) = 578.6 barn in the reactor

$\left(\frac{\mu_a}{\rho}\right)_{air}$ (*mass attenuation coefficient in air*) = 0.293 cm^2/g for 0.7 MeV in the air

Linear attenuation coefficient of lead (μ_{Pb}), water (μ_W), and aluminum (μ_{Al})

$\mu_{Pb} = 1.124$ cm^{-1}, $\mu_W = 1.711$ cm^{-1}, $\mu_{Al} = 0.1971$ cm^{-1} at 0.7 MeV

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Chapter 8
Spent Fuel Reprocessing and Nuclear Waste Transmutation

Abstract Reprocessing is a key activity in the scheme of spent fuel recycling. Reprocessing allows better uranium resource utilization and the opportunity for nuclear waste transmutation at the expense of higher fuel cycle cost and increased nuclear security and nonproliferation concerns. This chapter provides an overview of the processes involved with two major reprocessing technologies, PUREX and pyroprocessing. Technology of nuclear waste transmutation as a way to render nuclear waste less hazardous is also discussed.

Keywords Separation processes · PUREX · Pyroprocessing · UREX · Transmutation

The activities related to recycling of spent fuel are performed under the modified open or closed nuclear fuel cycle as part of spent fuel management. Reprocessing of spent fuel to separate fissile materials is a key activity in this recycling scheme. Reprocessing also present opportunities to improve efficiency in nuclear waste management through selective removal of radionuclides at the expense of higher fuel cycle cost and increased nuclear security and nonproliferation concerns.

8.1 Overview of Reprocessing

The necessary steps to extract uranium and plutonium for reuse from the constituents of spent fuel, include breaking of the cladding barrier, dissolution of the fuel, and partitioning of elements by using chemical and/or physical processes. Depending upon the separating agents used, the methods can be divided into aqueous separation processes or pyro-processes.

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https://doi.org/10.1007/978-94-024-2106-4_8

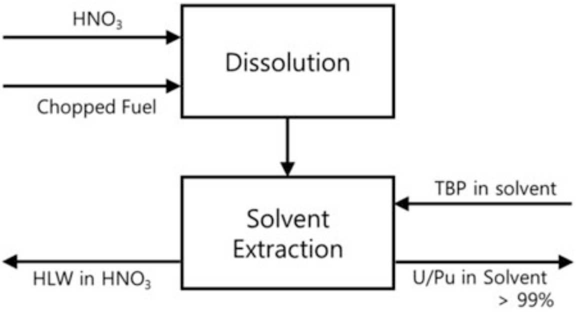

Fig. 8.1 A simple mass-flow diagram for the PUREX process

8.1.1 *Aqueous Processes*

Aqueous separation methods are low temperature processes (normally at 30–70 °C) in which highly dissolving acids (e.g., HNO_3) are used to convert fuel materials into aqueous solutions. Dissolved materials are separated using chemical compounds that form selective bonding with the materials of choice. Examples of the methods include solvent extraction, ion-exchange, and photochemical separations.

Solvent extraction is based on selective transfer of components between two immiscible liquids using differences in the solubility of compounds. These two immiscible liquids include an organic solvent and aqueous solution containing the compounds to be separated. The organic solvent forms selective bonds with the metallic element(s) to be separated. Once such selective bonds are formed, the metallic elements are soluble in the organic phase and the immiscibility of the organic phase in the aqueous phase enables the necessary separation. Uranium and other actinides can easily form an organic-soluble, electrically neutral complex compound through coordination bonds with the organic solvent and can be readily extracted from the aqueous phase.

The solvent extraction technique called the PUREX (Plutonium - URanium EXtraction) process has been the method of choice for spent fuel reprocessing. In the PUREX process, the nuclear fuels are chopped and dissolved in nitric acid. Organic solvent, TBP (Tri-Butyl Phosphate or Tri-n-Butyl Phosphate, $(C_4H_9)_3PO_4$), is then added to the system. Once TBP selectively forms bonds with the dissolved uranium and plutonium ions in the aqueous solution, they become part of the organic phase. As the organic phase material floats over the aqueous phase, they can be physically removed. By going through multi-stage operations of such removal, very high levels of purification can be achieved. Figure 8.1 represents a simple mass flow diagram of the PUREX process.

In the process of ion exchange, the ionized substances dissolved in water will move to a solid phase by being in contact with ion-exchanger solids (thus separated). In exchange, the stoichiometrically equivalent amount of ions of the same sign in the solids will move to the aqueous phase. Photochemical process is based on creating transient excited states of a molecule through selective absorption of a photon. The molecule in the excited states becomes vulnerable to chemical change and goes through chemical reaction for the necessary separation. The method of ion exchange or photochemical separation has not been commercially utilized for spent fuel reprocessing.

8.1.2 Pyro-processes

Pyroprocesses are based on high temperature processing (over several hundreds °C, e.g., > 450 °C) to perform the necessary separation (as the word, pyro means fire in Greek). Pyroprocesses could be either pyrophysical (sometimes called pyrometallurgical) or pyrochemical processes. Pyrophysical processes use the differences in physical properties of compounds such as vapor pressure/melting points or solubility for separation. For example, fractional distillation uses the differences in vapor pressure between compounds for separation. Liquid-liquid partitioning uses differences in solubility for partitioning of compounds in the liquid melt. Liquid-solid extraction is based on converting a component in a mix to a solid through precipitation out of liquid melt.

Pyrochemical techniques use the differences in relative thermodynamic stabilities of compounds for separation in oxidation-reduction reactions. The most common approach is the electrochemical method using molten salts (e.g. LiCl-KCl or $LiF\text{-}CaF_2$). Molten salts are used to dissolve the fuel materials and as electrolyte for electrochemical separation. Metals including liquid metals (e.g. cadmium, bismuth, magnesium) are also used as electrodes for separation. This process is often called “pyroprocessing” as the most widely used pyrochemical method.

8.1.3 History of Spent Fuel Reprocessing

In early 1940s in the U.S., various methods of spent fuel reprocessing were investigated as part of plutonium-based nuclear weapons development within the Manhattan Project. Among the efforts, bismuth phosphate precipitation was the first demonstrated technique. Developed in 1942, the process relied on adding bismuth nitrate and sodium phosphate for the precipitation of plutonium. Only plutonium was separated with the production of only micrograms from batch operations after going through numerous processing cycles and chemicals. The process produced large volumes of high-level waste. The nuclear weapon, exploded in Nagasaki, Japan, on August 9, 1945, was the product of the scale-up version of this operation at a facility in Hanford in the state of Washington (the first nuclear weapon exploded in Hiroshima on August 6, 1945 was a HEU bomb).

Subsequently, solvent extraction quickly emerged as the most successful method to enable continuous, large scale operation for the extraction of both uranium and plutonium. Initially, the solvent extraction technique explored was using diethyl esther as the organic solvent. However, as diethyl esther was quite volatile, less volatile hexone (methyl isobutyl ketone) was selected as the extractant (at Argonne National Laboratory). This is known as the Redox process. The process enabled recovering both U and Pu with high yield and high separation efficiency. A pilot plant of Redox was built in Oak Ridge, Tenesseee, in 1948–1949 followed by the construction of an industrial scale plant in Hanford, Washington, in 1951. It was also used for the recovery of highly enriched uranium (HEU) at a separate facility in the state of Idaho. However, hexone was still highly flammable and volatile and operation of the redox plant was also expensive as the process required large volume of fresh chemical consumption and waste storage. The U.K. and Canada jointly pursued the use of dibutyl carbitol as extractant in late 1940's which has lower vapor pressure than hexone. This is known as the Butex process. An industrial scale Butex process was built at the Windscale plant in UK in 1952 which performed reprocessing for military purposes until 1964.

Then the discovery of tri-n-butyl phosphate (TBP, $(C_4H_9)_3PO_4$)) came along. TBP was first invented at the Metallurgical Laboratory at the University of Chicago with the filing of a patent entitled, "Solvent Extraction Process for Plutonium" in 1947. TBP was less volatile and flammable than hexone and was also more chemically stable in a nitric acid environment. TBP also had better radiation resistance than hexone allowing repeated use of the solvent through recycling. Use of TBP for uranium extraction was demonstrated at Oak Ridge National Laboratory (ORNL) in 1949 and the related process flowsheet was developed by Knolls Atomic Power Lab in New York in the same year using the mixer-settler concept. After the pilot plant stage operation at ORNL from 1950 and 1952, an industrial scale plant was built at Savannah River, South Carolina, in 1954. The Redox process at Hanford was also replaced with PUREX in 1956. A modified PUREX process was also adopted at the Idaho Chemical Processing Plant in 1953.

The PUREX process allowed high throughput continuous operation while minimizing waste through recycling of water, nitric acid, and solvents which helped to lower the operating costs. With a somewhat parallel development, the U.K. replaced the Windscale operation with PUREX in 1964 which continued the operation until 1976. In the U.S., three reprocessing plants were built using the PUREX technique, i.e., the Hanford operation, the Savannah River Plant, and Idaho Chemical Processing Plant. The first two were for the recovery of plutonium from the fuels discharged from plutonium production reactors for nuclear weapons production. The Idaho Chemical Processing Plant was for reprocessing and reuse of HEU spent fuels from naval propulsion reactors (from nuclear submarines and aircraft carriers) and experimental reactors.

Commercial reprocessing of spent fuel based on PUREX also started in the U.S. In 1966, the Nuclear Fuel Services (NFS) Plant (300 MT spent fuel/year capacity) at West Valley, NY, began operation and reprocessed a total of 640 t of spent fuel in 6 years. However, operation of the West Valley facility met challenges as the previous reprocessing experiences were based on processing low burnup spent fuels while commercial spent fuels had much higher burnup. Over the changing safety and environmental regulations and related high retrofit costs to meet the regulations, the plant was shutdown in 1972. Other commercial plants built in the

U.S. include the Midwest Fuel Recovery Plant, Morris, Illinois and the Barnwell Nuclear Fuel Plant at Barnwell, South Carolina. However, these plants never operated. The Midwest Fuel Recovery Plant had difficulties with unit operations causing delays and eventually halting of construction. The Barnwell plant, in contrast, was the state-of-the art facility based on the experiences gained from West Valley NFS operations. The plant was nearly completed with startup testing in progress in 1980. Nonetheless, the announcement of the U.S. policy on banning spent nuclear fuel reprocessing in 1977 forced the plant to shutdown. All of the commercial reprocessing activities in the U.S. has ceased since then (OTA 1985).

In France, the Marcoule plant has been under operation since 1958. The La Hague plant began operation in 1967 for reprocessing of spent fuel from the UNGG (uranium natural graphite gas) reactor and in 1976 added UP2 for the treatment of LWR oxide fuels. Both have been upgraded routinely. In 1994, the La Hague plant was refurbished and expanded, doubling the existing capacity, with UP2 (800 MT/year, for the domestic spent fuel) along with the newly completed UP3 plant (800 MT/year, for the overseas services) in 1990, totaling 1600 t/year capacity. The La Hague plant reprocessed spent fuels from Japan, Germany, Belgium, the Netherlands, and Switzerland (reprocessing of German spent fuel stopped in 2005). In the U.K., Thermal Oxide Reprocessing Plant (THORP), a larger PUREX-based oxide fuel reprocessing plant, was built in 1994 and went into operation in 1997 with initial capacity of 600 MTHM/year. The plant was upgraded to 1200 t/year later. The THORP plant reprocessed spent fuels from customer countries like Japan, Germany, Switzerland, Spain, Italy, Sweden, Netherlands, and Canada. The operations ended in November 2018 due to lack of "economic case" for supporting commercial reprocessing. The Dourney reprocessing plant was also built to support U.K.'s fast breeder program in late 1950s and reprocessed also the spent fuels from overseas research reactors. The plant was closed in 2006 also due to lack of business. Germany has operated the Karlsruhe reprocessing plant (WAK) from 1971 with 35 t/year capacity. About 200 t of spent fuel was reprocessed until the end of the operation in 1990. Germany started building the new Wackersdorf reprocessing plant in 1985 with 350 t/year capacity. However, the project was cancelled in 1991 under intense protest from the public. Germany changed the government policy on spent fuel to direct disposal in 1994.

Japan started the construction of the Tokai-Mura reprocessing plant in 1970 which started commercial operation in 1981 at 200 MT/year capacity. Japan also decided to build a larger facility at Rokkasho-Mura with 800 MT/year capacity. Construction of the plant started in 1993 and was originally expected to be completed by 1997. The target start-up date was then postponed to 2008. The plant completed hot commissioning test in 2006, but extra efforts were needed to enhance nuclear safeguards. The site was then expected to begin operating in October 2013. However, post-Fukushima safety/security regulations caused additional delays. The current target for its operation is in 2022. In Russia, the RT-1 plant began civil reprocessing operation in 1976 at 400 MT/year capacity. Other countries such as Belgium, India, Argentina, Brazil, China, and North Korea also own reprocessing capability using PUREX in a smaller scale.

Table 8.1 List of industrial reprocessing plants in the world

Facility	Country	Capacity	Fuel Type	Start-up date	Current Status
Marcoule, UP1	France	400 t/ year	Thermal, metal	1964	Shutdown in 1997. Currently, under decommissioning.
La Hague, UP2	France	800 t/ year	Thermal, oxide	1976 (1994 upgrade)	In operation
La Hague, UP3	France	800 t/ year	Thermal, oxide	1990	In operation
Windscale	U.K.	1200–1500 t/ year	Magnox fuel (thermal, metal)	1964	Shutdown in 1973. Currently, under decommissioning.
Sellafield, THORP	U.K.	1200 t/ year	Thermal, oxide	1997	Planned to be shutdown in 2018
Dounreay	U.K.	0.025–0.05 t/ day	Fast, oxide	1960, 1980	Shutdown in 2001. Currently, under decommissioning.
Tokai Mura	Japan	200 t/ year	Thermal, oxide	1977	Head-end process shutdown in 2014. Separation in operation (decommissioning under planning)
Rokkasho Mura	Japan	800 t/ year	Thermal, oxide	2018, planned	Planned to start operation in 2018
Mayak, RT1	Russia	400 t/ year	Thermal, oxide	1977	In operation

Table 8.1 shows the list of industrial reprocessing plants in the world. As seen in the table, all industrial spent fuel reprocessing operations presently use the PUREX technology. These include the La Hague plant in France, the Sellafield facility in the U.K., the Mayak facility in Russia, and the newly built Rokkasho-Mura facility in Japan.

Pyroprocessing is a way to separate actinide elements from spent fuel based on the electrorefining process, i.e., electrolysis-based refining. E*lectrolysis* is a technique that uses a direct electric current (DC) to drive decomposition of chemical compounds which is a non-spontaneous chemical reaction. Once decomposed, the desired component is separated from the mixture through electrotransport and deposition on electrodes. Use of the electrorefining process has a long history in the minerals industry. The process was used to purify impure metals, first invented for refining copper and commercially applied for aluminum production in 1890. This was followed by commercial production of lead in 1905, nickel in 1910, and zinc in about 1915. In this refining process, an impure metal is made the anode and deposited at a cathode by electrotransport using a suitable electrolyte to produce greater purity of the metal. The electrolyte used in most commercial metal processing was aqueous acid-base solutions (for aluminum, nickel, zinc, and copper). Molten salt was also used for beryllium purification.

Use of electrorefining for the separation of uranium in molten salt (NaCl-KCl) was first reported in 1930. Electrodeposition of pure uranium from a BaCl-KCl-NaCl-UCl_3 melt was also confirmed in the early 1940s as part of the Manhattan project. However, due to the dirty nature of the product (i.e., lower separation efficiency), the process was not pursued as a method for plutonium separation. In 1950s, Argonne National Laboratory (ANL) began examining the fundamental aspects of uranium electrodeposition from molten salts for fast reactor applications by using LiCl-KCl eutectic as electrolyte. In the 1960s, the research work spread into U.K., France, Belgium, and the Soviet Union. Electrorefining of plutonium from impure plutonium feed was also investigated at Los Alamos National Laboratory (LANL) in the 1960s.

Since the early 1980s, the majority of work on uranium electrodeposition in the U.S. was conducted at ANL in relation to the Integral Fast Reactor (IFR) fuel processing program. Use of pyroprocessing was necessary as the fuel to be reprocessed was sodium-bonded fast reactor fuel. Presence of sodium prevents the use of aqueous process due to highly reactive nature of sodium in an aqueous solution. The work was focused on the development of electrorefining methods for separating actinide elements from fission products for recycling in a fast reactor. The molten-salt based electrochemical technology was also proposed for recovering TRUs and U from LWR fuels to support the Accelerator Transmutation of Waste (ATW) project in the late 1990 in the U.S. Currently, Idaho National Laboratory is using pyroprocessing to treat the spent fuels from EBR II reactor for their disposal. The process has been researched and developed in Russia, Japan, India, and South Korea for recycling or transmutation of actinides (Pu, Np, Am, Cm) (see Sect. 8.4) or reuse of uranium. No commercial scale pyroprocessing plant has been built.

8.1.4 Comparison of PUREX and Pyroprocessing

The PUREX processes and pyroprocesing present different features due to the differences in the materials used and their properties, the way the processes are implemented, and the resulting performance of materials separation.

The organic solvents used in PUREX processes, TBP experiences degradation through radiation damage. When high radiation fields are involved in the process (i.e., due to handling of high burnup fuels), degradation of organic solvents is expected resulting in poorer performance of separation. To prevent this, the PUREX process puts requirements on the period of spent fuel cooling to limit the irradiation effects on solvents. In contrast, the solvent used in pyroprocessing is molten salt which is highly resistant to radiation damage. Therefore, the process can be applied to the freshly discharged spent fuel without any cooling requirements and can be used for any spent fuel regardless of burnup and cooling time. Therefore, pyroprocessing provides the potential for recycling of total actinides in spent fuel through an integral fuel cycle with co-location of the irradiation, processing, and fabrication facilities on the same site. Also, the concern over criticality accidents is much reduced in pyroprocessing as molten salt does not include hydrogen and carbon which work as neutron moderators with potential nuclear criticality concern.

While the PUREX process is implemented through continuous operations of solvent extraction, the pyroprocessing technology relies on batch operations. Therefore, scaling up of the PUREX process for higher throughput is quite feasible. In contrast, the total throughput of pyroprocessing is inherently limited unless parallel deployment of multiple units is implemented. At the same time, the PUREX process involves the use of substantial tankage and reagents with long distance piping (involving several hundred kilometers of stainless steel piping). Therefore the system is very complex. The THORP facility in Sellafield, for example, was unable to detect leakage of 20 t of dissolver solution containing 200 kg of plutonium for more than 8 months in 1995.

The fact that pyroprocessing is based on batch operation means smaller and compact size of the plant which puts less requirement for the security of the plant. Monitoring of the movements of nuclear materials within the plant is also easier with pyroprocessing. However, it is difficult to accurately measure and keep track of the fissile materials inventory in pyroprocessing. This is due to a lack of input accountability tank and presence of large amount of material hold-up in process vessels along with the fact that the spent fuel does not completely dissolve in the electrorefining process.

Another major aspect that distinguishes PUREX from pyroprocessing is the degree of purification of the product. With PUREX, high purity plutonium product can be achieved by employing multiple separation stages. In pyroprocessing, however, the product's purity is inherently low due to the inherently contaminated nature of the products.

To better illustrate this point, let's define a factor called decontamination factor (DF). DF is the ratio of the activity of the nuclide to be separated in the feed to that in the product to represent the degree of separation achieved by the process, as shown below.

$$\mathrm{DF} = \frac{\text{Activity of the nuclide to be separated in the feed}}{\text{Activity of the nuclide to be separated in the product}} \tag{8.1}$$

In general, the PUREX process achieves DFs in the order of 10^6 to 10^8 with after six to seven separation stages. In contrast, pyroprocessing achieves ~100 of DF. DF is related to the separation efficiency as,

$$\mathrm{DF} = \frac{1}{1 - \text{Seperation Efficiency}(\text{in fraction})} \tag{8.2}$$

The low decontamination factor in pyroprocessing means higher level of contamination in the products, mostly by fission products and minor actinides. Therefore, the level of radiation is very high prohibiting the access to the product. In addition, the contaminated nature of the products make them much less attractive as materials for nuclear explosives. Presence of large decay heat also provides self-protection of nuclear materials and barrier against diversion or misuse of the technology.

8.2 PUREX

8.2.1 *The Overall PUREX Process*

To allow the treatment of spent nuclear fuels, the cladding of fuels needs first to be removed. The cladding removal and fuel dissolution are together called the head-end processes. Then the dissolved materials go through the actual processes of separation. Therefore, three steps, the preparation, the dissolution, and the separation operations, constitute the PUREX process.

In the head-end processes, spent fuel is chopped into small pieces. The pieces are then dropped into tanks of nitric acids. The nitric acid will dissolve the UO_2 fuel matrix. Off-gases released during these processes are treated to minimize the release to the environment. The head-end processes also include removal of suspended solids in the dissolver solution and conditioning of the dissolver solution to facilitate solvent extraction.

In the dissolution step, the UO_2 fuel is dissolved in the nitric acid, leaving Pu, U, TRU, and fission products in the nitric acid solution. The separation step uses TBP, the organic solvent. When the solution is mixed with TBP, uranium and plutonium are extracted to the organic phase leaving fission products behind in the nitric acid solution. Repeated treatment of the solution through mixing with TBP further removes U and Pu in the solution achieving high degree of separation efficiency. Through the chemical adjustment of the organic solution, U and Pu are also separated from each other. The remaining highly radioactive nitric acid solution containing fission products and other actinides as liquid waste becomes HLW. This HLW contains more than 99% of the fission products (nonvolatile) and less than ~0.5% of the U and Pu. Simplified flow sheet of the PUREX process is given in Fig. 8.2.

8.2.2 *Pre-processing Storage*

When spent fuel reprocessing is implemented, the freshly discharged spent fuels are stored for specified periods as a preparatory step for reprocessing and to simplify shipment through cooling. This is called a pre-reprocessing storage. This pre-reprocessing storage is to allow decay of fission-product activity, to reduce decay heat production, and to reduce radiation damage to the materials used in fuel reprocessing. The target radionuclides that decay away during the storage include: ^{131}I ($t_{1/2}$ = 8.04 d), ^{237}U ($t_{1/2}$ = 6.75 d), and ^{133}Xe ($t_{1/2}$ = 5.27 d) (they all emit both β and γ rays). Typically about 200 days are required for ^{131}I to decay to

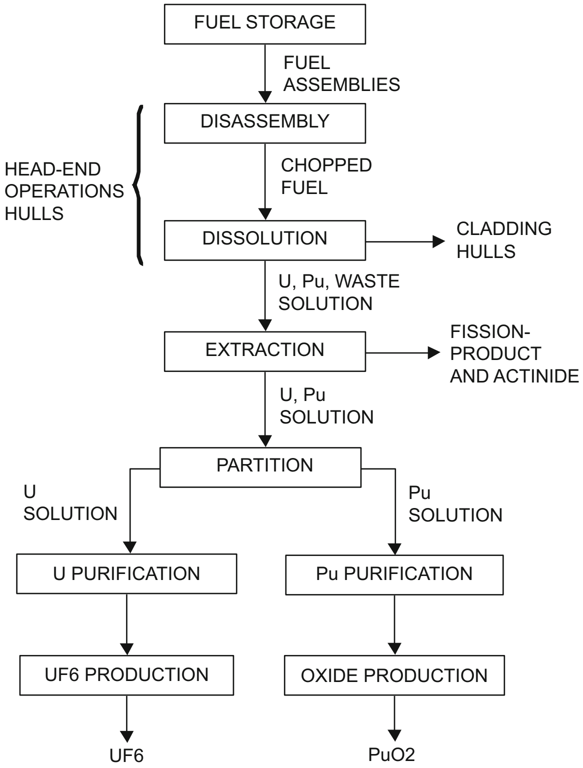

Fig. 8.2 Simplified flow sheet of the PUREX process for spent reactor fuel

the minute level of long-lived ^{129}I. This will avoid troublesome quantities of gaseous and dissolved radioiodine in fuel reprocessing. The decay of ^{237}U eliminates the need for remote handling of the purified uranium recovered by fuel reprocessing as ^{237}U decays to ^{237}Np while releasing 0.2 MeV and 0.06 MeV gammas. Also, presence of high activities of ^{237}U would interfere with monitoring for fission-product decontamination of the removed uranium. The decay of ^{133}Xe leaves ^{85}Kr as the only radioactive noble gas liberated in the fuel reprocessing process. One potential concern from this storage is the buildup of fissile ^{239}Pu from the decay of ^{239}Np ($t_{1/2} = 2.35$d).

8.2.3 Head-end Process

As a preparatory operation to permit fuel dissolution, mechanical shearing or sawing of fuel cladding is conducted along with chopping of UO_2 fuel into short segments. During this process, most of the volatile fission products (e.g., noble gases) inside the cladding is released as off-gases. The sheared short segments of UO_2 fuel are then exposed to air or oxygen at high temperature. This results in the conversion/ oxidation of UO_2 to mostly U_3O_8. This process is called voloxidation (volatilization

+ oxidation). In voloxidation, the fuel structure is broken up releasing gaseous or volatile products trapped in the UO_2 grain boundaries. Most of tritium produced in the fuel as fission product becomes a part of off-gases from the process.

As a preparation for separation, the dissolver solution is also conditioned for the solvent extraction stage by adjusting the acidity of the solution to pH of around 2.5.

8.2.4 Separation Processes

Separation of chemical substances in PUREX follows the principle of solvent extraction. The degree of separation of a compound in the process is represented by the term, distribution coefficient, D, which is defined as,

$$D = \frac{\text{Concentration of compound in organic phase}}{\text{Concentration of compound in aqueous phase}} \tag{8.3}$$

The distribution factor will be dependent on types of solvent used and the factors that affect the chemical reactions involved, e.g., the concentration of the species in the aqueous phase in their respective oxidation state, the concentration of the organic extractant, temperature, and the acidity of the system.

In PUREX, the organic phase is the solvent used to extract plutonium and uranium in a paraffinic hydrocarbon diluent (i.e., kerosene). The aqueous phase is the feed solution after the dissolution of UO_2 fuel in nitric acid.

The actinides have multiple oxidation states (valences, see Sect. 4.1.2) available in aqueous solution. Figure 8.3 shows various oxidation states of actinides available

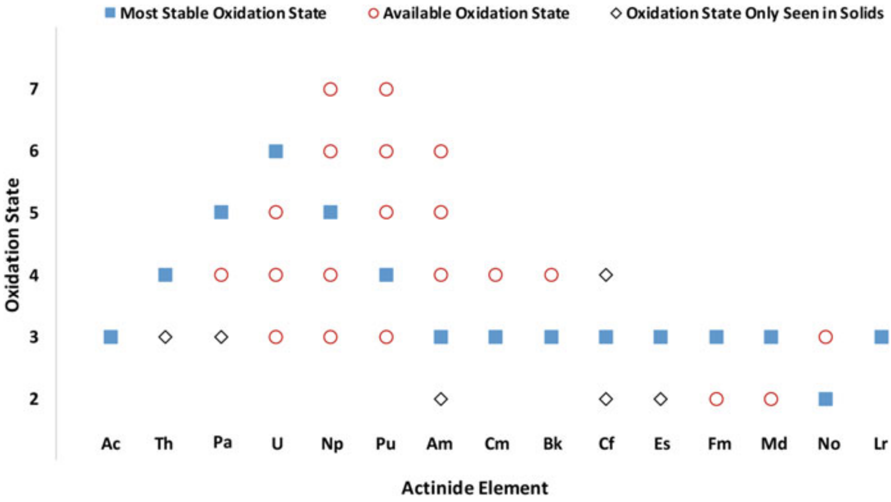

Fig. 8.3 Various oxidation states of actinides in aqueous solution. (Redrawn from Source: Jubin 2011)

in aqueous solution. Among the various oxidation states, the most stable state is shown in red which varies among the actinides (e.g., +6 for U and +4 for Pu in aqueous solution). The most stable state is also the most extractable state by readily forming chemical complexes with organic ligands. Depending upon the oxidation states taken in the solution, the extractability of actinides, i.e., the resulting distribution coefficient, would be different.

In the dissolution step, the fuel and cladding are reacted with hot nitric acid. Uranium as ceramic oxide and most other elements in the fuel are dissolved in the process. The metallic cladding does not dissolve and the remaining cladding hulls are washed with water, separately packaged, and shipped for storage. Removal of suspended solids in the dissolver is performed by typically using a semi-continuous centrifuge to reduce the radiation level and the heat inventory and to prevent criticality accident. The suspended solids include a variety of insoluble fission products (e,g, platinum, ruthenium, rhodium, technetium, palladium, and molybdenum), plutonium rich fuel grains, and mixed zirconium-molybdenum oxides.

When spent fuel is dissolved in nitric acid, both U and Pu are present mostly in the forms of UO_2^{2+} and PuO_2^{2+} in the oxidation state of +6. As the valence state of +4 (tetravalent state) is considered most extractable for Pu (while the +6 state is most extractable for U), the feed is conditioned for Pu with nitrous acid to produce more extractable Pu(IV) as shown below.

$$Pu^{(VI)}O_2{}^{2+} + N_2O_4 + 2H^+ \rightarrow Pu^{4+} + 2HNO_3$$

The steps of separation in PUREX include extraction, scrubbing, and stripping. Extraction refers to the separation of U and Pu from the feed solution into the organic phase. Scrubbing means removal of impurities from the separated organic phase by using a new immiscible liquid phase. Stripping is a step of back extracting Pu and U from the organic phase to the aqueous phase for their recovery. Use of scrubbing and stripping allows the recycle of organic solvents for reuse.

The following reactions take place during extraction.

$$\mathrm{UO_2{}^{2+}\ (aq) + 2NO_3{}^-\ (aq) + 2TBP\ (org) \Leftrightarrow UO_2(NO_3)_2 \cdot 2TBP\ (org)}$$

$$\mathrm{Pu^{4+}\ (aq) + 4NO_3{}^-\ (aq) + 2TBP\ (org) \Leftrightarrow Pu(NO_3)_4 \cdot 2TBP\ (org)}$$

In this step, 30 v/o (volume percent) TBP in kerosene (a normal-paraffin diluent) is used as extractant. The concentration of uncombined TBP in the organic phase or the aqueous nitrate ion concentration is controlled to change the direction of the reactions. For example, increasing the concentration of uncombined TBP in the organic phase will push the reactions to the right, increasing the relative amount of U or Pu cations in the organic phase. By reducing the nitrate or nitric acid concentration in the aqueous phase, the reaction can also be reversed.

For the extraction process, the degree of separation for the compound of interest, M, is given by the distribution coefficient D_M as,

$$D_M = \frac{\left[M(NO_3)_x \cdot nTBP\right]_{org}}{[M^{x+}]} = K_l\left[NO_3^-\right]_{aq}^x [TBP]_{org}^n \tag{8.4}$$

where K_l is an equilibrium constant given for the reaction equilibria involving a compound l. As Eq. 8.4 implies, increase in acidity (i.e., increasing nitrate level) will increase D_M until the competition between acid and TBP ($TBP + HNO_3 \rightarrow TBP{\cdot}HNO_3$) becomes significant. Also, increase in the loading of uranium in the solvent (i.e., $[M^{x+}]$ is increased) leads to decrease in the D_M values. This is due the decrease in the extraction capacity of TBP.

Example 8.1: Separation Factor in PUREX for U-Th Assume that an aqueous solution contains 0.2 M uranium mixed with 0.8 M thorium in 3 M nitric acid. If the solution is contacted with 1 M TBP of equal volume in kerosene, determine how much U will be separated from the solution in comparison to Th in a single stage solvent extraction.

Equilibrium constants for the formation of TBP complexes with $Th(NO_3)_4$, $UO_2(NO_3)_2$, and HNO_3 are as follows.

$UO_2^{2+}(aq) + 2NO_3^-(aq) + 2TBP(o) \leftrightarrow UO_2(NO_3)_2{\cdot}2TBP(o)$	$K_2 = 7.0$
$Th^{4+}(aq) + 4NO_3^-(aq) + 2TBP(o) \leftrightarrow Th(NO_3)_4{\cdot}2TBP(o)$	$K_1 = 0.7$
$H^+(aq) + NO_3^-(aq) + TBP(o) \leftrightarrow HNO_3{\cdot}TBP(o)$	$K_3 = 0.347$

Answers:

Separation of U from Th in the aqueous solution can be represented by using the separation factor. The separation factor is a measure of the system's ability to separate two different compounds. It is defined as the ratio of distribution coefficients (Eq. 8.3) of uranium and thorium in the problem. The separation factor in this case is defined as,

$$\alpha = \frac{\left(\frac{\text{mole fraction of } U_{organic}}{\text{mole fraction of } U_{aqueous}}\right)}{\left(\frac{\text{mole fraction of } Th_{organic}}{\text{mole fraction of } Th_{aqueous}}\right)} = \frac{\left(\frac{\text{mole fraction of U}}{\text{mole fraction of Th}}\right)_{\text{organic}}}{\left(\frac{\text{mole fraction of U}}{\text{mole fraction of Th}}\right)_{\text{aqueous.}}}$$

Since the volume of initial aqueous solution is mixed with an equal volume of organic TBP, after combining the aqueous solution with kerosene, the concentrations of the compounds at equilibrium can be assumed to be half of what was initially given:

$$HNO_3 = 1.5\ \text{M}, \text{Th} = 0.4\ \text{M}, \text{U} = 0.1\ \text{M}, \text{TBP} = 0.5\ \text{M}$$

(continued)

Example 8.1 (continued)

Using the equilibrium constants to find the concentrations of [$Th(NO_3)_4{\cdot}2TBP$] and [$UO_2(NO_3)_2{\cdot}2TBP$] gives,

$$\begin{aligned}\left[Th(NO_3)_4\cdot 2TBP\right] &= K_1\cdot[Th]\cdot[NO_3]^4\cdot[TBP]^2 = 0.7\cdot(0.4)\cdot(1.5)^4\cdot(0.5)^2\\ &= 0.354\ M\end{aligned}$$

$$\begin{aligned}\left[UO_2(NO_3)_2\cdot 2TBP\right] &= K_2\cdot[UO_2]\cdot[NO_3]^2\cdot[TBP]^2\\ &= 7.0\cdot(0.1)\cdot(1.5)^2\cdot(0.5)^2 = 0.394\ M\end{aligned}$$

Separation factor, α, is then:

$$\alpha = \frac{\left(\frac{mole\ fraction\ of\ U}{mole\ fraction\ of\ Th}\right)_{organic}}{\left(\frac{mole\ fraction\ of\ U}{mole\ fraction\ of\ Th}\right)_{aq\ (inorganic)}} = \frac{\frac{0.394}{0.354}}{\frac{0.2}{0.8}} = 4.45$$

The U/Th separation factor from a single stage solvent extraction is 4.45. Therefore, single extraction stage will remove ~4.5 times more of U than Th.

The process of extraction, scrubbing, and stripping is repeated to achieve the desired level of purification. This is shown in Fig. 8.4 as a schematic of a typical solvent extraction process. As shown, the system is designed for the flow of aqueous and organic phase materials to move in countercurrent directions in the system while the two phases are being contacted. This design is to allow multi-stage implementation of solvent extraction with recycling of TBP allowing multiple contacts between the two phases reducing the amount of solvent needed.

The aqueous phase remaining after U and Pu are extracted is called raffinate. Raffinate contains almost all of the nonvolatile fission products along with Am, Cm,

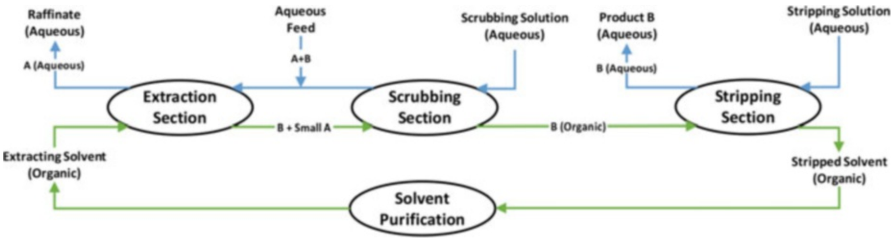

Fig. 8.4 Schematic of a typical solvent extraction process including extraction, scrubbing, and stripping stages

and corrosion products. Due to complexity in the chemical state, Np exists in both aqueous raffinate and organic extract.

Separation of U and Pu from the feed solution and subsequent purification of U and Pu from each other is done at the end of extraction and scrubbing, by controlling the oxidation state of each element. For this purpose, Pu(IV) is reduced to Pu(III), the organic-insoluble trivalent state. This is done by treating organic solution with reducing agents such as U^{4+} or ferrous sulphamate (Fe^{2+}). In the case of using U^{4+} as reducing agent, the reaction becomes,

$$2Pu^{4+} + U^{4+} + 2H_2O \rightarrow 2Pu^{3+} + UO_2{}^{2+} + 4H^+ \tag{8.5}$$

The Pu in the trivalent state is then partitioned into the aqueous phase and back extracted as plutonium nitrate. The U remaining in the organic solution is also converted to aqueous phase as uranyl nitrate. Then the back-extracted impure plutonium nitrate and uranyl nitrate solutions are purified and converted to the product in preferred chemical forms, e.g., oxides for the fabrication into mixed oxide (MOX) fuel (through either evaporation or precipitation followed by calcination).

To the maximum extent practicable, materials are recycled in these processes to minimize the volume of wastes or effluents and to reduce the cost of chemicals. For example, the evaporated nitric acids are collected, condensed and recycled. Low level aqueous wastes are concentrated for the recovery of water and nitric acid along with additional recovery of Pu and U. High level aqueous wastes, the raffinate from the process, are concentrated by evaporation while condensed nitric acid is recovered and recycled.

Fuel-cladding hulls, particulate filters, discarded equipment tools, and contaminated trash from the processes become high level or intermediate level wastes. They are compacted and packaged in special waste containers for final disposal. Filters, adsorber materials, scrubbing solutions, and other process wastes from off-gas treatment mostly become low level waste. They are also treated in a similar way for final disposal.

A related practical issue in PUREX operation is the degradation of TBP due to radiation damage. Under irradiation, TBP experiences thermal and radiolytic degradation to form di-butyl phosphate and mono-butyl phosphate. Both by-products complicate the flowsheet chemistry as di-butyl phosphate forms strong extractable complexes with plutonium and zirconium while mono-butyl phosphate tends to form precipitates. Their effects can be minimized by removing the degradation products in the solvent through washing with alkali solutions.

8.2.5 Off-gas Treatment

Off-gases are produced from the head-end processes including fuel mechanical chopping, the processes of voloxidation, and dissolution of the solid UO_2 fuel matrix. These off-gases contain up to 10% of krypton and xenon in the fuel and some of the $^{14}CO_2$, ^{3}H, and other volatile fission products (^{129}I, ^{106}Ru) along with oxides of nitrogen. These off-gases are treated to minimize the carry-over into the solvent and their releases as gaseous effluent to the environment. Such treatment includes removal of tritium, ^{14}C, ^{129}I, ^{85}Kr, and ^{106}Ru.

For its removal, tritium is reacted with dilute nitric acid (called aqueous scrubs) and captured as tritiated water in solid absorbents. The solid adsorbent then go through immobilization using grout to be rendered into waste form for final disposal. Carbon-14 is removed either physically or chemically and encapsulated into a waste form for disposal. Physical removal is through either physical adsorption or concentration on molecular sieves. Chemical removal is through reaction with sodium hydroxide ($CO_2 + 2NaOH \rightarrow Na_2CO_3 + H_2O$) followed by immobilization as $CaCO_3$. Removal of ^{129}I is through precipitation by using scrubbing liquids or by using sorbents (charcoal filters or silver impregnated zeolites). The removed iodine is encapsulated typically as silver iodide (AgI) and made into a waste form such as glass, ceramic, or cementitious material. Removal of ^{85}Kr is through cryogenic process. In the process, the carrier gas is compressed (e.g., to 8 atm), cooled (-170 °C) and passed through distillation column for collection of ^{85}Kr. The collected ^{85}Kr is kept in pressurized steel containers for decay-in-storage. Collection of xenon can be done by using the same technique. For the removal of ^{106}Ru, the off-gas condition is controlled to be reducing allowing the conversion of ruthenium into RuO_2 particulates. These particulates are removed by filtration. Along with these processes, water, nitrogen oxide, and hydrocarbons in the system are also removed to prevent explosion due to combination of hydrogen and oxygen in the system,.

8.2.6 Implementation of PUREX

Separation by solvent extraction requires mixing of two (i.e., aqueous and organic) phases well enough for efficient mass transfer between them. Such mixing can be achieved through various turbulence creating schemes. These schemes include mixer-settler, pulsed column, and centrifuge. In these schemes, mixing of two phases are first achieved and then the separation takes place by the gravity (or centrifugal force).

Due to the presence of fissile materials in the processes, criticality control is important in any of these schemes. One key consideration in this respect is the use of "favorable geometry". It is known that for a cylindrical geometry, criticality cannot be reached for any specified composition as long as the cylinder diameter is below a

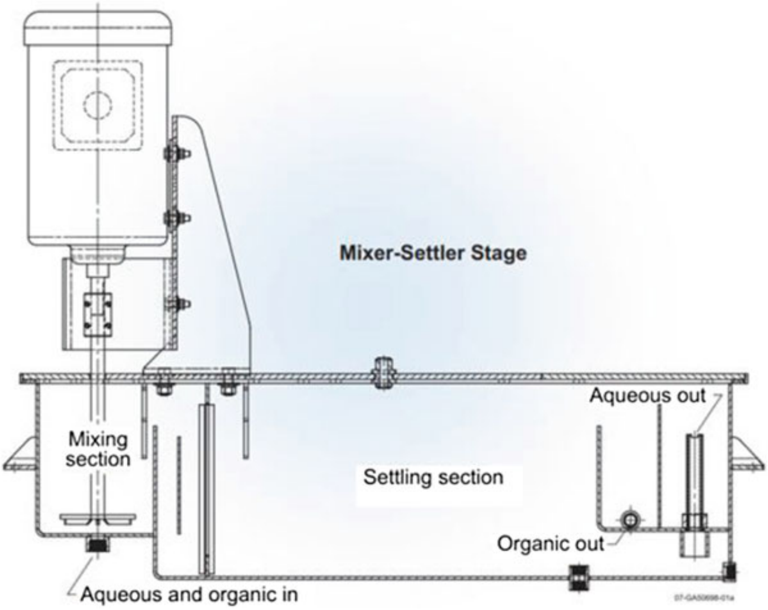

Fig. 8.5 Schematic of mixer settler separation concept. (Source: Law and Todd 2008)

certain value. For example, at any achievable concentration for highly enriched uranium nitrate, criticality is not possible if the employed cylinder diameter is less than ~15 cm (6 inch). This could be applied to vessels or any piping with the flow of fissile. Such "favorable geometry" could also be applied to locations where many vessels are in close proximity with possible interactions of neutrons between vessels. Control of criticality can also be achieved by adding soluble neutron absorber in the solution.

A mixer settler consists of a series of discrete stages of chambers. Each stage includes a mixing chamber to which the organic and the aqueous phases are pumped into for mixing. Once equilibrium is reached, they are moved to a settler chamber where the organic and aqueous phases are separated during the residence. Due to the large volume of liquids contained in the chambers, criticality control can be challenging. Therefore, the mixer-settler technique is used for spent fuel with low fissile (i.e., Pu) content. Use of neutron poisons may be needed for additional criticality control (Fig. 8.5).

The pulsed column uses hydraulic pulse mechanism for mixing. In each mixing section, the organic phases are pushed through the perforated plates while being contacted by the aqueous phase. Separation takes place at the top or bottom of the column. For one separation stage, the distance of pulsed column could be in several meters. The system requires small floor space but the structure is tall. Because of small space needed, the overall capital cost is low and the amount of inventory of solvent needed for operation is moderate compared to a mixer settler. Also, with the

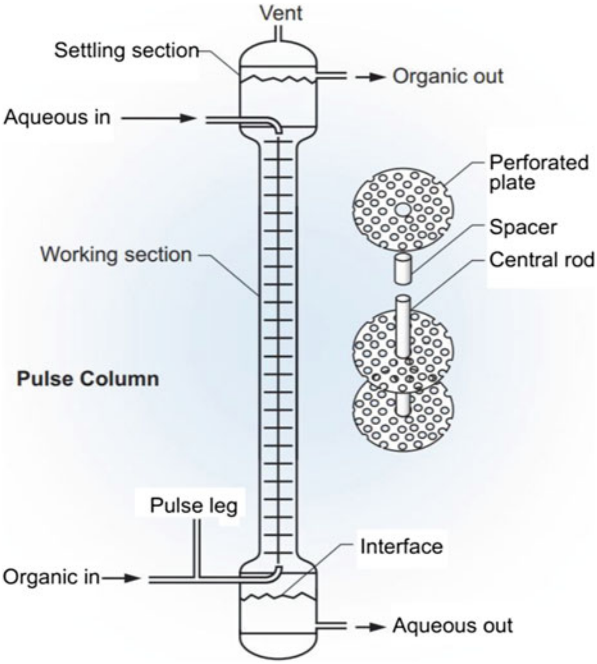

Fig. 8.6 Schematic of the pulsed column separation unit. (Source: Law and Todd 2008)

use of "favorable geometry", criticality control is easier allowing the processing of high plutonium content fuel (Fig. 8.6).

The centrifuge technique uses centrifugal force to mix the two phases. After mixing, the lighter organic phase containing uranium and plutonium moves around the rotator while the heavier aqueous phase moves toward outside, resulting in the separation of two phases. The technique requires small floor space and small solvent inventory but demands high capital cost and remote maintenance capability. The residence times are shorter compared to those of mixer settler or pulse column (Fig. 8.7).

Presence of high activity fission products and actinides presents difficulty in PUREX operations in addition to issue of criticality control. To provide radiation safety to operating personnel, the operation must be conducted within heavily shielded system with remote monitoring and control. Well-designed treatment of gaseous and liquid effluents is also needed to minimize the release of radioactive materials to the environment. The remaining fission products and actinides in the waste stream must be properly packaged and disposed of.

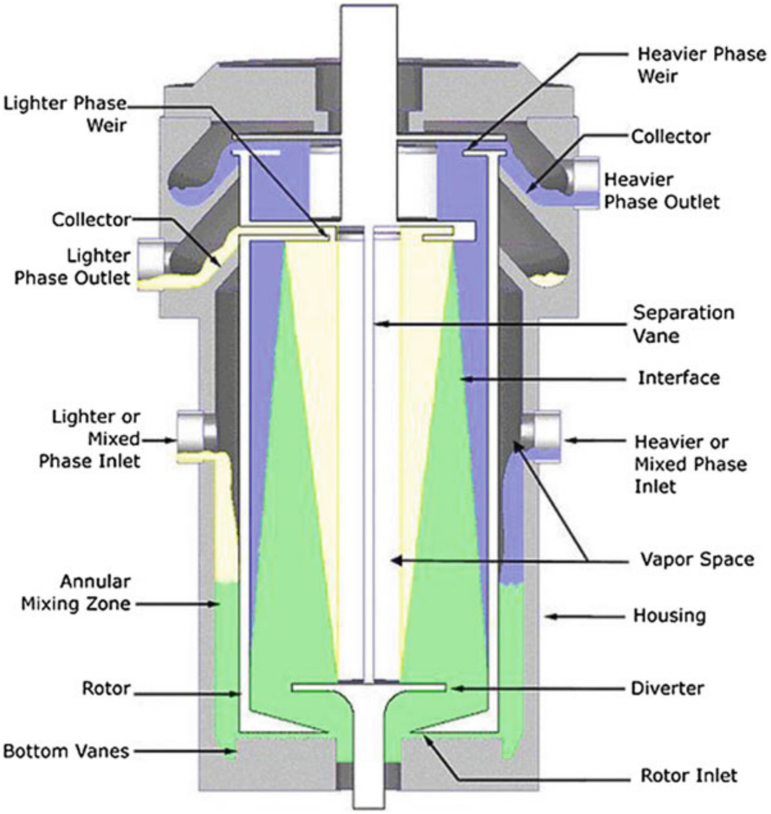

Fig. 8.7 Schematic of centrifugal contactors for solvent extraction. (Source: Wikimedia Commons 2006)

8.2.7 Treatment of HLW for Stabilization

The highly radioactive raffinate from the primary solvent extraction process in PUREX becomes high level waste (HLW). HLW is a mixture of nitric acid, more than 99% of the non-gaseous fission products, very low levels of uranium, plutonium, and virtually all of minor actinides present in the spent fuel along with activation products, corrosion products, and other chemicals added during reprocessing.

As a preparatory step for safe handling and disposal, HLW has been treated with stabilization technologies such as calcination, vitrification. These stabilization technologies involve feed preparation, waste immobilization, immobilized waste packaging, and treatment of secondary wastes.

Treatment of nuclear waste for stabilization is to provide better physical and chemical characteristics for its handling, transport, storage, and disposal of waste. With the resulting waste form (the stabilized product), HLWs are easier to handle, less dispersible during transportation and storage, and less mobile after disposal. In the end, the resulting waste is isolated from the biosphere over very long periods of time. Such long-term isolation requires not only the use of engineered barriers but also geologic isolation. These two aspects will be discussed in Chaps. 10 and 11.

8.2.7.1 Calcination

Calcination is a process in which the liquid waste is dried and converted into unconsolidated solid oxide powder or granules. The resulting granular product is called calcine.

The fluidized bed calciner has been used at Idaho Chemical Processing Plant since the 1970s for the treatment of liquid HLW. The rotary kiln calciner was also developed at the AVM (Marcoule Vitrification Facility) by France in connection with a melter to convert the calcine to glass. The calciner is a slightly sloped, externally rotated cylinder that is heated externally. As the cylinder has a slight slope, the waste is injected into the upper end and moves to the lower end, resulting in a finely divided oxide powder.

8.2.7.2 Vitrification

Vitrification is to convert HLW into a monolithic glass product. The process involves mixing of either original liquid waste or calcined waste with a glass frit. The mixture is melted in a special furnace, and the liquid glass may be cast into a mold or poured directly into a metal canister and left for cooling.

While there are alternative forms of glass for waste stabilization, borosilicate glass has been used worldwide as the product of vitrification. This will be further discussed in Chap. 9 (Sects. 9.4.1 and 9.4.5). The world's first industrial-scale vitrification operation began at AVM (Marcoule Vitrification Facility) in France in 1978 in-line with spent fuel reprocessing. The continuous glass making process implemented at the facility involves the use of an induction-heated continuous melter in which the glass calcined HLW are mixed and melted.

The French vitrification method can be summarized as,

- The HLW (liquid) sprayed into a heated chamber →
- The moisture and other volatile liquids are driven off →
- Waste material becomes calcine powder at the bottom →
- The calcine powder is mixed with glass making frit and tumbled into stainless steel canister →
- The temperature of the canister is raised (1100 °C) and the frit turns to molten glass →
- Molten glass is poured into a stainless canister and cools into a glass log →
- The canister moves remotely from the furnace to an automated welding machine where an end-cap is remotely welded on →

- The canister is placed in an overpack of cast steel →
- The canister with overpack is placed inside a metal or ceramic corrosion barrier (thus, ready to be placed in a repository)

Different types of vitrification operations have been developed. Most of them include calcination of HLW followed by mixing with glass frit for melting for glass making. The resulting glass melt can be poured continuously into canisters. When the glass melt in the canister cools, it become a monolithic glass product. An alternative is to mix the calcined powder with glass frit to be poured into a storage canister. The canister is then heated to 1000–1100 °C and cooled again to produce a monolithic glass product. The other alternative of vitrification is not to involve the step of calcination. In this case, liquid HLW is directly mixed with glass frit and goes through continuous melting and pouring into canisters for HLW glass production.

Generation of HLW in the U.S. started from the operations of Pu production reactors in Hanford, Washington for military purposes. A decision to build the first reactor was made in January 1943. The first reactor started the operation in September 1944. Total number of Pu production reactors built at the site was nine by 1963. The initial operation of spent fuel reprocessing was based on Bismuth phosphate precipitation which produced the plutonium used in Nagasaki on August 9, 1945. This operation was replaced by Redox (in 1951) and then by PUREX (in 1956).

To store the HLW from the reprocessing operation, single-shell mild steel storage tanks were built at Hanford in 1944. This continued at Hanford with the construction of last group of single-shell tanks in 1964 which went to service in 1966. In 1953, the U.S. Geological Survey gave warning that integrity of Hanford tanks is doubtful. During this period (in 1957), a panel of experts convened by National Academy of Science recommended that HLW be buried in bedded salt formations. In 1959, Joint Committee on Atomic Energy held a hearing on the issue but concluded that HLW stored at Hanford tanks did not pose an insurmountable challenge. At that time, the manager of Hanford predicted that the storage tanks would remain serviceable for "decades" and possibly as long as 500 yrs. However, it was later found that first tank leak already started in 1958 from a tank constructed in 1947. This resulted in release of 35,000 gallons of HLW. In 1965, in order to guard against further leaks from single-shell tank, in-tank solidification program was initiated.

In 1968, construction of new tanks as double-shell structure began and became operational in 1971. The program of extracting Sr and Cs from the liquid waste also begun. In 1970, groundwater monitoring well was drilled which led to detection of highly radioactive contaminants leaking from single-shell tanks. In 1972, a pumping program was started to transfer liquid from single to double-shell tanks. DOE stopped placing HLW in single-shell tanks in 1980. In the meantime, the largest release event took place in 1973 from tank 106-T releasing 115,000 gallons of HLW. Hanford's reprocessing operations ended in 1989 with a total of 2 million m^3 (or 525 million gallons) of HLW produced and stored in 177 tanks (149 sing-shell tanks and 28 double-shell tanks). Each tank contained about 1 million gallon of HLW. This is about two-thirds of the total HLW generated in the U.S. by volume.

According to the estimation by U.S. DOE, definite or possible leaks had occurred from 1959 through 1988 in 66 of the 149 single-shell tanks. These leaks resulted in releasing about 750,000 gallons of HLW. DOE transferred all of the remaining liquid HLW from the single-shell tanks to double-shell tanks. Moreover, DOE has built the Hanford Waste Vitrification Facility (call Hanford Vit Plant) which is under operational test in 2018 with target operation of the plant by the end of 2023. The HLWs in double-shell tanks will be vitrified at the plant for final disposal. The expected total cost of HLW removal and stabilization at Hanford is about $72.3 billion.

Another facility for spent fuel reprocessing built in the U.S. was the Savannah River Plant (SRS). It was part of the U.S. crash program to build the hydrogen bomb against the Soviet Union who conducted its first fission bomb nuclear test explosion on August 29, 1949. The Savannah River Plant was built between 1950 and 1952 with five plutonium production reactors. First HLW generation from the spent fuel reprocessing occurred in 1954. The resulting HLWs were neutralized and stored in mild steel tanks; tanks were isolated from environment by concrete vault. Its operation ended in 2011 but resumed for reprocessing of spent fuels from High Flux Isotope Reactor (HFIR) in 2016 with the operation expected to continue through 2024.

Approximately (100 million gallons) of HLW were produced at SRS (this volume was reduced to about 34 million gallons by removal of excess water through evaporator processing operations) and were stored at 51 large carbon steel tanks with 4 different designs. These include 24 single-shell tanks (12 Type I tanks, 4 Type II, and 8 Type IV) and 27 of double-shell Type III tanks. None of the tanks was equivalent to Hanford single-shell tanks. Each tank stored about ~750,000 to 1.3 million gallons. Seven of the 12 Type I tanks, all four of the Type II tanks, and two of the eight Type IV tanks have leaked with minor consequences. One tank (Tank 16, a Type II tank) was the only tank having a release of waste resulting in a few tens of gallons of waste escaped to the soil. The tank was cleaned and decommissioned and the contaminated soil was remediated. The waste from single-shell tanks were transferred to double-shell tanks in mid 1990s. For the immobilization of HLW, the Defense Waste Processing Facility (DWPF) was constructed in 1983 and began operation in March 1996 producing borosilicate glasses. The facility is projected to produce approximately 8170 canisters of vitrified HLW.

Another facility where spent fuel reprocessing was performed in the U.S. was Idaho Chemical Processing Plant. The plant was to reprocess spent fuels from U.S. Navy nuclear propulsion reactors for recycling of HEUs as fuel. The plant's construction began in 1950 with its operation starting in 1953 and its reprocessing operation stopped in 1992. Approximately 31,432 kg of uranium had been recycled. Every kilogram of purified uranium produced, on average, over 100 gallons of HLW. These wastes were maintained in an acidic solution (without neutralization) and stored in stainless steel tanks. A total of eleven 300,000 gallon tanks were built along with four smaller 30,000 gallon tanks. Starting in 1963, all of the HLW were stabilized into solid through the calcination process. This eliminated the need for long-term storage of liquid waste and reduced the volume by more than nine-fold.

With the waste as solid, it was also easier to handle and retrieve the waste if necessary. The calcined products were stored in underground storage bins made of stainless steel and will be made into glass for final disposal.

The West Valley reprocessing facility represents the only commercial reprocessing plant operated in the U.S. Construction of the facility began in 1963 with the reprocessing operation starting in 1966. The HLW generated was neutralized and stored in carbon-steel tanks. The West Valley was closed in 1972, after reprocessing 600 MT of fuel. This resulted in 600,000 US gallons of HLW which were stored in two of underground storage tank. There was no leakage involved. Vitrification of the waste began in 1999 and completed in 2002. The vitrified waste is stored on site awaiting final disposal.

8.2.8 Modifications of PUREX

One of the key concerns in the utilization of PUREX is the presence of pure plutonium as the separation product. Presence of pure plutonium product presents the risk of potential diversion for the production of nuclear weapons. Careful nuclear materials accounting and implementation of surveillance programs must be in place with close cooperation with International Atomic Energy Agency (IAEA). This issue is further discussed in Sect. 15.2.

To avoid the production of readily weapon usable nuclear materials, efforts have been made to modify the PUREX process. Such efforts are based on co-separating Pu with a contaminating material thus to complicate the use of plutonium for nuclear explosives. Such co-separation can be possible through the changes in the system chemistry.

One example of such approach is the UREX process (uranium extraction). The UREX process uses an organic reagent, aceto-hydroxamic acid (AHA), along with TBP. AHA is a reductant/complexant and forms complexes with Pu and Np and reduces them to inextractable Pu(III) and Np (V). So in the extraction stage, only U is extracted into the organic phase and Pu and Np remain in the aqueous phase. Then, plutonium and neptunium are extracted together in a later downstream step. As plutonium is always mixed neptunium, it becomes much less attractive for nuclear explosives. Also, presence of neptunium provides a strong gamma-ray signature thus increases real-time detection capability of plutonium movements. The flowsheet of UREX is shown in Fig. 8.8 covering fuel dissolution, extraction, scrubbing, product purification, and waste treatment.

A challenge with the UREX process is that AHA degrades over time (within several hours to a few days). The degradation products, such as acetic acid and hydroxylamine nitrate, can interfere with subsequent chemical processes. Also, the possibility of potential misuse still remains if intentional change of solvents in the UREX process is exercised for pure plutonium separation.

The UREX+ process is an extension of UREX, as an advancement. In UREX+, additional steps of selective removal of various elements are performed to enhance

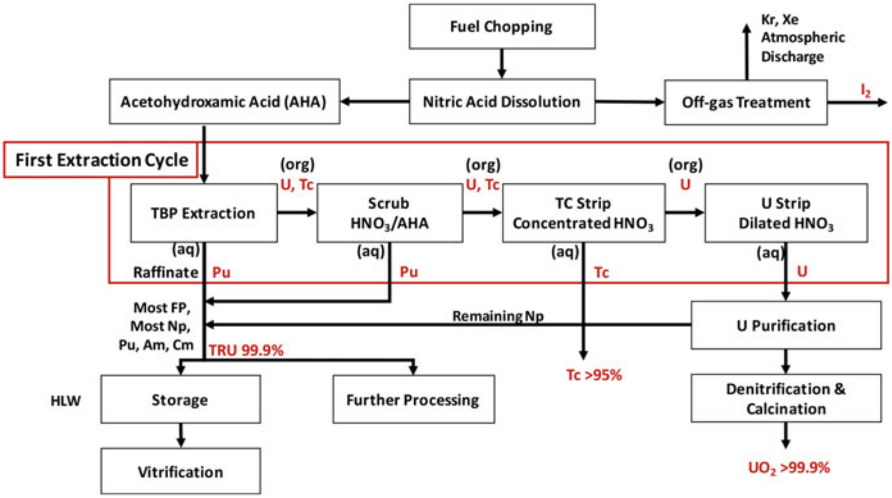

Fig. 8.8 A simplified flow sheet of the UREX process

the overall efficiency of spent fuel management. Such selective removal is to reduce the demands placed on final waste disposal and to minimize the resulting human health risk. The elements removed by UREX+ include uranium, iodine, technetium, cesium, strontium, plutonium, neptunium, americium, and curium.

Separation of uranium significantly reduces the mass and volume of the material to be disposed of as high level waste as uranium makes up the vast majority of the mass and volume of spent fuel. Uranium can then be recycled into fuel or disposed of as low level waste. Iodine-129 (^{129}I) and technetium-99 (^{99}Tc), the long-lived fission products, represent key contributors to dose to individuals from the disposal. With the removal of ^{129}I and ^{99}Tc, the projected long-term health effects from spent fuel disposal can be significantly reduced. ^{137}Cs and ^{99}Sr are the key contributors to the decay heat inventory of spent fuel in the near-term (within several hundred years). With ^{137}Cs and ^{90}Sr removed, the amount of heat that needs to be handled in a storage or disposal facility is drastically reduced. Plutonium, neptunium, americium, and curium are the nuclides that can be reused in the fuel for energy generation through refabrication into nuclear fuel and recycling in a reactor. Am-241, plutonium, and curium are also the key decay heat producers.

The UREX+ process consists of five unit processes of solvent extraction in series (see Fig. 8.9) for the separation of the above listed elements. These processes include: (1) UREX (where uranium and technetium are removed); (2) FPEX or CCD-PEG (where cesium and strontium are removed); (3) NPEX (where plutonium and neptunium are removed); (4) TRUEX (where americium, curium, and rare-earth-fission-product are removed), and; (5) TALSPEAK or Cyanex-301 (where americium and curium are separated from the rare earths). Americium can also be separated from curium through additional steps.

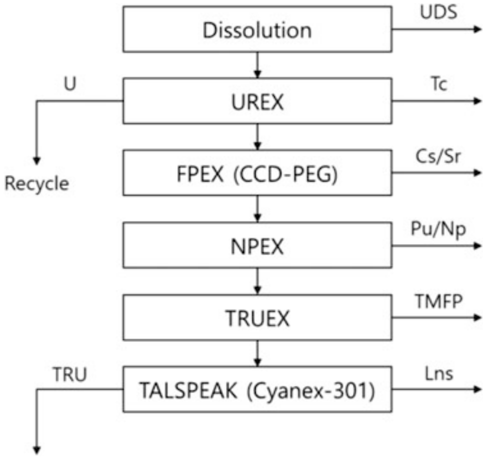

Fig. 8.9 A simplified flow sheet of the UREX+ process (UDS: undissolved solids; TMFP: transition metal fission products; Lns: lanthanides)

In connection with spent fuel dissolution in nitric acid, these processes are performed by using different solvents: TBP in UREX and NPEX, a mixture of chlorinated cobalt dicarbollide (CCD) and polyethylene glycol (PEG) in FPEX or CCD-PEG, carboylmethyl-phosphine oxide and TBP in TRUEX, and di-(2-ethylhexyl) phosphoric acid (HDEHP), lactic acid and diethylenetriamine-pentaacetic acid (DTPA) for TALSPEAK.

The separated technetium is converted to a metallic waste form for long-term disposal. The separated cesium and strontium are immobilized in a ceramic waste form for extended storage. Plutonium, neptunium, americium, and curium are recycled through MOX fabrication. The remaining waste from the processes is classified as HLW for final disposal.

8.3 Pyroprocessing

Pyroprocessing has emerged as an alternative method of spent fuel reprocessing. By noting some of the benefits (described in Sect. 8.1.4) in comparison to the PUREX process, active R&D effort is underway. No commercial scale of operating pyroprocessing plant exists yet,.

8.3.1 Electrochemical Cell as the Separation System

Separation of elements in pyroprocessing is based on partitioning of elements between one or more metal phases and a molten salt phase. Molten salt in pyroprocessing works both as solvent and electrolyte with high radiation resistance. As a solvent, molten salt provides fast kinetics for metal dissolution. As an electrolyte, molten salt allows electro-transport and separation of constituents of spent fuel. Along with molten salt, liquid metal is also used as a reacting media for the separation of different classes of materials again with high radiation resistance and fast kinetics. Chemical species, upon dissolution, exist as ions in either molten salt or liquid metal, or both, depending on thermodynamic stability of the compounds.

Typically chlorides are employed as molten salt. The relative stabilities of elements in the chloride system permit distinctive grouping of them into different classes for separation. The most commonly used chloride system is a mixture of lithium chloride and potassium chloride as a eutectic mixture (LiCl-KCl; 59.5:40.5) system. A eutectic mixture is a homogeneous mixture of two or more chemical substances having a lower melting point than that of any of its components. These substances usually do not interact to form a new chemical compound. At the same time, their mixture inhibits the crystallization process of one another (making them easier to melt), at certain specific ratio, resulting in lower melting point than either of the compounds. Therefore, the required energy consumption is lower with eutectic mixture to operate the molten salt system.

Fluorides can be also be used as molten salt electrolye in pyroprocessing. Compared to chlorides, the Gibbs free energy of formation between plutonium and other actinides neptunium is larger with fluorides. This means separation of plutonium from other actinides is more difficult in the chloride system. For example, the Gibbs free energy of formation of chlorides for plutonium ($PuCl_3$) and neptunium ($NpCl_3$) is −62.4 and −58 (kcal/mol), respectively at 500 °C. In the case of fluorides, the Gibbs free energy of formation for plutonium (PuF_3) and neptunium (NpF_3) is −456.314 and −440.856 (kcal/mol), respectively at 525 °C. With smaller difference in the free energy of formation between plutonium and neptunium in the chloride system in comparison to the fluoride system, the chloride system would be preferred over the fluoride-based one from the point of proliferation resistance. In terms of the cost of operation, the LiCl-KCl molten salt has the melting point of 350 °C compared to 454 °C of LiF-NaF-KF, 460 °C of $LiF\text{-}BeF_2$, and 769 °C of LiF-CaF2 molten salt. Therefore, the operational cost of using LiCl-KCl as molten salt would be lower than the system using fluorides. Most of the current efforts in pyroprocessing development are based on the use of LiCl-KCl.

8.3.2 Head-end Process and Oxide Reduction

As in the case of PUREX, the head-end process of pyroprocessing includes fuel mechanical chopping and off-gas treatment. If the fuel is in oxide form, conversion to metallic form is also necessary prior to fuel dissolution. For such conversion in the head-end process, ceramic oxide fuels are first physically treated, to be in the form of porous pellets or fragments. These porous oxides or fragments are then reduced to metal by direct ionization or reaction with lithium in a bath of molten lithium chloride.

8.3.3 Electrorefining

Along with thermodynamic stabilities of elements in the electrolyte, electrotransport (ions moving in an electrolyte in response to an electric field) drives the basic process of separation in pyroprocessing. This step of separation is called electrorefining, i.e., electrolysis-based refining. To support eletrorefining operations, an electrochemical cell is constructed with an anode (made of impure metal, i.e., reduced spent fuel as metallic form), a cathode (made of a solid and/or liquid metal), and an electrolyte (made of molten salt or liquid metal).

General anode and cathode reactions are as below.

$$\text{Anode process} : M^{0}\ (\text{metal}) \rightarrow M^{Zi}\ (\text{molten salt}) + z_i\ e \tag{8.6}$$

$$\text{Cathode process} : M^{Zi}\ (\text{molten salt}) + z_i\ e- \rightarrow M^{0}\ (\text{metal}) \tag{8.7}$$

where z_i is the charge number of species i.

The reaction at an anode involves removal of z_i electrons per atom from metal M. Thus the M^{Zi} ions are formed in the salt phase. The removed electrons are conducted through an external circuit to the cathode, where the metal is formed. As cathode, carbon steel is used for uranium separation and liquid cadmium is used for plutonium and actinides co-separation

The fuel in metallic form as small pieces are placed in a perforated steel basket which works as an anode for electrochemical operations. Therefore, the container is called “anode basket”. The anode is submerged into molten LiCl-KCl eutectic salt in an electrorefiner vessel at high temperature (500 °C, 773 K). The passage of electric current causes the dissolution of metal through oxidation.

In the eletrorefining cell, liquid metal bath lies at the bottom underneath molten salt to allow species to distribute between the two reacting media. Once dissolved, the distribution of elements in the electrorefiner pool is determined by thermodynamic stability of elements given by the Gibbs free energy of formation (see Eq. 8.9 below).

During the electrorefining operation, alkaline (e.g., Li, Na, K, Rb, Cs), alkaline earth (e.g., Mg, Ca, Sr, Ba) and lanthanide elements (e.g., La, Ce, Nd, Gd, Sm, Eu) tend to distribute in the molten salt phase. This is because these elements are most stable in molten salt. Noble and transition metal elements (e.g., Cr, Mn, Fe, Co, Ni, Y, Zr, Nb, Tc, Ru, Rh) tend to reside in the liquid metal as they are most stable in liquid metal. Actinide elements (e.g., U, Np, Pu, Am, Cm) tend to distribute in both molten salt and liquid metal as the thermodynamic stabilities of actinides are located between those of fission product lanthanides and transition metals. Therefore, these elements are somewhat unstable in both media. These distributions of elements in molten salt or liquid metals are depicted in Fig. 8.12.

Partitioning of elements between the liquid metal and molten salt phase (in the electrorefiner pool) is described by the following reaction. This reaction is rapid under the given temperature (~500 °C) of the system.

Table 8.2 Free energies of formation of fission products, actinides, and electrolyte chlorides at 500 °C (ΔG_f° is in kcal/mol of chlorine)

Relatively stable (salt phase)		Electrotransportable		Relatively unstable (metal phase)	
Compound	Free energy	Compound	Free energy	Compound	Free energy
CsCl	−87.8	$AmCl_3$	−64	$CdCl_2$	−32.3
KCl	−86.7	$CmCl_3$	−64	$FeCl_2$	−29.2
$SrCl_2$	−84.7	$PuCl_3$	−62.4	$MoCl_2$	−16.8
LiCl	−82.5	$NpCl_3$	−58	$TcCl_3$	−11.0
NaCl	−81.1	UCl_3	−55.2		
$LaCl_3$	−70.2	$ZrCl_4$	−46.6		
$PrCl_3$	−69.0				
$CeCl_3$	−68.6				
$NdCl_3$	−67.9				
YCl_3	−65.1				

Data sources: Pankratz (1984) and IAEA (1983)

$$yMCl_x + xN \leftrightarrow yM + xNCl_y \tag{8.8}$$

where, *M* and *N* are different metallic elements; *x* and *y* are the oxidation states of the stable chlorides of *M* and *N*, respectively. The free-energy change (*ΔG*) in the reaction is

$$\Delta G = x\Delta G_f^\circ(NCl_y) - y\Delta G_f^\circ(MCl_y) \tag{8.9}$$

where ΔG_f° is the Gibbs free energy of formation from the pure elements in their standard states. If *ΔG* is negative, the reaction proceeds spontaneously to the right, putting the metal *N* in the chloride form. If *ΔG* is positive, the metal *N* would remain in the metal phase. Selected values of free-energy of formation (ΔG_f°) of chlorides are given in Table 8.2.

The differences in the free energy of formation of the selected chlorides are also graphically represented in Fig. 8.10.

The equilibrium constant for the reaction can be represented as a function of the free energy change,

$$K_{eq} = e^{-\Delta G/RT} \tag{8.10}$$

with *R* as the gas constant. When the reaction is at equilibrium, concentrations of the metals and their chlorides can be determined from the equilibrium constant and the activity coefficients of metal and chloride species. This is shown by the following relationship.

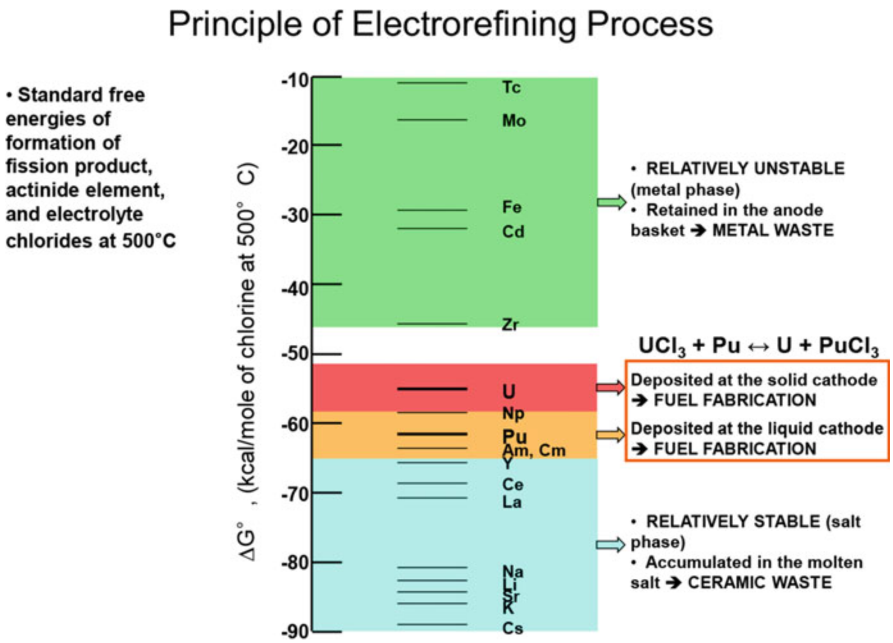

Fig. 8.10 Standard free energies of formation with chlorides at 500 °C through electrorefining. (Bobolea, 2009)

$$K_{eq} = \frac{(a_M)^y \left(a_{NCl_y}\right)^x}{(a_N)^x (a_{MCl_x})^y} = \frac{(N_M)^y \left(N_{NCl_y}\right)^x}{(N_N)^x (N_{MCl_x})^y} \frac{(\gamma_M)^y \left(\gamma_{NCl_y}\right)^x}{(\gamma_N)^x \left(\gamma_{MCl_x}\right)^y} \tag{8.11}$$

where, the α_i is the activity, the N_i is the mole fraction, and the γ_i is the activity coefficient of the species i, respectively .

Equation 8.11 can be rearranged to represent the distribution of a substance in molten salt and the liquid metal (Johnson 2001).

$$\frac{\left(N_{NCl_y}\right)^x / (N_N)^x}{(N_{MCl_x})^y / (N_M)^y} = K_{eq} \frac{(\gamma_N)^x (\gamma_{MCl_x})^y}{(\gamma_M)^y \left(\gamma_{NCl_y}\right)^x} \tag{8.12}$$

Use of this relationship is further explained in Example 8.3 for the distribution of U and Pu in molten salt and the liquid metal. Table 8.3 shows the activity coefficients of actinides and rare earths at infinite dilution in liquid cadmium and molten salt.

The dissolved metal in molten salt forms metal chloride and is transported by electric current toward a cathode and gets deposited on the cathode through reduction. Thus the target metals are separated from a mixture through deposition on a cathode. This is based on oxidation and reduction: Oxidation at an anode reduces the

Table 8.3 Activity coefficient of actinides and rare earths at infinite dilution

Element	Activity coefficient in liquid Cd	Chloride	Activity coefficient in LiCl-KCl
U	75	UCl_3	5.79×10^{-3}
Np	8.2×10^{-3}	$NpCl_3$	–
Pu	1.38×10^{-4}	$PuCl_3$	6.62×10^{-3}
Am	$\approx 2 \times 10^{-6}$	–	–
Cm	$\approx 3 \times 10^{-5}$	–	–
Ce	9.76×10^{-9}	$CeCl_3$	1.5×10^{-3}
La	3.58×10^{-9}	$LaCl_3$	4.7×10^{-3}
Pr	1.8×10^{-8}	$PrCl_3$	3.3×10^{-3}
Nd	$\approx 6 \times 10^{-9}$	$NdCl_3$	1.8×10^{-2}
Y	–	YCl_3	6.3×10^{-6}

Data source: Park (1999)

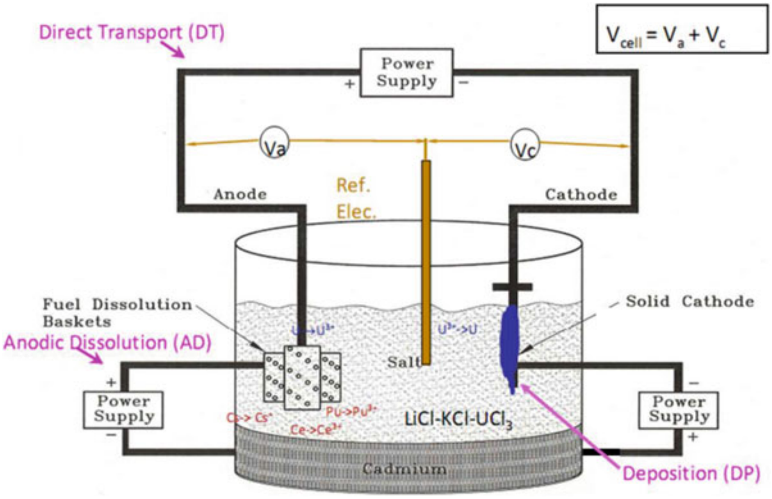

Fig. 8.11 Electrorefining process for spent fuel with a solid cathode. (Source: Simpson 2008)

quantity of the corresponding metal and increases the amount of metal chlorides in the salt. Reduction at a cathode removes metal chlorides from the salt and deposit the corresponding metals in a metallic phase onto the cathode. Through these reactions, uranium and the actinides elements are separated from the fission products present in the spent fuel and collected at cathodes. This process of electrorefining is captured in Fig. 8.11 where the cathode used for the collection of uranium is a solid cathode. Note, electric current is the flow of charge through surface. Electric current is generated as electrons move in an electrolyte in response to an electric field.

The amount of metals collected at the cathode is determined by the amount of charge passed. The amount of electric charge passed is the product of the electric current and time. In fact, the relationship for the amount of cathodic deposition is directly proportional to the molar mass of the element, the applied electric current, and the time of operation as represented below. This is called Faraday's first law of electrolysis:

$$m = \frac{M \cdot Q}{n \cdot F} = \frac{M \cdot I \cdot t}{n \cdot F} = \text{Constant} \cdot I \cdot t \tag{8.13}$$

where, *m* is the mass of metallic element deposited on the cathode, *M* is the molar mass of the element, *Q* is the amount of charge passed, *I* is the value of the applied electric current, *t* is the operation time, *n* is the number of electrons involved in the reaction, and *F* is Faraday's constant (96,485 C/mol). The unit of current is ampere (A) and 1 ampere is equivalent of 1 coulomb (C) of charge moving in one second.

Example 8.2: Amount of Electrotransported Actinitide Show that about 3 g of actinides are electrotransported to the cathode per ampere-hour.

Solution:

Since most of the actinides in spent fuel is uranium, the molar mass of the actinides can be assumed as the molar mass of uranium (238 g/mol).

Therefore, according to the Faraday's first law of electrolysis, the amount of electrotransported actinides is as follows, assuming the actinides deposited on the cathode are mostly uranium with $n = 3$ (from the presence of UCl_3 in the molten salt):

$$\mathrm{m} = \frac{M \cdot Q}{n \cdot F} = \frac{M \cdot I \cdot t}{n \cdot F} = \frac{238(g/mol) \cdot 1(C/s) \cdot 3600(s)}{3 \cdot 96485(C/\mathrm{mol})} \cong 3\ g$$

Distribution of elements in the salt and the metal phases will also depend on the cell chemical conditions. If cell condition is changed to more oxidized (e.g., by adding a chloride of relatively positive free energy of formation, such as $CdCl_2$), actinides are more likely to be in molten salt. If the condition becomes more reduced (e.g., by adding a metal, such as lithium), actinides are more likely to be in liquid metal.

In order to ensure co-collection of plutonium with the minor actinides (Am, Np, and Cm) for proliferation resistance, plutonium and minor actinides are electrotransported to liquid metal, where stabilization of actinides occurs by formation of intermetallic compounds with the liquid metal. In this case, activities of plutonium and minor actinides are greatly reduced.

Liquid cadmium is currently used for such purpose as cathode. In this case, plutonium activity coefficient is reduced from 1.38×10^{-4} to 4×10^{-6} indicating plutonium's activity is much reduced in the plutonium-cadmium intermetallic compound. Here a reduction in a metal's activity is equivalent to a reduction in the stability of the corresponding chloride or a reduction in the magnitude of free energy of formation of chloride. The free energy of formation of plutonium chloride, in this case, is lowered from -62.4 kcal/mol to -56 kcal/mol. Accordingly, plutonium and

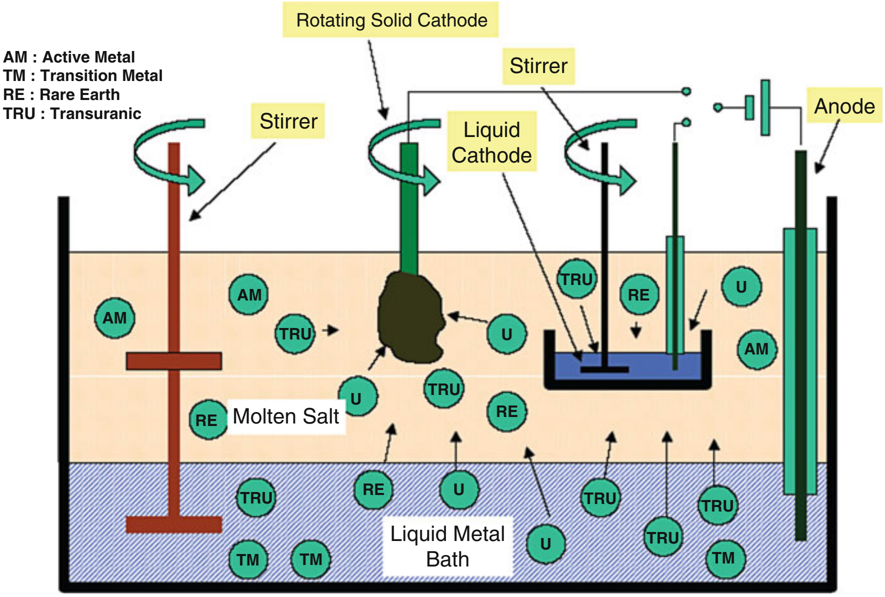

Fig. 8.12 Electrorefining process for spent fuel with the use of both solid cathode and liquid cathode. (Source: Park, 1999)

other actinides are always co-deposited on the surface of the liquid cadmium cathode along with some rare-earth fission products.

An example of electrorefining process using a liquid cathode is shown in Fig. 8.12. The crucible containing liquid cathode is suspended in the salt phase, and electrical contact is made to the liquid cathode. Electrotransport of actinides can be carried out either directly between the anode and the cathodes or through a liquid metal pool. In the latter case, the actinide elements in the fuel are first transported to a liquid metal pool (typically liquid cadmium) located at the bottom of the electrorefiner vessel and then electrotransported to the product-collection cathodes.

Example 8.3: Distributions of Key Elements in an Electrorefiner System In the electrorefining process, the distribution of elements is dictated by their thermodynamic properties. Based on the data given in Tables 8.2 and 8.3, determine how U and Pu are distributed between the liquid cadmium and the molten salt at equilibrium.

Solution:

To describe the distribution of U and Pu in molten salt and the liquid metal, consider the following reaction.

(continued)

Example 8.3 (continued)

$$UCl_{3(\text{in salt})} + Pu_{(\text{in liquid metal})} \leftrightarrow U_{(\text{in liquid metal})} + PuCl_{3(\text{in salt})}$$

From Eq. 8.12,

$$\frac{PuCl_3/Pu}{UCl_3/U} = K_{eq} \cdot \frac{\gamma_{Pu}}{\gamma_U} \cdot \frac{\gamma_{UCl_3}}{\gamma_{PuCl_3}}$$

The ratio UCl_3/U can be considered as the distribution coefficient of uranium (D_U): It is the ratio of the mole fractions in the molten salt solution to the mole fractions dissolved in the liquid metal solution.

The equilibrium constant can be calculated from Eqs. 8.9 and 8.10.

The standard Gibbs energy change for the reaction is

$$\Delta G = \Delta G_f^{\circ}(PuCl_3) - \Delta G_f^{\circ}(UCl_3)$$

At 298.15 K, this becomes

$$\Delta G = -213.296 - (-189.910) = -23.386(\text{cal/mol})$$

At 773.15 K (500°C) (Johnson 2001),

$$\Delta G = (-213.171 + 38.0T) - (-193.659 + 40.413T) = (19.512 - 2.413T)$$
$$= -21.378(\text{cal/mol})$$

(therefore, the reaction will proceed to the right spontaneously).

Then, by using Eq. 8.11, $K_{eq} = e^{-\Delta G/RT} = 1.1 \times 10^6$

In the case of the molten salt phase, the activity coefficients of U and Pu in the molten salt are 5.79×10^{-3} and 6.62×10^{-3}, respectively (Table 8.3). Assuming the liquid cadmium an ideal solution, the activity coefficients of U and Pu are 1 in the liquid cadmium.

$$\frac{D_{Pu}}{D_U} = K_{eq} \cdot \frac{\gamma_{Pu}}{\gamma_U} \cdot \frac{\gamma_{UCl_3}}{\gamma_{PuCl_3}} = 1.1 \times 10^6 \cdot \frac{1}{1} \cdot \frac{5.79 \times 10^{-3}}{6.62 \times 10^{-3}} = 0.96 \times 10^6$$

Therefore, as long as any UCl_3 remains in the molten salt, Pu will remain in the molten salt phase (i.e., Pu is not in the liquid metal phase) while uranium is electrotransported to a solid cathode.

(continued)

Example 8.3 (continued)

Now, when both uranium and plutonium are reacting and dissolved in liquid cadmium solution, the activity coefficients of U and Pu in liquid cadmium are 75 and 1.38×10^{-4}, respectively (as shown in Table 8.3).

$$\frac{D_{Pu}}{D_U} = K_{eq} \cdot \frac{\gamma_{Pu}}{\gamma_U} \cdot \frac{\gamma_{UCl_3}}{\gamma_{PuCl_3}} = 1.1 \times 10^6 \cdot \frac{1.38 \times 10^{-4}}{75} \cdot \frac{5.79 \times 10^{-3}}{6.62 \times 10^{-3}} = 1.8$$

Therefore, Pu is separated from U with the separation factor of 1.8 in the liquid cadmium.

8.3.4 Cathode Processing and Waste Treatment

The collected metals in the cathode from the electrorefining operation need to be processed to separate out the target metals while the deposits are mixed with chloride salts. To remove salts and recover the target metals, the cathode is loaded into a crucible and goes through a distillation process in a high-temperature (1200 °C) vacuum furnace. This process is called cathode processing. By using this high temperature processing, any contaminant including salt included in the cathode deposits is removed through vaporization. These vaporized salt materials become the distillates which are collected and treated for recycling into the electrorefiner. The metal deposits on the cathode are consolidated by melting and solidified into metal ingots.

The electrorefining and cathode processing operations must be performed remotely in a highly shielded hot cell facility due to the presence of large amount of fission products as irradiation sources.

At the end of operations, the waste from pyroprocessing includes salt waste, metal waste, and off-gases. According to the operational procedures adopted at Idaho National Laboratory, the salt waste goes through crushing and blending with zeolite and glass frit to be solidified into a ceramic waste form for final waste disposal. Metal wastes containing trace of actinides generated from the head-end process are consolidated into a metallic ingot for disposal. These metal wastes are comprised of cladding hulls and fuel structural parts. If the treated spent fuel is in an oxide form, cesium and strontium as high heat generating nuclides could be separated from the oxide reduction process and be treated into a waste form for long-term (in the order of 300 years) storage. Iodine and technetium that are released from the head-end processes can also be collected from the off-gases and treated for immobilization into a waste form for very long-term disposal. As pyroprocessing has not been commercially demonstrated, some of these works are still underway. Note that, in comparison to the waste generation from the PUREX process, the volume of wastes generated from pyroprocessing is much smaller.

8.4 Transmutation

Transmutation (or nuclear transmutation) is another way of HLW treatment. It is a way of rendering HLW less hazardous through conversion of constituent nuclide (s) by nuclear reaction. The reaction is through bombardment of subatomic particles such as neutrons or protons on the target nuclide(s).

For example, ^{129}I disintegrates into stable ^{129}Xe with a half-life of 1.57×10^7 years. If ^{129}I could absorb one neutron, it becomes ^{130}I which has a half-life of only 12.36 hrs and disintegrates to a stable ^{130}Xe. Similarly, ^{99}Tc could absorb a neutron and become ^{100}Tc, which undergoes radioactive decay into a stable ruthenium within minutes. The process can be considered a way of providing aids to natural decay by effectively shortening half-lives.

Most commonly, the proposed transmutation is based on neutron-induced nuclear reactions. The neutrons produced in nuclear reactors or accelerators are utilized as a projectile. Transmutation accomplished by a neutron being captured in an undesirable isotope can be described as:

$$^{A}X + {}^{1}_{0}n \rightarrow {}^{A+1}X \tag{8.14}$$

The new isotope ($^{A\,+\,1}X$) may:

- be stable
- have a shorter half-life
- spontaneously fission into fission products
- be subsequently transformed by further neutron captures

For the above reaction to be realized, the candidate nuclide must be separated from nuclear waste and incorporated into a target for neutron bombardment. The candidate nuclide must therefore possess relatively large cross section for the relevant nuclear reaction. As transmutation requires significant preparatory work, the nuclide must be of significant importance with respect to the long-term health risk from nuclear waste to justify the necessary cost.

According to these considerations, the candidate nuclides for transmutation include the actinides (neptunium, plutonium, americium and curium) and long-lived fission products. Table 8.4 below lists the examples of major nuclides of concern and their nuclear reaction characteristics. For example, ^{99}Tc and ^{129}I are very long-lived with high environmental mobility requiring special disposal strategies for long-term isolation from the biosphere. Carbon-14, another potential candidate as a long-lived nuclide with potentially significant health risk implications is not included as it does not have the large enough cross section for transmutation relevant nuclear reaction.

A prerequisite step for transmutation of nuclear waste is partitioning of target nuclides: The nuclide species under consideration must first be partitioned from the waste and then recovered in a form suitable for neutron bombardment. Partitioning involves the use of chemical treatment processes that are designed to extract or

Table 8.4 Candidate radionuclides in spent fuel for transmutation

Nuclide	Half life (yr)	Thermal capture cross section (c.x. in barn)	Thermal fission c.x. (b)	Thermal capture to fission ratio	Fast capture c.x. (b)	Fast fission c.x. (b)	Fast capture to fission ratio
$^{99}\mathrm{Tc}$	2.12×10^5	13.8	–	–	–	–	–
$^{129}\mathrm{I}$	1.57×10^7	3.2	–	–	–	–	–
$^{135}\mathrm{Cs}$	3×10^6	30.2	–	–	–	–	–
$^{239}\mathrm{Pu}$	24,400	58.7	102	0.58	0.56	1.86	0.3
$^{237}\mathrm{Np}$	2.1×10^6	33	0.52	63	1.7	0.32	5.3
$^{241}\mathrm{Am}$	430	110	1.1	100	2.0	0.27	7.4
$^{243}\mathrm{Am}$	7400	49	0.44	111	1.8	0.21	8.57
$^{245}\mathrm{Cm}$	8500	17	116	0.15	0.9	5.1	0.18

[a] Cross-sections for the actinides are from OECD-NEA 1999b, Table II.3, p. 148. Thermal refers to a typical thermal reactor rather than thermal neutrons at room temperature. NAS-NRC 1996 was used for Technetium-99, Iodine-129. The cross section for Tc and I is for the actual spectrum of neutrons in a pressurized water reactor (e.g. an LWR). The cross-section for Sr and Cs is for room temperature thermal neutrons (and would likely be lower for the slightly higher energy neutrons in a thermal reactor or moderated target in a fast reactor)

isolate the desired nuclides as discussed in Sect. 8.2. As to the form of target material for neutron irradiation, oxide, nitride, or metallic fuel form are candidates for actinides transmutation. For iodine, technetium, and cesium, a ceramic form is also considered.

For transmutation of actinides, the preferred nuclear reaction is fission (over radiative capture). In the fission reaction, an actinide splits into fission products. As most of the fission products are short-lived, potential hazard of the actinide is likely to be reduced after fission. In the case of a capture, an actinide will capture neutrons and becomes a heavier nuclide. The heavier actinide may undergo radioactive decay, or again capture another neutron to become even heavier, or undergo fission, depending upon the cross sections. Usually, heavier nuclides have sizable fission cross-section and thus go through additional transmutation.

The probability of fission per absorbed neutron is one way to represent the likelihood of transmutation. Figure 8.13 shows the comparison of fission probability by thermal vs. fast neutrons (as the ratio of the fission cross-section to the total cross-section (the sum of cross-sections for all reactions)). While the probability of fission is higher for fast neutrons for most actinides, $^{239}\mathrm{Pu}$ has higher fission probability per neutron absorbed for thermal neutrons than fast neutrons. Therefore, except for $^{239}\mathrm{Pu}$, using fast neutrons is much more effective for transmutation of actinides.

8.4.1 Transmutation Half-life

The time required to achieve the desired level of transmutation is of interest as a way of determining the efficiency of a transmutation process. For this purpose, the

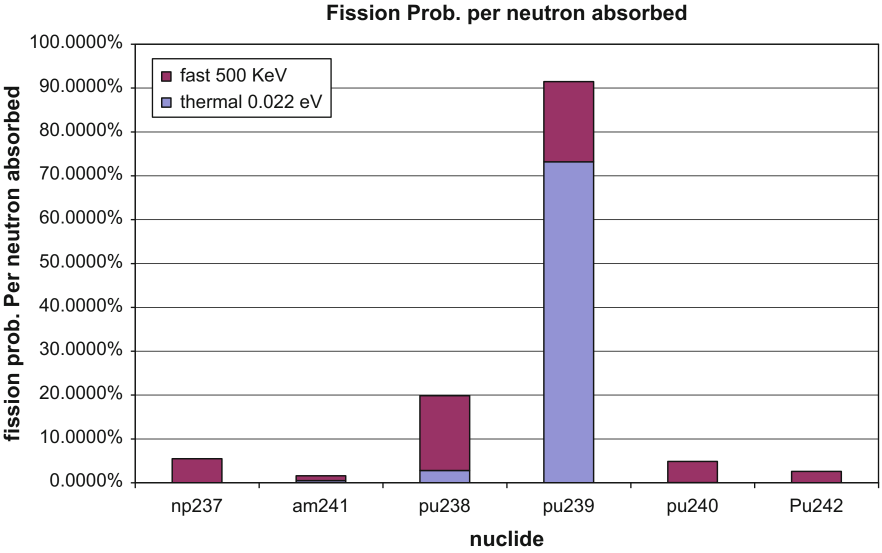

Fig. 8.13 Fission probability per neutron absorbed. (Data source: IAEA-NDS 2020)

transmutation half-life can be defined for the target nuclide of interest in a nuclear reactor system. The transmutation half-life is defined as the time required to make half of the material transmuted:

$$\mathrm{T}^{t}_{1/2} \equiv \text{transmutation half life} = \frac{\ln 2}{\sigma \phi} \tag{8.15}$$

where, σ is the cross section for the transmutation nuclear reaction and ϕ is the neutron flux.

Transmutation half-life depends on the rate of nuclear reaction of a nuclide with a neutron. The reaction rate is proportional to the reaction cross section and neutron flux. Different types (fission or capture) of cross-sections are used in the equation depending upon the type of nuclear reactions utilized for transmutation.

If the transmutation reaction is nuclear fission:

$$\mathrm{T}^{f}_{1/2} \equiv \text{fission transmutation half life} = \frac{\ln 2}{\sigma_f \phi} \tag{8.16}$$

If the transmutation is through neutron capture:

$$\mathrm{T}^{c}_{1/2} \equiv \text{capture transmutation half life} = \frac{\ln 2}{\sigma_c \phi} \tag{8.17}$$

Example 8.4: Transmutational Half-life Calculation Calculate the transmutation half life of ^{237}Np (through either fission or capture) in (1) a thermal reactor and (2) a fast reactor. Assume that the neutron flux level in the reactor is 3×10^{13} (n/cm^2−s) in the thermal reactor and 5.85×10^{15} (n/cm^2−s) in the fast reactor. Use the cross section data in Table 8.4.

1) In a thermal reactor

$\sigma_f = 0.52$ b and $\sigma_c = 33$ b

$$t^f_{1/2} = \frac{\ln 2}{0.52 \times 10^{-24}(\text{cm}^2) \times 3 \times 10^{13}(\text{cm}^{-2}s^{-1})} = 4.4 \times 10^{10}(s)$$
$$= 1.4 \times 10^3 \text{ year}$$

$$t^c_{1/2} = \frac{\ln 2}{33 \times 10^{-24}(\text{cm}^2) \times 3 \times 10^{13}(\text{cm}^{-2}s^{-1})} = 7.0 \times 10^{8}(s) = 22 \text{ year}$$

In this case, transmutation of ^{237}Np by capture produces ^{238}Np which decays (through β^- decay) to ^{238}Pu. Therefore, through transmutation, most of ^{237}Np turns into ^{238}Pu. Plutonium-238 can in turn be transmuted through fission. But the fission cross section of ^{238}Pu with thermal neutrons is very small indicating that thermal neutron based transmutation of ^{237}Np is inadequate.

2) In a fast reactor

$\sigma_f = 0.32$ b and $\sigma_c = 1.7$ b

$$t^f_{1/2} = \frac{\ln 2}{0.32 \times 10^{-24}(\text{cm}^2) \times 5.85 \times 10^{15}(\text{cm}^{-2}s^{-1})} = 3.7 \times 10^{8}(s)$$
$$= 12 \text{ year}$$

$$t^c_{1/2} = \frac{\ln 2}{1.7 \times 10^{-24}(\text{cm}^2) \times 5.85 \times 10^{15}(\text{cm}^{-2}s^{-1})} = 7.0 \times 10^{7}(s)$$
$$= 2.2 \text{ year}$$

In this case, the transmutation half-life of ^{237}Np through fission is 12 years. Also, ^{238}Pu, the product of the neutron capture reaction will go through fission with fast neutrons. Therefore, in a fast reactor, transmutation of ^{237}Np appears feasible. This serves an example of showing the feasibility of actinide transmutation in a fast reactor. For efficient transmutation of actinides in a fast reactor, higher ratio of σ_f/σ_c is desirable.

8.4.2 *Implementation of Transmutation*

The neutrons used for transmutation can be produced in either a nuclear reactor or a sub-critical accelerator system. Also, depending on the radionuclides to be transmuted, the neutrons used can be fast or thermal, or both. Therefore, both a thermal reactor and a fast reactor present opportunities for transmutation.

8.4.2.1 Thermal Reactors

The long-lived fission products such as ^{99}Tc and ^{129}I, can be transmuted to stable isotopes by the neutron capture reaction. The capture cross-sections for these nuclides are extremely small for fast neutrons but reach the level of a few barns for ^{129}I and ~ 14b for ^{99}Tc. Although these values may not be large enough for efficient transmutation, use of a high flux, i.e., increasing the number of neutrons, can make transmutation of these radionuclides feasible.

With the actinides, thermal neutrons have much higher overall reaction cross-sections than fast neutrons. Therefore, indirect transmutation through neutron capture can be pursued for long-lived actinides, such as ^{237}Np, ^{241}Am and ^{243}Am. Through capture, these actinides will form heavier nuclides, such as ^{238}Pu, ^{239}Pu, ^{242m}Am, ^{243}Cm and ^{245}Cm. These new nuclides have sizable fission cross-sections thus can be transmuted through fission.

Loading of minor actinides with high neutron capture cross-section into a reactor core for the purpose of transmutation will result in a decrease in the overall reactivity (reactivity is a measure of the state of nuclear reactor in relation to the criticality). In this case, the reactor core needs to contain higher inventory of fissile (e.g., higher fissile enrichment of the fuel to compensate for reactivity loss). During the life of the reactor core, the reactivity may also increase with the formation of fissile nuclides from radiative capture of actinides. This indicates the need for elaborate reactivity management during the core operating cycle.

8.4.2.2 Fast Reactors

While all actinides can be fissioned with fast neutrons, the overall reaction cross-sections (for both fission and capture) for fast neutrons are smaller than for thermal neutrons. This lower cross section effect is compensated by having high neutron flux in a fast reactor. In fact, fast reactors have neutron flux 100–1000 times higher than thermal reactors. This is shown in Fig. 8.14 for the case of a fast breeder reactor as an example.

Because of the differences in the cross sections, the build-up of actinides by neutron capture is much smaller in a fast reactor than a thermal reactor. Also, with fast neutrons, the fission-to-capture ratio of actinides is higher in a fast reactor. This implies more effective transmutation of actinides in fast reactors. The long-lived

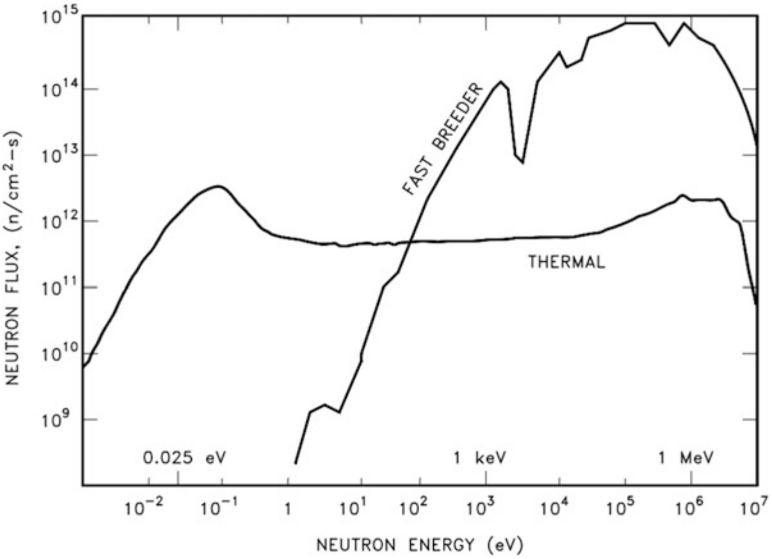

Fig. 8.14 Neutron flux spectra between a thermal reactor and a fast breeder reactor. (Source: DOE 1993)

fission products such as ^{99}Tc and ^{129}1 can also be transmuted in fast reactors by the use of moderators in the system, thus by creating thermal neutrons through fast neutron moderation.

8.4.2.3 Mixed-spectrum Reactor Concept

Given different transmutation effectiveness of thermal and fast neutrons, use of a mixed spectrum neutron system has been suggested. In this system, both fast and thermal neutrons are utilized in different annular zones: an inner zone operating with a fast spectrum and an outer zone operating with a thermal spectrum.

8.4.2.4 Accelerator Driven Transmutation Systems

When a very high energy (~ 1 GeV) proton hits a target such as lead or tungsten, a spallation reaction takes place, disintegrating the nucleus of the target atom and releasing large number of neutrons. Therefore, use of a high-energy proton accelerator with a spallation target offers capability for transmutation using the resulting neutrons. An example of this concept is shown in Fig. 8.15 (Abderrahim 2018) as an accelerator-driven waste transmutation system.

In this example, the proton beam from the accelerator hits the spallation target to generate neutrons. The generated neutrons from the target will drive the fission

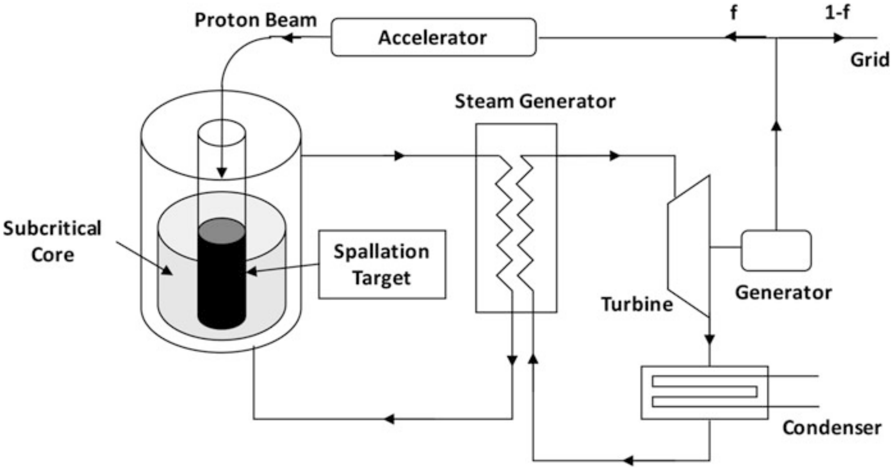

Fig. 8.15 Concept of an accelerator-driven system. (Abderrahim 2018)

reactions in the reactor core which contains the radionuclides to be transmuted. The heat from fission reactions is converted to generate electricity through the heat transfer using a coolant for steam production. Part of the generated electricity goes back to the system to support the operation of the accelerator.

Electron-based accelerator may also be considered for a similar application. However, because of the lighter weight of electron, the neutron generation efficiency for electron is much lower than proton.

8.4.3 Perspectives on Transmutation

The approach of nuclear waste transmutation requires partitioning of fission products or actinides from the wastes, followed by re-insertion of the separated target nuclide into a neutron environment for transmutation. This could significantly reduce the amounts of key long-lived, toxic species normally present in nuclear wastes. Transmutation also reduces heat loading in the waste leading to increased capacity of a geological repository (although this benefit is generic with any reprocessing treatments, see also Sect. 15.1.1.5). Transmutation also enhances utilization of resources for energy generation.

However, the time scale needed for transmutation to make an impact on nuclear waste management is usually very long. Thus the actual inventory of actinides or fission product is not reduced until a long period of time has passed. Toxicity index, the often used measure of toxicity to represent the benefit of transmutation is not a meaningful measure of the hazard of the waste in a geologic setting as the index ignores the migration and exposure aspect of the risk involved. Some of the benefits of transmutation can be realized through the current approaches of reprocessing by conducting partitioning of elements and using decay-in-storage. Also, implementation of transmutation is expected to be very costly.

Nevertheless, transmutation of nuclear waste may find new opportunities through Generation IV reactor development. The Generation IV reactor development by the Generation IV International Forum (GIF) is a cooperative international effort to commercialize the next generation nuclear energy system with the goal of achieving a competitive reactor construction cost, enhanced nuclear safety, minimizing nuclear waste generation, and further reducing the risk of weapons materials proliferation. Six different types of reactor concepts are being pursued in this effort. These six reactors include very-high temperature reactor (VHTR; a graphite-moderated, helium cooled, thermal reactor), molten-salt reactor (MSR; a molten-salt cooled, thermal, epithermal, or fast reactor), supercritical-water-cooled reactor (SCWR; a supercritical water cooled, epithermal reactor), gas-cooled fast reactors (GFR, a helium cooled fast reactor), sodium-cooled fast reactor (SFR), and lead-cooled fast reactor (LFR, a molten lead or lead-bismuth eutectic cooled fast reactor). These reactors are capable of performing transmutation of nuclear waste as part of the design goal of nuclear waste minimization as a dedicated burner reactor (see Sect. 15.1.1.3 for the definition a burner). Transmutation performance is found better with fast reactors through repeated recycling of actinides as buildup of actinides is constrained in thermal reactors due to reactor safety concerns (Taiwo and Hill 2005).

8.5 Conclusion

Reprocessing is a required step for recycling of spent fuel under the modified open or the closed cycle approaches. This chapter provided an overview of PUREX as well as pyroprocessing technology. While PUREX is the current method of choice for reprocessing, production of weapons usable pure plutonium product remains a major concern. Development of an alternative approach, the UREX family of technologies relieves the concern by the coexistence of plutonium with neptunium in the separated product. Remaining concern with UREX is the scenario of changing solvents in the process. Pyroprocessing enhances proliferation resistance in reprocessing by having the separated plutonium product always mixed with uranium, minor actinides and some fission products with high level radiation fields and heat. Challenges in the use of pyroprocessing is to accurately account and track the fissile materials in the process. Advancement to enhance nuclear safeguards in pyroprocessing are needed for its commercial development. Further discussions of nuclear materials safeguards and security are provided in Chap. 15.

Nuclear transmutation is appealing as a way of converting a radioactive material into a non-radioactive or short-lived one. While there in no real technological barrier to transmutation of key radionuclides, the cost required to make it happen is expected to be very high. Realization of the Generation IV reactor development could provide a viable alternative for nuclear waste transmutation once the goal of cost-effective reactor construction is achieved.

Reprocessing also presents opportunities for increasing effectiveness in nuclear waste management through partitioning of several key radionuclides of concern and target nuclide specific waste management scheme implementations. These opportunities include waste volume reduction, heat load reduction, and more importantly implementation of integrated waste management strategy. Integrated waste management strategy (Gombert and Roach 2007) is based on the goals of no nuclear wastes generation without proper paths for its safe disposition and no long-term storage of unstabilized or liquid wastes. The strategy is to enhance environmental stewardship in nuclear waste management by maximally utilizing recycling and reuse of materials to minimize the burden of disposal. Further development of integrated waste management strategy is on demand.

Homework

Problem 8.1 PUREX is the current commercial technology of spent fuel reprocessing. Pyroprocessing is an alternative technology for spent fuel reprocessing under development. Compare the two technologies in terms of their pros and cons.

Problem 8.2 By using the data in Table 8.3., determine how Pu and Np are distributed between the liquid cadmium and the molten salt in an electrorefining process.

Problem 8.3 Determine the transmutation half-life of ^{99}Tc and ^{129}I in a thermal reactor with a neutron flux of $3\text{x}10^{13}$ (n/cm^2-s).

Problem 8.4 Upon sampling of an acid tank in a spent fuel reprocessing plant, it was found that the acid solution contains $3.8\text{x}10^{-4}$ Ci/cc of ^{239}Pu. No other fissile materials were found in the tank. The tank is a right cylinder of 100 cm diameter.

(a) What is the concentration of ^{239}Pu in g/cm^3?
(b) Is the tank in a critical configuration? Calculate the k_{eff} of the tank using six factor formula.
(c) What is the safety factor of the tank? (Safety factor is defined as the ratio of the critical concentration of ^{239}Pu in the tank to the actual ^{239}Pu concentration in the tank.)

(Data):
^{239}Pu constants:

$t_{1/2} = 24{,}400$ years
$\sigma_{aF} = 1100$ b (absorption cross section)
$\eta = 2.035$ (fission neutrons per neutron absorption in ^{239}Pu)

Moderator constants (of the acid):

$\sigma_{aM} = 0.664$ b (absorption cross section)
$\tau_{TM} = 27$ cm^2 (age of moderator)

$L^2_{TM} = 8.1\ cm^2$ (square of thermal diffusion length)
$\rho_M = 1.0\ g/cm^3$
$M_M = 18\ g/mol$

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Chapter 9
Engineered Barriers for Nuclear Waste Management

Abstract Engineered barriers are used to achieve isolation and containment of nuclear waste thus to provide safety in nuclear waste management. This chapter discusses how various engineering materials are employed for the multiple barriers under the concept of defense in depth. Describes the use of man-made barriers in nuclear waste management with the focus on the final disposal phase. Starting with the basics of engineering materials, the types and behavior of materials used for waste forms and waste containers are described.

Keywords Nuclear waste package · Glass · Ceramic · Metal · Corrosion

"Engineered" (man-made) barriers play critical roles in achieving isolation and containment of nuclear waste during the handling, storage and permanent disposal of the waste. The performance of these man-made barriers largely depends on how the materials are employed/implemented and how they behave in the respective environments of the applications. This highlights the significance of the materials in engineered barriers in providing safety in nuclear waste management. This chapter describes the use of man-made barriers in nuclear waste management with the focus on the final disposal phase. Starting with describing the basics of engineering materials, the types and behavior of materials used for waste forms and waste containers are also discussed. The natural geological barrier, another important barrier for nuclear waste disposal, is discussed in the next chapter.

9.1 Basics of Engineering Materials

Understanding the characteristics and properties of engineering materials provide the basis for the selection of candidate materials in various technological applications in nuclear waste management.

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9.1.1 Introduction to Materials for Engineering Applications

Material is made up of atoms. Atoms, as basic building blocks, are combined together by chemical bonds to form a molecule. When atoms or molecules of different elements are combined, a compound is formed. When molecules, atoms, or mixtures of them as compounds are combined together, different materials are made. The properties of materials depend upon how constituent atoms, molecules or compounds are bound or combined.

When atoms or molecules are combined together, different materials are formed depending upon the processes taken (e.g., chemical reaction, melting, pressurization, etc.) and their conditions (i.e., temperature and pressure). They take the form of gas, liquid, or solid. For example, melting can be used to form an aggregate of atoms as molten liquid. The molten liquid also becomes a solid when the temperature is lowered below a certain point (freezing point).

The molecules in the liquid state are close to each other supported by strong interactions between them and the shape of a liquid changes with the shape of its container. In the gaseous state, molecules or atoms are unconnected to others, characterized by the long distance between the individual atoms or molecules. These atoms or molecules are rapidly moving within available spaces with no strong interactions between them. The gaseous state does not have definite shape or structure. In the solid state, molecules or atoms are tightly connected through chemical bonding. As a results of the bonding, either a regular lattice or an amorphous shape is formed. The shape or volume of a solid is relatively stable.

The materials employed in the engineered barriers for nuclear waste management are solids. Therefore, issues mostly related to the use of solid materials are covered in the following discussions.

The solid structure are classified into two categories, i.e., crystalline vs. non-crystalline. If the aggregate of atoms and molecules shows regular repeating pattern in the atomic arrangement with both short- and long-range order, it is a crystalline solid. If the aggregate has no long-range pattern but with short-range order in atomic arrangement, it is a non-crystalline solid. The majority of naturally occurring and artificially prepared solids are crystalline. Because the crystalline structure always has a larger binding energy (thus more stable) than the non-crystalline structure, solids tend to exist in the crystalline state.

There are five groups of solid materials used for engineering applications: metals (crystalline), ceramics (crystalline), polymers (non-crystalline), semiconductors (crystalline), and composites (crystalline or non-crystalline).

Metals are hard, malleable, and ductile material, made of elements that are good electrical conductors. Metals and alloys (combination of metals) are often used for structural or load-bearing applications. With strength, stability, thermal conductivity, and corrosion resistance, metals are the main materials for the containers of nuclear waste.

Ceramics are made through high temperature heating and solidification of non-metallic minerals. They have high strength, and are strong in compression, and are resistant to damage under high temperatures and corrosive environments. Use of ceramics has been suggested for immobilization of high level waste.

Polymers are produced by combining organic molecules through chemical reactions. Therefore, polymers are large organic molecules with repeating structural units. Polymers in general have low strength but provide good corrosion resistance and have been suggested for immobilization of low level waste.

Composites are the materials formed from mixtures of two or more materials. Glass (non-crystalline), concrete (crystalline) or cement (crystalline) are the examples. With composites, unusual combinations of properties, not found in a single material, can be obtained. Glass is an inorganic amorphous material. Glass is widely used for immobilization of radioactive waste for both LLW and HLW because of good chemical stability and the capacity to accommodate a variety of materials in the solid structure. Cement and concrete are aggregate material formed through chemical combination of different elements and compounds. They are widely used as a material for shielding, as structural barriers, or for stabilization or immobilization of waste.

Semiconductor is a material with electrical conductivity between a conductor and an insulator. Except semiconductors, all solid materials are widely used in nuclear waste management.

9.1.2 Overview of Materials Properties

For an application in nuclear waste management, the mechanical, physical, and chemical properties of a material must be considered with respect to the expected environment of application and the required performance. These properties depend on how atoms or molecules in the solid structured are connected, arranged and aligned together. Irradiation of a material by ionizing radiation can also induce changes to these properties.

9.1.2.1 Mechanical Properties

Mechanical properties describe how a material responds to an applied force or other imposed conditions such as temperature or abrasion. For instance, time-dependent damages to a material by the presence of loads or strains imposed will be dependent on its mechanical properties. These mechanical properties include strength, hardness, ductility, formability, and resistance to fatigue or creep.

Strength is the capacity of a material to withstand the breaking, bowing, or deforming under the action of applied mechanical loads. Hardness is the ability of a material to resist permanent shape change due to external stress. Ductility is the capacity of a material to deform plastically without being ruptured. Formability, a property related to ductility, is the ability of a material to be formed into a desired shape without being damaged. Fatigue is the weakening of material caused by the repeated loading of the material. Creep is the change of shape of a material as a very slow time-dependent process under the influence of a constant external mechanical stress. For the application of a material as waste container, properties such as high strength, good ductility, formability, and good resistance to creep will be desirable.

9.1.2.2 Physical Properties

Physical properties of a material include density, temperature of state change (i.e., melting point, boiling point, and freezing point), electrical conductivity, specific heat, thermal expansion, thermal conductivity, porosity, and weldability.

Density refers to the quantity of mass per unit volume. The melting point is the temperature at which a given solid melts. The boiling point is the temperature at which a given liquid becomes a gas. The freezing point is the temperature at which a given liquid become a solid. Specific heat is the amount of heat needed to increase the temperature of unit mass of a material by 1 °C. Thermal conductivity represents how easily heat can be conducted by a material. Electrical conductivity is to represent how well electricity can be conducted by a material. Porosity is a fraction of the volume of voids over the total volume of a material. Weldability is the ability of a material to be welded with other materials like metals.

Electrical conductivity characterizes the ability of electrons or other charge carriers to move within the lattice of a material. Such ability depends on how much energy is needed to raise electrons from the valance band into the conduction band. Here a band refers to a collection of energy levels which electrons in an atom occupy as discrete energy levels. The valance band is the energy levels filled by electrons in their lowest energy states. The conduction band is the unfilled energy levels into which the electrons in the valance band can jump up when excited. Once electrons or charge carriers are in the conduction band, they are considered free and move under electric potential.

Thermal energy transfer in a material is based on either free electron movements or thermally induced lattice vibration. The energy transfer due to free electron movement occurs as electrons move toward the colder areas of the material when they gain thermal energy and transfer the energy to other atoms. In this case, thermal conductivity is directly related to electric conductivity. In the case of lattice vibration, when additional energy is given to a material, the atoms in the material gain thermal energy and vibrate. The vibration of an atom creates an elastic wave (called phonon) through which energy is transferred to the surrounding atoms. Heat capacity is also controlled by lattice vibration and atomic structure. If a material has a tendency to easily create such elastic wave at high frequency, the material has high heat capacity.

Desired physical properties of a material for nuclear waste management vary depending on the specific applications.

9.1.2.3 Chemical Properties

Chemical properties are any of a material's properties related to chemical reactions. The tendency for chemical reactions is reflected by chemical reactivity of element, determined by the number of electrons in the outermost shell. The most inert material is the one in the group 0 with their outermost electron shells completely filled. These so-called noble gases have little tendency to gain or lose electrons. The most reactive metal is from Group 1 (alkali metals) which has only a single valence electron. The most reactive nonmetal element is the halogen group (e.g., chlorine). In general, the halogens, alkali metals, and alkaline earth metals (with two valence electrons) are highly reactive.

Metals in general have intermediate chemical reactivity often possessing multiple oxidation states. Metal with low chemical reactivity are called noble metals which can be narrowly defined as metal with a filled electron *d*-band (gold, silver, and copper) (see Sect. 4.1.1). A noble metal is also defined as one with resistance to oxidation and corrosion. Under this definition, noble metals include ruthenium, rhodium, palladium, silver, osmium, iridium, platinum, and gold but exclude copper. However, the list of noble metals often include copper, mercury and rhenium. Although not included as noble metals, titanium, niobium, and tantalum are also corrosion resistant metals.

To represent relative chemical reactiveness of metals in a given environment, a series called galvanic series is devised. Galvanic series represent a hierarchy of metals or other electrically conductive materials, including composites and semimetals. According to this galvanic series for seawater (Fig. 9.1), most noble one is graphite. From the center, more left tend to be active and gets corroded as an anode (a source of electron) in a galvanic cell. More right generally means nobler and tend to be protected from corrosion (a cathode in a galvanic cell).

9.1.3 Atomic Bonding and Material Properties

The properties of a solid depend on the types of composing atoms and how these atoms are bonded. As discussed in Sect. 4.1, there are mainly four different types of atomic bonding, i.e., the ionic bond, the covalent bond, the metallic bond, and the Van der Waals bond. The strength of a bond affects various physical properties such as the melting point, the boiling point, the hardness, electrical conductivity, etc.

If a material is formed through the ionic bond, the transfer of electrical charge requires the movement of entire ions. As these ions do not move as easily as electrons, ionically bonded material has low electrical conductivity. When a force is applied to an ionically bonded material, a layer of ions is pushed and repulsive force can occur between ions with the same positive or negative charge. Therefore, most ionically bonded materials are hard, non-malleable, non-ductile/fragile, and not volatile. The melting point and boiling point of a material with ion-binding are

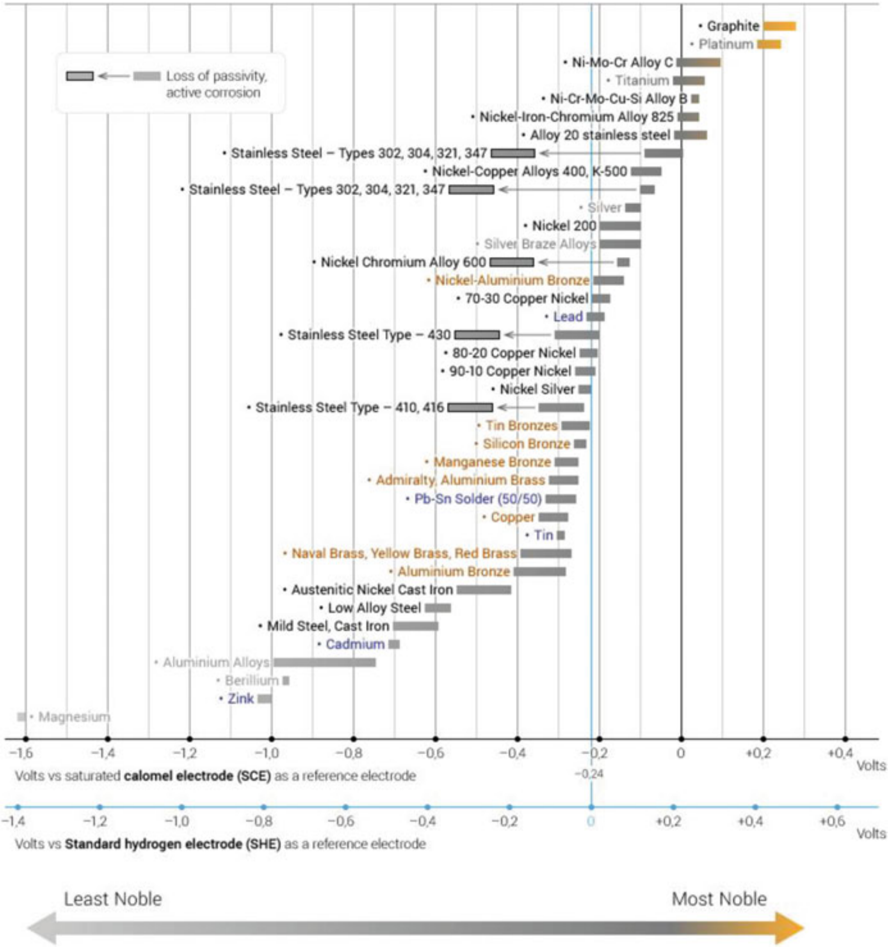


Fig. 9.1 Galvanic series for seawater (EngineeringClicks 2017)

relatively high compared to the materials with other bindings due to the strong attraction between ions. Examples of ionically bonded material are NaCl, BeO, MgO, LiF, $CaCl_2$ etc. The ionic bonds are found in many ceramics.

In covalently bonded materials, movement of electron requires the breakage of the covalent bond. Thus materials with covalent bond have poor ductility and poor electrical and thermal conductivity, therefore, make good insulating materials. Also the materials are not very hard although exceptions such as carbon, silicon, and diamond exist. Compared to ionically bonded materials, covalently bonded materials have much lower melting and boiling points. The materials in this category include polymer materials, ceramics, and semiconductors.

With ionic and covalent bonds, the strength of bonds is high and the materials tend to have a high strength, a high melting point, a high modulus of elasticity (i.e, stiff, experiencing very little stretching under elastic loads), low coefficient of thermal expansion, and brittleness.

In a material with metallic bonds, i.e., metals and alloys, the electrons can move freely with abundant presence. In this case, good electrical and thermal conductivity are expected. This is because electrons in a good electrical conductor material move faster as the electrons gain additional kinetic energy from the thermal energy. The atoms in the metallic bond are able to roll over each other without breaking the bond. Thus metals exhibit malleability and ductility. Since the force between the electrons and the metal cations is very strong, the melting points and boiling points are usually high. Nickel, copper, and iron are examples of metallically bonded materials.

The Van der Waals bond is a weak bonding through electrical interactions between atoms or molecules. Although weak, the Van der Waals bond can dramatically change the properties of materials. For example, the material formed by covalent bonds is expected to be brittle. However, with additional bonding by the Van der Waals bond, the material becomes much less brittle. The Van der Waals bond is particularly important in plastics and polymers.

Most materials are formed through a mixture of two or more types bonding between atoms.

9.1.4 Atomic Arrangement and Material Properties

9.1.4.1 Crystal Structures and Material Properties

In addition to the types of bonding between the atoms, the way the atoms are arranged in the structure also affects mechanical, physical and chemical properties of a solid. Such arrangement is related to the efficiency in the packing of atoms in the repeating pattern of the crystalline solid structure. The degree of packing efficiency within the structure depends on how the system minimizes the total intermolecular energy to achieve the stability of the crystal structure. This degree of packing efficiency, represented by the packing factor, also determines the microstructure and behavior of a solid. The packing factor is the volume occupied by atoms per total volume of a crystal structure.

The densest packing of atoms corresponds to the most stable crystal structure with large number of bonds per atom. For example, atoms can be packed together in a simple cubic (sc) array or in a close-packed cubic structure, or in close-packed hexagonal structure. Most common examples of these structures are body centered cubic (bcc), face centered cubic (fcc), or hexagonal close packed (hcp) (see Fig. 9.2 below).

The bcc arrangement has atoms at each of the eight corners of a cube (like the cubic unit cell) plus one atom in the center of the cube. So the corner atoms are shared among eight unit cells. The bcc unit cell has a packing factor of 0.68. In the fcc and the hcp structure, the atoms from one layer nest themselves in the gaps between the atoms of the adjacent layer. The difference between them is in the

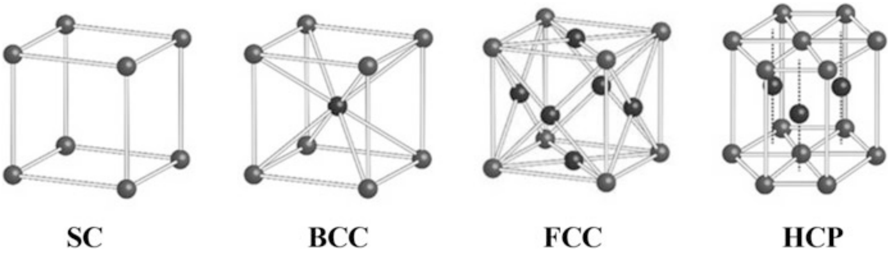


Fig. 9.2 Schematics of crystal structures of simple cubic (SC), body-centered cubic (BCC), face-centered cubic (FCC), and hexagonal close-packing (HCP)

Table 9.1 Examples of metals with bcc, fcc, and hcp crystal structures

Crystal Structure	Metals
BCC (body centered cubic)	lithium, sodium, potassium, rubidium, cesium, chromium, iron (alpha phase – the one stable at the lowest temperature), molybdenum, tantalum, and tungsten
FCC (face centered cubic)	aluminum, copper, gold, iridium, lead, nickel, platinum and silver
HCP (hexagonal close packed)	cadmium, cobalt, lithium, magnesium, potassium, sodium, titanium (alpha), and zinc

stacking sequence. The fcc layers cycle between three positions while the hcp layers cycle among the two equivalent shifted positions. The fcc and hcp structures both have a packing factor of 0.74. The sc structure, also known as primitive cubic, has one atom at each corner of a cube. Due to low-packing efficiency and low number of nearest neighbor around each atom, the sc structure is relatively rare in nature compared with other cubic structures.

If planes of atoms are closely packed, it is easier for the planes to slide by each other. Therefore, the lattice structures with closely packed planes experience plastic deformation more easily than those with less closely packed ones. Also, in comparison to non-cubic lattices, cubic lattice structures allow slippage more easily as their symmetry provides closely packed planes in several directions. Therefore, the bcc or fcc structures are less hard than the hcp structure. Between bcc and fcc, a fcc structure with a denser packing will exhibit less hardness and more ductility than a bcc structure.

The table below shows the types of crystal structures of various metals. Metals with the bcc structure include all alkali metals (Li, Na, K, Rb, and Cs), chromium, iron (alpha phase - the stable one at the lowest temperature), molybdenum, tantalum, and tungsten. Metals with the fcc structure include aluminum, copper, gold, iridium, lead, nickel, platinum and silver. The hcp structured metals include cadmium, cobalt, lithium, magnesium, potassium, sodium, titanium (alpha), and zinc (Table 9.1).

The size of each crystal, referred to as a grain, also affects the mechanical properties. A structure with larger grains have lower strength, hardness and ductility than the one with smaller sized grains. Size of a grain in a crystal is largely controlled by the rate of cooling when the solid was formed. Smaller grains, i.e., a fine grain structure, are the result of rapid cooling while larger grains are formed from slow cooling. The interfaces between the grains is called grain boundary. The orientation and the shape of grain boundaries also affect mechanical properties of a materials such as yield strength and ductility. Yield strength is the stress at which a solid material ceases to be elastic and begins to deform plastically.

9.1.4.2 Non-crystalline vs. Crystalline Solids

Non-crystalline solids such as glass are formed through the mixture of covalent bonds and ionic bonds. Such bonding do not allow the atoms in the structure to slip past each other. Thus non-crystalline solids cannot plastically deform but fracture under stress. Non-crystalline solids are a low strength, brittle material, and can be formed when liquids are rapidly cooled below the melting point. With rapid cooling, the atoms or molecules do not have enough time to rearrange themselves in a crystalline structure. Thus the solid has no long-range periodicity in atomic locations. The atomic arrangement could be in the form of a tangled mass of long-chained molecules or a 3-dimentional network of atoms with short-range order (but no long-range order).

While crystalline solids have a particular melting point, non-crystalline solids melt over a wide range of temperature. Also, non-crystalline solids gradually become soft as their temperature is increased. Plus, when their viscosity drops, non-crystalline solids begin to behave like ordinary viscous liquids. In a crystalline solid, while the structure has repeating pattern of atomic arrangement, arrangements of grains are different along the different directions. Therefore, along the different directions, the value of a physical property can be found different. In contrast, in non-crystalline solids, value of any physical property would be the same in any directions as the atomic arrangement is irregular in all directions.

9.1.4.3 Treatments of Metals for Property Modification

Some special treatment can be made to modify the properties of metals for better performance in their intended applications. This treatment includes alloying, mechanical working, and heat treatment such as annealing.

Alloying is mixing two (or more) metals through melting. By adding (alloying) metals such as manganese, silicon, copper, magnesium and zinc to a base metal, the strength or corrosion resistance of the metal can be increased.

Strain hardening (also called work-hardening or cold-working) is the process of making a metal harder and stronger through plastic deformation by the application of external forces. When a metal is plastically deformed, dislocations move and additional dislocations are generated. Here dislocation refers to irregularity within a crystal structure due to abrupt change in the arrangement of atoms. The more dislocations within a material, the more they will interact and these movements eventually get tangled in the crystal structure. As the mobility of the dislocations become reduced or the movements are halted, the strength of the material is increased. This type of strengthening is commonly called cold-working. It is called cold-working because the plastic deformation must occur at a temperature low enough that atoms cannot rearrange themselves (i.e., below the recrystallization temperature). Cold-worked areas in metals, however, become prone to corrosion due to the increase in dislocation concentration.

Another type of mechanical working is hot working, i.e., plastic deformation of metals at high temperature allowing rearrangement of atoms through crystallization. In this case, less stress is required than cold working and hardening of metals because plastic deformation is eliminated. Hot working is to improve mechanical properties of metals such as toughness and ductility by raising the temperature of metals above the recrystallization temperature and by eliminating porosity and refining grain structures. Oxidation of metals takes place during hot working. To eliminate the chance of embrittlement by oxygen, hot working of metals needs to be done at inert environment.

Annealing is the process of heating a material to a temperature below its melting point and then cooling it slowly. Through this process, the materials gets softened and becomes less brittle while internal stress is removed or reduced.

9.1.5 Radiation Effects on Materials

Materials exposed to ionizing radiation are susceptible to radiation-induced damages. The specifics of radiation damage depends on the type of radiation, the irradiated material, and the dose level. The level of radiation dose expected during the storage or disposal of nuclear waste becomes important in the consideration of radiation effects. For a reference, the maximum absorbed dose rate as a design basis for the standardized HLW/spent fuel container is <1 Gy/h in the U.K. (King 2014c).

In terms of radiation damage, low LET radiation (electrons, x-rays, and gamma rays) and high LET radiation (neutrons, alpha particles, and fission fragments) have different effects. While both low and high LET radiation causes extensive ionization of atoms in the target material, low LET radiation causes damage mainly through the rupture of chemical bonds and free radical formations. High LET radiation causes damage mainly through atomic displacements.

9.1.5.1 Effects of Low LET Radiation

The extent of radiation damage caused by low LET radiation depends largely on the type of material. In the case of metals, the electrons stripped from an atom via ionization are readily replaced by other electrons through free electronic movement in metals. Thus low LET radiation will have only a transient effect on metals. In the case of polymers and plastics, as organic molecules contain almost exclusively covalent bonding, breaking of covalent bonding by radiation is destructive and causes chemical changes (e.g., cross-linking of chains). The resulting damage can be significant and is manifested as changes in material properties (such as strength, hardness, conductivity). Materials with ionic bonding such as ceramic, its damage by radiation is not so destructive as the electrical charge balance can still be maintained in the presence of individual ionization events). In this case, structural properties of the material are virtually unaffected. Color change may occur to the material from the trapping of electrons in the crystal lattice. Glass contains extensive ionic bonds along with covalent bonds and shows similar irradiation behavior as ceramics.

9.1.5.2 Effects of High LET Radiation

In the case of high LET radiation, the incident heavy particles pass through the crystal lattice of the material, colliding with and knocking the atoms in the solid structure off their lattice sites (this is called "knock-on"). The ejected atoms can acquire sufficient energy to eject further atoms. Eventually the process terminates when an ejected atom has insufficient energy to displace another atom from its lattice site. Such displacement in turn affect neighboring atoms creating additional displaced atoms. This leads to production of displacement cascade. Eventually displacement of atoms is terminated but leaves defects in the structure as deviations from an orderly array. The number of displacements per atom (dpa) is commonly used as a measure of radiation damage to a material.

If the deviations occurring in the solid structure are localized to the vicinity of only a few atoms, it is called a point defect. Points defect can include interstitials, missing atoms from their regular sites, and vacancies. Interstitial means presence of extra atoms in the holes between atoms. Vacancies are created by the displacement of atoms to the interstitial locations. Line defects are also formed if the deviation extends through propagations along lines. In this case, the locus of defective points produced lie along a line in the lattice and these line defects are also called dislocations. Thus whole rows of atoms along certain directions in the structure are arranged anomalously. If the line defects spread over a plane or surface, they form a plane defect. Such plane defect includes defects in the boundary between two crystals or between regions of a crystal or defects in the alternate stacking procedure.

Accordingly, when crystal structure is irradiated by a high LET radiation, a series of defects can be produced in the form of point defects, line defects, or plane defects. As these defects accumulate, the microstructure of the material changes. These microstructure changes often lead to material hardening, embrittlement, or swelling. Increase in hardness due to irradiation can make the material brittle as introduction of defect clusters reduces the ductility of the material. Swelling is the increase in the volume of the material caused by the accumulation of voids in the structure. The voids are formed as the radiation-produced vacancies aggregate.

Irradiation also increases the rate of creep of a material. This increase is beyond what is expected from the normal temperature-dependent effect. Irradiation also causes creep in the temperature ranges where temperature effect on creep is expected to be negligible. The increase is due to the production of excess defects in the crystal structure through the formation and movements of vacancies or dislocations by irradiation.

In the case of anisotropic materials, radiation-induced growth of dislocations is also observed in the irradiated materials. Anisotropic materials include composites and some metal alloys such as zircaloy and have different physical properties in different directions. Growth of radiation-induced dislocations causes anisotropic dimensional change, affecting its structural integrity.

9.1.5.3 Effects on Gases or Liquids

In the case of gases or liquids, exposure to radiation causes chemical changes. Such chemical changes are through decomposition of a molecule and formation of free radicals. Formation of free radicals also creates various other species. As discussed in Sect. 5.1.2, when water molecule is ionized, hydrogen (·H) and hydroxyl (·OH) free radicals are formed. Recombination of them can lead to the formation of gaseous hydrogen (H_2), hydrogen peroxide (H_2O_2), oxygen (O_2), hydroxyl radicals (·OH), and peroxide radicals ($\cdot O_2{}^{-2}$).

$$H_2O \overset{a,\beta,\gamma,n}{\rightarrow} H, OH, e^-, H^+, H_2, H_2O_2$$

9.2 Nuclear Waste Package as Engineered Barriers

In the selection of a material for engineered barriers, chemical properties such as corrosion resistance are of primary importance for long-term nuclear waste isolation. In general, pure metals such as iron, copper, or aluminum gets corroded slowly in the atmosphere. To improve upon the corrosion characteristics or strength of these pure metals, alloys such as steel, stainless steel, brass, bronze, nickel chromium alloy, or nickel copper alloy are developed. These alloys use iron, copper, nickel, or aluminum as a base metal as the principal metallic component and are combined with one or more metallic elements.

9.2.1 Design of Nuclear Waste Package

A key component of engineered barriers for waste disposal is the waste package. The primary objective in designing the waste package is to prevent the release of radionuclides from the waste. Any expected failure must be within the prescribed limits for isolation and containment of nuclear waste. For example, the licensing

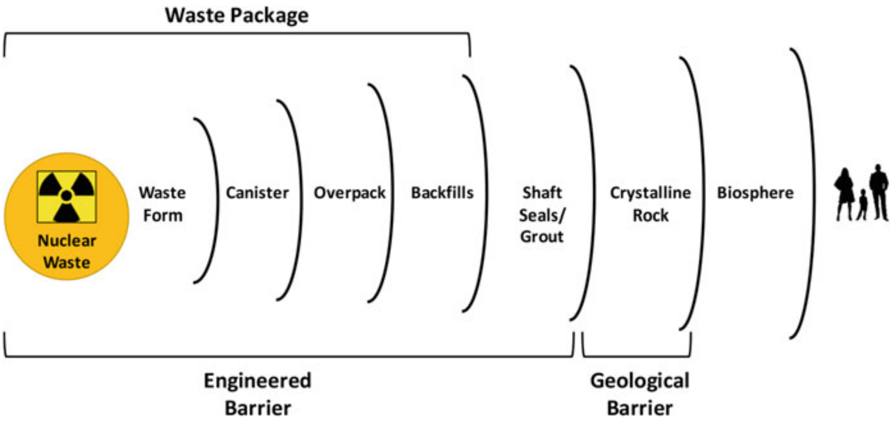


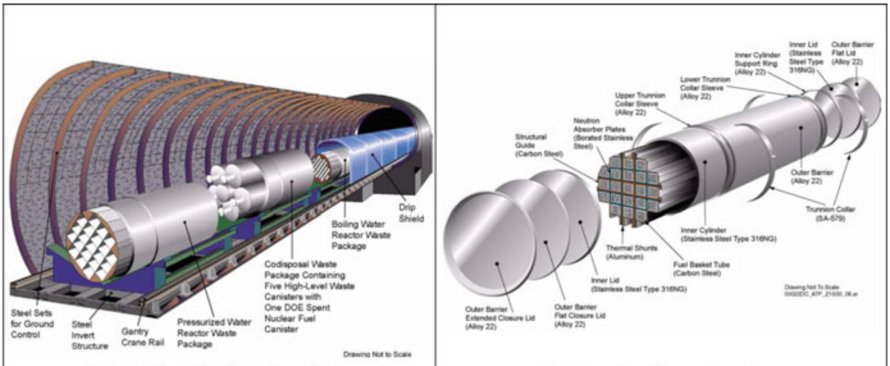
Fig. 9.3 The concept of multiple barriers for nuclear waste disposal

criteria for geological repositories of HLW and spent fuel in the U.S. specifies that containment of HLW must be "substantially complete" (assuming anticipated processes and events) for 300–1000 years after repository closure.

Given the uncertainties in the long-term behaviors of engineered systems, nuclear waste package is designed in the form of multiple barriers. These multiple barriers are to provide defense-in-depth in safety. Therefore, even if one barrier fails, other barriers can still remain effective in preventing the release of radionuclides. These barriers include the waste forms, the surrounding waste containers (canister and overpack), the backfills, and the emplacement holes/tunnels. Use of these barriers as part of the geological disposal system is depicted in Fig. 9.3. Each component of the multiple barriers comes with different characteristics and design objectives. Therefore, putting nuclear waste inside waste package is like placing diverse layers of protection against materials movement between the waste and the surrounding environment.

The waste forms, the first engineered barrier of the package, are to render the waste into physically, chemically, thermally stable one. For this purpose, the waste is encapsulated into an immobilized form. The resulting waste form becomes resistant to leaching, powdering, cracking and other modes of degradation. Here leaching refers to a process by which a liquid dissolves and removes the soluble components of a solid material. Materials such as glass or ceramics have been utilized as part of a waste form. An exception is spent nuclear fuel. Spent fuel is often disposed of as waste form in and of itself without special treatment. The materials for waste immobilization are further discussed in Sect. 9.4.

The next barrier is the container. The containers for HLW disposal are typically made of double barriers, called a canister and an overpack. They provide protection of the waste form against physical and chemical stresses during transportation, interim storage, and disposal. A canister is the primary container as an aid to handling and transportation of nuclear waste. Upon disposal of nuclear waste, the canister provides physical support and protection for waste form while preventing



Cutaway of a drift with waste packages
(Source: Figure 3-3 (DOE 2002))

Key Elements of the waste package
(Source: Figure 3-2 (DOE 2002))

Fig. 9.4 Use of waste package in the U.S. Yucca Mountain Repository and its key elements. (Source: Figure 3-3 DOE (2002); Source: Figure 3-2 DOE (2002))

groundwater intrusion and permitting retrievability of the waste if needed. An overpack is a hemetically sealed high integrity protective barrier and works as the primary barrier against corrosion under geologic disposal. Thus an overpack is mainly to provide long-term containment of nuclear waste. It is also a physical barrier against groundwater intrusion and aids to ensure retrievability of waste. Metals, alloys, ceramic, or composite materials are commonly considered as the container material.

The overpack is emplaced in the geological repository surrounded by the backfill materials insider the tunnels or holes excavated in the geologic medium. Backfills are mass transport barrier for the purpose of preventing direct water contact and acting as a buffer against external mechanical impact. Backfills also work as a chemical barrier, in particular, in the case of placing the waste packages in the saturated geologic medium. In this case, backfills work as a sealant of waste package and providing a pH buffer and a reducing chemistry environment to inhibit corrosion. They also work to minimize dissolution and migration of radionuclides.

The tunnels or emplacement holes (if the waste is placed vertically) are used for the application of backfills and provide stabilization and enclosement of waste packages. Using emplacement hole cavity to store the waste package increases the effectiveness of backfill.

Figure 9.4 shows an example of nuclear waste package emplaced horizontally in a tunnel, as envisioned for HLW disposal at the Yucca Mountain repository in the U.S.

9.2.2 *Predictability of Materials Performance for the Nuclear Waste Package*

Nuclear waste package must feature long-term resistance to environmental attack. Such resistance must also meet the legal requirements for licensing approval. This implies that an essential part of nuclear waste package design is predictability, i.e., the ability to demonstrate the expected performance of a nuclear waste package under the influence of degradation processes. The specific processes of waste package degradation includes gas-metal interactions/oxidation, aqueous corrosion, microbial corrosion, dealloying, hydrogen embrittlement, and mechanical effects. Here, dealloying refers to selective leaching (dissolution) of one component of the alloy. Hydrogen embrittlement is a damage as a result of hydrogen absorption. Mechanical effects are time-dependent damage of components due to the presence of loads or strains imposed from the sources within or outside of the waste package. These sources include hydrostatic pressure, pressure imposed by overlying material, waste form, thermal fatigue, and creep-rupture (NRC 1990).

Predictability in Nuclear Waste Management

Predictability in nuclear waste management does not mean exact prediction of the future. The purpose of prediction in this case is not to exactly predict the future evolution of the system but to provide enough technical basis to ensure that the provided design can meet the regulatory safety requirements.

9.2.2.1 Mechanistic Model Development

Predictability in terms of long term behavior of waste package barriers requires the understanding of fundamental mechanisms underlying the degradation processes. Such understanding leads to the development of so-called mechanistic models. Combined use of good mechanistic models and the data representing the conditions expected in the geological repository enables long-term material performance prediction. If good mechanistic models and the necessary data are absent, extremely conservative assumptions need to be made defeating the purpose of designing multiple barriers.

For the development of mechanistic models, first principles relationships (i.e., the fundamental core understanding of the relationships among only the most essential parameters) for the involved processes need to be identified. Experiments are also needed to observe the effect of key variables on the process over time under consideration. While these experiments are usually short-term and are often accelerated tests by having the system subjected to higher-than-usual levels of external influences from one of more variable, use of a mechanistic model to underpin the role of key parameters in explaining the process under consideration enables the prediction of system's long-term behavior. Other tests such as characterization tests or service condition tests may also be needed. Mechanisms can be proposed and evaluated for each specific step or process. The values of model parameters are extracted from the tests conducted and verified through comparisons.

9.2.2.2 Characterization of the Environment

To support mechanistic model development, a parallel effort of characterizing the surrounding environment is needed. Here the environment includes the conditions of the geological repository and their evolution over time with respect to chemistry, hydrogeology, and natural disruptions in relation to material degradation. Uncertainties in these characterization need to be identified as well.

The environment for a waste package includes both internal and external, i.e., inside and outside the waste package. Inside the waste package, the exposure conditions vary through irradiation and aging of the enclosed nuclear waste. Radiation will change the chemistry environment (redox potential of the system) through radiation-induced decomposition of the constituents and create species with the potential for accelerated degradation of waste package components. The decay heat from nuclear waste will increase the ambient temperature, potentially accelerating chemical reactions.

Outside the waste package, chemical factors and mechanical forces will affect the performance of a waste package. The principal chemical factors are the amount of oxygen and water available along with water salinity. In terms of mechanical forces, the levels of stress that arise from the surroundings, such as groundwater pressure, pressure from the surrounding rocks, or stresses from tectonic and seismic activities are important. Hydrology, temporal climatology, geology, geochemistry, presence of microbial species, and tectonics and seismic activities all play roles in characterizing the external environment.

Uncertainty is inherent in predicting the performance of waste packages. By gathering more data to support modeling through collection of field data, laboratory data, and natural analog data, some of the uncertainty can be reduced. Improving predictive models using site specific data and accelerated materials tests and performing validation analyses also reduces uncertainty. Further discussions on uncertainty is given in Sect. 12.2.3.

9.2.3 Fabrication and Monitoring of Nuclear Waste Package

The process of fabrication of various waste package components must follow quality control procedures. Exercise of quality control includes the use of qualified personnel and equipment, procedures, and in-process product examinations. Once fabricated, inspections and monitoring of the products follow. Inspections are to determine the acceptability of fabricated components and are performed through visual testing, non-destructive examination, and destructive testing. The acceptability of fabricated components for repository emplacement is determined based on pre-set inspection criteria.

Once nuclear waste packages are deployed at a site, in-situ monitoring is set up to address any issues related to observing any unsatisfactory behavior (defects or malfunctions) of the packages before permanently closing the repository. Through in-situ monitoring, any needs for retrieval and correction of nuclear waste package are identified. The retrieved packages are then replaced through repackaging. Non-destructive techniques such as ultrasonic, x-ray, or magnetic resonance imaging are used as part of a monitoring program. This is followed by long-term monitoring during the post closure period.

9.3 Spent Fuel as Waste Form

In the case of the open (one-through) fuel cycle, unreprocessed spent fuel serves as a final waste form for disposal. This is due to the expected durability of spent fuel materials including the fuel cladding. It should also be noted that changes have occurred to the properties of UO_2 fuel and the cladding during reactor irradiation. These changes become the source of concerns in long-term behavior of spent fuel during the final disposal phase.

9.3.1 Irradiation Induced Changes in UO_2

Irradiation induced changes in the UO_2 fuel matrix occur mainly from the presence of fission products and heat. Formation of gaseous fission-product bubbles and accumulation of solid fission products cause fuel swelling and increase its volume. High internal temperatures of the fuel and the cracks developed from thermal stresses during reactor operation also lead to expansion of fuel volume. This expansion causes contacts between the fuel and the cladding, as so-called pellet-cladding interactions (PCIs), potentially leading to fuel failures. These are explained in the next subsection (9.3.2).

Production of fission products with high kinetic energy (about 85% of the fission energy is carried by the fission products) leads to atomic displacements (as explained in Sect. 9.1.5). Each fission event may produce as many as 10^6 knock-ons. The fast neutrons released during fission may also cause atomic displacement (but far less effectively than fission products due to the smaller amount of energy carried). Atomic displacements create interstitials, vacancies, loops, and voids that prevent dislocations from moving or slipping. The resulting outcome is hardening of the material (lowering ductility).

The very high kinetic energies of fission products also allow them to move some distance away from their original positions and come to rest in the holes between the atoms (called interstitial positions). Diffusion may cause them to move further to become part of the crystal lattice. These atoms may join other atoms of the same element to form crystals of their own at crystal boundaries. They may also migrate to grain boundaries, cracks, or other void spaces such as the fuel-cladding gap. These radionuclides located at grain boundaries, at cracks, and in the cladding gap become available for release upon cladding breach.

Redistribution of fission products and other irradiation products (after atomic displacements or fission energy generation) in different regions or locations of nuclear fuel depends on their compatibility in the UO_2 crystal structures. The ones with the crystal structures compatible with that of UO_2 remain in solid fuel, replacing a uranium atom in the lattice. They do not migrate and cannot dissolve unless the whole fuel matrix dissolves. Pu and minor actinides (Np, Am, Cm) belong to this group. Therefore, inventory of these nuclides along with U are almost entirely (~99.2%) contained within the fuel matrix. There are also radionuclides whose crystal structures are incompatible with that of UO_2. Their atoms are trapped at grain boundaries and sometimes forms segregated micro-phases. Fission products such as Mo, Tc, Ru, Rb, and Pd may belong to this group. The nuclides in gaseous form are free to migrate and may diffuse from the hot center of the fuel toward the outer cooler regions, such as pores and fuel-cladding gaps. Fission products such as Kr, Cl, I, and Cs are in this group. This results in the distributions of the fission and other reaction products within fuel grains, at the grain boundaries, and in the fuel-cladding gap.

The process of nuclear fission consumes uranium and also releases oxygen in an amount corresponding to the amount of U consumed (about 136 grams of oxygen per kg of fissioned ^{235}U). These oxygen atoms are likely to remain in the UO_2 lattice and combine eventually with the atoms of fission products, actinides, or activation products in the crystal lattice, becoming impurities. If not as oxides, fission products also remain in the lattice as metallic or alloy inclusions.

9.3.2 Irradiation-Induced Changes in the Cladding

During reactor irradiation, thermal, chemical, and mechanical interactions occur between the cladding and UO_2 fuel. These interactions include ratchetting, bambooing, or chemical attacks and may cause fuel failures.

Ratchetting refers to plastic deformation of cladding due to exertion of cyclic mechanical stress. Cyclic mechanical stress can be the result of periodic PCIs due to swelling and stretching of UO_2 through thermal expansion during the period of reactor power increase. Geometrical mismatch between the cladding and the fuel created by the swelling-induced cracks in fuel surfaces can also be the cause of ratchetting.

Bambooing is related to diametral deformations of cladding induced by fuel expansion and interactions with the cladding through formation of ridges. During irradiation of oxide-fuel elements, formation of ridges may occur at regular intervals along the axis of the rods. These ridges can cause radial strain potentially leading to a cladding failure toward the end of fuel life.

Chemical attacks to the cladding come from fission products or oxygen. Attack from fission products such as Cs, Mo, and Te come from inside. The chemical agents, e.g., iodine, also accelerate crack propagation if the crack is initiated (e.g., through stress corrosion cracking, perhaps, during power transients). Excess oxygen oxidizes the surface of the cladding outside.

During reactor operation, the cladding also gets corroded (oxidized) on both the inner and the external sides. The external oxidation forms about 100 μm thickness oxide layer from the contact with the cooling water. The internal oxidation due to contact with the UO_2 fuel is much thinner (about 10 μm). Increase in cladding corrosion leads to thinning of cladding wall. Hydrogen is also produced from the corrosion of zircaloy which diffuses through the oxide layer and is absorbed in the metal. If excess hydrogen is absorbed by the cladding, zirconium hydrides may form. This causes embrittlement of the base metal which results in gross loss in ductility and the possibility of brittle failure. The reactions are as follows.

$$Zr + 2H_2O \rightarrow ZrO_2 + 2H_2$$

$$Zr + 2H \rightarrow ZrH_2$$

Presence of zirconium hydrides is known to accelerate corrosion of zircaloy cladding, in particular, at high burnups. Also growth of oxide layer causes the increase of temperature in the oxide-zircaloy interface through an insulating effect. This also accelerate the corrosion effect.

All of these changes will compromise the barrier characteristics of the cladding. These changes also make it difficult to predict the behavior of spent fuel as a waste form during the disposal phase. These difficulties may cause not counting fuel cladding as credible barrier against the release of radionuclides. This is albeit the experience of the spent fuel cladding integrity has been relatively good during dry storage.

Irradiation of the cladding by neutrons also produces various activation products as a source of radiation in particular during decommissioning of nuclear power plant or the final disposal phase. Examples of them include:

$$^{92}Zr + n \rightarrow\ ^{93}Zr\ \left(\mathrm{T}_{1/2} : 1\tilde{.}5 \times 10^{6}\ \mathrm{y}\right)$$

$$^{58}Ni + n \rightarrow\ ^{59}Ni\ \left(\mathrm{T}_{1/2} : 7\tilde{.}4 \times 10^{4}\ \mathrm{y}\right)$$

9.3.3 Radionuclide Release from Spent Fuel

Radionuclides in the UO_2 fuel are redistributed in various regions or locations with implications on possible release upon cladding failure. The ones remaining in UO_2 crystal lattice do not get released unless the whole fuel matrix dissolves. Dissolution of UO_2 fuel matrix is expected to be controlled by the solubility of uranium and the oxidation condition of the surface of the UO_2 fuel. The release characteristics of the

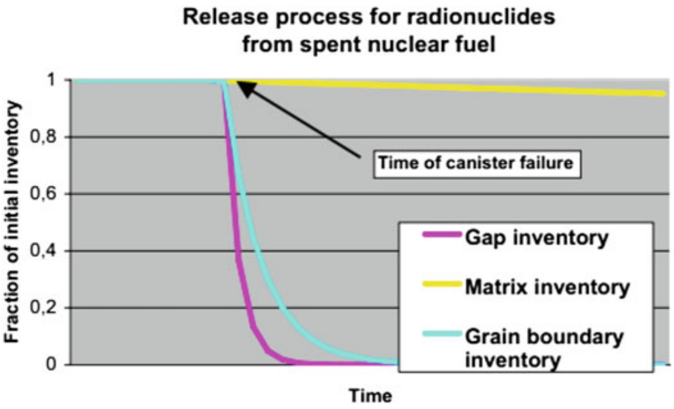


Fig. 9.5 Schematics of three stages in fission product release from used fuel (source: SKI Report 2007)

radionuclides trapped at grain boundaries depends on the conditions of the grains. For example, if the grains are tightly bound together at the boundary, release would be restricted. If the grains are loosely bound with sufficient space between them allowing solution access, release may occur rapidly. The gaseous radionuclides can be readily released upon the breach of the cladding.

If the barriers of nuclear waste package fail during disposal in a geological repository, three different phases of radionuclide release from spent fuel can be envisaged: (1) release from the gap, (2) release from grain boundaries, and (3) release from UO_2 matrix. This is shown in Fig. 9.5. While many nuclides have their inventory distributed in all three regions (the gap, the grain boundaries, and UO_2 matrix), determining the portion of the radionuclide inventory that is readily available for release is a critical issue in repository safety analysis. This portion is called the “Instant Release Fraction.” Radionuclides released in this category will eventually be part of migrating radionuclides and form the initial peak arriving at the biosphere and the locations of human habitat. Much effort has been made to determine the instant release fraction of key radionuclides and some of them as examples are shown in Table 9.2.

9.4 Materials for Waste Immobilization

Waste form, as a medium to stabilize nuclear waste through immobilization, serves as the first barrier to the release of radionuclides from nuclear waste. Key functions of a waste form include: (1) to provide physically, chemically, thermally stable form, (2) to immobilize the radioactive materials, thus slowing release even when contacted with water, and (3) to resist leaching, powdering, cracking and other modes of degradation.

Table 9.2 Examples of the values of instant release fraction (as percent) for key radionuclides of concern in repository safety assessment

	Switzerland (spent fuel at 48 GWd/MTU) [(1)]	Sweden[(2)]	U.S.[(3)]
	Best estimate	Best estimate	Lower, apex, upper (Triangle distribution)
I-129	4	3	0.5, 11, 34
Cs-135	4	3	0.16, 3.7, 11.3
Tc-99	2	0.2	0.05, 0.13, 0.28
Se-79	4	3	–
Cl-36	0	6	0.5, 11, 34
C-14	10	5	10, 10, 10

Source data: (1) Johnson and McGinnes 2002; (2) Johnson and Tait 1997; (3) Jain et al. 2004

In the case of HLW, glass or ceramics have been used as the main waste form material. In the case of LLW, typical waste form materials include cement, polymers, glass, metals, and sorbent materials. Discussions in this section will cover glass, ceramics, cement, and polymers.

9.4.1 Glass

From its natural presence, glass has been noticed as a very stable material. Examples of natural glass include the glass made from lighting strikes (called fulgurites), the glass made from volcanic origin (called obsidian glass), impact glasses, and natural glass of unknown origin called tektites. These natural glasses have demonstrated extraordinary durability over long time.

It has been also known that any substance can be made into glass, in principle, from a molten state through fast enough cooling. In terms of processing for waste form development, making glass requires lower melting temperature than making ceramics, thus is less costly. Compared to cement or polymers (as less costly option than glass), glass is a much more stable waste form.

Glass as a waste form for nuclear waste immobilization was investigated in early 1950s in Canada as well as in the U.S. Similar research was performed in the U.K. and France in the late 50s and later in Germany, Japan, and India.

As a product of fusion, glass provides the capacity to accommodate a variety of materials in the unordered amorphous solid structure. Up to 30 wt % of waste loading has been reported for glass. Glass also has an intrinsic resistance to radiation damage due to the unordered nature of the solid structure.

Glass currently represents smaller volume of HLW in the U.S. compared to spent fuel. However, it is the majority of waste form in U.K., France, Germany, and Russia. Glass technology is also used to immobilize contaminated materials as part of the effort for cleanup of radioactively contaminated sites.

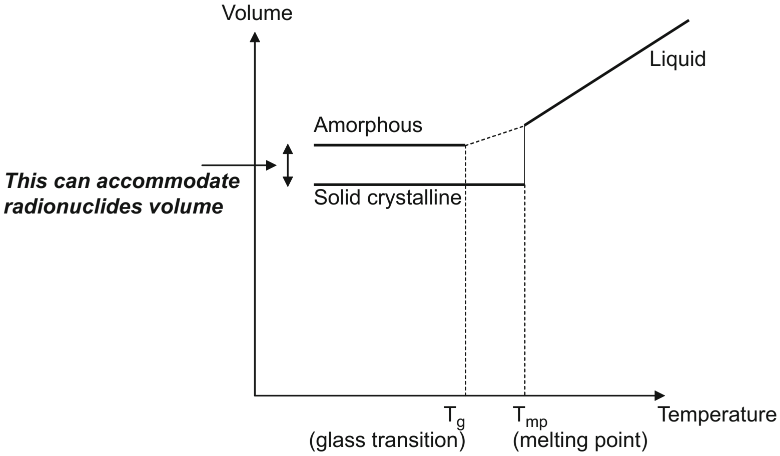


Fig. 9.6 A diagram depicting the capability of glass accommodating foreign waste materials within its volume

9.4.1.1 Characteristics of Glass as Waste Form

When a mixture of solid materials is heated above the melting point, it becomes a liquid melt mixture. When such mixture is cooled again, it forms a crystalline solid. However, if such liquid melt mixture is cooled rapidly, it does not have enough time to configure itself as a crystalline structure but becomes a rigid solid incapable of forming crystalline structure, i.e., an amorphous material with no long-range order. In this case, the volume of the amorphous solid formed is larger than that of the crystalline solid. This is depicted in Fig. 9.6. The difference in the volume due to larger space taken up by the unstructured amorphous solid can be utilized to accommodate other heterogeneous materials.

This points to the possibility of using glass as a waste form to accommodate foreign materials such as nuclear waste. It turns out that the process of glass making (called vitrification) can handle wide variety of waste feeds for their immobilization. Nuclear waste can then be in isolation from the surroundings through long-term stability of glass, except for the volatile radionuclides.

The concept of making glass by fast cooling is captured by the so-called TTT diagram (TTT: time, temperature, and transformation) shown in Fig. 9.7. As the TTT diagram shows, when the rate of cooling is fast enough (within days), there will be no crystallization in the glass matrix. However, if the rate of temperature decrease becomes slower (e.g., the cooling period is extended over weeks), then crystallization can take place. The most critical temperature range to watch is between 600 °C and 900 °C which represent the more likely temperature ranges of crystallization development. Depending upon how slow the rate of temperature decrease is in this range, higher fraction of crystallization could occur in the resulting glass matrix. Also, the resulting glass may have crystalline regions on a fine scale.

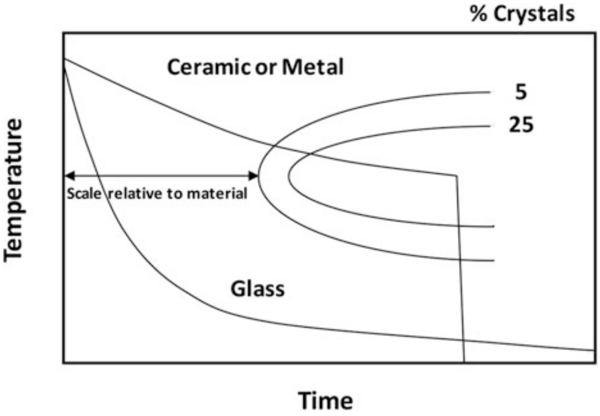


Fig. 9.7 Time, temperature, and transformation (TTT) diagram for glass making

9.4.1.2 Compositions of Glass as Waste Form

Key desired characteristics of glass as waste form is high chemical durability (for safety) and high waste loading (for economics). In the case of waste loading (i.e., how much (e.g., volume %) of nuclear waste is included in the glass), however, the highest value may not always be preferred as it affects radiation effect and heat generation of the glass. In terms of chemical durability, the chemical composition of the glass product, mainly the non-radioactive glass forming constituents, controls its long-term behavior.

Glass is made up of three main components, i.e., network formers, modifiers, and intermediates. The network formers form the backbone of a glass structure. They include SiO_2, B_2O_3 or P_2O_5. Modifiers include oxides such as Na_2O, K_2O, CaO, or BaO. They decrease the liquidus temperature of melts (i.e., the temperature where crystals can co-exist with the melt in thermodynamic equilibrium; the material is homogeneous above the liquidus temperature) and adds favorable processing properties to the glass. Higher content of modifiers, however, leads to degradation of chemical durability and thus only narrow range of variation is allowed for modifiers. Intermediates are to increase the chemical durability and viscosity of the melt and may also act as a network former in the presence of other network formers. Examples of intermediates are Al_2O_3, Fe_2O_3, MgO, or ZrO_2.

Details of which oxides are to be used depend on the type of glass. In terms of chemical durability, the most durable glass is a vitreous silica. However, making vitreous silica glass requires high processing temperature (>1700 °C) thus comes with very high cost (also with small waste loading). Therefore, adding other constituent materials should be considered.

Today, the main type of glass suggested for nuclear waste immobilization is borosilicate glass with processing temperature of 1200–1500 °C. It reflects a compromise between chemical durability and ease and cost of manufacturing. Use of boron helps to lower the processing temperature as boron stabilizes the glass structure at low temperature (500–600 °C). Boron also can have a beneficial effect on chemical durability. In 1981, France selected borosilicate glass for the vitrification of HLW generated from commercial spent fuel reprocessing. The U.S. also selected borosilicate glass in the same year for the vitrification of defense HLW. There is a tremendous momentum behind the borosilicate glass technology development in a number of countries.

The borosilicate glass is based on the Na_2O-B_2O_3-SiO_2 ternary system. The most basic borosilicate glass system adopted by the French vitrification program uses 56.1% SiO_2, 25.3% B_2O_3, and 18.6% Na_2O (in weight percent) (Manaktala 1992). Another example of borosilicate glass is the glass system developed at Savannah River Plant in the U.S. with 68.0% SiO_2, 13.0% Na_2O, 10.0% B_2O_3, 7.0% Li_2O, 1.0% MgO, and 1.0% ZrO_2. In this application, SiO_2 provides stable glass network structure. The oxide of boron, as part of network former, also reduces the melting temperature without much sacrifice of chemical durability. Using Na_2O and Li_2O as modifiers not only reduces the melting temperature and the viscosity of the melt but also provides sufficient electrical conductivity for the mixing of the constituents. Using MgO increases the chemical durability of the glass, and ZrO_2, even at small content, helps to increase the chemical durability, density, viscosity, glass transition temperature, and to decrease the thermal expansion coefficient.

The lead-iron phosphate glass is another potentially viable glass waste form. It is mainly based on the use of three major oxides: P_2O_5, PbO, and Fe_2O_3. Use of P_2O_5 as key constituents over SiO_2 in silicate glass is to lower processing temperature and provide a higher solubility for sulfates. The lead-iron phosphate glass provides good leach resistance and low melting temperature (~800 °C) but corrosion of refractory material and using lead as main ingredient remain a concern.

Making good glass for a given waste composition requires experiences based on trial and errors. The leaching characteristics of a glass are very sensitive to the changes in waste glass constituents. Significant variations in glass leaching can result due to small changes in glass composition.

9.4.1.3 Stability of Glass as Waste Form

For the stability of glass, potential degradation mechanisms that may affect the effectiveness of its use as waste form to need to be examined under the influence of chemical attack, heat, and radiation.

Chemical Stability of Glass: Glass Corrosion

While glass is chemically inert material, chemical durability of glass eventually depends on the reactions with water. When glass is in contact with water, water diffuses into the glass and go through the ion-exchange reactions with alkali ions (Na^+, Li^+). The following reaction is an example.

$$H_3O^+ + Na^+(g) \rightleftharpoons H_3O^+(g) + Na^+,$$

where (g) means glass.

With gradual removal of alkali ions from the surface of glass, a gel-like alkali depleted (reaction) layer is formed. This layer is somewhat open and allows the diffusion of water molecules and ions through it. Then the incoming water interacts with glass and starts dissolving the silica network of the glass ($2H_2O + SiO_2 \rightleftharpoons H_4SiO_4$). The reaction also breaks silicon-oxygen bonds (H_2O + Si-O-Si $\rightleftharpoons$ SiOH HOSi). The resulting hydroxyl ions also attack the Si-O bonds of the glass structure while some of the SiOH species may form Si-O-Si again. Eventually dissolution of the glass matrix takes place with the release of Si or other constituents to solution. The soluble constituents (e.g., Li, Na, Cs, B) of the glass are dissolved congruently with dissolution of glass matrix.

As the dissolved silica and other glass constituents accumulate in the solution, the release rate is reduced. When the solution is saturated with species dissolved, some elements released into solution precipitate on the surface layer and form a variety of secondary phases. Commonly, these secondary phases are crystalline ceramic materials (e.g., oxides, hydroxides, silicates, carbonates, etc.) and may act as a barrier to further dissolution of glass. Most of the less-soluble constituents (e.g., Fe, Al, Ca, actinides), upon the dissolution of the glass matrix, will precipitate at the glass surface as secondary solid phases. This indicates that solubility of elements largely controls the release of glass constituents.

These observations indicate that the processes of glass corrosion and their effects are interdependent. The released species may further go through sorption and colloid formation which in turn affect solubility. The chemistry and properties of the layer and the solution all affect the corrosion process. Differences in the type of bonding in the glass network also affect the leaching behavior of glass constituents. The long term leaching behavior of glass is controlled by the combined effects of these processes and the related factors such as glass composition, solution composition, pH, temperature, redox potentials, and the flow rate.

The chemical durability of a waste form is considered satisfactory if the normalized leach rate of the most intrinsically soluble species from the waste form is less than 1 $g/m^2/day$ in a comparatively large volume of hot deionized water (with the surface area to volume ratio being less than 0.1 cm^{-1}). The leach rate value of borosilicate glass is 1.12 $g/m^2/day$ (see Table 9.4) based on the leaching experiment performed as part of U.S. DOE's comparative waste form evaluation study (DOE 1982). The result is based on the measurement of cesium release (the most soluble species) from the glass.

Example 9.1: Glass Corrosion

Upon exposure to aqueous solutions, nuclear waste glass develops surface reaction layers. These reaction layers may work as a rate limiting diffusion barrier. An argument is also made that these layers provide little or no barrier to glass leaching. To examine these arguments, experiments were performed (Chick and Pederson 1983):

Experiment 1: A glass sample was placed in deionized water for 56 days following the specification of so-called MCC-1 (Materials Characterization Center static procedure) test. At the end of the 56 days, a relatively thick reaction layer was developed on the glass sample. Then the sample was removed from the water, rinsed, and placed in clean water for additional 7, 14, 28, 42, and 56 days. Placing the sample in the fresh, under-saturated water was to show the role of pre-built surface reaction layer in additional leaching of glass.

For the major elements of the glass structure, such as silicon, boron, and sodium, the results of their leaching were found to be very similar. Figure 9.8 shows an example of the results as cumulative release for boron. The behavior of the curve through the first 56 days shows the buildup of surface reaction layer and its effect on subsequent leaching. After the day 56, when the sample with built reaction layer was placed in clean water, we can postulate three different curves: (1) Surface layer controls subsequent leaching as a diffusion barrier, (2) No effect of surface layer, (3) Solubility of the released substance controls the leaching behavior. Figure 9.8 shows the corresponding curves to each of these assumptions.

Curve (1) will be followed if the first assumption holds, i.e., the surface reaction layer built during initial 56 days continues to control the subsequent leaching.

Curve (2) will be followed if the second assumption is true, i.e., preleaching during the first 56 days had no effect on subsequent leaching. In other words, if the effect of putting the preleached glass with built-in surface reaction layer is negligible, the new leaching behavior in the new fresh water will start a new leaching process while building another layer of surface reaction.

What actually was observed was Curved (3). The actual release behavior was close to Curve (2) but showed somewhat reduced release of boron. This indicates that the role of built-in surface reaction layer exists but was minor. The somewhat reduced level of release from the new saturation model may show the retardation effect of the surface reaction layer reducing the release or the increased concentration of the leached boron in the solution affecting the glass release behavior. In the latter case, the reduction may reflect the effect of solubility limit on the release.

(continued)

Example 9.1 (continued)

Experiment 2: Anther glass sample was placed in deionized water for 28 days to build up a reaction layer (again following the specification of the MCC-1 test). At the end of the 28 days, the glass sample was removed from the water and a new glass sample was placed for consequent leaching test in the same water. The objective of this second experiment was to measure the leach rate with no reaction layer present, but with the solution already partly saturated.

The triangles in Fig. 9.9 shows the observed results of leaching for silicon as the cumulative result. If leaching were limited by diffusion through the reaction layer, the concentration would rapidly rise when the new glass sample was placed with no pre-built surface reaction layer. Then, the resulting total release from the two glass samples should be greater than the release from a single sample. The fact that the total release from the two glass samples, to some extent, simply extended the release result from the first sample indicates that the concentration of silicon in the solution from the first sample had a strong effect on the release behavior of the second sample. This behavior indicates the importance of the effect of solution saturation in glass leaching.

The observations from both experiments show that diffusion may not be too important in glass corrosion and the influence of the reaction layer on leach rate is expected to be minimal/small. Instead, the results indicate the larger role of solubility of the leached substance.

Diffusion layers can play a role if the flow rate is high where the solution saturation effect becomes negligible. However, this is not likely in geological repository conditions due to low water infiltration rate in the system. Accordingly, solubility limited release appears to best describe glass leaching.

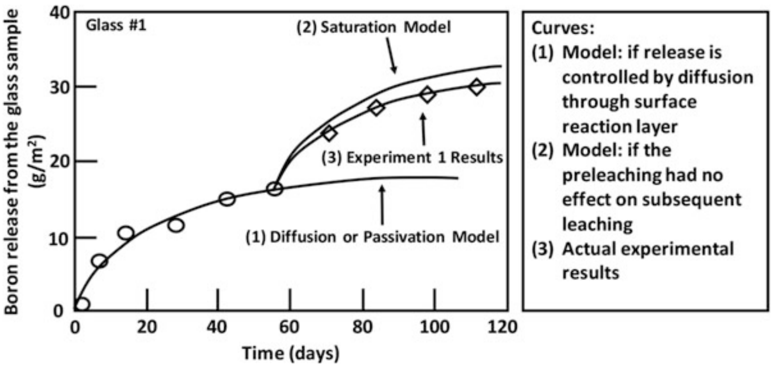


Fig. 9.8 Comparison of Experiment 1 results of glass leaching to models

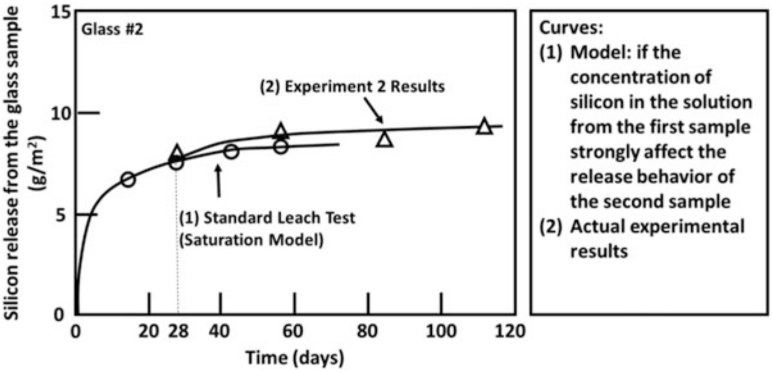


Fig. 9.9 Comparison of Experiment 2 results of glass leaching to models

Thermal Stability of Glass

As heat is released from the decay of fission products, glass waste form containing fission products could experience significant temperature increase. Increase in glass temperature can cause the transformation of the amorphous structure into more ordered crystalline lattice. Development of crystallization in glass due to temperature rise is called devitrification. Devtrification causes deletion of silica and a lowered chemical durability of glass, thus reduction in the waste loading capacity. Devitrification also leads to localized microcracking of glass.

The temperature range of concern for devitrification of glass is typically above 500 °C. As the expected temperature of geological repository is below 300 °C, devitrification is not expected to be a concern. In fact, most national waste management programs specify extended cooling periods of spent fuels before their emplacement in a geological repository to minimize the temperature excursion within a repository during post emplacement periods.

Another potential issue related to temperature rise of glass is phase separation. Under elevated temperature conditions (above 600 °C), borosilicate glass can separate into a silica-rich phase and a borate-rich phase. The segregated boron rich phase (depleted in silica) is chemically less durable. Again, due to the much lower temperature conditions, phase separation is not expected to occur in geological repositories.

Radiation Stability of Glass

Major sources of radiation in HLW glass are alpha particles from actinides decay and beta and gamma decays of the fission products. As discussed in Sect. 9.1.5, radiation damage from the low LET radiation of beta and gamma decays is negligible in glass. Radiation damage from high LET alpha particles is mainly in the form of atomic displacements which could lead to changes in volume, leach rate, stored energy, and mechanical properties of glass. However, studies conducted to date indicated that radiation effects on glass are not significant.

9.4.2 Ceramic

Ceramic, non-metallic inorganic solid, is one of the oldest man-made materials around, often made of clay minerals as raw materials. With the recognition of very long-term stability of certain mineral phases containing radioactive elements in nature (e.g., monazite), use of synthetic mineral phases, i.e., ceramics, for long-term stabilization of nuclear waste has been considered. In fact, in early 1950s in the U.S., using crystalline ceramic was considered to immobilize HLW. Along with good chemical durability, use of ceramics as waste form also comes with the benefit of high waste loading, high thermal conductivity, very high thermal stability, and capability to accommodate wide chemical variability.

However, the process of ceramic waste form fabrication is not straight forward. In comparison to glass where the waste materials are simply added to the glass precursors in its making, making ceramic waste requires an integrated approach to chemical design and the processing method. The chemical valence of the waste ions must match that of the host ions to maintain electrical balance of the host crystal phase. The cost of fabrication is also comparably high as the required processing temperature for ceramic is high (1700 °C) along with the need for hot pressing under high pressure.

9.4.2.1 Compositions of Ceramics as Waste Form

In a ceramic waste form, the constituents of nuclear waste occupy specific atomic positions in the ceramic's crystalline structure. Therefore, to incorporate various elements present in nuclear waste, appropriate host mineral phases must be provided. With the availability of such host phase, a solid mixture containing waste elements is formed. The results is the uniformly distributed elements of waste within the crystal lattice of the host component.

There are a number of host phases used for a ceramic waste form. This is because single-phase crystal structures don't usually contain all the host sites needed for a wide variety of elements present in nuclear waste. (there are also exceptions with monazite or sodium zirconium phosphate as discussed below). In addition, the major nuclides to be immobilized in the waste behave as ions in a variety of sizes. Thus the constituent mineral phases must provide host sites to incorporate these ions in the waste. These sites can be provided by the use of additives, such as Al^{3+}, Zr^{4+}, Si^{4+}, Ti^{4+}, Ca^{2+}, and RE^{3+} (rare earth). These additives are to alter or modify the actual waste composition to produce the desired combination of crystalline phases. Based on these considerations, three major classes of oxide ceramic have been suggested: silicate-based, alumina-based, and titanate-based. In addition, given the difficulty in matching the chemical valence state between the waste ions and the host ions, an "extra" phase(s) is used to accommodate the variations in the chemistry of the waste form.

Silicate-based ceramic was extensively investigated earlier in the U.S. (at Penn State) to immobilize nuclear waste. The so-called "supercalcine" waste form was developed from this effort including nine mineral phases. These nine phases include pollucite, ($CsAlSi_2O_6$), scheelite ($CaMoO_4$), fluorite $(U, Zr, Ce)O_2$), apatite ($Ca_5(PO_4)_3OH$), monazite ($REPO_4$), zirconia (ZrO_2), corundum (Al_2O_3), spinels ($NiFe_2O_3$), and various perovskites. This approach was expanded and continued for designing tailor-made ceramic waste forms.

Alumina-based ceramic waste form was examined at the Savannah River Plant for the immobilization of defense HLW. The mineral phases in the waste form include (1) uraninite ($(U, Th)O_2$) as a host for U, the actinides, and some RE, (2) magnetoplumbite (normally $X(Al, Fe)_{12}O_{19}$ where X is Sr, Ba, or mix of Cs and La) as a host for Sr and Cs, (3) nepheline ($NaAlSiO_4$) in the case of processing waste with high sodium concentration, (4) alumina, and (5) spinel (normally $MgAl_2O_4$). The alumina and the spinel phases serve as an extra phase to provide chemical stability and for microstructural isolation of the two host phases.

Development of titanate-based ceramics was also made at the Sandia National Lab of the U.S. as waste form for HLW. Titanates offer the possibility of higher *waste* loading than glass and have been most actively investigated. The waste form consisted primarily of rutile (i.e., TiO_2) along with zirconolite, perovskite, hollandite, and metal alloys. Similar phases were selected and further developed into a waste form known as "Synroc" at Australian National University.

Synroc is based on four main titanate minerals, i.e., zirconolite ($CaZrTi_2O_7$), hollandite ($Ba_{1,2}(Al,Ti)_8O_{16}$), perovskite ($CaTiO_3$), and titanium oxide (Ti_nO_{2n-1}). These minerals provide the capacity to incorporate nearly all of the elements in HLW into the crystal structure: Zirconolite is the host phase for U, the actinides, and RE. Hollandite is used as the host for large cations such as Cs, Rb, Sr, K, Ba, and various medium-sized cations such as Mo^{4+}, Ru^{4+}, Fe^{3+}, Fe^{2+}, Ni^{2+}, and Cr^{3+}. Perovskite is the host for a wide range of elements including Sr, U, Na, and RE. The titanium oxide is the majority phase (57%) and provides leach resistance. For synroc, waste loading of about 20% (by weight) was reported. The original form, synroc-C, was developed mainly to immobilize liquid HLW from spent fuel reprocessing. As borosilicate glass was selected for HLW vitrification, development of synroc continued by the Australian government for other applications such as plutonium immobization. U.S. DOE selected hot isostatic pressing with synroc to process calcine waste at the Idaho National Laboratory.

Another development on titanate-based approach produced a waste form for defense HLW, commonly called "tailored ceramics" by Lawrence Livermore National Laboratory and Rockwell International Science Center. The waste form uses five principal phases such as zirconolite, perovskite, magnetite spinel, magnetoplumbite, and nepheline. While zirconolite and perovskite provide similar roles as in Synroc, the magnetoplumbite incorporates Na, Sr, and Cs. The magnetite spinel is for the transition elements, and nepheline is to provide leach resistance as an extra phase. The main feature of this waste form is high waste loading in excess of 60%.

Natural monazites are also an interesting waste form candidate. They are known for ability to contain significant amounts of the actinides ions (thorium and uranium) along with the lanthanides, indicating the capability of the crystal structure to incorporate heavy actinides. These observations led to consideration of monazite as nuclear waste form. Monazite as a single phase ceramic waste form can be prepared with structures to incorporate all elements in nuclear waste. Such incorporation is enabled by appropriately synthesizing various structures within a mixed lanthanide orthophosphate phase ($LnPO_4$, with La, Ce, Nd, ... as Ln (lanthanide)). Monazites also feature very high chemical stability and excellent radiation damage resistance. With the characteristics of a negative temperature coefficient of solubility, monazite is also expected to show increase in chemical durability with increase in temperature.

Development of glass ceramics as waste form has also been pursued. Glass ceramics is a fine-grained ceramic product containing both amorphous phase and one or more crystalline phases. To produce a glass ceramic waste form, nuclear waste is first made into a calcined product and subsequently vitrified into a homogeneous glass. The glass is then reheated to intermediate temperatures of melts (between 500 and 800 °C) producing crystalline phases. In this process, nucleation agents such as TiO_2 and ZrO_2 are added to the base composition to control the crystallization process and to tailor the glass composition. Creation of the crystalline phases is to partition the waste radionuclides into the more durable host phases. The effort for synroc development as discussed above has also evolved into glass ceramic waste form.

Metal matrix is another substitute waste form for the vitrification of particulate HLW materials such as calcines. In this case, lead, zinc, aluminum, iron, iron alloys, or copper is used as the base metal. Its preparation is based on either by mixing the calcine with molten metal or by mixing the material with metal powder. Mixing is followed by pressing and sintering at around 950 °C. In the process, the calcines are dispersed in the continuous metal matrix with up to 70 vol % of the final product.

Inorganic ion exchange was also considered as a waste form in the past. In the inorganic ion exchange process, the liquid HLW is neutralized and flows through the inorganic ion exchanger sodium titanate ($Na_2Ti_3O_7$). The process removes nearly all of the chemical constituents of the waste except ruthenium, cesium, and technetium. These unremoved elements are removed either by zeolites (cesium) or by anion exchange (technetium and ruthenium). The resulting ion exchange media are mixed, dried, pressed, and sintered at about 1100 °C into a waste form.

9.4.2.2 Stability of Ceramics as Waste Form

Ceramic is a chemically inert refractory material. As ceramics are manufactured under high temperature and pressure, the resulting ceramic waste form is almost non-porous and provides very high chemical and radiation stability. Nevertheless, water can get into the ceramic waste form through the pores and loose the soluble portions of the ceramic bonds resulting in dissolution of the materials. Experimental studies on ceramic leaching reported the normalized elemental leach rate in the order of 1 $g/m^2/day$ for most soluble species like cesium (DOE 1982).

Theoretically, irradiation of ceramics can cause atomic displacements with possible volume expansion and microcracking. Accumulation of atomic displacements in the ceramics could also change the crystalline structure into an amorphous one by destroying long-range and local crystalline order of the structure. Such change would lead to degradation of thermo-mechanical properties and reduced chemical durability. However, studies on the effect of radiation on the leach behavior of ceramics showed such effects are insignificant. Nevertheless, some host phases may show increase in leach rate under irradiation. If the leachant is very corrosive, leaching may be enhanced due to irradiation.

9.4.3 Cement

Cement is a binder that hardens through chemical interactions with water. The hardened cement has been widely used for immobilization of low level or low and intermediate level waste. Among various types of cement available (e.g., Portland cement, blast furnace slag cement, pozzolanic cement, aluminous cement, and masonry cement), Ordinary Portland cement (OPC) is most commonly used due to ready availability, low cost, low temperature processibility, and good waste form performance.

9.4.3.1 Characteristics of Cement as Waste Form

Cement becomes a binder to solidify waste when mixed with water. Unlike concrete, which uses aggregate materials like sand and gravel to form solid structure, cement uses water to establish the bonds between the cement paste compounds and to harden the mixture. Cement comprises a class of materials and tailoring of its properties can be made to meet specific requirements. The waste materials are incorporated in the crystal matrix either by being physically trapped or by chemical reactions. Additives are often used to make processing easier or to improve the solidification properties (e.g., to control the viscosity of the mixtures or to improve the stability of the product). Additives include alkaline additives (hydroxides, oxides, carbonates) or nonalkaline salt. Cement has also been used as part of grout. Grout, as a mixture of Portland cement and various sands, is widely used to encapsulate nuclear waste in bulk form in containers, tanks, or structures.

Cement is readily available at low cost. It provides ability to solidify a wide variety of wastes with tolerance in the variations in the waste chemistry. It is also relatively safe and easy to handle and has long shelf life. However, cement provides low waste loading and presents the material compatibility issue with organic materials or high salt content waste. Also, presence of porosity in cement can allow water to cause leaching of radionuclides. U.S. NRC specifies that low and intermediate waste solidified in cement should have less than 0.5% by volume of the waste as free liquids to minimize the effect on compressive strength and leaching characteristics (NRC 1991).

9.4.3.2 Compositions of Cement as Waste Form

The chemically active portion of OPC is termed paste. It includes three main oxide (SiO_2, CaO, Al_2O_3) along with Fe_2O_3, SO_3 and H_2O, comprising more than 95 wt% of cement. MgO, Na_2O and K_2O are also added in smaller amounts. A water/cement ratio of 0.22–0.24 (by weight) is used to achieve complete hydration of the compounds. Hydration refers to the process of a substance being chemically combined with water. The excess water not consumed by hydration remains in the pore under high pH (in the range 12.5–14) conditions.

The hydrate phases found in cement pastes include portlandite (calcium hydroxide, $Ca(OH)_2$), ettringite ($3CaO.Al_2O_3.3CaSO_4.36H_2O$), monosulphate ($3CaO.Al_2O_3.CaSO_4.12H_2O$) and the calcium silicate hydrogel (C-S-H) phase. The first three phases are crystalline but the CSH phase is relatively noncrystalline gel-like component. The CSH phase is the most abundant phase and plays an important role for the stability of cement. The CSH phase largely controls the cement pore/paste chemistry and the Ca/Si ratio in the C-S-H phase (typically in the range 1.7–2.0) has a major effect in the sorption of anions in the waste. The additives used also influence the pore/paste chemistry.

9.4.3.3 Stability of Cement as Waste Form

Degradation of cement as a waste form involves both chemical and physical processes. In these processes, chemical reactions between the dissolved carbonate species and the cement play key roles. These reactions, called carbonation, reduces the Ca/Si ratio of the CSH gel (decalcification) and eventually destroys the CSH gel producing hydrous silica and calcium carbonate. Then increase in volume from the formation of calcium carbonate can cause microcracking of cement. Such volume increase also reduces the permeability of cement, thus increasing resistance to diffusion-related chemical degradation mechanisms. Carbonation can also immobilize radionuclides through formation of insoluble carbonate solids (such as Cs and Sr) or increase the leaching rate of elements or compounds (Cd, Co, Ca, Pb, and nitrate) through microcracks. Therefore, carbonation has both positive and negative effects on the stability of cement. The rate of carbonation depends on the moisture content of the material, cement mixture composition, and wetting-drying cycles experienced.

Another important reaction related to cement stability is the reactions between sulfate ions in solution and the compounds in hydrated cement. This is called sulfate attack. The sulfates dissolved from the monosulphate phase diffuse into the saturated pores of the cement and react with calcium hydroxide and calcium aluminate hydroxide. This results in the formation of gypsum and ettringite. Formation of gypsum reduces compressive strength of the waste form compromising the ability to withstand weight loadings. Formation of ettringite leads to volume increase, thus creating cracks and resulting in mass loss. As this sulfate attack is from the diffusion of sulfate ions into the saturated pores of the cement, its rate will be controlled by the permeability of the material in relation to carbonation as described. The results of these deleterious effect are disruptive expansion, strength loss, and/or disintegration of the material. Within several 100 years, the cement waste form may structurally degrades.

However, beyond this physical failures, effective chemical barrier characteristics of cement can be maintained for long periods. Studies on the evolution of pH over time indicated that the pH of cement will remain above 10.5 for very long time (e.g., millions of years). This high pH condition is maintained by the dissolution of excess amounts of portlandite and alkali present in hydrated cement. The exact time period will be dependent on the cement content of the system and infiltrating water flow (Atkinson 1985). Initially the pH (above 13 in a high alkali cement) will be controlled by the alkali hydroxides in the pore fluid lasting for the first 100 to 10,000 years. Following the dissolution of the alkali hydroxides, the pH will be buffered by the solubility of portlandite (at about 12.4) until all the free $Ca(OH)_2$ has undergone dissolution. This environment may last from 100–10,000 years to 1000–100,000 years. After this phase, the C-S-H gel material controls the pH until all the gel material has dissolved (maintaining the pH at about 10.5). Maintaining a high pH environment in the cementitious materials allows the retention of certain radionuclides such as ^{14}C.

Leaching of radioactive elements from cement waste form takes place through the combination of diffusion and dissolution. When the solution in the cement pore is contacted by external water with different ionic composition, the resulting concentration gradient will drive the diffusion of dissolved ions. Dissolution takes place in the pore water according to the solubility of the cement hydrate phases. The solubility of these phases is the highest in portlandite, followed by monosulfate, ettringite, and C-S-H gel. Presence of dissolved CO_2 can cause accelerated dissolution of these phases, thus enhancing leaching.

Radiation damage to cement occurs through radiolysis (i.e., radiolytic decomposition) of cement-pore water or any hydroxyl group in the cement. Such radiolysis leads to gas generation, principally hydrogen, which may increase the pore pressure in cement. Therefore, cement as a waste form is used mainly for low level or low and intermediate level waste but not for high level waste.

9.4.4 Polymers

Cement as waste form has an issue of materials compatibility with oils, organic liquids, and sometimes ion-exchange resins. This led to the development of polymer as an alternative for the immobilization of low and intermediate level waste.

9.4.4.1 Characteristics of Polymers as Waste Form

Physical and chemical properties of polymers are compatible with various types of radioactive wastes over a wide range of conditions including wet and dry wastes. In comparison with cement, the polymeric waste form has, in general, improved leach resistance, chemical inertness, and compatibility with organic based wastes (e.g., organic ion exchange resins). However, it comes with low radiation stability and slightly higher fabrication cost.

9.4.4.2 Compositions of Polymers as Waste Form

Main groups of polymer for waste form application include thermoplastic polymers and thermosetting polymers. Thermoplastic polymers are a plastic polymer material that becomes liquid when heated and solidifies upon cooling. An example is bitumen (asphalt). Bitumen becomes a viscous fluid when heated (to ~145 °C) but rehardens when cooled and becomes an elastic solid. Liquid bitumen can be mixed with waste solids and then cooled to form a solid matrix where wastes are mechanically incorporated into the solid structure.

Thermosetting polymers are a polymer material polymerized *in situ* from monomers or pre-polymers through cross-linking between polymer chains. The polymerized thermosets form permanent bonding between the linear chains of monomer molecules, as a very rigid three-dimensional chain structure. The wastes are again mechanically trapped in the structure. The energy to drive the process of polymerization can be provided by heat, catalysts, ultraviolet lights, etc. Vinyl ester-styrene (VES) is the principal thermosetting polymers used by the nuclear industry. VES is often referred to as Dow binder as the product of the Dow Chemical Co. Other thermosetting polymers also include polyester resins and epoxy resins. VES process, unlike other polymeric waste forms, does not require heating in waste processing thus minimizes volatilization and release of radionuclides such as ^{14}C.

The polymerization process with VES starts with the addition of a catalyst to the waste/binder mixture. A catalyst or an initiator is a relatively unstable material (e.g., benzol peroxide) that decomposes to form free radicals. Free radicals are used to open the double bond of a monomer and monomers are added successively to the growing chain (in seconds). Promoters (e.g., cobalt naphthenate or dimethyl aniline) are used to accelerate the decomposition of the catalyst before the waste/binder mixture sets (IAEA 1988). When all the monomer is consumed or when the growing chains meet end to end, the growth of the chains ends. Polymerization is complete in ~1 h. The rate of polymerization, the molecular weight and the molecular weight distribution can be controlled by varying the catalyst concentration.

9.4.4.3 Stability of Polymers as Waste Form

Polymers as a non-porous waste form exhibit generally good chemical resistance. However, polymers are most susceptible to radiation damage among the waste forms considered. Under irradiation, chemical bonds are broken through ionization and bond rearrangements occur leading to various chemical changes. These changes include cross-linking (bonding between chains), forming larger polymer chains, fragmentation into shorter chains, or gas evolution. Such changes cause alterations in physical and mechanical properties, such as viscosity, conductivity, tensile strength, hardness and flexibility. For example, fragmentation into shorter chains results in the formation of a new polymer with degraded physical properties. Cross-linking often produces a more rigid and brittle polymer. Accordingly, polymer is not recommended as waste form for high radiation conditions.

9.4.5 Comparisons of Materials for Waste Immobilization

Depending on the type of nuclear waste and its characteristics, different materials can be employed and configured for the development of waste forms. As discussed, cement or polymers are mainly considered for low level or low and intermediate level waste. For liquid high level waste, glass has been adopted. Use of glass is based on comparative studies of a variety of waste form materials. For example, in the U. S., as many as 17 materials were considered as potential HLW waste form media in the early 1980s (DOE 1982). The list includes three main types, glass, ceramics, and concrete (with high temperature or pressure processing) and was narrowed to include borosilicate glass and high-silica glass for glass, super-calcine ceramic and SYNROC for ceramic, and FUETAP (formed under elevated temperature and pressure) concrete and hot-pressed concreate for concrete. Comparisons of these materials regarding product performance as well as process factors are shown in Table 9.3.

In terms of product performance, ceramics were very good, in particular with respect to waste loading and long-term stability. Glass had an average performance in general. High-silica glass has the highest waste loading among all of the materials compared. Borosilicate glass received the highest score in development status and thermal stability (along with high-silica glass and super-calcine ceramic). Concrete, in general, showed inferior product performance. Leachability of concrete was the worst among the three main types.

In terms of chemical durability, more detailed comparison is shown in Table 9.4. The results are based on cesium release. The average daily release rate from the best performing high-silica glass was 0.405 (g/m^2/d). This is followed by 0.806 (g/m^2/d) of SYNROC and 1.35 (g/m^2/d) of borosilicate glass. The measured release rate from FUETAP concrete was 45.6 (g/m^2/d). It can be said that, except the concrete, the differences observed between glass and ceramic were not significant. Based on

Table 9.3 Evaluation of candidate waste forms for immobilization of HLW at U.S. Savannah River Plant (DOE 1982)

			Glass		Concrete		Ceramic	
		Weight of category	Borosilicate Glass	High-silica Glass	FUETAP[a] Concrete	Hot-pressed Concrete	Super-calcine Ceramic	SYNROC
Product factors	Development status	2	5	2	3	2	2	3
	Waste loading	2	3	2	1	3	5	5
	Leachability	5	3	5	1	1	4	4
	Long-term stability	5	3	3	2	4	5	5
	Thermal conductivity	1	2	2	2	2	2	2
	Thermal stability	2	5	5	2	3	5	5
	Transportation safety	2	4	4	2	3	5	5
	Weighted sum		66	68	33	49	81	83
Process factors	Complexity	6	4	3	5	3	2	2
	State of development	3	5	2	4	1	1	1
	Quality assurance	3	3	3	3	3	1	1
	Yield and recycle	3	3	2	2	2	1	1
	Process safety	3	4	4	4	3	3	2
	Weighted sum		69	51	69	45	30	30
Overall score			**135**	**119**	**102**	**94**	**111**	**110**

[a] *FUETAP* Formed under elevated temperature and pressure

Table 9.4 Chemical durability comparison of waste forms in US DOE product performance evaluation (using the leachate rate of Cs ($g/m^2/d$)) (DOE 1982)

	Borosilicate Glass	High-silica Glass	SYNROC-D	Tailored Ceramic	FUETAP[a] Concrete
Deionized water at 90C	1.12	0.028	0.75	4.50	48.
Silicate water at 90C	0.73	0.121	0.38	2.25	37.
Silicate water at 150C	2.28	1.02	0.740	8.14	37.
Brine solution at 90C	0.35	<0.20	0.20	5.46	53.
Brine solution at 150C	2.28	0.654	1.96	5.64	53.
Average	**1.35**	**0.405**	**0.806**	**5.20**	**45.6**

[a] *FUETAP* Formed under elevated temperature and pressure

product performance, both ceramics and glass would be suitable with ceramics having better performance over glass. Concrete would be an inferior waste form material.

In terms of process factors, borosilicate glass clearly outperformed ceramics as well as concrete in all category, i.e., process complexity, process safety, quality assurance, yield and recycle of materials, and state of development.

The final candidates chosen through the screening work were borosilicate glass and synroc. They both provide compatibilities with wide range of materials from various waste streams. However, given the maturity of the technology and readiness for industrial scale applications, borosilicate glass has been chosen for vitrification of defense and civilian HLW in the U.S. (DOE 1982). Similar decision was also made in France, Russia, and other countries. This was largely due to industrial readiness of borosilicate glass with economic viability driven by the imminent needs for solidification of liquid high level waste from both civilian and military reprocessing operations. Recently, synroc has also been chosen for immobilization of plutonium in the U.S.

9.4.6 Modeling Waste Form Leaching

Release of radionuclides from waste form takes place due to chemical degradation of materials. Such degradation is initiated by migration of water into the waste form, leading to hydrolysis (surrounded by water molecules) and dissolution of materials. Through dissolution, radionuclides become part of the aqueous phase. How much dissolution takes place is controlled by solubility limits imposed by the primary waste form or through formation of new alteration phases in the solution. Migration of the dissolved radionuclides will occur through diffusion, driven by the concentration gradient. If there is a pressure gradient or groundwater flow, the movement is also driven by advection (bulk movement). Modeling of waste form leaching is thus based on describing the process of diffusion and dissolution.

9.4.6.1 Release by Diffusion

Diffusion of a substance in a medium occurs through random movement of molecules from a high concentration area to a low concentration area. In a waste form, diffusion is the rate controlling mass transport mechanism until network dissolution becomes dominant.

Assume that release of a non-radioactive substance from a waste form is described by the following diffusion equation: The waste form is a semi-infinite slab medium (infinite in the y and z directions). The change in the concentration of a diffusing substance is given by,

$$\frac{\partial C}{\partial t} = D\frac{\partial^2 C}{\partial x^2} \tag{9.1}$$

The initial conditions of the concentration are: $C = C_0$ *(0 < x < l, within the waste form), C = 0 (x > l, outside the waste form)*, and the boundary conditions of the concentration are: $C(x = \infty) = 0$, $\int_{-\infty}^{\infty} C(x)dt = M$, where M is the total mass of the substance in the waste form.

The solution of the diffusion equation as the concentration of the substance outside the waste form is,

$$C(x,t) = \frac{M}{\sqrt{4\pi Dt}}e^{-\frac{x^2}{4Dt}} \tag{9.2}$$

The rate of release, i.e., loss of diffusing substance from the semi-infinite waste form medium per unit cross-sectional area is given by

$$J(t) = -\left(D\frac{\partial C}{\partial x}\right)_{x=0} = C_0\sqrt{\frac{D}{\pi t}} \tag{9.3}$$

The term, *J(t)*, represents the diffusive leach rate per surface area (g/cm^2/sec) of a nonradioactive substance into a leachant (a liquid used in leaching) from an idealized, semi-infinite medium. The rate of release from the waste form over the entire surface are is then given by,

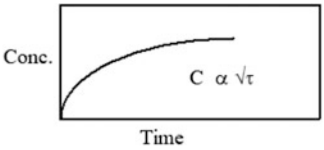


Fig. 9.10 The characteristic of diffusion controlled release

$$q(t) = SA \cdot C_0 \sqrt{\frac{D}{\pi t}} \tag{9.4}$$

where, $q(t)$ is the total diffusive leach rate (e.g., g/sec) of the substance, SA is the geometrical surface area of the waste form (cm^2), C_0 represents the initial bulk concentration of the leached species in the waste form (g/cm^3), and D is the effective bulk diffusion coefficient (cm^2/sec) in the waste form.

To represent the total cumulative release, we take an integration of $J(t)$ over the given time period and multiply it by SA.

$$\begin{aligned} \textit{Cumulative release over } t &= \left[\int_0^t J(t')dt'\right] \times SA = \left[\int_0^t C_0\sqrt{\frac{D}{\pi t'}}dt'\right]SA \\ &= 2C_0 \cdot SA\sqrt{\frac{Dt}{\pi}} \end{aligned} \tag{9.5}$$

This indicates that, as the characteristic of diffusion controlled release, mass transport by diffusion through a layer or medium is dependent of the square root of time. This is depicted in Fig. 9.10.

Also, the total cumulative mass released from the waste form per surface area is,

$$\begin{aligned} \textit{Total mass released per surface area} &= Q(t) \equiv \frac{M^{element}_{leached\ out}}{SA} \\ &= 2C_0 \cdot SA\sqrt{\frac{Dt}{\pi}} \cdot \frac{1}{SA} = 2C_0\sqrt{\frac{Dt}{\pi}} \end{aligned} \tag{9.6}$$

The fractional cumulative release is given by dividing the cumulative release by the initial mass ($M = C_0 * V$).

$$F(t) = \frac{2C_0 \cdot SA\sqrt{\frac{Dt}{\pi}}}{V \times C_0} = 2\frac{SA}{V}\sqrt{\frac{Dt}{\pi}} \tag{9.7}$$

where V represent the specimen volume.

9.4.6.2 Release by Dissolution

The process of dissolution-based mass release from the waste form is usually described by using empirically derived dissolution rate. Then, the rate of release, *q (t)* (g/cm^2/day), is directly controlled by the dissolution rate as,

$$q(t) = j_0 \times \frac{M_{waste\ form}}{SA} \tag{9.8}$$

where, $M_{waste\ form}$ is the total mass of the element being released, *SA* is the surface area, and j_0 is the dissolution rate, defined as:

$$Dissolution\ rate = j_0 = \frac{Cumulative\ amount\ of\ released\ mass}{Total\ element\ mass \times leaching\ period} \tag{9.9}$$

Example 9.2: Leach Rate and Effective Bulk Diffusion Coefficient
A series of static leach tests has been performed on identical samples of simulated borosilicate glass. The samples are cylindrical, with a radius of 5 cm and a length of 10 cm. The density of the glass is 2700 kg/m^3. The samples are doped with non-radioactive molybdenum (3% by weight) and strontium (1.5% by weight) to simulate the leaching of these fission product nuclides. As leachant, a dilute brine solution and deionized water were used in the experiment, each at 20 °C. The measured quantities of molybdenum and strontium in solution after 28 days of leaching are given in the following table.

Table Results from 28-day cumulative leach tests, as mass in solution (g) ($M_{leached\ out}^{element}$).

Element	Deionized water	Brine
Sr	0.0012	0.062
Mo	0.192	0.139

1) Calculate the glass leach rate, *q(t)*, in both brine and deionized water, normalized to each element. (Note: It is reasonable to assume that the samples are large enough to use the leaching expressions derived for semi-infinite slab geometry.) Use the data in the table and assume that leaching takes place as a result of dissolution of the glass matrix.
2) For the case of leaching in deionized water, assume that the leaching mechanism is diffusion and that the glass matrix is effectively insoluble. Use the data in the table and estimate the values of diffusion coefficients for each element in deionized water.

(continued)

Example 9.2 (continued)

Solution:

1. Use:

$Dissolution\ rate = j_0 = \frac{Cumulative\ amount\ of\ released\ mass}{Total\ element\ mass \times leaching\ period}$ from Eq. 9.9

$Glass\ leach\ rate = q(t) = j_0 \times \frac{M_{glass}}{SA}$ based on pure dissolution assumption and Eq. 9.8)

Surface area, $SA = 2\pi rh + 2\pi r^2 = 4.71 \times 10^{-2}\ m^2$
Volume, $V = \pi r^2 h = 7.85 \times 10^{-4} m^3$
Mass, $m = \rho V = 2700 kg/m^3 \cdot 7.85 \times 10^{-4} m^3 = 2.12\ kg$

Initial Sr mass $= M_0^{Sr} = 0.015 \cdot 2.12\ kg = 3.18 \times 10^{-2}\ kg$
Initial Mo mass $= M_0^{Mo} = 0.03 \cdot 2.12\ kg = 6.36 \times 10^{-2}\ kg$
Leaching period $= 28$ days $= 2.419 \times 10^6\ s$

Dissolution in deionized water:

$$Dissolution\ rate\ of\ Sr = j_{0DW}^{Sr} = \frac{0.0012 \cdot 0.001\ kg}{3.18 \times 10^{-2} kg \times 2.419 \times 10^6 s}$$
$$= 1.56 \times 10^{-11}\ s^{-1}$$

$$Dissolution\ rate\ of\ Mo = j_{0DW}^{Mo} = \frac{0.192 \cdot 0.001\ kg}{6.36 \times 10^{-2} kg \times 2.419 \times 10^6 s}$$
$$= 1.25 \times 10^{-9}\ s^{-1}$$

Dissolution in brine:

$$Dissolution\ rate\ of\ Sr = j_{0brine}^{Sr} = \frac{0.062 \cdot 0.001\ kg}{3.18 \times 10^{-2} kg \times 2.419 \times 10^6 s}$$
$$= 8.06 \times 10^{-10} s^{-1}$$

$$Dissolution\ rate\ of\ Mo = j_{0brine}^{Mo} = \frac{0.139 \cdot 0.001\ kg}{6.36 \times 10^{-2} kg \times 2.419 \times 10^6 s}$$
$$= 9.03 \times 10^{-10} s^{-1}$$

(continued)

Example 9.2 (continued)

Glass leach rate in deionized water:

$$q_{DW}^{Sr} = j_{0DW}^{Sr} \cdot \frac{M_{glass}}{SA} = 7.0 \times 10^{-10} \left[\frac{kg}{m^2 - s}\right]$$

$$q_{DW}^{Mo} = j_{0DW}^{Mo} \cdot \frac{M_{glass}}{SA} = 5.6 \times 10^{-8} \left[\frac{kg}{m^2 - s}\right]$$

Glass leach rate in brine:

$$q_{brine}^{Sr} = j_{0brine}^{Sr} \cdot \frac{M_{glass}}{SA} = 3.6 \times 10^{-8} \left[\frac{kg}{m^2 - s}\right]$$

$$q_{brine}^{Mo} = j_{0brine}^{Mo} \cdot \frac{M_{glass}}{SA} = 4.1 \times 10^{-8} \left[\frac{kg}{m^2 - s}\right]$$

2. Use:

$Q(t) = 2C_0\sqrt{\frac{Dt}{\pi}} = \frac{M_{leached\ out}^{element}}{SA}$ from Eq. 9.6

where, C_0 is the initial concentration of a nuclide in the glass (kg/m^3), and since $C_0 = \frac{M_0^{element}}{V}$

$$\frac{2M_0^{element}}{V}\sqrt{\frac{Dt}{\pi}} = \frac{M_{leached\ out}^{element}}{SA}$$

$$\rightarrow D = \left(\frac{M_{leached\ out}^{element} \cdot V}{2M_0^{element} \cdot SA}\right)^2 \cdot \frac{\pi}{t}$$

$$\begin{aligned} D_{DW}^{Sr} &= \left(\frac{M_{leached\ out}^{Sr} \cdot V}{2M_0^{Sr} \cdot SA}\right)^2 \cdot \frac{\pi}{t} \\ &= \left(\frac{1.2 \times 10^{-6} kg \cdot 7.85 \times 10^{-4} m^3}{2 \cdot 3.18 \times 10^{-2} kg \cdot 4.71 \times 10^{-2}\ m^2}\right)^2 \cdot \frac{\pi}{2.419 \times 10^6 s} \\ &= 1.3 \times 10^{-19}\ [m^2/s] \end{aligned}$$

Similarly, $D_{DW}^{Mo} = 8.2 \times 10^{-16}\ [m^2/s]$.

9.5 Corrosion of Metals

Metals are susceptible to degradation through various stress factors. Presence of elevated temperatures, high levels of radiation, oxygen, water, and other species in the host media becomes stress factors driving degradation of metals as waste package materials in a geological repository. Corrosion is the most significant mechanism among various forms of degradation in altering the nature of metals.

Corrosion is the gradual deterioration of metals (or other materials) due to chemical/electrochemical reactions with their surroundings. Long-term performance of nuclear waste packages will largely depend upon corrosion characteristics of the component metal or metal alloy materials.

9.5.1 Basic Understanding of Corrosion

Corrosion is a specific type of redox reactions. It proceeds with a loss of electrons of metals reacting with water and oxygen. The driving force for corrosion is the energy stored in the metals. This energy was absorbed as part of the metal manufacturing process through refining of natural ore materials. Metals, as a product of refining, are in temporary state and, through the release of the absorbed energy, will eventually return to the natural state. That process is corrosion. Corrosion is a natural consequence of metals being in a temporary form.

Consider an example of iron. Iron, a naturally occurring material, commonly exists as hematite, an oxide of iron (Fe_2O_3). Heating the iron oxide drives off oxygen and leaves a base metal, steel. With the added energy, steel is thermodynamically unstable. Under normal environmental conditions with no additional energy addition, steel will go back to its natural state, iron oxide (i.e., rust) by releasing energy.

The amount of energy needed to extract a base metal varies depending on the element. The energy is relatively high for magnesium, aluminum, and iron. For copper and silver, the energy is relatively low. The higher the energy needed to extract the base metal, the more susceptible the resulting metal to corrosion.

Most metals or metal alloys corrode merely from exposure to moisture in the air. Exposure to high temperature gases that contain oxygen also causes corrosion. The process of corrosion is also strongly affected by the presence of other substances to which the metal is exposed. Factors that affect the tendency for corrosion of metal include presence of reactive ions (e.g., chlorine, sulfur ions), variations in local chemistry conditions (e.g., redox potential, pH, and temperature), small variations in composition, stress fields, and presence of microbe. In addition, radiation can play a role in corrosion of metals.

Corrosion has various types and includes uniform corrosion, localized corrosion (pitting or crevice corrosion), galvanic corrosion, intergranular corrosion, stress corrosion cracking (SCC), and microbiologically influenced corrosion. Figure 9.11 shows the schematics of various types of corrosion.

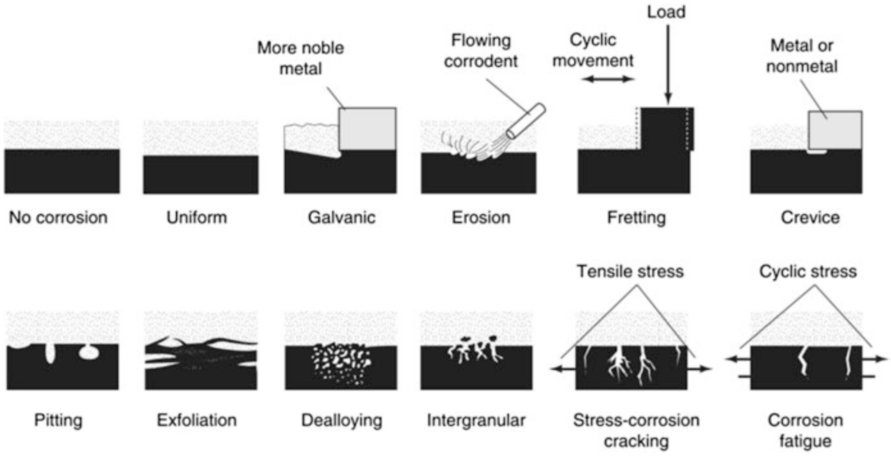


Fig. 9.11 Schematic summary of various forms of corrosion (Davis 2000)

9.5.1.1 Corrosion Cell

For corrosion to occur, corrosion cell must be present. A corrosion cell is composed of an anode, a cathode, and an internal and an external circuit that electrically connects the electrodes. Normally, water provides the internal circuit and works as the carrier of ions and electrons (i.e., as an electrolyte). The external circuit is given by the metal. The anode and cathode are adjacent to each other on a same piece of metallic surface.

At the anode, the metal dissolves and releases electrons into the bulk of the metal (e.g., $Fe \rightarrow Fe^{2+} + 2\,e^{-}$). The electrons migrate through the metal and react with H^+ in the solution at the cathode to form H_2 (e.g., $2H^+ + 2e^- \rightarrow H_2$) or are consumed to form hydroxyl ion (e.g., $\frac{1}{2}O_2 + H_2O + 2e^- \rightarrow 2OH^-$). Therefore, oxidation (at the anode) and reduction (at the cathode) reactions are essential part of corrosion. Difference in potential between the anode and the cathode is the driving force for corrosion indicating the degree of thermodynamic instability in the metal.

The corrosion cells are present as the metal surface is not uniform due to variations in composition, local environment, orientation of grain structure, and differences in the amount of stress and surface imperfections. Thus, a piece of metal can be covered with many tiny corrosion cells (Fig. 9.12). These tiny corrosion cells are also called “local action cells” and drive the corrosion process.

The products of the anodic and cathodic processes frequently migrate through the solution and further react with each other to produce corrosion products. For example, when iron is present in water, the ferrous ions released from the surface of iron (anode) migrate through the electrolyte and meet the hydroxyl ions from the cathode which were moving in the opposite direction. They combine to form ferrous hydroxide ($Fe(OH)_2$) as corrosion product.

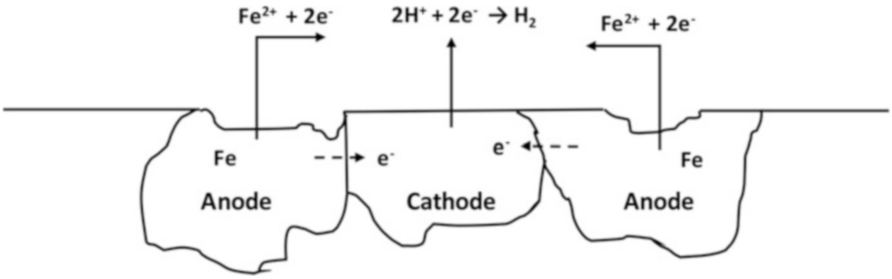


Fig. 9.12 Corrosion cells on metallic surface

$$2Fe + 2H_2O + O_2 \rightarrow 2Fe^{2+} + 4OH^- \rightarrow 2Fe(OH)_2$$

Ferrous hydroxide further reacts with oxygen in the solution to form ferric hydroxide ($Fe(OH)_3$, rust).

$$2Fe(OH)_2 + H_2O + ½\,O_2 \rightarrow 2Fe(OH)_3 \text{ (rust)}$$

Oxidized or hydrated cations may also precipitate as oxide films (Fe_2O_3 (hematite or rust) or Fe_3O_4 (magnetite)) on the surface of the metal. These oxide films may provide the protection of the base metal.

$$4Fe + 3O_2 \rightarrow 2Fe_2O_3, \text{or}$$
$$Fe(OH)_2 \rightleftharpoons FeO + H_2O$$
$$Fe(OH)_3 \rightleftharpoons FeO(OH) + H_2O$$
$$2\,FeO(OH) \rightleftharpoons Fe_2O_3 + H_2O$$

9.5.1.2 Passivity

In early studies of metal corrosion, investigators noted an unusual behavior of metals showing corrosion resistance: When a small piece of iron or steel is immersed in a concentrated nitric acid at room temperature, no corrosion reaction was observed. It turns out that when metals corrode, it quickly forms a surface oxide layer. This oxide layer provides protection of the surface against corrosion.

Passivity refers to a loss of chemical reactivity by metal under certain environmental conditions. Metals that normally corrode will sometimes exhibit a passivity to corrosion. In the passive state, the corrosion rate of a metal is very low, 3–6 orders of magnitude lower than in the active state.

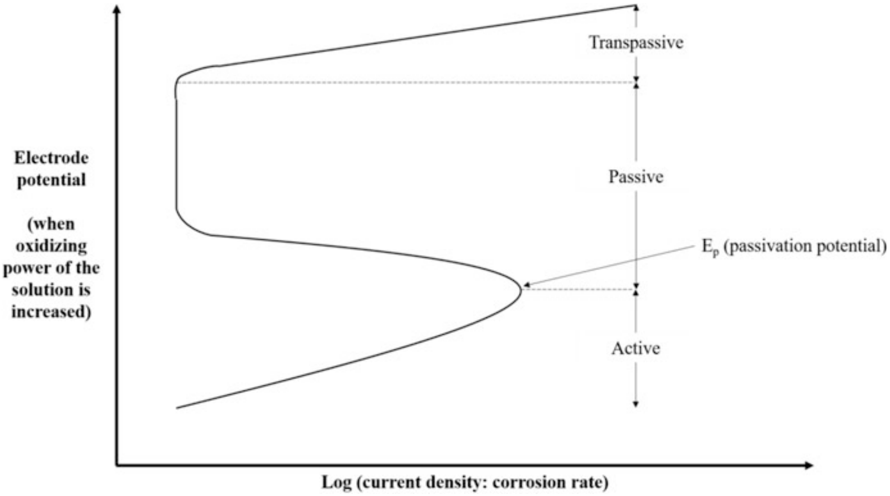


Fig. 9.13 Polarization diagram of a passivable system (Evans diagram)

The corrosion rate is typically represented as the current density, i.e. the current per unit area of a specimen used for the corrosion test. The electric current per unit area is determined by measuring the total current in the corrosion cell and the surface area of the specimen.

Polarization diagrams of corroding metals, sometimes called Evans diagrams (Fig. 9.13), are graphs of potential versus log current or log current density. To establish a polarization diagram, changes in the corrosion rates are experimentally determined with the increase in the electrode potential. Increasing the electrode potential can be through the increase in the oxidizing power of the solution, e.g., by adding oxygen. (note that changes in oxidizing power are directly related to electrode potential change, as shown in 4.3.3.4 (Eq. 4.23)). As shown in Fig. 9.12, the behavior of the given metal under the influence of corrosion can be divided into three regions as a function of electrode potential: active, passive, and trans-passive.

In the active region, when the oxidizing power of the solution is increased, a rapid increase in corrosion rate (measured as current density) occurs. Then above some critical potential E_p, even when the oxidizing power is increased, corrosion rate decreases. During this transition from the active to the passive region, very large reduction in corrosion rate is observed. This corrosion resistance above E_p is defined as passivity. In this passive region, corrosion rate remains the same when the oxidizing power is further increased. However, under extremely strong oxidizing conditions, corrosion rate increases again. This region is termed the trans-passive region.

Passivity occurs due to the buildup of a stable, protective metal oxide layer on the surface of the metal. Once the layer, as insoluble corrosion products, is formed, it acts as a barrier to mass transport separating the metal surface from the environment.

Therefore, in the passive region, corrosion either markedly decreases or stops. For further corrosion to occur, the reactants must diffuse through the oxide film. Metals such as zirconium, chromium, aluminum, titanium, and the stainless steels form thin, tenacious oxide films when exposed to water or to the atmosphere. The film is extremely thin but very effective in producing passivity in a metal.

While in the passive region, if the passive film is breached (called the breakdown), so-called localized corrosion can start. Two types of breakdown processes exist: electrochemical breakdown and mechanical breakdown. Electrochemical breakdown takes place at a potential above a specific "breakdown potential." Mechanical breakdown occurs when the passive film is ruptured as a result of stress or abrasive wear. All breakdown mechanisms involve a damaging species such as the chloride ion.

9.5.1.3 Pourbaix (Eh-pH) Diagrams

The conditions necessary for passivation are recorded in a diagram called Pourbaix diagrams. A Pourbaix diagram is where possible stable (equilibrium) phases of an aqueous electrochemical system are shown. An example of a Pourbaix diagram is given in Fig. 9.14 for the corrosion of copper (in water at 25 °C with chloride concentration 0.001 M). The region of passivity as well as immunity are shown in the figure along with the region of electrode potential and pH where corrosion of copper is expected

The lines in the Pourbaix diagram are the boundaries of predominant ions. Therefore on these lines, the activities of ions are equal. The diagram is a compact summary of specific conditions of electrode potential and pH under which the metal either can react or does not react (immunity). It also show the reaction products that will be present when equilibrium has been attained as a function of electrode potential and pH. The information in Pourbaix diagrams can be used to adjust potential and/or pH for the prevention of corrosion thermodynamically.

9.5.2 Uniform Corrosion

Uniform corrosion (or "general corrosion") is the regular, uniform removal of metal from a surface through oxidation. This is the most common form of metal corrosion in aqueous environments. In uniform corrosion, a nearly infinite number of microcells are established uniformly distributed over the metallic surface. The result is a uniform attack on the metal surface. Uniform corrosion is characterized by a chemical or electrochemical reaction that proceeds uniformly over the entire exposed surface or over a large area. This results in an even, general wasting away of metal from the corroding surface. Therefore, under uniform corrosion, measurement of

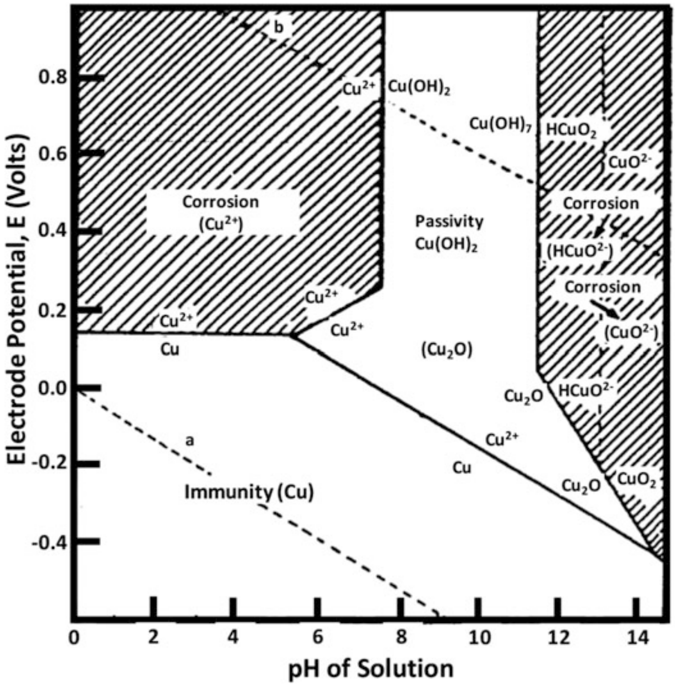


Fig. 9.14 Pourbaix diagram for the Cu-H_2O system at 25 °C with chloride concentration 0.001 M, showing the domains of corrosion behavior. Line a is the potential for hydrogen evolution and Line b is the potential for oxygen reduction. The shaded area for general corrosion (marked with diagonal lines) marks the pH and potential inside an active region (source: Guy and Rhines, 1962)

corrosion penetration rate can be used to predict the life of equipment or structure. Typically corrosion is measured in mm/year or mpy (melts per year, 1 melt = 1/1000 inch).

The important factors controlling the uniform corrosion behavior of metals are pH, redox potential, ionic compositions, and temperature (and radiolysis, if radiation is present). In uniform corrosion, anodic areas (where metal dissolution occurs) and cathodic areas (where hydrogen evolution or oxygen reduction occur) frequently alternate, in a microscopic scale. If impurities (e.g., carbide precipitates) are present on the metal surface, corrosion can become localized around the impurities. If corrosion is unavoidable, the most desirable form is uniform corrosion.

Example 9.3: Rate of Corrosion Calculation

A sheet of carbon steel 2 m wide by 2 m long has lost 50 g to corrosion over the past 6 months. Determine the penetration rate of the steel by corrosion in mm/year. Use 7.87 g/cm^3 as the density of carbon steel.

(continued)

Example 9.3 (continued)
Solution:

Since the surface area is 4 m^2 and the exposure duration is 6 months we have a corrosion rate of 50 g/(4 m^2 × 6 months × 30 days/month) or 0.069 g m^{-2} day^{-1}.

To converting into mm/year,

0.069 g m^{-2} day^{-1} $\times 10^{-4}$ (m^2/cm^2) × 365 (day/year) / 7.87 (g/cm^3) × 10 (mm/cm) = 0.0032 mm/year.

9.5.2.1 The Pilling-Bedworth Ratio

When an oxidation layer is formed on a surface of metal through corrosion, depending on whether the amount of oxide produced is enough to cover the metal consumed by corrosion, the layer can be protective or non-protective. This aspect is captured by so-called the Pilling-Bedworth (PB) ratio. The PB ratio is the ratio of the volume of reaction product (oxide) to the volume of metal from which the product forms. If the ratio is greater than one, the oxide product is protective. If this ratio is less than one, insufficient amount of oxide is produced to cover the metal and the oxide is non-protective. For example, for calcium and magnesium, the ratios are 0.64 and 0.79, respectively, and their oxides are nonprotective. For aluminum and chromium, the ratios are 1.3 and 2.0, respectively, and the oxides are protective.

Example 9.4: The Pilling-Bedworth Ratio Calculation for Oxidation
Zirconium oxidizes to ZrO_2. Calculate the Pilling–Bedworth ratio for the oxidation and indicate whether the oxide layer would be protective.

Solution:

The Pilling–Bedworth ratio = Volume of oxide/Volume of metal =$(M_o/\rho_o)/(M_M/\rho_M)$where M_o is the molecular weight of ZrO_2, (123.22 g), M_M is the weight of Zr (91.22 g), ρ_M is the density of Zr (6.5 g/cm^3) and ρ_o is the density of the oxide (5.9 g/cm^3).

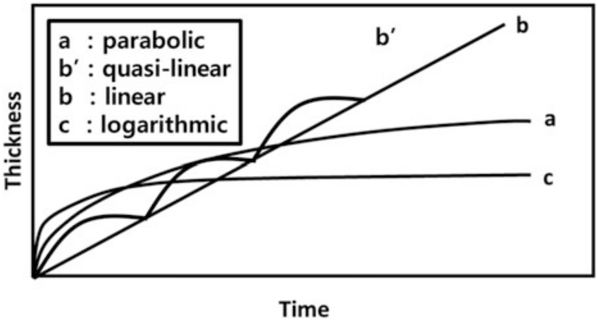
Thus, P–B ratio = (123.22/5.9)/(91.22/6.5) = 1.5

Since the ratio > 1, the oxide is protective.

9.5.2.2 Quantitative Description of Oxide Product Development Under Uniform Corrosion

To describe how metals corrode over time uniformly, different models have been suggested. These models explain formation of oxide film on any metal from uniform corrosion as a function of time.

Fig. 9.15 Metal corrosion thickness vs. time



If the growth of oxide is due to diffusion of ions (or electrons) through the oxide layer, the rate of growth is inversely proportional to the metal thickness. In this case, oxide growth follows parabolic time dependency (the curve a in Fig. 9.15): The oxide growth occurs while the oxidation rate decreases. The oxide film limits the rate of oxidation by limiting the diffusion of oxidant to metal surface or electrons to solution.

If the oxide film is not protective or if corrosion product is either liquid or volatile, the reaction rate is constant at an interface. Then, the oxide growth relationship is linear (the curve b in Fig. 9.15). From time to time, the oxide layer can also crack due to pores developed in the layer while the rate of oxidation is parabolic. In this case, the oxide growth behavior is quasi-linear (the curve b' in Fig. 9.15). Also if the oxide layer develops parallel flaws inside, then the effects of oxide on diffusion is reduced. In this case, the oxide growth follows logarithmic behavior (the curve c in Fig. 9.15).

In summary, the relationship to describe oxide buildup by uniform corrosion can be (a) parabolic, (b) linear, (b') quasi-linear, and (c) logarithmic as shown in Fig. 9.15.

Parabolic (a in Fig. 9.15)

The parabolic curve occurs through diffusion controlled mass transport in metal.

Consider Ficks 1st Law in steady state ($\frac{\partial C}{\partial t} = 0$) :

$$J(x,t) = -D\frac{\partial C(x,t)}{\partial x} \tag{9.10}$$

Then from Ficks 2nd Law:

$$\frac{\partial J}{\partial x} = -\frac{\partial C}{\partial t} = 0\ (i.e., steady\ state) \tag{9.11}$$

As J is constant in x, Eq. 9.10 gives,

$$\frac{dC}{dx} = -\frac{J}{D}\ (constant) \tag{9.12}$$

Integration of Eq. 9.12 gives, $C(x) - C(0) = -\frac{J}{D}x$, or:

$$J = -\frac{D(C(x) - C(0))}{x} \tag{9.13}$$

Now, we introduce *L(t)*, the thickness of oxide product, to describe the rate of change of the thickness. The thickness is proportional to J with R as the proportionality constant:

$$\frac{dL}{dt} = RJ \tag{9.14}$$

where R (cm^3/g) is the ratio of the volume of oxide formed to the gram of oxygen reacted.

Using Eqs. 9.13 and 9.14:

$$\frac{dL}{dt} = \frac{R \cdot D(C(0) - C(L))}{L(t)} = \frac{k}{L(t)} \tag{9.15}$$

where,

$$k = R \cdot D(C(0) - C(L)) \tag{9.16}$$

So for the parabolic equation, the rate of oxidation is inversely proportional to oxide thickness.

Integration of Eq. 9.15 through separation of variables gives $L(t)^2 - L(0)^2 = 2kt$, or:

$$\mathrm{L}(t) = \sqrt{2kt} \tag{9.17}$$

This is the parabolic growth law where *C(0)* and *C(L)* are given as boundary conditions ($D \sim 10^{-8}$ to 10^{-9} cm^2/s).

Linear (b in Fig. 9.15)
In the linear growth case, the rate of oxidation is constant, or $dL/dt = k$.

Quasi-Linear (b' in Fig. 9.15)
This quasi-linear case represents parabolic growth with the oxide film developing cracks from time to time. This case may have relevance in describing the long-term behavior of the film:

Logarithimic (c in Fig. 9.15)
The logarithmic changes of oxide thickness is given by, $L(t) = k\ log(t)$ (Fig. 9.16).

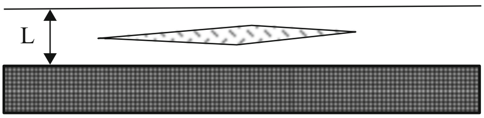


Fig. 9.16 Logarithmic metal oxide film development

Example 9.5: Corrosion Behavior Examination

This example is to examine the corrosion behavior of a metal overpack to be placed in a geological repository.

Assume that the overpack is made of iron and corrodes with atmospheric oxygen via the reaction: $4\ Fe + 3\ O_2 \rightarrow 2\ Fe_2O_3$ where the density of the iron metal is 7.7 g/cc and the density of hematite is 5.9 g/cc. The thinnest section of the overpack is 8 cm thick and corrosion is uniform over the entire overpack.

(a) Determine the thickness of oxide formed for 1000 years by assuming parabolic kinetics of the reaction.

(b) Determine how much Fe is corroded for a given thickness of oxide film during 1000 years.

The diffusion coefficient for oxygen through a hematite film is temperature dependent, given as

$$D(t) = D_0 e^{-\frac{E_a}{R \cdot T(t)}}$$

where E_a is the activation energy (measured to be 7 kilojoules per mole), and R is the gas constant (8.314 Joules per mole per K) and T is the absolute temperature as a function of time. D_o has been experimentally determined to be 3×10^{-6} cm^2/sec. Assume that any oxygen reaching the metal surface is consumed immediately. The repository temperature profile is described by $T(t) = T_0 e^{-bt}$, where $b = 0.0005$ year^{-1} and T_0 is the initial temperature of the overpack surface, assumed at 623 K.

Solution:

(a) Determine the thickness of oxide formed for 1000 years by assuming parabolic kinetics of the reaction.

Use molecular weight scaling to determine the volume of oxide produced per gram of molecular oxygen reacted.

$$\begin{aligned} R &= \frac{Volume\ of\ oxide\ formed}{g\ of\ O_2 reacted} \\ &= \left(\frac{1 cm^3}{5.9 g\ oxide}\right)\left(\frac{159.6 g\ Fe_2O_3 oxide}{mole\ oxide}\right)\left(\frac{2\ mole\ of\ oxide}{3\ mole\ of\ oxygen}\right)\left(\frac{1 ml\ O_2}{32 g\ O_2}\right) \\ &= 0.564 \left[\frac{cm^3\ oxide}{g\ O_2}\right] \end{aligned}$$

From Eq. 9.14, $\frac{dL}{dt} = RJ = -RD(t)\frac{dC}{dt}$

$$L(t)dL = -RDdC$$

(continued)

Example 9.5 (continued)

$$L^2(t) = 2\int_0^T RD(t)C_0 dt$$

where, $C_0 = 2.37\text{x}10^{-4}$ g/cm^3 (oxygen concentration in the air)

Thickness of oxide is found by

$$L(t)_{ox}{}^2 = 2\int_0^{1000} RD(t)C_0 dt$$

This can be solved numerically. $=> L_{ox} = 2.13$ cm oxide. This is the total thickness of oxide formed.

(b) Determine how much Fe is corroded for a given thickness of oxide film.

$$4Fe + 3O_2 \rightarrow 2Fe_2O_3$$

$$\begin{aligned} R_m &= \frac{thickness\ of\ Fe\ corroded}{cm\ oxide\ film\ formed} \\ &= \left(\frac{5.9g\ Fe_2O_3 oxide}{1cm\ oxide \times 1cm^2}\right)\left(\frac{1\ mole\ oxide}{159.6g\ oxide}\right)\left(\frac{4\ mole\ of\ Fe}{2\ mole\ of\ oxide}\right)\left(\frac{55.8g\ Fe}{1mol\ Fe}\right) \\ &\quad \times \left(\frac{1cmFe \times 1cm^2}{7.7g\ Fe}\right) \\ &= 0.536\left[\frac{cm\ Fe}{cm\ Oxide}\right] \end{aligned}$$

Corroded length of Fe is

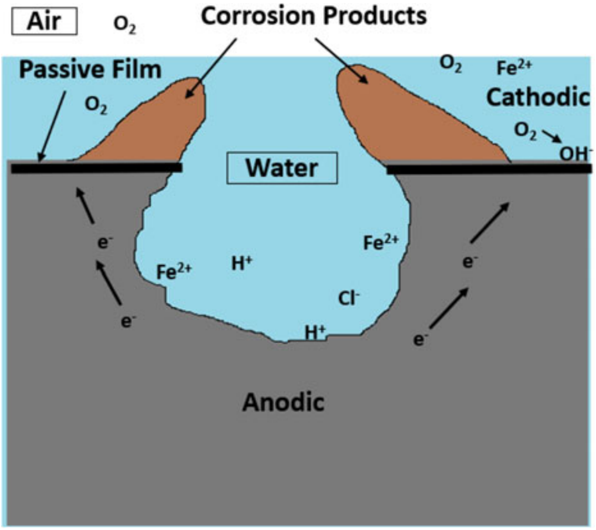
$$\mathrm{L_m} = \mathrm{R_m} \cdot L_{ox} = 0.536\left[\frac{cm\ Fe}{cm\ Oxide}\right] \cdot 2.13\ [\text{cm Oxide}] = 1.14\ [cm\ Fe]$$

The corroded length of Fe is 1.14 cm which is less than the thickness of the thinnest section of the waste package (8 cm) → The waste package will not fail.

9.5.3 Localized Corrosion

As discussed in 9.5.1.2, passivation of metal is important to alleviate corrosion damage. However, through localized accelerated attacks, breakdown of the

Fig. 9.17 Depiction of pitting corrosion



protective surface films can take place leading to localized corrosion. The occurrence, intensity, and location of these localized attacks are generally random in nature.

9.5.3.1 Pitting Corrosion and Crevice Corrosion

Pitting corrosion is a form of extremely localized attack that results in holes in the metal. When pitting corrosion occurs, the pit is the anode and the non-pitted surface is the cathode (Fig. 9.17). Causes of pitting corrosion include specific surface conditions such as scratches, surface compositional heterogeneities, or alloy chemistry combined with specific environmental variations including the presence of damaging species (e.g., chloride ions). It is difficult to detect, predict, and prevent pitting corrosion and the resulting failure of metal occurs suddenly. A small, narrow pit with minimal overall metal loss can lead to the failure of an entire engineering system.

Crevice corrosion (Fig. 9.18) is another localized form of corrosion occurring in the gap between two joining surfaces (generally called crevices). Crevice corrosion results when a portion of a metal surface is shielded in such a way that the shielded portion has limited access to the surrounding environment (e.g., oxygen). It is a special case of pitting corrosion near crevices formed by either two metal surfaces or metal and a nonmetal surface.

Crevice corrosion and pitting corrosion are related and similar because they both require stagnant water, chloride, and oxygen. They differ mainly in geometric considerations. Metals or alloys that depend on oxide films for corrosion resistance

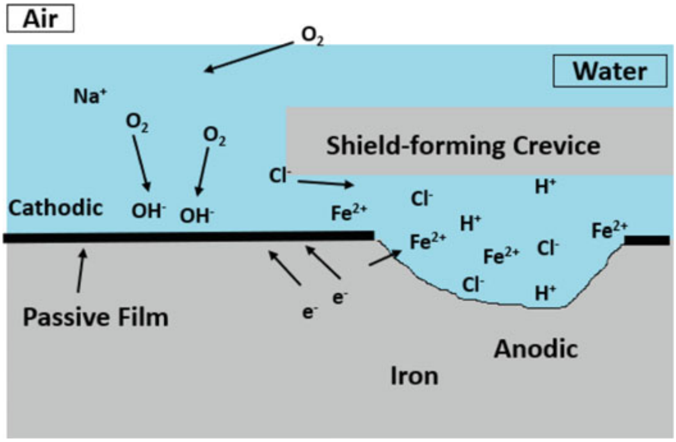


Fig. 9.18 Crevice corrosion

are particularly susceptible to pitting or crevice corrosion. These films are destroyed by high concentrations of chloride or hydrogen ions.

Oxygen depletion has an important indirect influence on the development of crevice or pitting corrosion. After oxygen is depleted in a local area, no further oxygen reduction occurs, even when the dissolution of metal continues. This tends to produce an excess of positive charge in the solution, which is balanced by the migration of chloride ions into the crevice or pit. Therefore, the concentration of metal chloride can be increased within the crevice or pit. The chloride ion acts as a catalyst in pitting and crevice corrosion (i.e., increases the corrosion rate, but is not used up in the reaction). The chloride ion tends to be absorbed on the metal surface or the passive films and polarize the metal, initializing localized corrosion. In this case, presence of other anions such as carbonate, sulfate, and nitrate works also as inhibitors of localized corrosion as their presence can mitigate the role of chloride ion.

Once initiated, oxygen reduction or hydrogen evolution may start in the crevice or the pits. Large surface areas will become cathodic and pits or crevices will become anodic and corrode. Metal dissolution will thus be concentrated in small areas and will proceed at much higher rates than the uniform corrosion. Large crevices are less likely to corrode because water movement causes mixing and replenishes oxygen, hydrogen ions, and other ions.

9.5.3.2 Intergranular Corrosion

Intergranular corrosion (IGC) is a localized attack at grain boundaries due to precipitates along grain boundaries. Typically when a metal corrodes, the attack is more or less uniform between the metal matrix and grain boundaries. However, under special circumstances, grain boundaries become more reactive leading into intergranular corrosion.

The best known form of IGC occurs in austenitic stainless steel when heat treatments deplete chromium in the grain boundaries or nearby structure by forming carbide ($Cr_{23}C_6$). Stainless steel, an alloy of chromium, nickel and iron, requires at least 12% Cr for passivity. If stainless steel is heated to a high temperature (in the temperature range between 425 and 815 °C), chromium carbide precipitates start to form along grain boundaries, leaving a zone depleted of chromium at the grain boundary. Below about 10% Cr, these areas lose resistance and corrode preferentially.

As a zone depleted of chromium will preferentially dissolve away, post-welding heat treatment or the use of low-carbon varieties is needed to prevent intergranular corrosion. When heated above 850 °C and fast cooled (quenched) back to room temperature, the precipitates will dissolve back into the grain structure.

Exfoliation as shown in Fig. 9.11 is a type of intergranular corrosion occurring at grain boundaries under the surface. Exfoliation raises surface grains from the metal.

9.5.4 *Environment Assisted Cracking – Stress Corrosion Cracking*

Stress corrosion cracking (SCC) is the most important form of environment assisted corrosion (it is sometimes considered a localized corrosion as well). SCC refers to the propagation of cracks induced by mechanical stress with the assistance of one or more active chemical agents. Therefore, SCC is an outcome of chemical and mechanical interactions between the material and its environment (Fig. 9.19).

SCC occurs in alloys with a static tensile stress in the presence a passive surface film under oxidizing conditions. A specific dissolved species is often required as well (such as, hot chloride for stainless steel, ammonia solutions for brass, and nitrates for carbon steel). An austenitic stainless steel is known to be susceptible to SCC under the presence of Cl^- and stress.

9.5.5 *Galvanic Corrosion*

Galvanic corrosion occurs when two dissimilar metallic materials are in contact with each other in an electrolyte solution. The driving force for galvanic corrosion is the potential difference between the two metals. One of the two metals is corroded preferentially; this metal is the anode and the unattacked metal is the cathode.

Each metal or alloy has a unique corrosion potential, E_{corr}, when immersed in a corrosive electrolyte. The most active alloy (anode) is always attacked preferentially by galvanic corrosion, whereas the more noble metal becomes cathodic. Some

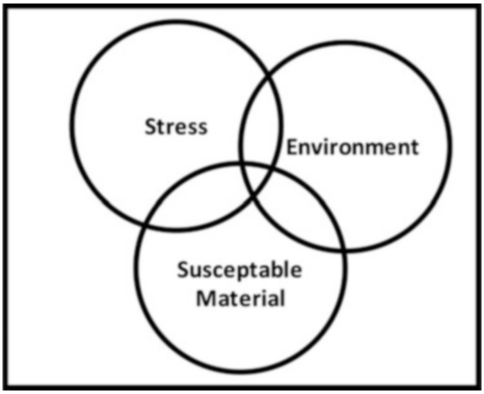


Fig. 9.19 Conditions to be met for stress corrosion cracking

metals are more intrinsically resistant to corrosion than others, either due to the fundamental nature of the electrochemical processes involved or due to the differences in how reaction products form. The Galvanic Series as shown in Fig. 9.1 gives a list sorted by corrosion potentials for various alloys and pure metals in sea water. In a galvanic couple, more noble metal will be a cathode and least noble (most active) metal will work as an anode. Galvanic corrosion can be prevented or its occurrence can be reduced by electrically insulating the two metals from each other or by choosing metals that have similar potentials according to the figure.

9.5.6 Microbiologically Influenced Corrosion

Microbiologically influenced corrosion (MIC) is a corrosion caused or promoted by the microorganisms. Presence of microorganisms at the surface of metals can render the local chemistry condition more corrosive through their metabolic activity. Such activity can change the conditions such as pH, redox potential, and concentration of oxygen or chlorine.

9.6 Candidate Materials for Waste Containers

The material employed to nuclear waste containers must feature good corrosion resistance for long-term isolation of nuclear waste,. The material must be strong enough to withstand the loads without failure during handling and storage. The cost of using the material should not be excessively high (e.g., naturally abundant) and the material must be amenable to the process of manufacturing (e.g., sealing should not be difficult). For the purpose of seeking license approval, the degree of uncertainty in the prediction of the long-term performance of the material should be low. In this regard, the materials must not be susceptible to highly localized (non-uniform) attack or it should be possible to provide conditions that avoid the conditions leading into localized attack. Use of clay backfills to tightly enclose metallic surfaces of the waste package to produce reducing environment is an example of providing such condition. In the end, the selection of material for waste container is based on the overall consideration of corrosion resistance, cost, fabricability, availability, and licensability.

A material that is not subject to corrosion under all foreseen environments does not exist. Nevertheless, we can select materials that show good corrosion resistance with predictable corrosion rate to allow the design of waste package as successful candidates. Current candidate materials for nuclear waste containers or packages include copper, carbon steel, stainless steels, titanium alloys, and nickel-based alloys. Ceramics or graphitic materials (graphite, silicon carbide) which show good corrosion resistance may also be considered if they can be combined with other materials to provide structural strength. High density polymers have also been used for the containers for the disposal of low and intermediate level waste (this is discussed in Chap. 13).

9.6.1 Carbon Steel

Steel is an alloy of iron with carbon and other elements (e.g., tungsten, chromium, and manganese). Depending on the types and amounts of alloying elements used, various steels are produced such as carbon steel, cast iron, low alloy steel, or stainless steel.

9.6.1.1 Types and Use of Carbon Steel

Carbon steel is an iron alloy with carbon content less than 2 wt.% and typically contains Mn (<1.65 wt.%), Si (<0.60 wt.%), Cu (<0.60 wt.%), and other minor alloying elements such as Cr, Co, Mo, Ni, Nb, Ti, W, V, or Zr (with no specified minimum content). The term carbon steel is also used to refer to mild steels with carbon content of 0.16–0.29 wt.%. Mechanical properties of carbon steel such as hardness, ductility and tensile strength vary as the composition changes.

Use of carbon steel started in the eighteenth century and it is now by far the most widely used steel (e.g., more than 85% of the steel produced and used in the U.S. is carbon steel). Carbon steel containers are widely used for the disposal of low and intermediate level waste. Use of carbon steel has also been considered as a container material for HLW disposal in Switzerland, France, Belgium, Japan, Germany, Canada, and the U.S. Its benefits include strength and ductility of the material, predictable corrosion rate, extensive experiences in fabrication, relatively good natural abundance, and low cost.

In contrast to carbon steel, cast iron contains 2–3% carbon with the remainder being iron. Cast iron is a fairly brittle low malleability metal. Cast iron was also considered for HLW container in the early U.S. program and in Germany. Corrosion characteristics of cast iron is similar to carbon steel, but perhaps involves lower possibility for localized corrosion.

9.6.1.2 Chemical Degradation Characteristics of Carbon Steel

Although carbon steel does corrode, it is less prone to catastrophic failures. Both the natural occurrences and archaeological analogs (including the samples from the Roman Empire days) exhibited low corrosion rates of iron. Carbon steel corrodes uniformly under oxidizing conditions with its rate in the order of 10–100 μm/year. Localized corrosion of carbon steel (i.e., crevice corrosion or pitting corrosion) is less likely although it could occur if breakdown of an oxide layer occurs. Conditions for localized corrosion include presence of Cl^-, non-uniform wetting of the surface resulting in the spatial separation of anodic and cathodic sites, and reductive dissolution of Fe(III) corrosion products during the time of transition from oxidizing to reducing conditions (King 2014b). Under reducing conditions, uniform corrosion of carbon steel takes place at the rate of 0.1–1 μm/year. No localized corrosion is expected to carbon steel under reducing conditions.

Carbon steel is not susceptible to SSC due to lack of cyclic loading under the conditions of geological disposal. Carbon steel, however, would go through galvanic corrosion if contacted by copper, Ni- or Ti-based alloys, the more-noble materials. Under both oxidizing and reducing conditions, carbon steel can be susceptible to microbially-induced corrosion depending on the location and duration of microbial activity in the environment. Increase in corrosion rate due to irradiation from HLW/spent fuel is not expected under the geological disposal conditions. The likelihood of hydrogen related damage to carbon steel is also low (King 2014b).

9.6.2 Stainless Steel

Stainless steel is an iron-based alloy with high content of chromium (minimum of 11–13 wt.%). Stainless steel also includes Ni, Mo, Mn along with Si, C, P, and S at low content. With much lower carbon content, stainless steel has a higher melting point and lower malleability and durability than carbon steel. During the manufacturing process, chromium forms a stable oxide/hydroxide film on the surface of the steel which provide corrosion resistance.

9.6.2.1 Types and Uses of Stainless Steels

Different classes of stainless steel exist depending on the presence of predominant crystal structure(s). Austenitic stainless steels (e.g., Types 304) are the largest family of stainless steels (making up about two-thirds of all stainless steel production) containing high levels of chromium (17.5–19.5%) and nickel (8–10.5%) and low levels of carbon. They have face-centered cubic (fcc) crystal structure and maintain the microstructure at all temperatures even after heat treatment. Austenitic stainless steels are most weld-able, non-magnetic, and not heat-treatable. Ferritic stainless steels have body-centered cubic (bcc) structure and have trace amounts of nickel (0.5–0.75%) but contain varying amounts of chromium (10.5–30%) for corrosion resistance. Though not as strong or corrosion-resistant as austenitic steels, ferritic stainless steels generally have better engineering properties (e.g., ductility and formability). They are generally very weldable, but some have limitations by being prone to sensitization of the weld heat-affected zone and weld metal hot cracking. Accordingly, they are often used as thin layers. Ferritic stainless steels are magnetic, not heat-treatable, can be strengthened by cold working. Martensitic stainless steels have a body-centered tetragonal or bcc crystal structure and are magnetic and heat-treatable. With a wide range of properties, they are used as engineering steels, tool steels, and creep resisting steels. Preheating and post-weld heat treatment are required to obtain useful properties. Duplex stainless steels contain approximately equal proportion of austenite and ferrite stainless steel with the mixed microstructure. They have higher yield strength and greater stress corrosion cracking resistance than austenitic stainless steels.

Variations in each class of stainless steel are also available. For example, basic austenitic stainless steel is called Type 304. By adding Cr and Ni to the basic austenitic stainless steel, Types 309 and 309S is made with high-temperature oxidation resistance (and increased strength). Mo can be added to the basic type (304) to increase resistance to localized corrosion (Type 316). To reduce the susceptibility to IGC (i.e., precipitation of carbides, $Cr_{23}C_6$), Types 304 L and 316 L is made with low carbon alloys as a base metal, or Type 316Ti can be made by adding Ti to precipitate C as a Ti carbide thus preventing chromium carbide precipitation. The most commonly used ferritic stainless steels are Type 409 and Type 430. Type 409 is stabilized with titanium and niobium in combination with low carbon content. Type 430 has higher chromium and slightly higher nickel content with no titanium. Other ferritic stainless steels include 410 L, 430Ti, 434, 436, 439, 441, 444, 445, 446, 447. Matenistic stainless steel include types 403, 410, 410NiMo, and 420. The content of Ni and Mo can both be increased to improve corrosion resistance, forming the so-called super-austenitic alloys (Types 904 L and 926).

Stainless steel has been considered as part of a container material for HLW/SF disposal. In particular, austenitic stainless steels show better radiation shielding performance than ferritic stainless steels. In the U.S. DOE's HLW package design, austenitic 316 alloys is the inner barrier material (composition of 316 SS is 16–18% chromium, 10–14% nickel, 2–3% molybdenum, 2% (max) Mn, 1% (max) Si, 0.03% (max) C, 0.045% (max) P, 0.03% (max) S, 0.10% (max) N and 65–69% Fe (the remaining balance)).

9.6.2.2 Chemical Degradation Characteristics of Stainless Steels

With passive film, rate of general corrosion is very low with stainless steel. Under oxidizing conditions at neutral pH, the rates are a few μm/year. Under reducing conditions at alkaline pH, the rates are a few nm per year. The passive film can dissolve under oxygen environment with low pH and/or high concentrations of Cl^- or various S^- containing species, leading to pitting or crevice corrosion. Crevice corrosion occurs under less-aggressive conditions (i.e., at more-negative potentials and/or at lower Cl^- concentrations) than pitting as it is associated with the restricted mass transport of species into and out of the occluded region. Under the reducing conditions, localized corrosion would not occur to stainless steel except when the corrosion potential exceeds the critical potential for film breakdown or re-passivation of pit or crevice. Of the alloys considered, the duplex alloy (alloy 2205) provides the higher pitting resistance, followed by the super-austenitic Type 904 L, and the austenitic 316 L and 304 L alloys.

In the presence of Cl^-, stainless steel, especially the austenitic grade, is susceptible to stress corrosion cracking (SCC). The susceptibility to SCC is related to development of pitting corrosion as cracks most likely initiate from pits and increases with increase in temperature and Cl^- concentration and decrease in pH. As the conditions become reducing, the probability of SCC diminishes. Also, stainless steels are subject to various forms of MIC when exposed to active microbial communities and biofilms. Susceptibility of stainless steel to galvanic corrosion is low, as Ni alloys are relatively noble.

In the case of 316 SS, the general corrosion rate is in the order of $10^{-1} \sim 10^{2}$ μm/year under aqueous condition and 10^{-3} and 10^{-1} μm/year under humid air conditions (which is of interest in the U.S. Yucca Mountain). The localized corrosion rate is expected to be between 10^{3} and 10^{4} μm/year.

9.6.3 Copper

Copper, a fcc crystalline metal, is one of the few metals found widely in native deposits. In these natural deposits, copper has been shown stable for very long periods. For example, the copper deposits in the state of Michigan in the U.S. remained stable since its formation between 500 and 800 million years ago.

9.6.3.1 Types and Uses of Copper

The observed long-term stability of copper in natural deposits led to suggestion of copper as nuclear waste container material in countries like Sweden, Finland, Canada, Switzerland, U.K., and Japan.

The copper material considered for nuclear waste container is either a class of commercially pure copper or the oxygen-free phosphorus-doped (OFP) copper. The OFP copper also contains oxygen (<5 ppm to improve weldability), phosphorous (at 30–70 ppm to provide adequate creep ductility), hydrogen (<0.6 ppm to improve ductility), and sulfur (<8 ppm to improve tensile strength and ductility). Use of copper as a cladding of a waste container, instead of being implemented as solid container, was also suggested (such application would result in cost saving and flexibility in container design).

9.6.3.2 Chemical Degradation Characteristics of Copper

Copper is a thermodynamically stable material. This is true even in the presence of Cl^- as long as oxygen and sulfide are absent. Therefore, use of copper is advantageous in an oxygen- and sulfide-free environment. Under the oxidizing conditions, the rate of uniform corrosion is low with the reported value of 0.94 μm/year and under the condition of geologic disposal within the compacted backfill, the rate of uniform corrosion of copper is expected to be less than 1 μm/year (King 2010).

Although localized corrosion is observed in certain sulfide-containing environments, pitting corrosion, SSC, or galvanic corrosion of copper is not expected in a clay-backfilled geological repository (i.e., in O_2-free environments in the absence of sulfide). Also, as copper does not form hydrides, hydrogen related degradation mechanisms are not a concern for copper.

Predicted total damage to copper due to general corrosion and localized corrosion under geological repository conditions is expected to be up to 1 m over 10^6 years period (King 2014a). This means a 10 cm thick canister would remain intact for more than one million years, even under the near-surface oxidizing conditions.

9.6.4 Titanium Alloys

Titanium alloys are strong and very corrosion-resistant. Their corrosion resistance comes from the formation of a stable TiO2 passive film which is thermodynamically stable over a relatively wide range of Eh and pH.

9.6.4.1 Types and Uses of Titanium Alloys

Titanium alloys can be either in single-phase or two-phase. Two phase alloys are to increase the strength of the alloy and used for aerospace applications. For waste container applications, single phase alloys are considered with variations in the composition. With the basic commercial Ti alloy as Grade 2, variations of Ti alloys include Grade 5, 7, 11, 12, 16, and 17. Grade 5 is to increase strength with addition of aluminum and vanadium. Grade 7 and 16 have enhanced resistance to crevice corrosion with addition of palladium. Grade 12 is a more cost effective approach for the same purpose with additions of molybdenum and nickel instead of palladium. Grades 11 and 17 has lowered oxygen content with improved ductility while maintaining the crevice-corrosion resistance.

As a nuclear waste package material, Grade 2 was considered in Canada, Belgium, and Sweden. Grade 5 and 7 were considered in Belgium. Grade 12 and 16 were also considered in Canada. In the U.S., Ti Grade 7 is considered as the material for drip shield at the Yucca Mountain repository. The composition of Grade 7 includes 0.03% (max) N, 0.10% (max) C, 0.015% (max) H, 0.25% (max) O, 0.30% (max) iron, 0.12–0.25% (max) Pd, 0.4% Residuals, and 98.7–98.8% Ti (the balance).

9.6.4.2 Chemical Degradation Characteristics of Titanium Alloys

The rate of uniform corrosion of Ti alloys is expected to be about 1–20 nm/year with the maximum up to 0.325 μm/year in oxidized aqueous solutions. Under the geological repository conditions, pitting corrosion is not expected. Ti alloys are also generally resistant to stress corrosion cracking. Ti alloys are not subject to microbiologically influenced corrosion.

In the use Ti alloy as the drip shield material (in the U.S.), one potential concern is the uptake of hydrogen for hydride formation. Such hydride formation can lead into hydrogen-induced cracking. However, the available hydrogen concentration in the repository condition (~ less than 120 mg per gram) is much lower than the critical hydrogen concentration level of several hundred mg per g for hydrogen-induced cracking (e.g., 400 mg/g in alloy Ti-7). Therefore the concern of hydrogen-induced cracking can be disregarded. Galvanic corrosion of Ti alloy is a possibility if coupling to more-active metals, such as Fe, Cu, Al, occurs in the repository setting. This could result in increased hydrogen absorption and hydride formation in the Ti alloy. Such coupling can be prevented by the use of backfills.

9.6.5 Nickel-Based Alloys

9.6.5.1 Types and Uses of Nickel-Based Alloys

Nickel-based alloys are known to be very strong and have very good corrosion resistance. There are six major groups of Ni alloys, with different principal alloying elements: Ni-Cu, Ni-Mo, Ni-Cr, Ni-Cr-Mo, Ni-Fe-Cr, and Ni-Fe-Cr-Mo alloys.

Among them, the Ni-Cr-Mo group (i.e., Alloys 625 and 22, Hastelloy C-4 and C-276) or the Ni-Fe-Cr-Mo group (Alloy 825) have been considered for nuclear waste container applications. The countries considering the use of nickel-based alloys include the U.S. (Alloys 625, 825, and 22), Canada (Hastelloy C-276 and Inconel 625), Germany (Hastelloy C-4), Belgium (Hastelloy C-4), and Argentina (Alloy 22).

In the U.S., Alloy 22 has been most widely investigated as candidate for the overpack of HLW package at Yucca Mountain. The composition of Ally 22 is 20.0–22.5% chromium, 12.5–14.5% molybdenum, 2.0–6.0% iron, 2.5–3.5% W, 2.5% (max) Co, and 50–60% nickel (the balance). Nickel-base alloys are easier to weld than titanium but are more costly. Challenge in the use of Nickel-based alloys is prediction of its long-term performance.

9.6.5.2 Chemical Degradation Characteristics of Nickel-Based Alloys

Corrosion resistance of Nickel-based alloys is attributed to the presence of chromium (under oxidizing conditions) and molybdenum (under reducing conditions). The chromium oxide film (Cr_2O_3) is stable over a wide range of Eh and pH while the molybdenum oxide film (MoO_2) provides additional stability under acidic reducing conditions. In Alloy 22 and Hastelloy C-276, molybdenum also provides improved resistance to localized corrosion with tungsten.

Studies showed that the mean corrosion rate of Alloy 22 is 5–10 nm/year with its maximum up to 0.073 μm/year under the expected conditions of Yucca Mountain pore waters at a temperature of 60 °C (DOE 2008).

Occurrence of crevice corrosion or pitting corrosion is possible with Nickel-based alloys through film breakdown. If crevice corrosion occurs, the estimated range of corrosion rate is between 12.7 and 1270 nm/year (with 127 nm/year as median). Also while nickel-based alloys are much less susceptible to SCC and hydrogen embrittlement in comparison to stainless steels, certain aggressive conditions could lead to SCC and hydrogen embrittlement. These conditions include elevated temperatures (>150-200C), high acidity (pH <4), presence of H_2S, and presence of lead. Therefore, use of nickel-based alloys in geological disposal requires control of temperature and measures against the presence of chloride, sulfide species, or lead (King 2014c). In the presence of large quantities of nutrients, the general corrosion rate of Ni-based alloys could double due to microbially-induced corrosion. As Ni alloys are relatively noble, the concern of galvanic corrosion would be minimal. Analysis of corrosion data and characterization of uncertainty in long-term performance behavior of nickel-based alloys is still underway.

9.7 Backfills and Seals

As introduced earlier, backfills are used as the last enclosure barrier of nuclear waste package. Backfills are to contain nuclear waste and to control/limit the release of radionuclides by being a physical and chemical barrier against water and other species transport. Backfills also provide good ion exchange capacity for cations (Sr^{++}, Cs^{+}, Pu^{++}) if these ions are released from the waste package. Accordingly, backfills function as a getter material to prevent the migration of these radionuclides. However, for anions (TcO_4^{-}, I^{-}), backfills are poor in capturing them with low anion exchange capacity.

9.7.1 Bentonite Clay in Water Saturated Repository

Common materials used for backfill are clays such as bentonite, zeolites, mineral mixtures, and cements. Among them, bentonite is most widely used. Bentonite is a geological name for a smectite-rich clay with chemical form of $(Ca\text{-}Al\text{-}Si)O_x \bullet (H_2O)_x$ or $(Na\text{-}Al\text{-}Si)O_x \bullet (H_2O)_x$. The smectite gives the clay a large surface area, high ion-exchange capacity, and a strong affinity for water. More detailed discussions on these clay materials are given in Sect. 10.3.

When contacted by water, the clay will swell and fill any cracks and seal the waste package. Therefore in a water saturated medium, backfills provide impervious layer to water flow. In this case, backfills can effectively form an insulation around the waste package. Then, backfills effectively disconnect the waste package from the surrounding environment and provide a reducing chemistry environment. As discussed in the previous section, corrosion of metals under the reducing chemistry environment is significantly reduced including near elimination of localized corrosion. In isolating nuclear waste from the surrounding environment, backfills play a major role as mass transport barrier, enhancing safety of nuclear waste disposal.

9.7.2 Backfills in Water Unsaturated Repository

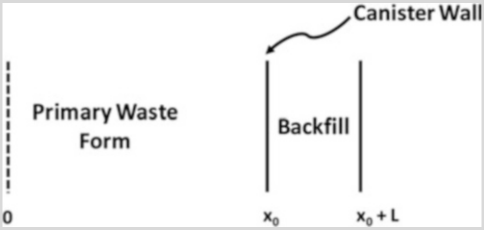
One of the drawbacks in the use of bentonite as backfill is that bentonite dehydrates at temperature 100–500 °C: Once dehydrated passing 300–400 °C, it is difficult to rehydrate the material. At that point, bentonite cannot function as a barrier against mass transport. Therefore, if the temperature of the backfill area is expected to be high, an alternative material is needed for backfill. This is possibly the case in a water unsaturated repository like Yucca Mountain of the U.S. In a water unsaturated repository, backfills become mainly a mechanical buffer to protect the waste packages from external loads or to prevent galvanic corrosion. The expected material for backfill in this case is the excavated rock materials.

9.7.3 Shaft Seals/Grouts

Prior to the closure of a geological repository, underground tunnels and shafts, boreholes, or any intersected areas or open areas within a geologic repository are sealed to prevent migration of radionuclides in case of their release into such spaces. The material used to seal these open spaces includes grouts or other cementitious materials, clays, polymers, resins, and mineral precipitates. These seals form the last part of engineered barriers before the initiation of radionuclides migration into geological rock system.

Example 9.6: Mass Transport through Backfill

To understand the benefit of using backfill as mass transport barrier in a geologic repository, consider the following example. A cylindrical high-level waste canister of radius x_0 is surrounded by a backfill layer of thickness L as shown below. Inside the canister, nuclear waste as immobilized waste form is contained.



If the repository is located in a zone of groundwater saturation, the backfill layer will be saturated with water. At some point during the post-repository closure period, when the container wall is breached, release of radionuclides from the waste form can take place. In this case, for the released radionuclides to migrate through the groundwater system, they must pass through the backfill layer. Movement of radionuclides through the backfill can be described by the following diffusion-advection equation.

$$\frac{\partial C}{\partial t} = \frac{D_h}{R}\frac{\partial^2 C}{\partial x^2} - v\frac{\partial C}{\partial x} - \lambda C$$

Detailed discussions on the use of this equation are given in Sect. 11.3.3.

Consider a radionuclide i with no decay precursor. Suppose that the concentration of i in a solution at the interface with the canister wall rapidly reaches the solubility limit C_{is} and remains at this level throughout the leaching period. Far away from the waste package, the concentration should be zero. By solving the equation using the given conditions, the steady-state flux of i across the external surface of the backfill is given by

$$q_i(L) = \frac{\varepsilon D_d^{(i)}\sqrt{\frac{\lambda_i R_i}{D_d^{(i)}}}C_{is}}{sinh\sqrt{\frac{\lambda_i R_i}{D_d^{(i)}}}L}\left[\frac{\mathrm{g}}{\mathrm{cm}^2 - s}\right]$$

where ε is effective porosity of backfill layer, $D_d^{(i)}$ is molecular diffusion coefficient of isotope i in aqueous solution in the backfill material [cm^2/s], λ_i

(continued)

Example 9.6 (continued)
is radioactive decay constant [s^{-1}], and R_i is retardation coefficient of i in backfill material.

(a) Derive the equation for the steady-state flux of i.

Assume

- There is **no** net flow of water through the backfill in either direction during leaching
- The concentration of i in solution at the outer surface of the backfill, $C_i(x_0 + L)$, is zero
- The backfill layer may be treated as a slab, infinite in the y and z directions

(b) Consider the release of ^{90}Sr ($t_{1/2} = 29$ y) from the waste package. Calculate how much reduction in the steady-state release rate of this radionuclide could be achieved by increasing the thickness of the backfill from 15 to 30 cm.

Use:

Effective backfill porosity, $\varepsilon = 0.1$
Backfill solid phase density, $\rho_b = 2.5$ g/cm^3
Distribution coefficient for Sr-90, $K_d = 1000$ cm^3/g
Aqueous phase molecular diffusion coefficient for Sr-90 in the backfill = 10^{-3} cm^2/s

Note (These equations are further explained in Ch11):

The mass transport through diffusion in a medium is proportional to the concentration gradient, as given below:

$q_i^h = -D_h \varepsilon \frac{\partial C_i}{\partial t}$
(see Eq. 11.52)

The retardation coefficient can be calculated using the following equation:

$R_i = 1 + \frac{1-\varepsilon}{\varepsilon} \rho_b K_d$
(see Eq. 11.9)

Solutions:

(a) **Find the steady-state flux of *i* across the external surface of the backfill**

From Eq. 9.19, with no net flow of water

$$\frac{\partial C_i}{\partial t} = \frac{D_h}{R_i} \frac{\partial^2 C_i}{\partial x^2} - v \frac{\partial C_i}{\partial x} - \lambda_i C_i \Rightarrow 0 = \frac{D_h}{R_i} \frac{\partial^2 C_i}{\partial x^2} - \lambda_i C_i$$

(continued)

Example 9.6 (continued)

There is no mechanical dispersion with no net flow of water, $D_h = D_d^{(i)}$ (see Eq. 11.56)

$$\frac{D_d^{(i)}}{R_i}\frac{\partial^2 C_i}{\partial x^2} - \lambda_i C_i = 0 \Longrightarrow \frac{\partial^2 C_i}{\partial x^2} - \frac{\lambda_i R_i}{D_d^{(i)}} C_i = 0$$

Solution of the above 2nd order differential equation is:

$$C_i(x) = A\cosh\sqrt{\frac{\lambda_i R_i}{D_d^{(i)}}}x + B\sinh\sqrt{\frac{\lambda_i R_i}{D_d^{(i)}}}x$$

Boundary conditions are:

(i) At $x = x_0$, $C_i(x_0) = C_{is}$
(ii) At $x = x_0 + L$, $C_i(x_0 + L) = 0$

If we let $x_0 \rightarrow 0$ and $x_0 + L = L$ for the solution in the backfill,

(i) $C_i(0) = C_{is}$
(ii) $C_i(L) = 0$

Applying these boundary conditions,

$$C_i(0) = C_{is} = A(1) + B(0) \Longrightarrow A = C_{is}$$

$$C_i(L) = 0 = C_{is}\cosh\sqrt{\frac{\lambda_i R_i}{D_d^{(i)}}}L + B\sinh\sqrt{\frac{\lambda_i R_i}{D_d^{(i)}}}L \Longrightarrow B = -C_{is}\coth\sqrt{\frac{\lambda_i R_i}{D_d^{(i)}}}L$$

Thus,

$$C_i(x) = C_{is}\left[\cosh\sqrt{\frac{\lambda_i R_i}{D_d^{(i)}}}x - \coth\sqrt{\frac{\lambda_i R_i}{D_d^{(i)}}}L \cdot \sinh\sqrt{\frac{\lambda_i R_i}{D_d^{(i)}}}x\right]$$

And

$$q_i(L) = -\varepsilon D_d^{(i)}\frac{\partial C_i}{\partial t}\bigg|_{x=L}$$

(continued)

Example 9.6 (continued)

$$q_i(L) = -\varepsilon D_d^{(i)} C_{is}\left[\sqrt{\frac{\lambda_i R_i}{D_d^{(i)}}}\cdot\, sinh\,\sqrt{\frac{\lambda_i R_i}{D_d^{(i)}}}L - coth\sqrt{\frac{\lambda_i R_i}{D_d^{(i)}}}L\cdot\sqrt{\frac{\lambda_i R_i}{D_d^{(i)}}}\cdot\, cosh\,\sqrt{\frac{\lambda_i R_i}{D_d^{(i)}}}L\right]$$

$$q_i(L) = \varepsilon D_d^{(i)} C_{is}\sqrt{\frac{\lambda_i R_i}{D_d^{(i)}}}\left[\frac{-\,sin\,h^2\sqrt{\frac{\lambda_i R_i}{D_d^{(i)}}}L + \,cos\,h^2\sqrt{\frac{\lambda_i R_i}{D_d^{(i)}}}L}{sinh\,\sqrt{\frac{\lambda_i R_i}{D_d^{(i)}}}L}\right]$$

$$\boxed{\therefore q_i(L) = \frac{\varepsilon D_d^{(i)}\sqrt{\frac{\lambda_i R_i}{D_d^{(i)}}}\,C_{is}}{sinh\,\sqrt{\frac{\lambda_i R_i}{D_d^{(i)}}}L}}$$

(b) Calculate the reduction in the steady-state release rate of this radionuclide by increasing the thickness of the backfill from 15 to 30 cm.

$$R_i = 1 + \frac{1-\varepsilon}{\varepsilon}\rho_b K_d = 1 + \frac{1-0.1}{0.1}\left(2.5\text{g/cm}^3\right)\left(1000\text{cm}^3/g\right) = 22501$$

$$D_d^{(i)} = 10^{-3} cm^2/s; \lambda_i = \frac{ln\,2}{28\times 365\times 24\times 3600} = 7.85\times 10^{-10} s^{-1}$$

$$\therefore\sqrt{\frac{\lambda_i R_i}{D_d^{(i)}}} = \sqrt{\frac{\left(7.58\times 10^{-10}\right)(22501)}{10^{-3}}} = 0.1306\, s^{-1}$$

$$q_i(15) = \frac{0.1\cdot 10^{-3}\cdot 0.1329\cdot C_{is}}{sinh\,(0.1329\cdot 15)} = 3.63\times 10^{-6} C_{is}$$

$$q_i(30) = \frac{0.1\cdot 10^{-3}\cdot 0.1329\cdot C_{is}}{sinh\,(0.1329\cdot 30)} = 4.85\times 10^{-7} C_{is}$$

(continued)

Example 9.6 (continued)

$$\boxed{\textbf{The reduction fraction} = \frac{q_i(15) - q_i(30)}{q_i(15)} = 0.866}$$

9.8 Conclusion

When nuclear waste packages are exposed to the natural environment, they will go through the process of degradation. Use of technology is to significantly delay such natural processes. The degree of time delays achieved by applying technologies becomes an important consideration in achieving safety in the disposal of nuclear waste. Engineered barriers are important examples of such technology. These engineered barriers include waste forms, waste canisters, overpacks, backfills, and the waste package enclosure spaces, as part of multiple defense in depth system. The waste forms through immobilization of the waste into glass or ceramics minimize the possibility of release of radionuclides from nuclear waste. Even the spent fuel as waste form provides a stable solid material for the retention of radionuclides. The technology for waste canisters or overpacks is to utilize corrosion resistant metallic barriers to prevent the release of radionuclides to the environment should any release of radionuclides takes place from the waste form. Long-term integrity of these metallic barriers must be assured through selecting materials with proven long-term corrosion resistance and good predictability. Backfills provide further redundancy in waste isolation by providing geochemically stable environment surrounding nuclear waste and effectively decoupling nuclear waste package from nature. Thus, backfills are a way to control the external environmental conditions for the overpack. As long as nuclear waste packages serve as effective barriers, long-term isolation of nuclear waste is achievable. Further discussions on the interactions of the engineered barriers with the natural barriers of a geological repository systems are given in Chap. 12.

Homework

Problem 9.1:
Describe key desirable properties of materials for each component of a nuclear waste package, i.e., waste form, waste canister, waste overpack, and backfills. Assume that the nuclear waste package is placed in a water saturated geologic medium.

Problem 9.2: Suggested the details of your design of a nuclear waste package for long-term disposal of spent fuel in a water saturated geologic medium. Specify the types of material and the geometry of the system.

Problem 9.3: Consider a long glass cylinder of radius 25 cm and length 2 m. The glass contains a radionuclide *j* with a half-life of 30 years. The weight fraction of the radionuclide *j* in the glass is 1%. The density of the glass is 3 g/cm^3. The rate of dissolution of the glass network in brine solution is 10^{-5} g/cm^2-day, normalized to radionuclide *j*.

(a) Suppose that the cylinder is placed in a large, well-stirred pool of brine. Calculate the maximum activity of the radionuclide *j* in solution and the time at which this occurs.
(b) By how much would the peak activity of j in solution be reduced if the cylinder was coated with a 1 mm thick layer of glass containing none of the radionuclide j but with otherwise identical properties to the glass core.
(c) Discuss the relevance of such results to the task of predicting waste form behavior in an actual repository situation.

Note:

1. Ignore the contribution of diffusion mechanisms to the transport of j into solution.
2. State clearly any other assumptions you make.

Problem 9.4: Consider the case of a semi-infinite slab of waste matrix containing a radionuclide initially distributed throughout the slab with a uniform concentration C_0, answer the following.

(i) Show that in the asymptotic limit of *t* tending to infinity, the amount of the radionuclide to have diffused into solution (i.e., the time-integrated flux) approaches the value

$$C_0\left(\frac{D}{\lambda}\right)^{1/2}$$

where D is the diffusion coefficient and λ is the decay constant of the radionuclide.

(ii) Give a physical interpretation of the factor

$$\left(\frac{D}{\lambda}\right)^{1/2}$$

(iii) Show that the maximum amount of the radionuclide present in the liquid phase is given by the expression.

$$\frac{C_0}{2}\left(\frac{D}{\pi}\right)^{1/2}\frac{1}{\sqrt{2\lambda}}e^{-1/2}$$

Problem 9.5: Assume that the waste packages in the repository are buried at 130 m from the surface. Waste packages are 0.6 m in diameter and 3.2 m long and have 10 cm thick walls. Assume that the corrosion takes place only through uniform corrosion. Ignore the oxygen contributions from below the repository. Canister pitch is 25 m on a square grid.

Initially the waste package will be corroded by the amount of oxygen available from the rocks between the waste package and the surface. Assume that half of the oxygen in the rock causes corrosion of the waste package. After the oxygen in the rocks is consumed, the iron overpack in the repository will be corroded by the supply of gaseous oxygen from the surface.

Estimate how long it would take for the waste package to be corroded through. Assume that the canisters consume all oxygen that reaches the repository.

Density of air is 0.001185 g/cc and O2 is 20% by weight.

Rust occurs via: $4Fe + 3O_2 \Rightarrow 2Fe_2O_3$
Density of Fe is 7.7 g/cc.
Diffusion coefficient of oxygen in rocks is 4×10^{-3} cm^2/s
Effective porosity of rocks is 0.02.
Assume steady state conditions, no velocity or sorption of oxygen.

Problem 9.6: The purpose of this problem is to demonstrate some of the pitfalls associated with the long-term extrapolation of experimental leaching data obtained over a short period. Consider a solid waste form containing a single non-radioactive isotope with an initial concentration C_0 kg/m^3. It can be shown that if the solid matrix dissolves at a constant rate a_1 m/year, the release rate of the isotope into solution for a semi-infinite plane is given by

$$f_1(t) = a_1 C_0 \ \frac{kg}{m^2 \cdot yr} \quad (1)$$

Alternatively, if the isotope is released into solution as a result of diffusion and the solid matrix is insoluble, then, of the solution concentration is kept negligibly small (i.e., the solution is 'well-stirred'), the release rate of the isotope is given by

$$f_2(t) = \frac{a_2}{\sqrt{t}} C_0 \frac{kg}{m^2 \cdot yr} \quad (2)$$

where a_2 is proportioned to the square root of the diffusion coefficient.

A 144-day dynamic leach experiment is conducted with a large borosilicate glass cylinder doped with a non-radioactive cesium isotope. Strictly speaking, Equations (1) and (2) are not valid for cylindrical geometries, but if the cylinder is large enough the error introduced by applying them will be small. Here, (1) and (2) are each fitted to the experimental leach data. Tolerable fits are obtained in both cases, yielding the following empirical values for the constants

$a_1 = 2.5 \times 10^{-5}$ m·year^{-1}
$a_2 = 1.5 \times 10^{-5}$ m·year$^{-1/2}$

If the borosilicate glass is now loaded with radioactive ^{137}Cs ($t_{1/2} = 30$ years) at the same initial concentration, use these results to predict the amount of ^{137}Cs in solution as a function of time after the onset of leaching assuming the leaching mechanism is bulk dissolution, if leaching begins at 1, 10, and 100 years after glass fabrication.

Perform analogous calculations assuming that diffusion is the leaching mechanism. Plot the results. Then plot the ratio of the two predictions of the amount of ^{137}Cs in solution as a function of time after the onset of leaching.

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Chapter 10
Geological Barriers for Disposal of Nuclear Waste

Abstract Very long-term isolation of nuclear waste in deep geological formations is the final approach in nuclear waste management. Deep geological formations provide protection of engineered barriers and significantly delays in the development of potential impacts upon the failure of engineered barriers. This chapter describes key desired characteristics of a geologic disposal system in terms of physical and chemical properties of rocks as part of the discussion of disposal site selection process.

Key words Methods for permanent disposal · Geological repository · Rock properties · Site selection · Thermal design limits

Disposal of nuclear waste, the final phase of nuclear waste management, is to provide very long-term isolation of the waste in remote areas. Various methods have been proposed for this purpose. These methods are to move nuclear waste far away from people and keep the waste in an isolated condition in a specially prepared medium. The medium considered for this purpose include deep underground geological formations, the floor of the deep ocean, geological formations under the deep ocean floor, the bottom areas of glaciers, and the outer space. After evaluating each of these options, disposal in deep underground geological formations has been selected as the method of choice by the international community. This chapter first reviews various approaches considered and then discusses the use of geological repositories for final disposal of nuclear waste.

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10.1 Methods Considered for Permanent Disposition of Nuclear Waste

10.1.1 Disposal in the Ocean

The ocean is vast and deep. Disposal of nuclear waste in the ocean was practiced in the past during the 1940s and 50s. The Soviet Union disposed of their naval submarines in the open sea. The U.S. routinely dumped their low level radioactive waste in the ocean during the Manhattan Project. These practices continued worldwide through 1960. In the U.S., such practice was banned in 1972. The last known dumping operation was in 1982.

Disposal of nuclear waste in the ocean was also considered in the form of so called, “subseabed” disposal. The concept, proposed in the 1970s, is to place the waste on the floor of seabed after lowering the waste package to the bottom area of the sea. This disposal method involves the emplacement of nuclear waste beneath the ocean floor within the thick clay sediments over large expanses of the deep and stable mid-oceanic regions. The deep-ocean areas are remote thus minimize the likelihood of human intrusion and also spacious enough to handle all high level waste produced worldwide. The ocean itself provides isolation of the waste from the land-based ecosystems with huge dilution capacity in the case of any radioactivity release. Two options considered in this approach in terms of how to guide the waste to reach the seabed are depicted in Fig. 10.1. These are expected to be relatively low cost options of disposing HLW. The first one is dropping the waste package from a ship in missile-shaped penetrators. Burying the waste packages up to 70 meters deep

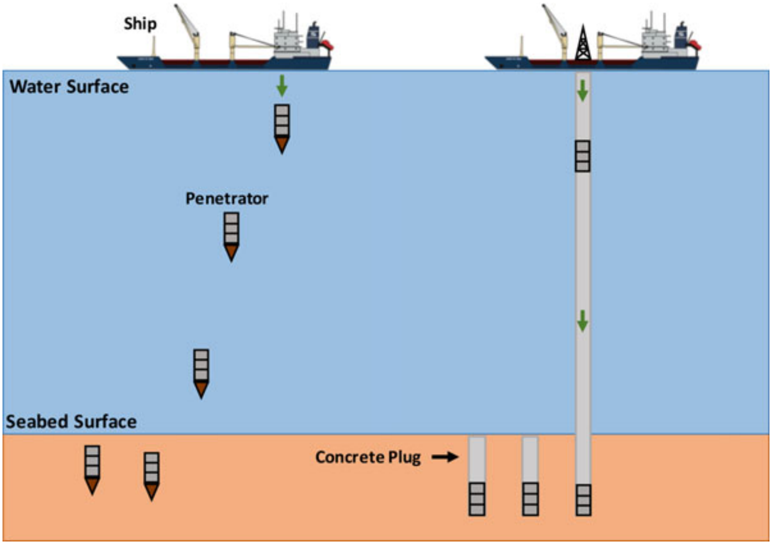

Fig. 10.1 Depiction of subsea-bed disposal of nuclear waste

in seabed sediment was found feasible through field trials of the method. The second option is to lower the waste package through predrilled boreholes of about 800 m in depth. After waste emplacement, the top 200 m of the holes will be sealed with cement or grout (Schneider 1974). The approach was once regarded as the most promising alternative to the method of mined geologic disposal.

One of the key concerns with these proposed approaches is technical uncertainty. The desirable area should be stable, tranquil, remote, and should not be a target of resource explorations in the future. However, finding such oceanic areas will be a challenge. Even if such area is found, uncertainty prevails in predicting the behavior of the waste packages placed in the area with respect to their failure and the resulting migration of radionuclides. In addition, there will be concerns over locating a harbor as a potential interim storage site for the handling, packaging, and shipping of spent fuels/HLW. Siting such facility would arouse public opposition over the likelihood of potential accident during transportation and/or storage. Retrieval of the waste, once emplaced, would be difficult and very costly.

With “subseabed” disposal, significant legal problems exist at both the national and international levels. In the U.S., the option was banned under the Ocean Dumping Act (officially, the Marine Protection, Research, and Sanctuaries Act) in 1972. At the international level, the countries who did not operate nuclear power plants expressed their concern as early as 1958 in Article of the Law of the Sea stating “that every State shall take measures to prevent pollution of the seas from the dumping of radioactive waste”. The IAEA provided guidance and recommendations since 1957 for ensuring that “disposal of radioactive wastes into the sea will not result in unacceptable hazards to human health and marine organisms” (Calmet 1989). In 1975, the London Dumping Convention (LDC) came into force regulating the dumping of waste at sea on a global scale. In 1983, ban on the dumping of all radioactive waste at sea was proposed at a consultative meeting of the LDC. The LDC also proposed to prohibit spent fuel/HLW from being placed on the surface of the seabed. This was followed by an international seabed working group (SWG) conducting research on subseabed disposal under the auspices of Nuclear Energy Agency from 1977 to 1987. When the third United Nations Convention on the Law of the Sea (UNCLOS) (which lasted from 1973 through 1982) came into force in 1994, consideration of subseabed disposal officially ended.

10.1.2 Disposal in Ice Sheets

The ice-sheet disposal is to use a nuclear waste package and its decay heat to melt through a thick sheet of ice, called ice sheet, for the emplacement of the waste. The waste package will naturally move down through the ice and stops on top of a bedrock or at a location inside the ice sheet. In the latter case, the waste package can be attached to cables anchored at the ice surface and suspended in the middle of the ice sheet. Refreezing of water above the waste would seal the emplacement opening and isolate the package. Also another way of ice sheet disposal is storing a waste package in a surface facility which would gradually sink toward the bedrock under the weight of naturally accumulating snow and ice (Schneider 1974). A remote, low-temperature environment in the continental glaciers of Antarctica was suggested as a medium for the method. An ice sheet covers an area greater than 50,000 km^2 (19,000 sq. miles) with ice thickness reaching several hundred meters. The time required for spent fuel/HLW to move down to the refreezing area is expected to be in the order of 10 years.

Main challenge with ice sheet disposal is uncertain performance of the disposal system: The general nature, evolution, and the behavior of the ice system are uncertain. The dynamics of interactions between the ice sheet and waste package are also uncertain, especially if the water produced from melting of ice works as lubricant between the bedrock and the ice sheet. Furthermore, the effect of climate change on the stability of the ice sheet system will add an additional dimension to uncertainty. There are also potential international disputes over the use of the glacier area if the practice is exercised on the Antarctica. An international group of glacier scientists, recognizing these uncertainties, recommended in 1974 that the Antarctic ice sheet not be used for waste disposal.

10.1.3 Disposal in the Space (Extraterrestrial Disposal)

Shooting nuclear waste into the space appears an attractive way of getting rid of nuclear waste. The destinations considered include high Earth orbits, solar orbits, solar-escape trajectory (through a direct injection to solar system escape), or the Sun (impacting it into the Sun) (Thompson 1974). To minimize the volume and payload, only the most troublesome part of spent fuel/HLW such as long-lived actinides and ^{129}I, ^{99}Tc, and ^{14}C after going through spent fuel reprocessing would be disposed of in this way. Such materials require relatively little shielding for unmanned space trip, reducing the payload requirement.

Placing the waste into solar orbits received more attention from the perspective of cost and technical feasibility. A space shuttle would carry the waste into orbit around the earth. An orbital transfer vehicle will then take over the waste from the shuttle to carry the waste into the desirable position in the solar orbit. Both the orbital transfer vehicle and the space shuttle will be reused with the space shuttle returning to earth. This approach requires extreme attention to avoid release of radioactive materials in the event of malfunction or any launch failure. Uncertainties were also noted regarding orbit stability implying the possibility of the waste package returning to earth while the waste remains radioactive. Sending the waste into the sun or using a solar-escape trajectory, as an alternative, would be too costly due to the very high energy requirement. In addition to the concern over uncertainty or cost, the option of space disposal leaves significant international political debate over the issue of space use.

10.1.4 Surface Disposal

Surface disposal is to bury nuclear waste at a facility on or near the surface of the land. The approach has been used for interim retrievable storage of spent fuel and disposal of low and intermediate level waste (LILW). In the case of LILW disposal, waste containers are emplaced in a facility with either soil or cement backfills. This approach, however, is considered inappropriate for spent fuel/HLW disposal as it presents easy accessibility by the public. Especially, if the institutional memory of the facility is lost, the waste becomes readily accessible through the inadvertent intrusion scenario.

10.1.5 Disposal in Geological Formations

Disposal of nuclear waste in geological formations has been most widely investigated and practiced among the alternatives of HLW disposal. This category of disposal method includes various options, such as, disposal in very deep holes, injection of liquid into deep wells, geologic disposal on small, uninhabited islands, injection through rock melting into porous or fractured strata, and disposal in mined geological repository.

10.1.5.1 Deep Boreholes

The deep borehole concept involves drilling a borehole about 5 km down into the Earth's crust and disposing of waste-filled canisters at the bottom of the holes. For spent fuel disposal, each borehole is expected to take 100 to 200 assemblies. The depth reached by the borehole is below the maximum depth of typical groundwater movement and thus provides chemically reducing conditions. Therefore, the possibility of waste package degradation and release of radionuclides is minimized. The depth of the disposal also significantly delays the transport of radionuclides from the waste into the biosphere, while minimizing the potential for disturbance by human intrusion or natural surface phenomena.

Large uncertainties remain with the approach in characterizing the hydrogeologic environment and the interactions between the waste and the rock in such deep locations. It will also be very difficult to verify the degree of isolation provided by the method, raising challenges in the licensing processes involved. Current borehole technology limits the diameter of the borehole to less than 50 centimeters. Thus, the waste currently stored in large containers would need to be repackaged in smaller containers. The number of boreholes to be drilled would be very large for actual implementation of the method.

The cost of deep borehole (~$158/kg-HM, (Brady 2011)) is expected to be lower than that of geological repository (DOE 2008). However, since the concept of deep borehole disposal is relatively new compared with that of geological repositories, more research is needed to better estimate the cost with the consideration of transportation and licensing issues.

10.1.5.2 Deep Well Injection

Deep well injection is a method of disposal for liquid radioactive waste. In this method, suitable rock formations (such as shale) are first fractured at depths of about 100–150 m by injecting fluid under high pressure down a borehole. Then a mixture of liquid radioactive waste and self-hardening grout is injected into the fractured rock, resulting in the emplacement of the waste in an immobile and irretrievable form. Thus the method neither uses a waste package nor a geological repository. The method was practiced in several countries in the past including the U.S. and the former Soviet Union. In the U.S., the method was utilized to dispose of industrial wastes with about 300 industrial waste-disposal wells. The Oak Ridge National Laboratory in the U.S. used the method in the early 1970s to dispose of ~6800 m^3 (1.8 million gallons) of low-level liquid defense waste at a single well fractured shale site. The approach was abandoned due to uncertainties about how the grout flowed within the subsurface system. The Soviet Union injected close to 50 million m^3 of intermediate-level liquid waste mostly into confined sandstone layers several 100 m below the surface in 3 locations (Krasnoyarsk, Tomsk, and Dimitrograd). Still, there is only limited field data on the long-term containment of the wastes. The option was phased out with the rise of environmental concerns and also as better practices of waste solidification and packaging became available.

10.1.5.3 Injection Through Rock Melting

Rock melting injection involves pumping freshly generated liquid high level waste from reprocessing into a conventionally mined cavity at depths of 1500–1800 m. In theory, the high levels of heat produced by the waste would melt the rock within several decades of time frame. Subsequent cooling would result in solidification of the waste within rock/waste mixture. After conceptual examination, this option was abandoned, given the concern over the uncertainties in waste handling, in emplacement, in the interactions between the wastes and the host rock, and potential migration of radionuclide after emplacement. Difficulties with verification of the method were also noted.

10.1.5.4 Geologic Disposal on Small, Uninhabited Islands

Efforts for effective isolation of radioactive waste also suggested the waste to be disposed of in small, uninhabited islands, as an alternative to conventional mined geologic disposal. The option provides similar benefits of deep geological disposal for waste burial but also gives additional safety measure by reducing the risk of unintentional human intrusion and allowing dilution of any radioactivity release through ocean water. The repository construction would start from the surface of the island but the waste emplacement areas will be located away from the island at depth below the ocean. Any rock type suitable for on-land disposal can be selected for the option. Building an artificial island for the application of the method has also been considered.

10.1.5.5 Mined Geological Repositories

Disposal of nuclear waste in mined geological repositories has been the most studied method. The method is based on emplacing waste in a mined cavity in deep geological formations and sealing the cavity with backfills and then eventually closing the mine. This approach received primary emphasis throughout the world since 1970s. A number of operating LLW/ILW disposal facilities in the world belong to this category. In the U.S., this method was formally selected in 1980 as the central part of the high level waste management strategy by the U.S. DOE (DOE 1980). International consensus also exists on the use of mined geological repository for high level waste disposal. Operation of licensed geological repositories at ~400–500m depth is expected in a very near future in Finland and Sweden.

The approach uses both natural and engineered barriers for isolation of nuclear waste. The natural barrier is the host rock at a site and the engineered barrier is the waste package. The isolation is mainly against the infiltrating groundwater. The engineered barrier will protect the waste from corrosion to prevent release of radionuclides into the host rock. If the release does take place, the host rock through its chemical and physical characteristics limits the rate of migration of the radionuclides. Through retention of radionuclides in the host rock system, there will be very long-term delays before the contaminated groundwater reaches the biosphere. Therefore, the natural and engineered barriers play a complementary role. The engineered barriers provide an assurance for the isolation of nuclear waste until waste package fails. The natural barrier covers the rest of isolation after the failure of engineered barriers eventually to meet the required level of safety. As long as both barriers serve the purpose of long-term isolation of nuclear waste, redundancy is provided in assuring safety in nuclear waste disposal, minimizing potential impact of radiation exposure to humans and the ecological system.

10.2 Host Medium of Geological Disposal

The host medium for geological disposal of nuclear waste is rock. Rocks are solid inorganic aggregates of naturally formed minerals. Minerals are the building blocks of rocks as individual grains of free uncombined elements or elemental compounds

Table 10.1 The average chemical composition of continental crust

Chemical composition	Continental crust
SiO_2	60.6
Al_2O_3	15.9
Fe_2O_3 $_+$ FeO	6.7
CaO	6.4
MgO	4.7
Na_2O	3.1
K_2O	1.8
TiO_2	0.7
P_2O_5	0.1
MnO	0.1

from the earth crust. Various minerals are bound, blended by melting, or cemented through natural processes to form rocks.

The earth crust is the outer layer of the earth covering about 35 km (ranging from 5 to 70 km) from the surface underlain by the mantle, the region between the earth core and the crust.

The earth crust is mainly made up of oxygen, silicon, and then aluminum. Behind them, other elements of importance include iron, calcium, magnesium, sodium, and potassium. These eight elements make up nearly 98% (in terms of atom fraction) of the total. Among them, oxygen is most dominant (~47% by weight). As the oxygen ion is quite large compared to silicon and aluminum, the earth crust can be viewed as a vast accumulation of oxygen compounds (as the combination of oxygen ions with interstitial cations of silicon and aluminum). The oxygen valences which are not satisfied by silicon and aluminum are neutralized by the other cations. The average composition of the earth crust in the continent is given in Table 10.1.

10.2.1 Rock-Forming Minerals

Minerals are chemical compounds classified by the nature of the anion which comprises the mineral. Minerals belong to various groups such as oxides (O^{2-}), hydroxide (OH^-), silicates (SiO_4^{2-}), sulfides (S^{2-}), carbonates (CO_3^{2-}), sulfates (SO_4^{2-}), halides (F^-, Cl^-, Br^-, I^-), and phosphates (PO_4^{3-}). There are also free uncombined elements among minerals called native minerals such as silver, gold, and copper. In terms of the number of minerals in each group, there are over 250 minerals in the oxide and hydroxide group, over 300 in sulfides, over 500 in silicates, about 200 in carbonates, about 100 in halides, and about 50 native elements. Further subdivisions may be based on the nature of the attached cations. In the case of silicates, the mode of cross-linkage between various silica and alumina groups provides further subdivisions.

10.2.1.1 Silicate Minerals

While the number of minerals making up rocks is more than a thousand, the major rock-forming minerals are generally a small number of silicon oxide or silicate minerals. This small number of silicon oxide or silicate minerals comprise over 99% of the earth crust. This is again because the earth crust is covered mostly by oxygen and silicon. These silicate minerals include quartz, feldspar, mica, pyroxene, amphibole, and olivine.

Quartz, also called silicate, is essentially silica (SiO_2) crystal and is one of the most abundant minerals along with feldspar. Its crystal structure is a continuous framework of SiO_4 silicon tetrahedral. Quartz is mostly crystallized from molten magma but is also chemically precipitated from hot water circulation near the volcanic activity. Quartz is light colored and hard and very resistant against mechanical and chemical weathering. It also includes wide variety of semiprecious gemstones.

Feldspar is the most abundant mineral making up as much as 60% of the earth crust and refers to aluminosilicates of Na, K, Ca, or Ba. The name feldspar derives from the German *Feldspat* meaning "a field rock that does not contain ore". The general formula can be stated as $XAl(Al,Si)Si_2O_8$, where X may be Na, K, Ca, and Ba. Compositions of feldspars can be expressed in terms of three end-members: potassium-feldspar endmember ($KAlSi_3O_8$), albite endmember ($NaAlSi_3O_8$), and anorthite endmember ($CaAl_2Si_2O_8$). Solid-state mixture between potassium-feldspar and albite are called alkali feldspar and includes minerals such as orthoclase, sanidine, and microcline. The mixture between albite and anorthite is called the plagioclase feldspars and includes oligoclase, andesine, labradorite, bytownite, etc. As one of the important precursors of clay minerals, feldspars are subject to chemical decomposition through prolonged weathering.

Mica is known as sheet silicates as it forms distinct layers. The general formula is $W(X,Y)_2\text{-}3Z_4O_{10}(OH,F)_2$, where W is generally potassium (or sodium or calcium), X and Y represent Al, Li, Mg, Fe^{2+} and Fe^{3+}, and Z represents Si and Al with the Si to Al ratio at about 3:1. Mica is fairly light, relatively soft, and heat-resistant. Thirty seven different mica minerals exist including lepidolite, biotite, phlogopite, and muscovite.

Pyroxene (sometimes called augite) is a group of single chain-silicates as silicon-aluminum oxides with Ca, Na, Fe, Mg, Zn, Mn, or Li. The general formula is $XY(Si,Al)_2O_6$, where X represents Ca, Na, Fe^{2+}, Mg (and sometimes Zn, Mn, and Li), substituting for Si and Al. Y represents Cr, Al, Fe^{3+}, Mg, Co, Mn, Sc, Ti, and V. The name pyroxene is derived from Greek and means 'fire stranger'. It was named so because of their presence as crystals embedded in volcanic glass, perceived to be impurities in the formed glass. In reality, pyroxenes are early-forming minerals crystallized before the lava erupted.

Amphibole (meaning (Greek) 'ambiguous') is a group of double chain-silicates (Si_4O_{11}) generally containing ions of iron or magnesium in the structure. It is generally similar to pyroxene but different in that it has double chain structure containing hydroxyl (OH) or halogen (F, Cl). A general formula can be written $(WXY)_7$-$8(Z_4O_{11})2(O,OH,F)_2$ where W is generally Ca or Na, X is Mg or Fe^{2+}, Y is Ti, Al, or Fe^{3+}, and Z is Si and Al. Amphiboles are dark-colored and form prism or needlelike crystals. Four of the amphibole minerals are commonly called asbestos.

Olivine (named for its typically olive-green color) is a group of magnesium iron silicate minerals. The composition of olivine is generally $(Mg, Fe)_2SiO_4$. Olivine minerals are weaker and weather rather quickly on the surface.

Ferro-magnesians are another name of a silicate minerals group in which cations of iron and magnesium form essential chemical components. Olivine, pyroxene, amphiboles, and the micas biotite and phlogopite belong to this group.

10.2.1.2 Clay Minerals

The interaction of the acids (CO_2, SO_2, and NO_x) of rainwater with the bases of the rocks causes dissolution of minerals and results in degradation of the rocks. In the process, soluble cations and silica (SiO_2) are leached out leaving behind a clay mineral. Thus, the clay minerals have lower content of silica than the parent rock.

Clay minerals of various types are important in sediments, sedimentary rocks, and soil. They are the hydrous aluminosilicates formed by the decomposition of other aluminosilicates, such as orthoclase, plagioclase feldspar, and mica (thus, they also belong to the group of silicates). Clay minerals include kaolinite, montmorillonite, illite (hydromica), and chlorite. They have the layer-lattice structure and have very small grain size (generally less than 0.002 mm in diameter).

Kaolinite is a name as a modification of “Kauling”. Kauling is a high ridge town near Jauchau Fu, China, where a white kaolinite clay was found several centuries ago. Kaolinite has the general formula as $(OH)_8Al_4Si_4O_{10}$ and consists of alternating layers of silica and alumina. It has considerable strength and stability but with little tendency to take on water and swell.

Montmorillonite was the name given to a clay mineral found at Montmorillion in France. The general formula is $(OH)_4Si_8Al_4O_{20}nH_2O$, where nH_2O is the interlayer of adsorbed water. It has layers consisted of one aluminum-hydroxyl unit sandwiched between the two (Si_4O_{10}) sheets. These layers are stacked one above the other with the water interlayer in between them. This provides an expanding lattice with varying water content and its characteristic swelling properties (in the presence of water). The name, smectite, is also used for this clay mineral group. Bentonite is also a clay in this group with high water absorption capacity. Bentonite is found in partially weathered volcanic deposits. At present, clays with the property of swelling on contact with water are loosely called “bentonite”. Montmorillonite clay minerals often produces kaolinite clay upon weathering.

Illite, sometimes called mica clay, is essentially clay-sized muscovite (a type of mica mineral) and has the following general formula, $(OH)_4K_y(Si_{8-y}\ Al)(Al_4Mg_6Fe_4Fe_6)O_{20}$, where y is between 1 and 1.5. Some of illite mineral is a mixed-layer muscovite-montmorillonite and some is a mechanical admixture of muscovite and montmorillonite, often with some clay-sized quartz.

Chlorite is derived from montmorillonite as a product of inserting a (Mg,Al)(OH) layer between the two montmorillonite layers.

The primary factors in determining the nature of clay are the chemical character of the parent material and the physicochemical environment. For example, formation of kaolinite is favored in an acid environment as the structure of kaolinite does not accommodate cations other than silicon and aluminum. Formation of montmorillonite is favored in a neutral or slightly alkaline environment as other cations such as magnesium and iron are essential to its formation. Montmorillonite has a close structural relationship to illite and chlorite and readily changes to these minerals, especially in the marine environment.

10.2.1.3 Other Minerals

There are other non-silicate minerals as components of rock. These include native elements and mineral of sulfides (compounds with a sulfur), oxides (i.e., simple oxides, hydroxides, and multiple oxides), halides (with fluorine, chlorine, iodine, or bromine as the main anion), sulfates (with the sulfate anion, $[SO_4]^{2-}$), carbonates (with carbonate, $[CO_3]^{2-}$), and phosphates (with $[PO_4]^{3-}$).

Examples of such minerals are: calcite ($CaCO_3$) and dolomite ($CaMg(CO_3)_2$), common carbonate minerals found in rocks; gypsum ($CaSO_4{\cdot}2H_2O$, hydrated sulfate), an important sulfate mineral, and; apatite ($Ca_5(PO_4)_3F$), $Ca_5(PO_4)_3Cl$), $Ca_5(PO_4)_3(OH)$) and monazite (ATO_4, T: phosphorus or arsenic; A: often a rare-earth element (REE)), important phosphate minerals. There are also various oxide minerals including corundum (Al_2O_3), hematite (Fe_2O_3), rutile (TiO_2), and magnetite (Fe_3O_4) which is the most magnetic of all the naturally-occurring minerals. Pyrite (FeS_2) is an example of sulfide mineral.

10.2.2 Formation and Properties of Rocks

Rocks present in nature are classified into three groups, i.e., igneous rock, sedimentary rock, and metamorphic rock. The classification depends on how they are formed. The following figure (Fig. 10.2) shows the taxonomy of rocks.

10.2.2.1 Rock Types

Igneous rocks are formed by the cooling and crystallization of once molten volcanic material, i.e., magma. Magma is molten rock deep within the earth, as molten silicate. Igneous rocks are further divided into two groups, i.e., extrusive (volcanic) and intrusive (plutonic) rocks. If magma stays, cools, and is solidified at considerable depth beneath the earth surface, it becomes intrusive rock. If magma erupts onto the surface from volcanoes (this is called lava) and cools, it becomes extrusive rock.

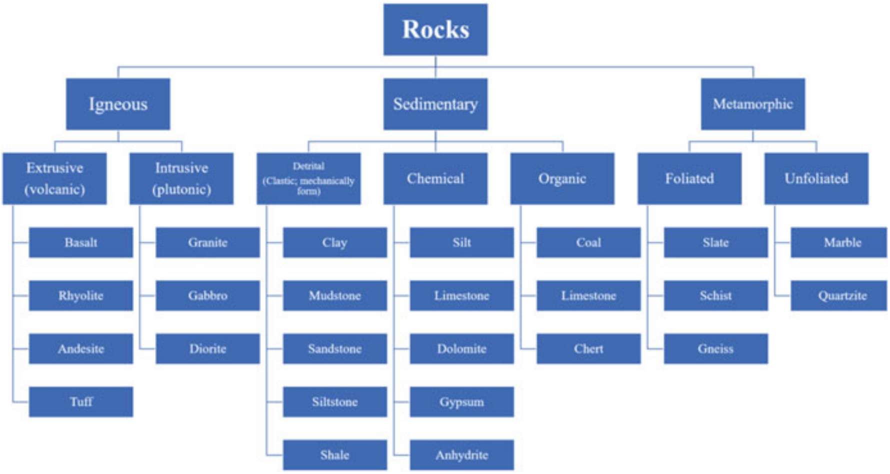

Fig. 10.2 Taxonomy of rocks

Intrusive rock is also called plutonic rock and includes granite, gabbro, and diorite. Plutonic rocks are crystalline and coarse-grained, formed through slow cooling of magma in deep earth. Extrusive rocks are fine-grained and include rhyolite, basalt, and andesite. If lava cools very quickly, it solidifies with no crystal formation and, instead of forming rocks, produces glass-like formation, such as obsidian.

The size of grains in rocks is related to the cooling rate of magma. When magma cools slowly, the individually formed crystal nuclei (the tiny crystal that forms by folded chain at the onset of crystallization) have time to grow to appreciable size. Also as water is more easily retained in an intruded magma than in an extruded lava, an intruded magma has a high proportion of water in the melt inhibiting the formation of nuclei but accentuating the development of large crystals. Therefore, coarse-grained crystals are formed in plutonic rocks. In the case of an extruded lava, its cooling is much faster than that of magma in deep earth and new individual crystal nuclei can be formed faster than the growth rate of crystals. This results in fine-grained crystals in extrusive rocks.

Also, plutonic rocks tend to show a partial crushing and shearing of the component grains by being subject to high pressures. Therefore, plutonic rocks, e.g., granite, tends to have many fractures in the system. This is not the case with extrusive rocks as they are not subject to high pressures. For example, the basaltic rocks contain numerous small, slightly spherical, holes ranging in size from very tiny to diameters of approximately 2.5 cm. These holes are referred to as vesicles and represent bubbles of water vapor which have separated from the lava. These vesicles are not present in granite as the rock have been developed under a high pressure.

The tuff rock is an interesting case. It is an igneous rock in terms of origin but sedimentary in terms of process. Tuff is formed when the materials out of a volcano are blown and accumulated on the ground. Therefore, a combination of the materials released through volcanic eruption and lava fragments, through eventual cooling, forms a tuff rock. The volcanic materials released are the small solid particles and liquid droplets. Tuff is a fine or medium-grained rock, as consolidated volcanic ashes and lava fragments with masses of crystals set into an ash matrix.

Metamorphic rocks form from the alteration of a pre-existing rock. Alteration could be from the process of mountain-building or seismic activities. Such alteration activities apply intense heat and/or pressure through squeezing on rocks. Rocks produced from such processes include slate, schist, gneiss, and marble. Metamorphic rocks are classified based largely on structures and in part by composition. Slate is a fine-grained rock with clear foliation feature where individual minerals can rarely be distinguished. Schist is a well-foliated rock with the mineral grains considerably coarser than in a slate. Gneiss is a foliated rock characterized by the segregation of light and dark minerals into parallel planes. Typical gneiss generally contains more feldspar than schist. Marble is a metamorphosed limestone in which the grain size of calcium carbonate is increased, resulting in shiny surfaces in which individual calcite crystals are easily recognized.

Sedimentary rocks are formed through a very long term process. They are formed from weathering of igneous and metamorphic rocks through gradual accumulation and hardening of sediment of particles. Sedimentary rocks can be divided into three groups, detrital rocks, chemical rocks, or organically formed rocks. Detrital (or sometimes called clastic) rocks are mechanically produced from the breakdown, movement, and accumulation of pieces of igneous rocks. Clay, shale, mudstones, siltstones, and sandstones are detrital rock as the product of the breakdown of preexisting rocks. Chemical rocks are formed through chemical precipitation and sedimentation in the site of deposition. They include rock salt, limestones (calcite rock), dolomites, gypsum, and anhydrites. Chemical rocks consist largely of the broken shells of organisms that existed near the site having been affected by the depositional environment. There are also organically formed rocks. They are similar to chemical rocks but are formed by the remains of plants and animals buried under sediments due to heat and pressure from overlying layers. They include carbonaceous rocks like coal and limestones. Limestone can be of either chemical or organic origin. Chemically formed limestones will be from evaporation of water with $CaCO_3$ while organically formed limestones will be from evaporation of water containing living organisms with $CaCO_3$. The particles in sedimentary rocks include minerals, shells, sand, pebbles and other fragments of material including fossils. Approximately 75–80% of the land area of the earth is covered by sedimentary rocks.

While most of earth surface is sedimentary rock, igneous rock constitutes the major portion of the volume beneath the surface, with granite or closely related rock types. Most continents have a core of very old rocks consisted primarily of igneous rocks admixed with some metamorphic rocks.

Rock formations are in cycle. Once igneous rocks are formed from volcanic activities, they are exposed to weathering (i.e., physical disintegration or chemical decomposition) and erosion, resulting in breaking down of rocks into mineral particles. These mineral particles are transported by rivers, wind, and glaciers, and deposited as sedimentary layers in dry land, river beds, lakes, deltas, dunes, and on the sea-bed. Through burial and compression, they eventually form sedimentary rocks. Some sediment is carried to the greater depths of the ocean floor by ocean currents. When sedimentary and igneous rocks are subjected to intense heat and pressure from such events as earthquakes or mountain-building, they become metamorphic rocks. If very high temperature and pressure are applied to the rocks, they may return to the molten state. This closes the rock cycle. Also, soil is the result of weathering of various types of rocks reflecting what was contained in the rock.

10.2.2.2 Chemical Properties of Rocks

Igneous rocks are mostly made up of silicate minerals such as quartz, feldspars, micas, and ferromagnesian. Magnetite also occurs in small amounts in almost all igneous rocks. The phosphate mineral apatite ($Ca_5(PO_4)_3Cl$), although in small amount, is also extremely common in most igneous rock. Plutonic rocks, e.g., granite, have higher content of silica compared to basalt. This is because highly silicic magma, by being more viscous (due to higher degree of cross-linking) among the most numerous silica tetrahedral, will not reach the surface as easily as the less viscous ones. Therefore, the silicic granite magmas tend to crystallize within the deep earth while less viscous magma reaches the surface and form basaltic lavas before cooled. Tuff includes the minerals such as feldspars, pyroxenes, and amphiboles.

As metamorphic rocks are derived from the alteration of parent rocks, their composition largely depends on what was in the parent rock. Typical metamorphic rock consists of a small number of minerals with grain sizes on the order of 1 to 2 mm along with a distinct layered structure. The small number of minerals indicates that the rock has come to an equilibrium with the given temperature and pressure. In a metamorphic rock, all of the grains have formed roughly at the same time at about the same size through interference with each other during their growth.

As the process of sedimentation involves variations and diversification, the sedimentary rocks have highly variable chemical composition. Therefore, determining the average composition of sedimentary rocks is not simple. While almost any minerals of igneous or metamorphic rock can be present in sedimentary rock, the elemental composition of average sedimentary rock closely resembles the composition of the parent igneous rock except the presence of CO_2, H_2O, and HCl. Therefore, the common minerals of sedimentary rocks include quartz, feldspar, calcite, dolomite, and clay minerals. For example, silicates and silicate-rich alumino-silicates (mostly feldspars) are characteristic of igneous rocks but carbonates and silicate-poor alumino-silicates (mostly clays) are abundant in sedimentary rocks. Salt rock, as an exception, is an evaporitic rock (i.e., formed from the evaporation of saline water). It is made up mostly of the mineral halite (NaCl) with impurities of clay minerals and iron oxide. Salt rock is usually massive and either bedded or domed.

The difference in silica content in rocks is sometimes discussed in terms of acidity of the rock. "Acid" rocks simply refers to rocks with high silica content. Granite is an example of an acidic rock with the silica content over 65%. Basalt is called a basic rock with lower silica content (~53%). Tuff is either acidic or basic. However, use of such term is phasing out as it originated long time ago when it was thought that silica was present in the rocks as silicic acid (which is false). In general, rocks with high contents of sodium oxide (Na_2O) or potassium oxide (K_2O) are called alkaline rock.

Effects on Groundwater Compositions

The body of rock acts as the principal chemical buffer to all solid-water interactions. Thus rock compositions directly affect chemical reactions taking place in the presence of water. In this case, groundwater is a solvent that is in contact with various earth materials dissolving them. Therefore, water becomes essential part of geochemical cycle of rocks providing important medium for chemical reactions.

That same property works against the very principle of isolation of nuclear waste by the penetration of groundwater to the waste isolation barriers through chemical reactions. Understanding how water and rocks and their constituents interact during the disposal phase of nuclear waste is, therefore, very important to project long-term safety of nuclear waste disposal.

Table 10.2 shows examples of groundwater compositions found in several candidate rocks of geological repository in the U.S. The table illustrates large variations in the composition in different rock types. Significant differences in groundwater composition may also exist between different rock formations of the same type, and even between different locations within the same formation.

The types of chemical and biological reactions taking place in the medium of nuclear waste disposal are controlled by the composition of the groundwater and host soil along with the prevailing pH and Eh conditions. These reactions also control the distribution of species of radionuclides in solution. Table 10.3 shows the speciation of several important radionuclides in groundwater as probable aqueous species. In addition to the simple cations and anions, there are a number of complex ions formed with OH^-, Cl^-, HCO_3^- (or CO_3^{2-}), SO_4^{2-}, F^-, and $H_2PO_4^-$ (or HPO_4^{2-}). Species are also present in combination with humic and fulvic acids. Humic and fulvic acids are acid derived from decayed vegetables matter or other organic substances. They are nutrients for plants and increase the ability of soil to retain water. Humic/fulvic substances are chemically organic polyelectrolyte macromolecules and are defined operationally by their solubilities: 1) Fulvic acids (FA): the fraction which is soluble at all pH levels, 2) Humic acids (HA): the fraction soluble above a pH of approximately 3.5, 3) Humin: the fraction insoluble at all pH levels.

Table 10.2 Compositions of deep groundwater

	Concentration in (mg/l)				
	Tuff*	Granite*	WIPP BrineA**	WIPP BrineB**	Basalt*
Temp,°C	30	30	--	25	45
pH	8.5	9.8	6.5	6.5	9 ~ 10
Eh(V)	--	0.17	--	--	−0.5
Ca^{2+}	13	59	600	900	1.3
Mg^{2+}	2	0.5	34,000	10	0.04
Na^+	49	125	42,000	115,000	250
K^+	4.7	0.4	30,000	15	1.9
Cl^-	7.6	283	190,000	175,000	148
SO_4^{2-}	21	19	3500	3500	108
Fe^{3+}	0.16	0.02	2	2	--
ΣCO_3	--	3	700	10	46
SiO_2	50	11	--	--	145
$B(BO_3^{3-)}$	--	--	1200	10	--
F^-	2.3	4	--	--	37
Li^{2+}	0.05	--	20	--	--
Sr^{2+}	0.05	--	5	15	--
Ba	0.2	--	--	--	--
Br^-	--	--	400	400	--
I^-	--	--	10	10	--
Cs^+	--	--	1	1	--
Rb^+	--	--	20	1	--
V	--	--	--	--	0.01
PO_4	--	0.1	--	--	--

Source: *Moody (1982); **Powers (1978)

Table 10.3 Probable aqueous species in groundwater

Ion	Species
Simple anions	HCO_3^-, CO_3^{2-}, I^-, IO_3^-, HSe^-, SeO_3^{2-}, SeO_4^{2-}, TcO_4^-, MoO_4^{2-}, RuO_4^{2-}
Simple cations	Cs^{2+}, Ni^{4+}, Sr^{2+}, Sn^{2+}, Pb^{2+}, Pd^{2+}, Th^{4+}, Pu^{4+}, Zr^{4+}, Pa^{4+}, U^{4+}, Np^{4+}, UO_2^{2+}, PuO_2^{2+}, NpO_2^+
Complexation ligand	**Simple cations forming mono- or polynuclear species with ligand**
OH^-	Ni^{2+}, Sr^{2+}, Sn^{2+}, Am^{3+}, Eu^{3+}, Zr^{4+}, Th^{4+}, Tc^{4+}, U^{4+}, Pu^{4+}, Pa^{5+}, UO_2^{2+}
Cl^-	Ni^{2+}, Sn^{2+}, Pd^{2+}, Pb^{2+}, Pt^{2+}, U^{4+}, Pu^{4+}
HCO_3^-/CO_3^{2-}	O_2^{2+}, PuO_2^{2+}, Th^{4+}, NpO_2^+, Ni^{2+}, Tc^{4+}
SO_4^{2-}	Eu^{3+}, U^{4+}, Pu^{4+}, Am^{3+}, UO_2^{2+}, PuO_2^{2+}
F^-	U^{4+}, Pu^{4+}, UO_2^{2+}, PuO_2^{2+}, NpO_2^+
$H_2PO_4^-/HPO_4^{2-}$	UO_2^{2+}, PuO_2^{2+}, NpO_2^+, U^{4+}, Pu^{4+}, Th^{4+}
Humic and fulvic acids	U^{4+}, UO_2^{2+}, Pu^{4+}, PuO_2^{2+}, Ni^{2+}, Pb^{2+}, Th^{4+}

Source: Moody 1982

Example 10.1: pH in Tuff Rock

For the rock composition given in Table 10.2, find the pH in the tuff rock.

Solution:

The major ions that affect the charge balance in a tuff rock are: [Ca^{2+}], [Mg^{2+}], [Na^+], [K^+], [Cl^-], [SO_4^{2-}], and [F^-]. This assumes that the ones below the concentration of 0.2 ml/g have negligible effect on pH of the tuff rock.

The resulting charge balance of the rock composition is,

$$\sum_{i}^{cations}\left(\frac{mol}{l}\right)_i - \sum_{j}^{anions}\left(\frac{mol}{l}\right)_j = 2\,[Ca^{2+}] + 2[Mg^{2+}] + [Na^+] + [K^+] - [Cl^-] - 2\left[SO_4^{2-}\right] - [F^-] = 2\left(\frac{13\left[\frac{ml}{g}\right]}{40.08\left[\frac{g}{mol}\right]}\right) + 2\left(\frac{2}{24.3}\right) + \frac{49}{23} + \frac{4.7}{39.1} - \frac{7.6}{35.5} - 2\left(\frac{21}{96.1}\right) - \frac{2.3}{19.0} = 2.29\left[\frac{mol}{L}\right]$$

Using Example 4.3, the pH of groundwater in the tuff rock is determined by solving,

$$2.29 \times 10^{-3} + \left([H^+] - 2\frac{10^{-21.54}}{[H^+]^2} - \frac{10^{-11.22}}{[H^+]} - \frac{10^{-14}}{[H^+]}\right) = 0$$

Where, terms in parenthesis are from Example 4.3 (Sect. 4.3).

Solving for [H^+],

$[H^+] = 2.457 \times 10^{-6} = 10^{-5.61}$, thus the pH value is 5.61

10.2.2.3 Physical Properties of Rock

As a potential host medium for geological repository, physical properties are important in relation to structural stability of rocks. These properties include density, strength, thermal expansion coefficient, thermal conductivity, specific heat, porosity, and permeability.

As igneous rocks are formed through cooling and crystallization of high temperature molten volcanic material, they present well structured, strong material to provide good structural support for geological repository. Metamorphic rocks, as a product of high temperature and pressure alteration, are weak in physical strength and thus not appropriate as a host medium for geological repository. Strength of sedimentary rock depends on what was mainly included in the rock but in general is low.

Table 10.4 shows the values of physical strength of selected rocks as the ultimate strength, the maximum stress that a rock can withstand before it breaks. Strength of a rock is related to its density which reflects the degree of open void spaces within the rock. The values of the density of selected rocks are also given in Table 10.4 as well as in Fig. 10.3. In general, the bulk density of a rock varies between 1.6 and 3.5

Table 10.4 Values of ultimate strength and dry bulk density of selected rocks

Rock type	Ultimate strength (kilobars)	Dry bulk density in g/cm3 (standard deviation)
Granite	11.5 (at 500 °C)	2.66 (0.06)
Basalt	10 (at 500 °C)	2.74 (0.47)
Tuff		2.36–2.57
Marble	5.5 (at 24 °C), 2 (at 500 °C)	2.67–2.75
Limestone	5.5 (at 24 °C), 3 (at 500 °C)	1.55–2.74
Dolomite	7 (at 24 °C), 6.5 (at 500 °C)	2.72–2.84
Shale	2.5 (at 24 °C)	2.06–2.67
Rock salt	1 (at 24 °C)	2.10–2.20

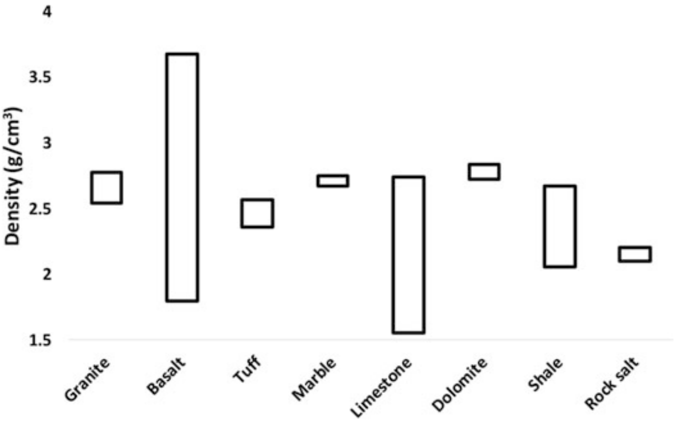

Fig. 10.3 Distribution of bulk density of selected rocks

(2.6 g/cm^3 as average) and density is in general high in high strength rocks. A related quantity, porosity, is discussed in Sect. 11.2.4.

Figure 10.3 also shows the distribution of density of different rocks.

Thermal conductivity is an important property in terms of rock selection for the stability of a geological repository. High thermal conductivity is desirable to minimize the impact of heat from nuclear waste on the stability of rocks. Thermal expansion coefficient is another physical property of importance in relation to temperature distributions within a repository. Thermal expansion of rocks is closely related to the surface uplift at a geological repository site which has to be constrained as part of thermal design limits. Table 10.5 shows the values of thermal conductivity and thermal expansion coefficient for selected rocks.

Porosity and permeability are important in terms of potential impact of groundwater flow and waste package degradation and migration of radionuclides. These hydrology related properties are discussed in Sects. 11.2.3 and 11.2.4.

Table 10.5 Values of thermal properties of selected rocks

Rock type	Thermal conductivity (W/m/K)	Volumetric thermal expansion coefficient $\alpha = \left(\frac{1}{V_0}\right)\left(\frac{\Delta V}{\Delta T}\right)$ (10^{-5} per °C)	Specific heat (10^3 J/kg K) (50–65 °C)	Volumetric heat capacity (J/m^3-K)
Granite	2.8 (2.3–4.5)	2.4	0.95	2.23E6
Basalt	1.7 (0.88–2.6)	1.6	1.04	
Tuff (Nevada)	1.85 (0.99–2.6)			
Clay/shale	1.8 (1.1–2.7)		0.94	2.5E6
Salt rock	4.9 (3.9–5.4) @ 100 °C 3.2 (2.7–3.7) @ 200 °C			2.0E6

Source: Hardin (2012), Robertson (1988), Clark (1966)

10.3 Candidate Rock Types for Geological Repository

Mechanical, thermal, hydrogeological and geochemical properties of rocks are examined to select host medium for geological repository. Desirable physical properties include good mechanical strength, resistance to creep deformation, high thermal conductivity and high heat capacity, and small thermal expansion coefficient to minimize thermally induced disruption. In terms of hydrogeology or geochemistry, desirable properties are low water content, low permeability, high degree of groundwater flow dispersion, and high ion retention capacity. Common candidates as host medium for geological repository include granite, salt, clay/shale, basalt, and tuff.

10.3.1 Granite

Granite is the most common type of rock among the candidates for geological repositories. Granite has high degree of physical strength, low water permeability, and is not associated with natural resources (metallic ore, oil or gas). In granite, both water content and solubility of most species are low. Thus the dissolved solids content and the corrosion potential of the associated water is generally low. As a disadvantage, the granite rocks can have a high secondary permeability as a result of fracturing and with low ductility. Therefore, fracture flow of groundwater is a concern in granite. Significant variations in fractures within individual rock units may also exist. The cost of mining hard rock is higher than for mining salt. Also, as thermal conductivity of granite declines rapidly with increasing temperature, higher thermal stresses exist in the repository vicinity when the decay heat inventory of nuclear waste is high.

10.3.2 Salt

Salt easily dissolves when contacted with water. However, the very existence of the salt bed or dome as large rock body implies that flowing water has been absent for very long periods of time (e.g., over millions of years). Salt beds are laid down through the evaporation of prehistoric seas whereas salt domes are formed when salt beds of lesser density than adjacent rock "bubbled" upward through the overlying strata under stress. The formation of salt domes is a manifestation of the quasi-viscous behavior of salt.

Salt medium is available in many countries and extensive mining experiences are available. Salt has very low permeability and low water content and salt formations are often located in areas of low seismicity. Also, salt rock plastically deforms under pressure, which leads to self-sealing of any fractures or openings. This ensures high degree of isolation of waste buried in salt. However, this also means that it is difficult to maintain the stability of underground openings after the excavation or during operations. The cost of developing disposal space is relatively low as compared to other rock types. With high thermal conductivity, the heat transfer properties of salt are good compared to other rock types.

In case of being contacted with moving water, salt would be dissolved and removed rapidly. Also salt rock has low sorptive capacity for contaminants, thus the degree of retardation in contaminant migration is low. Salt water is also highly corrosive to many materials. Salt is often found in conjunction with hydrocarbons. Therefore, use of salt rock formations for geological repository raises the possibility of human intrusion (for the exploration of petroleum or gas which may allow the entry of water). With high thermal expansion coefficient of salt, the degree of surface uplift at the site needs to be investigated.

10.3.3 Clay/Shale

Argillaceous rocks such as shales or clay are fine-grained sedimentary deposits. Advantages in using these rocks are their wide availability, low permeability, relatively high plasticity, high ion-exchange capacity thus providing significant retardation capability for contaminants, and great homogeneity of the medium facilitating modeling effort. However, as clay rocks have relatively low strength, potential instability of the mine structure (e.g., heaving and roof collapse) exists and should be addressed in repository construction. Clay rocks are also usually associated with the presence of contiguous aquifer implying larger potential health impact if the groundwater gets contaminated.

The rocks are often associated with the presence of hydrocarbons, thus presence of boreholes in adjacent strata is a possibility with potential concern of human intrusion.

10.3.4 Basalt

Basalt occurs widely in continental areas, and is the principal rock of the ocean floor. With their fluidity, basalt may form very thick lava sheets. Basalt is also a very strong rock with low permeability due to secondary mineralization within fractures. Typically with reducing chemistry, minerals in fractures are highly sorptive. There is little or no natural resource potential involved with basalt strata. However, with variations in properties and lateral and vertical extent of beds, basalt presents complex geology and hydrology. Some layers (interbeds) may have high permeability. Basalts also have relatively low thermal conductivity. With high physical strength, the cost of excavation is very high. Basalts are located in seismically active areas raising concerns over earthquake.

10.3.5 Tuff

Tuff rocks are usually present in significant thicknesses above the water table, i.e., in the unsaturated zone (see Sect. 11.1.2). Therefore, influence of water on the repository system can be minimized. If located in arid regions, water will have very low flow rate. Tuff also include highly sorptive materials constituting large portion of the bed providing a natural chemical barrier against migration of radionuclides. Little or no mineral or energy resource potential is involved with the tuff rock site. However, it is often difficult to characterize and model the tuff strata due to its presence in the unsaturated zone. Also seismic activity may be high in the region. If located in arid regions, the underlying aquifers may be attractive to future generations increasing the chance of human intrusion.

10.3.6 Comparisons of Rocks

Advantages and disadvantages of the major candidate rocks are summarized in Table 10.6. As indicated, no single rock comes with all of the desirable properties. Due to practical limitations in the available rock types in a country, selection of rock type is often based on what is available. One observation to note is that clay rock and rock salt can provide rather substantive barriers for the isolation of nuclear waste buried in the medium based on their natural characteristics. In the case of granite, basalt, tuff, the degree of isolation of nuclear waste is comparatively lower due to the presence of fractures in the system. This implies that the role of engineered barriers

Table 10.6 Summary comparisons of advantages and disadvantages of candidate rocks for geological repository

Rock types	Advantages	Disadvantages
Granite	- Low permeability for water - Not associated with natural resources - High degree of physical strength - Low solubility - Low water content	- High secondary (i.e., after fracturing) permeability - High excavation cost - Lower ductility - Fracture flow of groundwater - Significant variations within individual rock units - Higher thermal stresses due to increased thermal gradient in the repository vicinity (thermal conductivity declines rapidly with increasing temperature)
Basalt	- High mechanical strength - Low permeability due to secondary mineralization in fractures - Minerals in the fractures are highly sorptive - Little or no natural resource potential	- Complex hydrology - Most expensive to excavate - Complex geology - variations in properties and lateral and vertical extent of beds - Some layers (interbeds) have high permeability. - The basalts are located in seismically active areas. - Relatively low thermal conductivity
Tuff	- Highly sorptive materials constitute large portion of the rock bed - Little or no mineral or energy resource potential - Present in significant thicknesses above the water table - Very low water flows in arid regions	- Difficult to characterize and model the strata - Seismic activity may be high - Aquifers in arid regions may be attractive to future generations - Complexity of describing in unsaturated zone hydrology
Salt	- Low water content - Fractures tend to be self-sealing. - Low excavation cost - The heat transfer properties are good. - Very low permeability - Abundant availability - Extensive mining experiences - Often located in areas of low seismicity - High degree of nuclear waste isolation	- Dissolve rapidly, if contacted with water. - High solubility - Difficult to maintain the stability of underground openings - Salt water is highly corrosive. - Salt is often found in conjunction with hydrocarbons. - High thermal expansion coefficient - Low sorptive capacity
Clay/ Shale	- Wide availability - Low permeability - Relatively high plasticity potentially leading to self sealing of openings - High ion-exchange capacity - Homogeneity of rocks - High degree of nuclear waste isolation	- Relatively low strength - Structural instability of the mines - Presence of gases that can result from carbonaceous shales - The presence of contiguous aquifer - High water contents - Possible presence of boreholes in adjacent strata containing hydrocarbons

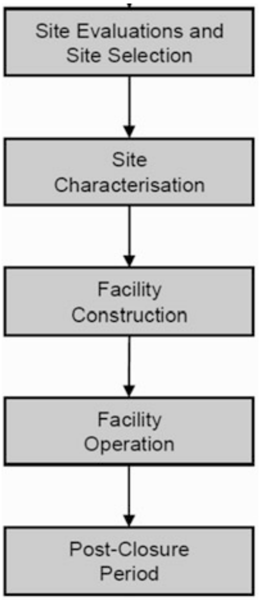

Fig. 10.4 Key steps in geological repository development. (Source: IAEA 2014)

becomes significant in granite, basalt, and tuff in comparison to clay or rock salt for long-term isolation of nuclear waste.

10.4 Development of Geological Repository

Presence of geological formations that have been physically and chemically stable for millions of years implies that these formations would remain stable for a long time in the future as well. This offers the potential for long-term isolation of nuclear waste.

Development of geological repository is a very complex task. It requires the examination of a variety of issues and their implications including science, technology, risk perception and acceptance among the public, the local and national politics, and credibility of the entity for project management. The activities to be carried out include site evaluation and selection, site characterization, facility construction, preparation/packaging/shipment/emplacement of nuclear waste, and closing the facility. These are captured in Fig. 10.4.

10.4.1 Site Evaluation

Identifying a potential host site is a key milestone in geological repository development. Along with the consideration of rock properties, various other factors need to be taken into account in this step including site stratigraphy, hydrological and geochemical features, meteorological conditions, and stability with respect to earthquake or volcanism.

In terms of stratigraphy, the rock body at the site should be deep enough. This is to rule out or minimize the impacts of surface disruptions, such as floods, storms, rain, weathering, or glacial phenomena to effectively separate the repository from the biosphere. This requires the repository to be located several hundred meters down from the surface. The area should also be large enough to accommodate the physical footprint of the facility. Typically several square kilometers of area are needed. The rock body should be reasonably homogeneous both vertically and horizontally to facilitate site characterization and hydrogeological analysis. It is also desirable to have a thick rock body (~ 100 m) with relatively flat inclination for the stability of the system and long-term isolation of radionuclides.

As to hydrologic characteristics, the movement of groundwater should be slow and preferably follow long path lengths before reaching the biosphere. For example, if two possible locations of geological repositories are identified that are rather closely located, one that has a very long path length for the groundwater before coming out to the surface has to be chosen. Also, under an ideal situation, the groundwater should be isolated from the aquifers accessed by the local population for water consumption. Presence of reducing chemical conditions within the rock is desirable to minimize degradation of engineered barrier materials and to maximize the degree of radionuclides isolation (through low solubility and large sorptive capacity as discussed in Chap. 11).

The desirable meteorological conditions of a site are low level precipitation to minimize water infiltration and low pressure variations to reduce pressure-induced gaseous release of radionuclides.

The stability of the earth's crust (i.e., tectonic stability) in the region should be also considered. If the region is tectonically unstable, deformation of rocks is likely at the site from the movements of rock bodies relative to each other, e.g., through earthquake. Such deformation include faults and folds. A fault is a crack or planar fracture in a volume of rock caused by relative movement of rock surfaces. A fold is a bend in rocks.

Tectonic instability could result in broad regional uplift or subsidence affecting the integrity of the repository system and the waste packages. Also, presence of possible volcanic activities or any disturbances compromising the stability of the repository or flow of mobile material should be examined.

Another important factor to consider is the presence of valuable natural resources such as oil, gas, or metals. A desirable site is away from such resources as people in the future generation may be drawn to the region with an intent to have an access to the resources.

Non-technical (societal) factors must receive serious consideration in site evaluation. These non-technical factors include population density in the region, socioeconomics of the local population, and the level of social acceptance of the repository in the region. Any cultural significance of the site or presence of artifacts with cultural significance in the area should also be examined. These non-technical factors can be potential show-stoppers to the process of site selection. Ease of access to the proposed facility, through ground transportation (trucks or trains) should also be considered as part of cost consideration of repository construction.

Consideration of these non-technical issues along with technical investigations becomes the basis of recommendations regarding whether the effort will move to the next phase of repository development, i.e., site selection. Such consideration will minimize potential detrimental impacts from social and political influences before major efforts of site development are made. Stakeholder engagement becomes a very important part of the effort in preparation for the next phase.

10.4.2 Site Selection

Perhaps the biggest difficulty in geological repository development lies with site selection. Consider the extent of effort made for the U.S. Yucca Mountain project. There were 126 public hearings held and $240 million was spent in support of the state and local governments for site selection. Although the site was approved and announced as the nation’s HLW disposal site, the project was later cancelled due to the objection from the host state. This case clearly demonstrate the challenge of site selection.

Different approaches can be taken to site selection. These approaches include the technical approach, the public participation approach, the market approach, and the distributive justice approach. The technical approach is to select “optimal” sites by technical experts using objective criteria. The public participation approach is to allow the public to select sites through participatory decision making process. The market approach is to select the site based on compensation mechanism as the basis of gaining acceptance from the candidate communities. The distributive justice approach is to select the site based on the consideration of fairness in the selection decision.

The technical approach is based on a belief that a complete, objective analysis is the best way to find a site. Such approach is defended on the ground that the public does not possess the necessary expertise for the selection. Therefore the approach ignores public participation in the decision process. The so-called “Decide-Announce-Defend” (DAD) model is a traditional technical approach. According to the approach, first, national search is made to find desirable candidate sites and decisions are made based on examining technical acceptance criteria. As the decision is made with technical judgments with little public input, the chosen site’s local community would feel helpless. In this case, the only power the community can exercise in the decision process is to object. In particular, with strong negative public perception of nuclear waste, the opposition is very much likely to be very intense. The technical approach was widely used around the world in the 1970s and 1980s only to witness failures. These failures include the Yucca Mountain project of the U. S., the initial Swedish and Finnish efforts, the South Korean HLW siting work, etc. A typical misconception in the approach is that if the experts can show the risk is low and the site is good, the public should trust and accept the decision. If the public does not accept or oppose, it is because they don't understand.

The public participation approach is the opposite one. It is based on a premise that if we bring everyone together to talk about the project, we will come to an agreement. In reality, bringing everyone together does not solve the problem. This is because what each person expects or wants may be different. Such approach may lead to selection of technically inferior sites or no agreement at all. One of the common tools used for public involvement is public hearings. Public hearings would work as long as the stakeholders (e.g., the public, government, and the industry) share similar concerns and remain open-minded and the public is well informed. Yet, in most cases, the public lack technical expertise and appropriate information to make an informed judgment. If major decisions have already been made and the public hearings are to obtain the legal right to proceed, the public will feel that they are deceived and just being taken advantage of. The meeting will not bear a meaningful outcome in that case.

In the market approach, any concern of or opposition to a project is believed to be assuaged by providing money. Therefore, the approach uses monetary compensation as a way to solve problems. The approach certainly has merits but compensation is not a panacea. Compensation is unlikely to gain acceptance of local residents if the recipients feel that they are being asked to forsake their fundamental moral value or obligation to future generations. In this case, monetary offers are considered a bribery and will have negative effects such as causing hostility.

The distributive justice approach appears ideal as it allows meaningful participation of the public based on fairness. However, as people's stakes in a decision differ widely, achieving consensus will be very challenging. The approach may not provide an acceptable solution to all the stakeholders involved even after much effort is expensed.

What is needed is an integrative approach that combines the strengths of different approaches while compensating the weaknesses of the respective approaches. Regardless, performing a good complete, objective technical analysis is important and necessary. However, technical analysis should not be the sole basis of site selection. Public participation is also an important part of the process. Nonetheless, the process should be carefully coordinated for the public to feel that their participation is meaningful. Monetary compensation for the affected public is not just appropriate but obligatory in any project involving the disposal of nuclear waste. It is part of procedural standards of fairness. In this regard, the level of compensation should be larger than the social costs imposed on people who live near the site. To effectively address distributive justice, multiple sites should always be considered. Such approach will reduce the sense of inequality by having only one local area taking the burden of accepting all of the nation's waste. Having only one candidate site also weakens the position of the government in the negotiations. With only single site under consideration, the candidate community has the huge leverage in the project by either opposing or delaying the process until the community gets what they want. Minimizing the required footprint size of the repository could help by possibly expanding the list of potential candidate sites.

The siting process could begin by sending a list of environmental requirements to all communities by the siting authority. To draw interest of potential communities, the amount of compensation should also be specified and announced. If necessary, the compensation amount should be increased if no interests emerge. The amount of compensation should be based on genuine consideration of the welfare of the host community. An ideal form of compensation is direct money payments to local residents as well as direct financial benefits to local governments (i.e., reduced tax rates) to directly ameliorate the negative impact from the facility. Conditional compensation should also be considered to cover the uncertain costs of development such as guaranteeing property values, or any relevant insurance program. Different levels of compensation could be designed depending upon whether the candidate sites can be confirmed to be acceptable from the site characterization studies. Along with the dealings with compensation, the entity responsible for the project must engage and develop collegial personal relationships with potential host communities by establishing and maintaining the culture of trust. This requires listening to the concern of the community residents with open communication efforts Please refer to the cases of Finland and Sweden where success in site selection was achieved after initial failures (discussed in Sect. 10.6).

10.4.3 Site Characterization

Site characterization is a detailed technical investigation of a site to examine the adequacy of the site regarding the necessary characteristics for nuclear waste isolation. Such characteristics require in-field investigations of physical rock properties, site hydrogeology and geochemistry, and their interactions. Site characterization is often performed in parallel with preliminary safety assessment. In preliminary safety assessment, site specific hydrogeological features are integrated with facility design concepts with respect to the suitability in meeting licensing requirements. The assessment has to also identify processes and phenomena at the site that potentially have large impact on waste isolation. Construction and operation of underground research laboratory (URL) could be part of the site characterization effort. If the site is found adequate in meeting the regulatory requirements for a geological repository, a request for authorizing repository construction is made.

A URL is any underground facility developed for the purpose of supporting the development of geological repository. A URL allows underground experimentation to analyze the potential behavior of repository system over long time spans. Such experimentation provides data to support model development to describe system component behaviors of a repository and to test the performance of models. A URL is used also to demonstrate the feasibility of the technologies (e.g., waste packages) proposed for implementation in geological repositories. Therefore, a URL supports technology and methodology development needed for a geological repository.

A URL can be directly tied to the mission of charactering a site or its mission is for general research purposes. If the facility directly supports characterization of a specific site, the URL is called site-specific URL. If a URL is developed for general research and testing purposes without a plan for waste disposal at the site, it is called a generic URL. More than twenty URL facilities have been built in the world, mostly as generic URLs. The number of site-specific URLs is increasing recently.

10.4.4 Facility Construction

Upon successful site selection and site characterization, construction of a repository begins once the required disposal capacity and the layout of waste emplacement in the repository are determined along with the specification of the design of the repository. It starts with building above ground facilities and excavating underground tunnels using tunnel boring machines. Details of the repository design could be modified during excavation to accommodate variations and perturbations in local lock mass and presence of faults.

Different design concepts of a geological repository consider how wastes are emplaced in the facility. These concepts include in-tunnel emplacement, borehole emplacement, and cavern emplacement. The distance between the tunnels or emplacement boreholes is determined mainly based on thermal design requirement for dissipation of heat (see the discussion in Sect. 10.5). In the case of in-tunnel emplacement (considered in Switzerland and USA), waste containers are placed horizontally along the axis of an emplacement tunnel. Waste packages are placed sequentially in each tunnel. Typically, the open spaces outside waste packages are backfilled and the tunnel is sealed. This approach has the advantage of easy constructability and simplicity of operation but requires potentially large amounts of backfill. Also backfill emplacement operation needs to be done remotely due to the presence of radiation field surrounding nuclear waste packages.

In the case of borehole emplacement (used in Sweden and Finland), additional boreholes within the floor (for vertical emplacement) or walls (for horizontal emplacement) of tunnels are excavated. One or more waste containers are then loaded into these boreholes and backfilled. Vertical borehole emplacement allows the opportunity to characterize the rocks exposed in the boreholes and provides shielding from waste packages to facilitate access after waste emplacement. The disadvantage of the approach is high cost and complexity of drilling and emplacement operations being complex. When horizontal boreholes are used, emplacement operations are simpler but drilling for the borehole is still complex and its cost is high.

In cavern emplacement, multiple waste containers are stacked together within one large silo of engineered barriers.

Installation of ventilation system is an important consideration in repository design. Ventilation system uses forced or natural convection to remove heat during the operational period and controls the temperature within the emplacement tunnels. Typically ventilation system removes up to 80% or more of heat generated by waste packages.

To obtain the license for the initiation of the construction/operation of the repository, safety assessment of the repository, called performance assessment, per “as built” conditions of the repository design is made. Performance assessment also allows early determination of the suitability of a site (as part of site characterization) and provide necessary insights in repository designs to meet the safety requirements for protecting humans and the environment.

10.4.5 Facility Operation and Site Closure

Once the facility begins operation, transportation and emplacement of nuclear waste take place. Emplacement follows the spacing requirement between the packages per the thermal design limits to control the temperature of the waste packages and the repository system. High-burnup (i.e., high heat) spent fuel assemblies can be combined with low-burnup (i.e., low heat) assemblies or other cooler waste types within waste packages for the purpose of maintaining evenly distributed heat loads. Emergency response plan must be in place until the full disposal capacity is reached to provide protection of the workers and the waste packages against any accidents or security threats during the repository operations. Depending upon the requirement on waste retrievability, the repository may remain open during a specified period. Retrievability is to correct any observed failures during the operation of the repository. Permanent closure of the repository requires the use of backfills and seals, along with setting up of security and monitoring systems. The final phase of safe assessment is performed before repository closure to support the decision for repository closure. Once the repository is closed, human access to the disposed nuclear waste is strictly prohibited. Special markers are set up in the perimeter of the repository on the surface to warn the people of the hazards in the area.

10.4.6 Post-closure Period

Institutional control of geological repository is needed to implement security and monitoring systems for the repository. The monitoring is to identify any failures of waste packages and release of radioactive materials. The period of institutional control typically ranges between 100 and 300 years. Permanent special markers are set up to alert or warn future generation to prohibit attempts to gain access to nuclear waste even in the case of loss of institutional memory. Such markers need to be carefully designed without relying on particular language(s) by taking into account various scenarios of institutional memory loss.

10.5 Consideration of Thermal Limits in Geological Repository Design

The thermal design limits are to prevent the failure of waste packages due to excessive temperature increase and to preserve the integrity of rocks and engineered barriers surrounding nuclear waste during the disposal stage. These limits as thermal constraints will affect waste package size, repository layout design, and operation of repository. These limits also effectively control the total amount of nuclear waste to be disposed of at a geological repository, therefore control the disposal capacity of the repository.

10.5.1 Thermal Design Limits

The thermal design limits of a geological repository are imposed by limiting the temperatures of the engineered barrier system and the rocks both near and at distance away from the engineered barriers (Hardin 2012).

Limiting the temperatures of rocks is to minimize thermally induced stresses or displacements in the rocks, which would minimize thermal degradation of rocks and the mined openings. Limiting rock temperatures also constrains formation of new hydrologic flow paths, thermally driven coupled processes, and large-scale thermal expansion. This also helps preventing induced fracturing or displacements of rocks along faults or fractures. Limiting rock temperatures also helps to minimize micro-cracking or mineralogical changes of rocks near the engineered barriers, where temperatures are much higher.

Temperature limits are also imposed on the waste package surface temperatures or waste centerline temperatures. For example, the cladding temperature of spent fuel should be lower than 350 °C during permanent disposal to prevent the cladding failure. The limit for cladding temperature can be higher (400 °C) during the operational phase. Also the peak centerline temperature of borosilicate glass waste forms should be below 500 °C at all times to avoid devitrification.

The U.S. Yucca Mountain repository design is based on so-called "high temperature operating mode" (HTOM). This approach is to keep the temperature of the repository system higher than the boiling temperature of water in order to prevent water infiltration into the repository system. Under HTOM, the temperature limits used at Yucca Mountain are: 1) The peak rock temperature midway between adjacent drifts (tunnels) must remain below the local boiling point (96 °C): This limit is to allow passing of water that is boiled away from the drifts by flowing through the repository without contacting the waste packages; 2) The peak rock temperature at drift walls must remain below 200 °C: This limit is to prevent fracturing of rocks and to minimize potential migration of radionuclides. Along with these two limits, the generic limit of spent fuel cladding temperature below 350 °C must also be kept.

Table 10.7 Thermal design limits associated with proposed geological repository

Country	Rock type	Temperature constraints
USA	Salt	Fuel cladding 375 °C max Salt 250 °C max HLW glass 500 °C max
Germany	Salt	Salt 200 °C max
Belgium	Clay	Backfill 100 °C max
France	Clay	Argillaceous host rock 100 °C max
Switzerland	Clay	Clay-based buffer 125 °C max
Sweden	Granite	Clay-based buffer 100 °C max
France	Granite	Canister surface 100 °C max
Finland	Granite	Clay-based buffer 100 °C max
USA	Tuff	Between tunnel temperature < 96 °C Tunnel wall temperature < 200 °C

Thermal conductivity of clay-based buffer material = ~0.4 W/m-K in dry compacted form and 1.35 W/m-K in saturated form
Source: Hardin (2012, Table 1.1)

Using the HTOM approach is feasible at Yucca Mountain because the repository is located within a dry unsaturated rock body. Such approach, however, is not applicable to other saturated rock repositories (the concept of saturated versus unsaturated rock and water table is discussed in Chap. 11).

In general, rock temperature limits are to prevent microcracking of rocks through thermal degradation. The corresponding target values are 200 °C for crystalline rocks (granite, basalt, or tuff) or 200° ~ 250 °C for salt rock, and 90 ~ 100 °C for clay/shale rocks. Thermal degradation of rocks is caused by the decrease in thermal conductivity and the associated swelling of rocks. In the case of clay rocks, temperature increase also leads to mineralogical changes (e.g., cementation) of the rock. In the use of clays as backfill surrounding the waste packages, similar limits (90 or 100 °C) are also applied. Table 10.7 shows the summary of thermal design limits adopted by different countries.

Also, as a temperature related measure, a limit on the maximum uplift at the surface over the repository due to thermal expansion was considered. This limit, in the case of the U.S., was 1.5 meter (Russell 1977).

10.5.2 Implementation of Thermal Design Limits

Demonstration of compliance with the imposed temperature limits requires heat-transfer analysis of the repository system. Typically, such analysis is made by using computational models describing 3-dimensional temperature distributions of a repository. An alternative way of compliance demonstration is to use the concept of linear averaged loading (or simply called, line loading) or areal power density (APD). The linear average loading represents the total thermal energy of the emplaced waste in a tunnel divided by the total length of the tunnel. The APD is defined as the concentration of thermal energy produced by the emplaced waste, which is averaged over the area of the repository and expressed in watts per square meter (or kilowatts per acre). Both concepts facilitate the loading of nuclear waste onto a geological repository while meeting the imposed temperature limits. These concepts assume that the SNF waste canisters are spaced nearly end to end acting as a uniform (line) source of heat. In the case of the Yucca Mountain repository, the desired APD was estimated at 57 kilowatts/acre (NWTRB 1992) to limit the centerline temperature of the waste package to be less than 350 °C.

While the APD or the line loading provides a simple way to accommodate thermal design limits in a repository, these approaches ignore the varying nature of decay heat in the emplaced waste. To overcome this limitation, using integrated decay heat over time is utilized through simplified temperature distribution calculation. The following equation (Eq. 10.1), a simplified model for rock temperature T in the vicinity of heat source, gives an example of calculating temperature distributions in the repository by quantifying integrated decay heat from the waste. In the equation, the temperature of the rock increases beyond the ambient rock temperature, T_{amb}, as a function of the heat generation rate of a waste package, via conduction heat transfer through the backfill. This relationship can then allow the estimation of the total amount of waste loading in a repository per the given temperature limits.

$$T(t) = T_{amb} + \frac{Q(t)}{2\pi K_{backfill}} \ln\left(\frac{r_2}{r_1}\right) + \int_0^t \frac{Q(\tau)}{4\pi K_{rock}(t-\tau)} \mathrm{e}^{-\frac{r_2^2 \rho C_P}{4K_{rock}(t-\tau)}} d\tau \tag{10.1}$$

where, K_{rock} = thermal conductivity of rock (W/m-K), ρC_p = volumetric heat capacity of rock (J/m^3-K), $K_{backfill}$ = thermal conductivity of backfill (W/m-K), $Q(t)$ = linear heat generation rate of the waste package (W/m), r_1 = the radius of waste package (m), r_2 = the radius of backfill buffer (m), T_{amb} = the ambient rock temperature (°C).

Example 10.2: Rock Temperature Estimation for the Geological Repository

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Assume that waste packages containing 8 PWR spent fuel assemblies are going to be emplaced horizontally in a geological repository. These spent fuels have been cooled for 30 years after reactor discharge.

1) By using Eq. 10.1, determine the maximum temperature of the rock surface outside the backfill.

2) If the maximum allowed temperature of the rock body between two emplacement tunnels is 96 °C, determine the spacing distance between the tunnels. (assume steady state heat transfer, q'' (W/m^2) = $k_{rock}(\Delta T/\Delta x)$)

Use:

The total decay heat in the waste package at the time of emplacement $Q(t) = 3300 \cdot e^{-0.0156t}$ (W) where t is in years.

The thermal conductivity of rock = 1.1 (W/m-K)

The thermal conductivity of backfill = 0.4 (W/m-K)

The radius of waste package = 0.33 (m)

The length of waste package = 6 (m)

The radius of backfill = 0.35 (m)

The volumetric heat capacity of rock = $2.2\text{x}10^6$ (J/m^3-K)

The ambient rock temperature = 20 °C

Solution:

1) The simplified model given in Eq. 10.1 indicates that the rock temperature $T(t)$ at time t (after spent fuel is placed in a geological repository) is the sum of three main components: Ambient rock temperature, heat conduction through backfill, and heat accumulated at the rock from $\tau = 0$ to t. Mathematically speaking, maximum temperature of $T(t)$ occurs either when $T'(t) = 0$, $t = 0$, or $t = \infty$.

Note that $q(t)$ = linear heat generation rate $= \frac{Q(t)}{L} = \frac{3,300 \cdot e^{-0.0156t}\ [W]}{6\ [m]}$ or $q(t) = 550 \cdot e^{-0.0156t}\ [W]$

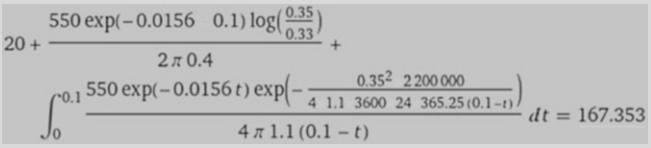
Let's first examine the last term in Eq. 10.1, accounting for the heat accumulated in the rock (the integral term). When $t = 0$, the integral term becomes zero. When $t = \infty$, the integral term also becomes zero (which can be found using the fundamental theorem of Calculus and finding the limit as $t \to \infty$ using L'Hospital's rule). Accordingly, the integral term becomes zero either when $t = 0$ and $t = \infty$.

Next, looking at the backfill heat conduction term, it can be easily deduced that its maximum occurs at $t = 0$ since it is a simple exponential term with negative exponent and $\lim_{t\to\infty} Q(t) = 0$. Thus the maximum additional temperature it contributes to the model is ~13 °C, as shown below:

$Q(0) \cdot \frac{1}{2\pi K_{backfill}} \ln\left(\frac{r_2}{r_1}\right) = \frac{550e^0}{2 \cdot \pi \cdot 0.4} \ln\left(\frac{0.35}{0.33}\right) = 12.88^\circ\text{C}$

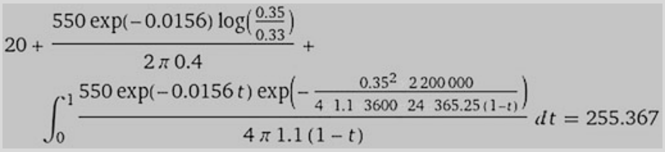
Therefore, one can tell that T_{max} occurs at neither $t = 0$ nor $t = \infty$. The time of T_{max} can be found by taking the derivative of the equation and forcing $T'(t_{max}) = 0$. Now, although finding the derivative of the backfill heat conduction term is simple, it is not easy to find the derivative of $\int_0^t \frac{Q(\tau)}{4\pi K_{rock}(t-\tau)} e^{-\frac{r_2^2 \rho C_P}{4K_{rock}(t-\tau)}} d\tau$ analytically because of the t term inside the integral. Instead of using analytical method, we can find T_{max} numerically through trial and error by substituting multiple values for t.

Note that the exponential term $-\frac{r_2^2\rho C_P}{4K_{rock}(t-\tau)}$ is unitless. Since $(t-\tau)$ has a unit of [y], K_{rock}'s unit of [W/m-K] must be multiplied with (3600 × 24 × 365.25) to convert the unit [W] or [J/s] into [J/y].
Below are few examples of using a calculation program to calculate $T(t)$:
For t = 0.1 y,



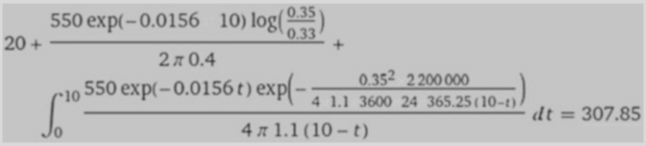
$$20+\frac{550\exp(-0.0156\ \ 0.1)\log\left(\frac{0.35}{0.33}\right)}{2\pi\,0.4}+\int_0^{0.1}\frac{550\exp(-0.0156\,t)\exp\left(-\frac{0.35^2\ 2200000}{4\ 1.1\ 3600\ 24\ 365.25\,(0.1-t)}\right)}{4\pi\,1.1\,(0.1-t)}\,dt=167.353$$

For t = 1 y



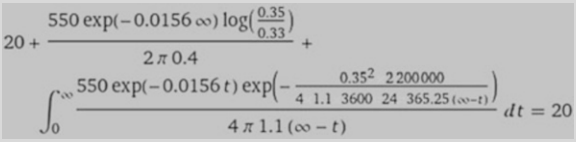
$$20+\frac{550\exp(-0.0156)\log\left(\frac{0.35}{0.33}\right)}{2\pi\,0.4}+\int_0^{1}\frac{550\exp(-0.0156\,t)\exp\left(-\frac{0.35^2\ 2200000}{4\ 1.1\ 3600\ 24\ 365.25\,(1-t)}\right)}{4\pi\,1.1\,(1-t)}\,dt=255.367$$

For t = 10y



$$20+\frac{550\exp(-0.0156\ \ 10)\log\left(\frac{0.35}{0.33}\right)}{2\pi\,0.4}+\int_0^{10}\frac{550\exp(-0.0156\,t)\exp\left(-\frac{0.35^2\ 2200000}{4\ 1.1\ 3600\ 24\ 365.25\,(10-t)}\right)}{4\pi\,1.1\,(10-t)}\,dt=307.85$$

For t = ∞ (when the rock temperature is expected to reach ambient temperature)



$$20+\frac{550\exp(-0.0156\,\infty)\log\left(\frac{0.35}{0.33}\right)}{2\pi\,0.4}+\int_0^{\infty}\frac{550\exp(-0.0156\,t)\exp\left(-\frac{0.35^2\ 2200000}{4\ 1.1\ 3600\ 24\ 365.25\,(\infty-t)}\right)}{4\pi\,1.1\,(\infty-t)}\,dt=20$$

The results of trial and error calculations are shown in the table:

t [y]	T(t) [°C]	t [y]	T(t) [°C]	t [y]	T(t) [°C]	t [y]	T(t) [°C]
0.1	167.35	6	305.47	12	306.15	50	218.18
1	255.37	7	307.14	13	304.88	100	129.17
2	279.03	8	307.96	14	303.39	500	26.22
3	291.00	9	308.15	15	301.73	1000	22.74

4	298.16	10	307.85	20	291.58	5000	20.52
5	302.65	11	307.16	30	267.21	∞	20

Thus, $\boxed{T_{max} \approx 308.15^{\circ}\mathrm{C}}$

2) Assume 1D heat flux in radial direction and steady state heat transfer with $q'' = q''_{max}$. To account for the maximum allowed temperature between the tunnels to be 96 °C,

$$q''_{\mathsf{max}} = \frac{3,300\cdot e^{-0.0156t}[W]}{2\pi r_{\mathsf{backfill}}L\ [m^2]} = k_{\mathsf{rock}}\frac{(T_{\mathsf{max}}-96)}{\Delta x}$$

At steady state, the required minimum distance between the tunnels at each time period can be found as shown below (no need to find when the maximum surface temperature of the rock is less than 96 °C):
At 10 years,

$$\Delta x = \frac{1.1(307.85-96)\cdot 2\cdot \pi \cdot 0.35\cdot 6}{3,300\cdot e^{-0.0156\cdot 10}} = 1.09[\mathrm{m}]$$

At 15 years,

$$\Delta x = \frac{1.1(301.73-96)\cdot 2\cdot \pi \cdot 0.35\cdot 6}{3,300\cdot e^{-0.0156\cdot 15}} = 1.14[\mathrm{m}]$$

t [y]	Δx [m]	t [y]	Δx [m]	t [y]	Δx [m]	t [y]	Δx [m]
0.1	0.31	6	1.01	12	1.11	50	1.17
1	0.71	7	1.04	13	1.13	100	0.69
2	0.83	8	1.06	14	1.13	500	–
3	0.90	9	1.07	15	1.14	1000	–
4	0.95	10	1.09	20	1.18	5000	–
5	0.98	11	1.10	30	1.20	∞	–

Therefore, the required minimum spacing required becomes approximately 1.2 m (i.e. the maximum value in the table above).

10.6 Status of Geological Repository Development

As shown in Table 10.8, the status of geological repository development varies from country to country. While many countries have not initiated site selection process, the most successful cases are with Finland and Sweden.

In Finland, construction of a geological repository is underway after completing the process of site selection and characterization. The site selected for HLW disposal is Olkiluoto, a granite site. The site also hosts two operating nuclear power plants. It was

Table 10.8 Summary status of major geological repository development projects

Country	Facility name	Location	Waste	Geology	Depth	Status
Finland	Onkalo	Olkiluoto	Spent fuel	Granite	400 m	Under construction (target: mid 2020 operation)
Sweden		Forsmark	Spent fuel	Granite	450 m	License review by 2020 (target: mid-late 2020 operation)
USA	Yucca Mountain	Nevada	SF/HLW	Tuff	200–300 m	Site selected in 2002, canceled 2010
Germany	Gorleben	Lower Saxony	HLW	Salt dome		Proposed, on hold
France	Cigeo	Bure	HLW	Mudstone	~500 m	Siting (pilot industrial phase since the selection of Bure site in 2006)
Switzerland			HLW	Clay		Siting process started in 2008 with investigation of 6 regions. 2 sites are to be further investigated in greater detail (anticipated start of repository no sooner than 2040).
Argentina	Sierra del Medio	Gastre	HLW	Granite		Under discussion
Belgium			HLW	Plastic clay		Formal siting process has not been initiated.
Canada			Spent fuel			Siting process initiated in 2008. 23 communities expressed interest. Based on site suitability evaluation, 9 communities are under consideration.
China						Preliminary investigation underway at Beishan in the Gobi Desert with additional sites under consideration (anticipated start of repository in roughly the 2050 timeframe).
Japan			HLW			Siting process initiated in 2002 is stalled. The government is in the process of developing a new approach.
Korea			Spent fuel	Granite		Formal siting process has not been initiated.
Spain			Spent fuel			Formal siting process has not been initiated (the implementer proposed 2063 as target for repository operation).
Taiwan			Spent fuel			Under discussion

(continued)

Table 10.8 (continued)

Country	Facility name	Location	Waste	Geology	Depth	Status
U.K.			HLW			Government established a consent-based siting process in 2008. The process was halted in 2013. Government announced a new process in 2014.

Source: NWTRB (2016)

one of five potential sites proposed in 1987 but at that time the local council firmly opposed the proposals. Since then, much effort was made by the nuclear-power company TVO, operator of the nuclear power plants at the site, through various community-engagement programs. Significant financial benefits were also presented to the local community. At the final selection phase (in 1999), the local council effectively volunteered to be the host. Expected operation of the repository is mid-2020.

Sweden is another country with a major progress in repository development. The Swedish government made initial attempts to identify potential host sites in several communities in 1980s. These attempts were stopped when citizens blockaded access roads against any site investigation attempts. A revived effort began in 1992 with emphasis on social engagement. Such reorganized effort made a difference resulting in the emergence of volunteer communities. After making pilot studies in five different communities, two sites, Oskarsham and Forsmark (both granite sites), became the candidates and were further investigated from 2002 until 2007. These investigations led to the selection of Forsmark in 2009 as the host site. A final decision for the construction of repository is expected in 2020.

Another major project of importance is the Yucca Mountain project of the U.S. The project is at a tuff rock site located inside the Nevada Test Site, where over 800 nuclear weapons tests had occurred. The site is far from major population centers (over 90 miles from Las Vegas) and features arid climate. The site was approved as the nation's geological repository for spent fuel and HLW disposal in 2002 but project has been cancelled in 2010 (as discussed in Sect. 2.2.6).

Other major development efforts of HLW geologic repositories are in France and Switzerland where active siting activities are underway. Germany had an active program in a salt dome which was suspended in 2000. Discussions are being continued for geological repository development in Argentina, Belgium, Canada, China, Japan, Korea, Spain, Taiwan, and U.K.

10.7 Conclusion

The natural geological barriers provide the ultimate medium for long-term isolation of nuclear waste. They provide protection of engineered barriers by minimizing and delaying the infiltration of groundwater into the repository systems. If any release of radionuclides takes place from nuclear waste packages, the natural barriers are to minimize the impacts of such releases by slowing down the dissolution of radionuclides, delaying their migration, and diluting their concentration in the geologic medium. While there is no such thing as a perfect rock, the candidate rocks considered for geological repository present significant merits for long-term waste isolation. The goal is to use the natural barriers along with the engineered system of geological repository to render the risk of nuclear waste into the level equivalent to (or below) natural background risk. In addition, the repository system provides a means of protecting humans against inadvertent intrusion during the service life of the repository.

In the selection of a site for a geological repository, technical features must be carefully examined through site evaluation and characterization. At the same time, consideration of non-technical factors, such as population density, socio-economics and public acceptance of the local community, and cultural significance of the site, must be given high priority. These nontechnical issues could prove to be show-stoppers in any site selection efforts. While different approaches are suggested for the site selection process, working closely with the local community with respect, credibility, and technical competence is essential for the success in geological repository development.

Homework

Problem 10.1: The following options were considered seriously for final disposal of nuclear waste in the past. Summarize the key reasons why they were not selected.

(a) Disposal in the subsea-bed
(b) Disposal in ice sheets
(c) Disposal in the space

Problem 10.2: Briefly summarize the rock cycle in relation to the presence of rock-forming minerals.

Problem 10.3: Summarize the desirable features of a site as a candidate for geological repository.

Problem 10.4: The following table gives the summary of the result of US DOE's study of five candidate sites (the rock types) for the nation's high level waste repository (US DOE 1986). The numbers represent the base-case values of performance measures for each category with possible ranges. Suggest your ways of using the data to make a recommendation on site selection.

	Richton Dome, Mississippi (Salt)	Deaf Smith, Texas (Salt)	David Canyon, Utah (Salt)	Yucca Mountain, Nevada (Tuff)	Hanford, Washington (Basalt)
Repository worker radiological fatalities	2 (<1–4)	2 (<1–4)	2 (<1–4)	4 (<1–9)	9 (2–17)

(continued)

	Richton Dome, Mississippi (Salt)	Deaf Smith, Texas (Salt)	David Canyon, Utah (Salt)	Yucca Mountain, Nevada (Tuff)	Hanford, Washington (Basalt)
Public radiological fatalities	0.7 (0.3–1.5)	0.5 (0.1–1)	<0.1 (<0.1–0.2)	<0.1 (<0.1- < 0.1)	0.7 (<0.1–1.5)
Repository worker non-radiological fatalities	27 (17–36)	29 (19–39)	27 (17–36)	18 (12–24)	43 (28–58)
Public non-radiological fatalities	0 (0–0)	0 (0–0)	0 (0–0)	0 (0–0)	0 (0–0)
Transportation worker radiological fatalities	0.52 (0–0.73)	0.64 (0–0.90)	0.73 (0–1.0)	0.81 (0–1.1)	0.9 (0–1.3)
Public radiological fatalities from transportation	2.4 (0–3.4)	2.9 (0–4.1)	3.5 (0–4.9)	4.1 (0–5.7)	4.3 (0–6.1)
Transportation worker non-radiological fatalities	1.3 (0.6–2.1)	1.6 (0.73–2.6)	2.1 (0.96–3.4)	2.5 (1.1–4.0)	2.7 (1.2–4.3)
Public non-radiological fatalities from transportation	5.3 (2.4–8.5)	6.7 (3.1–10.8)	8.4 (3.9–13.5)	10.2 (4.7–16.4)	11.0 (5–17.7)
Aesthetic impacts	4 (1–5)	4 (3–5)	6 (6–6)	4 (1–5)	1 (1–2)
Archaeological, historical and cultural impacts	0.5 (0–1)	1 (0–2.5)	3 (2.5–5)	2 (2–3.5)	0.5 (0.5–3)
Biological (ecological) impacts	2.67 (2–3.5)	2.33 (1.5–3)	3.5 (2.67–4.5)	2 (1–2.67)	2.33 (1–3.5)
Socioeconomic impacts	2 (1–3)	1.67 (1–3)	2 (1.33–3)	0.67 (0.33–2)	0.33 (0–0.67)
Repository cost (million $)	9000 (5850–12,150)	9500 (6175–12,825)	10,400 (6760–14,040)	7500 (4875–10,125)	12,900 (8385–17,415)
Transportation cost (million $)	970 (260–2040)	1120 (300–2350)	1240 (330–2600)	1400 (380–2940)	1450 (390–3040)

Problem 10.5: You are hired as a consultant to your government to provide a siting plan for the nation's HLW repository. The scope of the siting plan includes:

- How to select the site;
- How to conduct public education to support the project;
- How to develop and sustain the culture of trust between the local public and the government, for this project;
- How to bring procedural justice to the decision making process;
- How to address the issue of compensation;

Write your siting plan to the government.

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Chapter 11
Movements of Radionuclides in Groundwater

Abstract Movements of groundwater and the dissolved radionuclides therein connect geological repository with the human biosphere controlling long-term public health impacts of nuclear waste disposal. This chapter describes how groundwater transport and radionuclide migration in the porous rock medium occur subject to the natural site conditions of hydrology, geology, and geochemistry. Supporting mathematical equations are also derived and described for quantitative analysis along with the discussions of the importance of sorption and solubility of radionuclides in different geochemical environments.

Keywords Groundwater system · Hydraulic head and conductivity · Hydrodynamic dispersion · Kd (distribution coefficient) · Solubility

Success in long-term isolation of nuclear waste in a geologic disposal system largely depends on the role of groundwater in relation to potential release and migration of radionuclides from nuclear waste packages. This chapter presents a general overview of groundwater movement and contaminant transport in the underground system to provide a basis of performing safety assessment of a geological repository.

11.1 Groundwater System

11.1.1 Groundwater as Water Body in Hydrologic Cycle

Migration of radionuclides in the underground system requires presence of groundwater. Groundwater is water in rocks or sediment below the earth's surface. It is formed through the infiltration of water from the ground surface. The water on the ground surface becomes available through precipitation as rain or snow from the sky. The water falling as precipitation becomes part of streams, river, lake, or ocean but a small fraction of it infiltrates below the surface through soils and rocks to feed

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Table 11.1 Estimates of water balance (UNESCO 1971)

Water body type	Volume (km^3)	Volume (%)	Average residence time
Ocean and seas	1370×10^6	94	~ 4000 yrs
Fresh-water lakes and reservoirs	125,000	<0.01	~ 10 yrs
Swamps	3600	<0.01	1–10 yrs
River channels	1700	<0.01	~ 2 wks
Moisture in soil and the unsaturated zone	65,000	<0.01	2 wks- 1 yr
Groundwater	4–$60,000 \times 10^6$	4	2 wks – 10,000 yrs
Frozen water (icecaps and glaciers)	30×10^6	2	10–1000 yrs
Atmospheric water	13,000	<0.01	~ 10 days
Biological water	700	<0.01	~ 1 wk

groundwater. Some of this infiltrating water is lost to evaporation or taken up by vegetation and the remainder becomes a part of the groundwater system.

Groundwater represents the second largest water body accounting for 4% of the total water volume in the globe. As shown in Table 11.1, the largest water body on the earth is the oceans and seas. The water contained within the oceans and seas takes up about 94% of the total water volume. Besides the oceans and seas, groundwater accounts for about two-thirds of the remaining water resources. In terms of the total utilizable freshwater resources (i.e., excluding the water in the icecaps and glaciers), groundwater takes up 95% of the total, followed by 3.5% in lakes, swamps, reservoirs, and river channels and 1.5% in soil moisture.

Groundwater is just one element in the hydrologic cycle. The cycle ties together the processes that cause water to change state (vapor, liquid, solid) as it moves between different elements of the earth system. This indicates that ground water is a renewable resource. The hydrologic cycle (Fig. 11.1) includes evaporation, condensation, run-off, infiltration, percolation, and transpiration. The bulk of the earth's water remains in the oceans for thousands of years before being circulated through the hydrologic cycle. Most precipitation on land returns to the atmosphere by evaporation and transpiration.

Ground water is a very important resource for human living. It is used for human consumption through drinking, irrigation for agriculture, and various other public uses. In particular, the majority of water used for self-supplied domestic and livestock purposes comes from groundwater sources. In general, countries with abundant surface water resources will rely less on groundwater. In the U.S. about half of the U.S. population relies on ground water for its drinking water supply (this dependence varies among the states ranging from 2% (Montana) to 86% (Kansas)).

Although groundwater is renewable, its flow occurs slowly (at rates from meters per day to millimeters per year). Sometimes the groundwater in the underground rocks is isolated and not replaced once it is used. Radioactive materials contained in nuclear waste can become a source of groundwater contamination through their release from nuclear waste packages. The consequence of groundwater

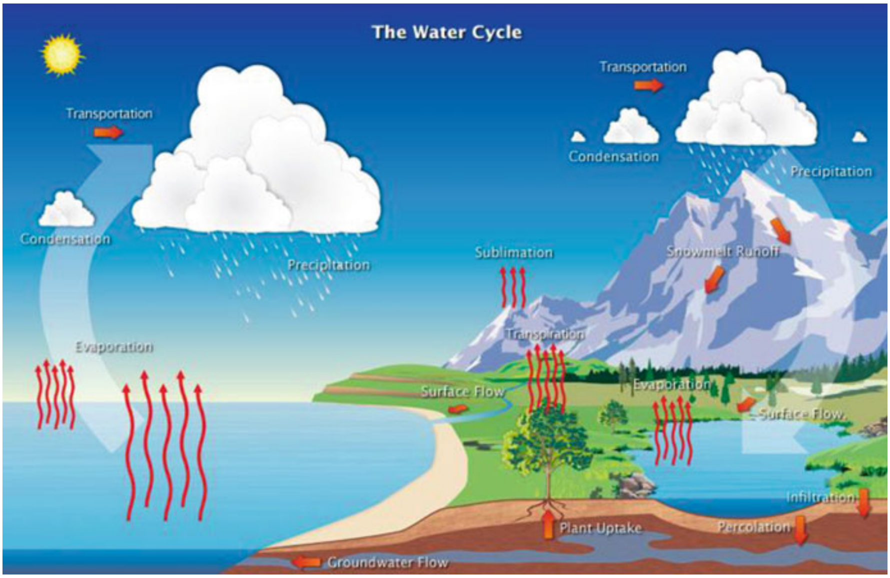


Fig. 11.1 Hydrologic cycle. (Source: NASA 2020)

contamination will depend on the type or degree of isolation of the groundwater body or the patterns of water consumption by the affected population.

11.1.2 Groundwater Systems

Groundwater is part of the underground system. The underground system is a porous medium made up of various types of soils and rocks with the pores (partially or fully) filled with groundwater. Here pores refer to the open spaces between soil grains. Depending on the level of water saturation in soil pore spaces, the underground system is divided into two zones, i.e., the unsaturated zone and the saturated zone.

The saturated zone is where the pore spaces are fully filled with water. This zone is also called the phreatic zone. The unsaturated zone, in contrast, is where the pore spaces are only partially filled with water. This zone sometimes is called the vadose zone or the zone of aeration. The boundary between the two zones is called the water table. The water table is where the pressure of water is equal to the atmospheric pressure. The pressure of water inside the unsaturated zone is lower than the atmospheric pressure and the pressure of water in the saturated zone is higher than the atmospheric pressure.

Within the saturated zone, various types of aquifers are located. Aquifers are rocks or sediment that act as storage reservoirs of groundwater and are typically characterized by high porosity and permeability. Within the saturated zone, less permeable or more or less impermeable regions also occur. These are called aquitard

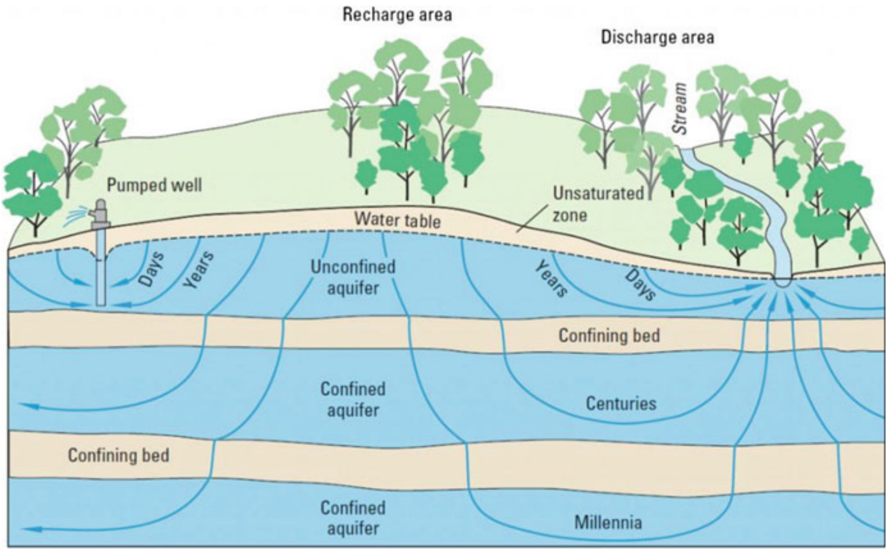


Fig. 11.2 Groundwater systems. (Source: Winter et al. 1998)

or aquiclude. Aquitard is a semi-pervious geological formation that allows some water to pass. An aquiclude is composed of a low permeability rock or sediment that essentially acts as a barrier to groundwater flow. Most geologic strata in the saturated zone are classified as either aquifers or aquitards. Very few formations fit the category of an aquiclude.

Aquifers are divided into unconfined aquifers and confined aquifers (see Fig. 11.2). An unconfined aquifer (also called an open aquifer) is an aquifer bounded by the water table as the upper boundary and a confining layer as the lower boundary. In an unconfined aquifer, water infiltrates from the surface through permeable soils and rocks or sediment in the unsaturated zone into the aquifer. A confined aquifer is an aquifer that is confined between two confining beds of rocks (such as aquitards). A confined aquifer is also called an artesian aquifer or a closed aquifer. A perched aquifer, a special case, can also be formed in the unsaturated zone. A perched aquifer is a temporary unconfined aquifer within the unsaturated zone formed during periods of heavy infiltration into soil due to the presence of a relatively low permeability layer (called a perching layer) (see Fig. 11.3). Such layer blocks downward movement of the infiltrated water forming a perched groundwater “pond” on the layer with a perched water table as upper boundary.

The water table forms the base of the unsaturated zone and the top of the saturated zone and its shape mimics that of the land surface as it is higher under hills and lower in valleys. The elevation of the water table also fluctuates with the variations in the levels of precipitation on the surface. In a humid region, the water table is located near the surface but in a dry climate region the water table is often far below the ground surface.

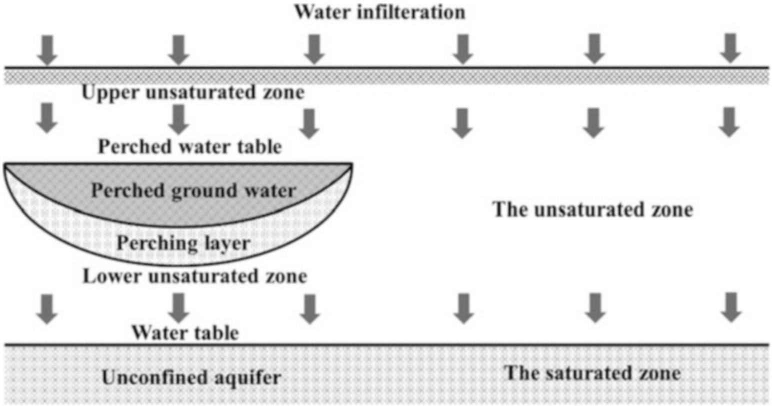


Fig. 11.3 Groundwater system terminologies

11.2 Describing Groundwater Flow

11.2.1 Hydraulic Head and Direction of Groundwater Movement

Groundwater moves because of energy. As the movement of groundwater through porous media is a mechanical process, the forces driving the movement must overcome the frictional forces between the moving fluid and the surfaces of the porous solid medium. This means that groundwater moves from the regions of higher mechanical energy to the regions of lower mechanical energy. When water percolates into the soil, it moves downward. This is because the water in the upper soil has more mechanical energy that the one in the lower soil. However, sometimes groundwater moves upward coming out to the surface at a spring. In this case, the water underneath the spring has more mechanical energy than the one in the surface.

The mechanical energy of water is a combination of potential energy (from the elevation in the presence of gravity), kinetic energy (from moving velocity) and pressure energy (from the contained pressure). Here energy refers to the ability to do work.

To explain this further, consider a movement of water mass from a standard state at elevation ($z = 0$) and at the atmospheric pressure (p_0) to some position at elevation z and fluid pressure of p. The mechanical energy of the water mass in the new position is the sum of the initial mechanical energy of the water mass at the initial position and the energy needed to move the water mass to the new position. The energy needed to move the water mass includes three components: (1) the energy needed to lift the mass from elevation zero ($z = 0$) to elevation z (elevation energy); (2) the energy needed to accelerate the fluid from velocity 0 to velocity v (kinetic energy), and; (3) the energy needed to raise the fluid pressure from P_0 to P (pressure energy). These components are expressed in mathematical terms as follows.

$$\begin{aligned} \textit{Mechanical Energ} = mgz\ (\text{elevation energy}) + \frac{mv^2}{2}(\text{kinectic energy}) \\ + m\int_{P_0}^{P}\frac{dP}{\rho}(\text{pressue energy}) \end{aligned} \tag{11.1}$$

where, mechanical energy is in [J], m is mass [kg], g is gravitational acceleration [m/s], z is new vertical position [m], v is the velocity of the fluid [m/s], P_0 is initial fluid pressure [Pa], P is final fluid pressure [Pa], and ρ is the density of the fluid [kg/m^3]. Ones in brackets are SI units, but other units may be used as long as they are on consistent basis.

Let's define fluid "*potential*" as the work done per unit mass to move the water to the new position. Here work is the outcome of expending energy (as energy is the ability to do work). Then the fluid potential is,

$$\textit{Fluid Potential} = \Phi = gz + \frac{v^2}{2} + \int_{P_0}^{P}\frac{dP}{\rho} \approx gz + \frac{P - P_0}{\rho} \tag{11.2}$$

Here, the fluid in a porous medium is assumed to be incompressible.

Here, the kinetic energy component is very small and can be ignored because groundwater moves very slowly. We can further simplify the equation by dividing fluid potential by the acceleration due to gravity. This results in a quantity called hydraulic head. The quantity "hydraulic head" is defined as follows.

$$\textit{Hydraulic head} = h = z + \frac{P - P_0}{\rho g} = z + \frac{\rho g \psi}{\rho g} = z + \psi \tag{11.3}$$

where z is the elevation head, and ψ is the pressure head. The atmospheric pressure is represented as gauge pressure (i.e., the pressure relative to atmospheric pressure). Here the term, head, is used to reflect that the quantity under consideration is a measure of length or height.

The hydraulic head can be readily measured as the sum of the elevation of the point of measurement and the pressure of water. Thus hydraulic head has two components: the elevation head, z, and the pressure head, ψ. Accordinly, we can state that groundwater moves through from a region/point of high hydraulic head to a region/point of low hydraulic head.

11.2.1.1 Hydraulic Head

As defined in Eq. 11.3, energy of groundwater is essentially the result of elevation and pressure. Groundwater moves in the porous media driven by the gradient of hydraulic head of the fluid (i.e., from a high head point/region to a low head point/region).

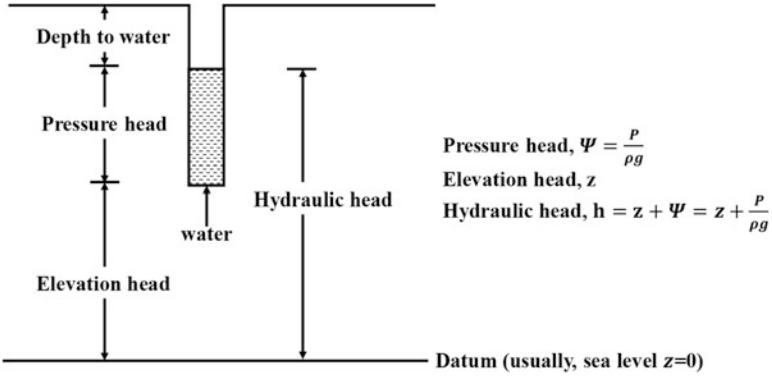


Fig. 11.4 Definition of hydraulic head

The dimensions of the head terms (h, z, and ψ) are those of length [L]. They are expressed as "meters of water" (or "feet of water"). The specification of "water" emphasizes that head measurements are dependent on fluid density shown as,

$$P = \rho g \psi \tag{11.4}$$

Hydraulic head is measured by using a device called piezometer. Piezometer is a hollow pipe or tube used to measure fluid pressure by being inserted into a saturated zone of the groundwater system. The height to which water rises in the piezometer represents the pressure head of water at the base of the piezometer, and the elevation of the point of measurement at the base of the piezometer is the elevation head. The common reference point used for the measurement of elevation is the sea level (zero elevation). An example of measurement of hydraulic head is shown in Fig. 11.4. The quantity $P/\rho g$ represents the rise of the water in a piezometer as shown in the figure.

With the distance between the piezometers specified, the hydraulic gradient dh/dl can be calculated as the difference in the measured hydraulic head divided by the distance between the piezometers. If the vertical distribution of hydraulic gradient is of interest, a piezometer nest is utilized where two or more piezometers are installed side by side at the same location at different depths (see Fig. 11.5). The simple standpipe piezometer can be replaced by more complex designs utilizing pressure transducers, pneumatic devices, and electronic components to facilitate the measurement.

Example 11.1 shows how the measurements using piezometers can be used to determine hydraulic heads, pressure heads, and elevation heads of groundwater at different points. The resulting information can be used to estimate the direction of groundwater flow. Figure 11.5 indicates that groundwater moves from left to right in the system shown by the first figure. In the system represented by the lower figure, groundwater moves upwards at the given location.

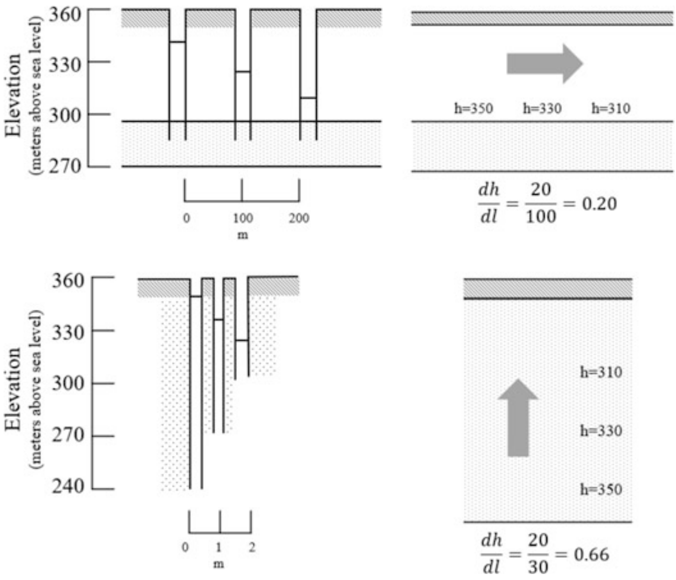


Fig. 11.5 Depiction of hydraulic gradient determination by using piezometer installations

Example 11.1: Calculation of the Hydraulic Head

At a single site, a field test using a set of piezometers (installed side by side) provided the following observations. Let P_1, P_2, and P_3 refer to the points of measurement of piezometers 1, 2, and 3.

Piezometer	1	2	3
Elevation at surface (m.a.s.l.)*	400	400	400
Depth of piezometer (m)	125	100	75
Depth to water (m)	25	40	35

* For elevation at surface, the unit m.a.s.l. stands for "metres above sea level"

Determine:

(a) The elevation head at P_1, P_2, and P_3 (m)
(b) The pressure head at P_1, P_2, and P_3 (m)
(c) The hydraulic head at P_1, P_2, and P_3 (m)
(d) The fluid pressure at P_2 (N/m^2)
(e) Direction of groundwater flow indicated by the data, and the hydraulic gradient between P_1 and P_2 and P_2 and P_3.

(continued)

Example 11.1 (continued)
Solution:

(a) Elevation head at P_1: 375–100 = 275 m
At P_2: 360–60 = 300 m
At P_3: 365–40 = 325 m

(b) Pressure head at P_1: 125–25 = 100 m
At P_2: 100–40 = 60 m
At P_3: 75–35 = 40 m

(c) Hydraulic head at P_1: 400–25 = 375 m
At P_2: 400–40 = 360 m
At P_3: 400–35 = 365 m

We can see that the values of hydraulic heads are the sum of the elevation heads and the pressure heads.

(d) For fluid pressure at B:

$$p_B = \rho g \psi_B = 1000\frac{kg}{m^3} \times \frac{9.81m}{s^2} \times 60m = 5.9 \times 10^5 \frac{kg}{m \cdot s^2} = 5.9 \times 10^5 Pa$$
$$= 0.59\ MPa$$

(e) Groundwater flows from a higher to a lower hydraulic head region. Between P_1 and P_2, it flows in direction P_1 to P_2.

Hydraulic gradient between P_1 and $P_2 = -\frac{375-360}{125-100} = -\frac{15}{25} = -0.6$
Between P_2 and P_3, groundwater flows in direction P_2 to P_3.
Hydraulic gradient between P_2 and $P_3 = -\frac{360-365}{100-75} = +\frac{5}{25} = 0.2$

One important application of hydraulic head measurements is to determine the direction of groundwater movement by installing piezometers in different locations. For such determination, the data of hydraulic head at least three different locations are needed. With three data points given, the following steps are used (see the second figure in Example 11.2 as well):

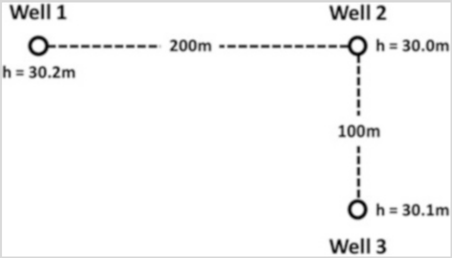
- Identify the point with the lowest head (Point 1).
- Draw a line connecting the highest head point (Point 2) with the lowest head point (Point 1) (Line 1).
- Draw another line connecting the second highest head point (Point 3) with the lowest head point (Point 1) (Line 2).

- On Line 1 (i.e. the line with the greatest head loss), determine the location where the head is expected to be the same as the second highest head measured (Point 4). Here we assume that the head gradient is linear along Line 1.
- Draw a line connecting Point 4 to the second highest head point, Point 3 (Line 3).
- Groundwater moves from Line 3 to Point 1 in a direction perpendicular to Line 3.
- Determine the hydraulic gradient.

An example showing these steps is given in Example 11.2.

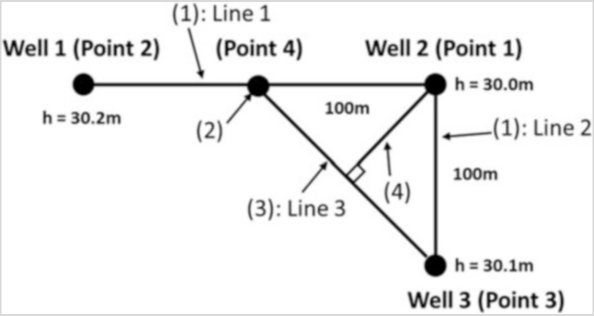
Example 11.2: Graphical Determination of Flow Direction and Hydraulic Gradient Calculation

Assume hydraulic head data from three wells as shown.



1. Draw lines connecting each well with the well with the lowest head.
2. Determine a point on the line with the greatest head loss where the head would be expected to be equal to that at the other well. (Assume a constant gradient exists along this line.)
3. Draw a line connecting this point to the other well.
4. The direction of flow will be perpendicular to this line.
5. Determine the hydraulic gradient.

Solution:



(5) From looking at line drawn in (4) in the above figure,

$\frac{dh}{dL} = \frac{30.1 - 30.0}{100/\sqrt{2}} = 0.0014$

11.2.2 Darcy's Law

As we understand why groundwater moves (and why in certain directions), the next question to consider is how to determine the rate of movement or the flow of its movement.

The movement of groundwater was first characterized in a study by Henry Darcy, a French hydraulic engineer, in 1856. He investigated the flow of water through a bed of permeable sand and found that the flow rate through porous media is proportional to the head loss and inversely proportional to the length of the flow path. This observation is called ***Darcy's law***. It serves as the basis of describing groundwater flow and is described by the following relationship.

$$Q = -KA\Delta h/\Delta L \tag{11.5}$$

where, Q is the flow rate, K is the hydraulic conductivity, A is the cross sectional area of the medium, and $\Delta h/\Delta L$ is the hydraulic gradient.

According to the Darcy's law, flow rate of groundwater is proportional to hydraulic gradient. The proportionality constant is defined as hydraulic conductivity. If the flow rate is divided by the cross sectional area of the porous medium, it becomes a velocity term and is called Darcy velocity.

The proportionality in the Darcy's law comes from the fact that a loss in the energy of groundwater per unit length of movement is due to the friction in the medium. However, such proportionality only applies to the flow in the laminar flow region (Fig. 11.6). Thus the Darcy's law is valid only with the laminar flow (the fluid with Reynolds number less than ~1).

Darcy velocity is also called specific discharge or superficial linear velocity and can be written as:

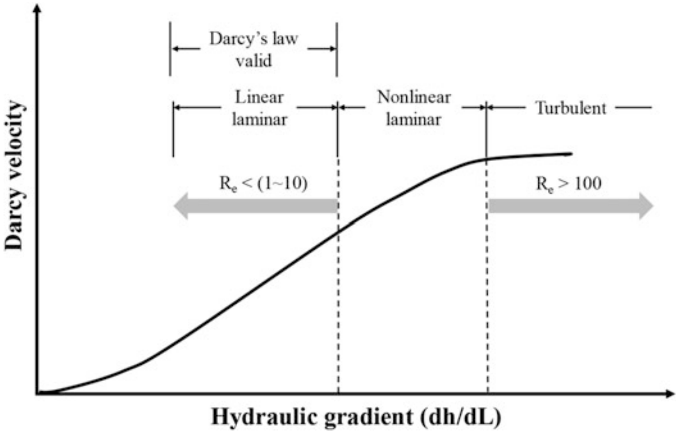


Fig. 11.6 Range of validity of Darcy's law

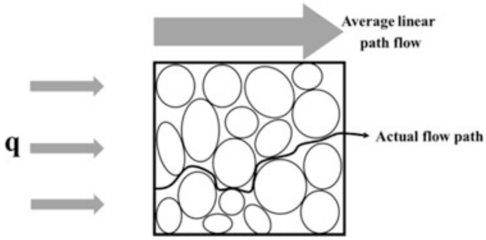


Fig. 11.7 Concepts of porosity and average linear velocity

$$v_0 = Q/A = -K\ dh/dL \tag{11.6}$$

Darcy velocity, v_0, represents the flow of groundwater per unit bulk area (including both pore spaces and the solid areas) per unit time. As the full cross sectional area of the porous medium is not available for flow (as shown in Fig. 11.7), Darcy velocity does not represent a physical reality. An alternative term called 'average linear velocity' is used as a more physically observable quantity. This average linear velocity, as a macroscopic quantity, represents the linear rate of groundwater movement in a defined direction. The average linear velocity is obtained by dividing Darcy velocity by the porosity of the medium as follows.

$$v = v_0/\varepsilon \tag{11.7}$$

The porosity (ε) is the volume of void divided by the total bulk volume. Thus average linear velocity takes into account the fact that water cannot pass through the solids. It is also called average pore velocity or seepage velocity.

Please note that the average linear velocity is still not an actual velocity of groundwater in the porous medium. As depicted in the following figure (Fig. 11.7), actual movements of groundwater in the pore spaces are multidirectional and at a rate that is much higher than what is represented by the average linear velocity. We can see that groundwater movements take place in the very tortuous paths of the pore spaces through interactions with the solid surfaces.

11.2.3 Hydraulic Conductivity

As explained, hydraulic conductivity is the proportionality constant in the Darcy's law. It represents a measure of the ease of water movement through a porous medium. Therefore, it can be expected that hydraulic conductivity depends on both the properties of the fluid (such as density and viscosity) and the characteristics of the medium (such as the size of the pores). It turns out that hydraulic conductivity is proportional to the permeability of the medium, the density of the fluid, and the gravity and is inversely proportional to the viscosity of the fluid. The following relationship captures this.

$$K = k\rho g/\mu \tag{11.8}$$

where k is the permeability of the porous medium with unit in [L^2], ρ is the fluid density, and μ is the fluid viscosity. The permeability is the average size of the pore spaces available in the medium for flow and is sometimes called the intrinsic permeability. The size of pore spaces varies depending on the type of soil texture (or classification) as discussed in the next subsection.

One of the units of permeability is darcy. 1 darcy is defined as the permeability of a medium that transmits fluid of a viscosity of 1 centipoise (1 gm/s-cm) with a specific discharge of 1 cm^3/cm^2-s in a hydraulic gradient of 1 atm/cm. This corresponds to 9.8717×10^{-9} cm^2.

As shown below, the unit of hydraulic conductivity is equal to that of velocity:

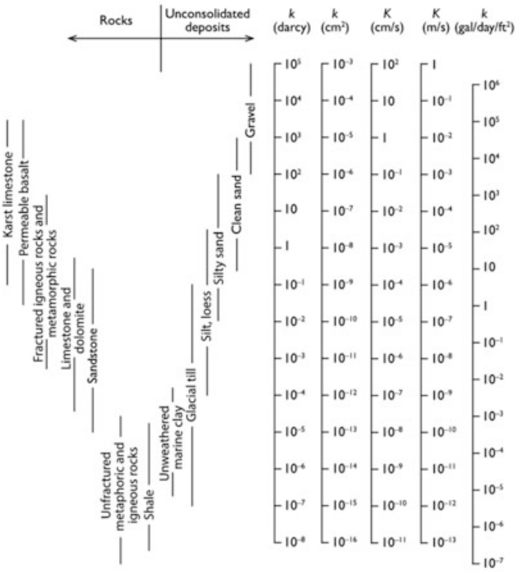
$$K = k\left[cm^2\right] \cdot \rho\left[\frac{g}{cm^3}\right] \cdot g\left[\frac{cm}{s^2}\right] / \mu\left[\frac{g}{s - cm}\right] \rightarrow K\ in\ \left[\frac{cm}{s}\right] \tag{11.9}$$

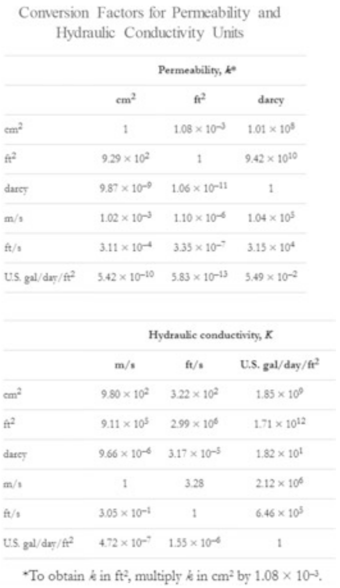
Representative values of hydraulic conductivity for various types of rocks are shown in Table 11.2. They are also captured in Fig. 11.8. As seen in the figure, hydraulic conductivity varies widely depending upon the type of rocks or soils. Even within the same type of rocks or soils, hydraulic conductivity varies within three or four orders of magnitude. This indicates the difficulty in characterizing the values of hydraulic conductivity at a site and the importance of addressing variability and uncertainty in the use of hydraulic conductivity. Here variability refers to natural variations of the quantity and uncertainty refers to potential errors caused in the measurements or due to lack of understanding.

Table 11.2 Representative Values of Hydraulic Conductivity (Note: 1 cm/s = 1×10^{-2} m/s = 1.04×10^3 darcys)

	Hydraulic Conductivity (m/s)
Unconsolidated sedimentary deposits	
Gravel	$3.0 \times 10^{-4} \sim 3 \times 10^{-2}$
Coarse sand	$9 \times 10^{-7} \sim 6 \times 10^{-3}$
Medium sand	$9 \times 10^{-7} \sim 5 \times 10^{-4}$
Fine sand	$2 \times 10^{-7} \sim \times 2 \times 10^{-4}$
Silt, loess	$1 \times 10^{-9} \sim 2 \times 10^{-5}$
Clay	$1 \times 10^{-11} \sim 4.7 \times 10^{-9}$
Sedimentary and crystalline rocks	
Karst and reef limestone	$1 \times 10^{-6} \sim 2 \times 10^{-2}$
Limestone, dolomite	$1 \times 10^{-9} \sim 6 \times 10^{-6}$
Sandstone	$3 \times 10^{-10} \sim 6 \times 10^{-6}$
Siltstone	$1 \times 10^{-11} \sim 1.4 \times 10^{-8}$
Basalt	$2 \times 10^{-11} \sim 2 \times 10^{-2}$
Fractured crystalline rock	$8 \times 10^{-9} \sim 3 \times 10^{-4}$
Weathered granite	$3.3 \times 10^{-6} \sim 5.2 \times 10^{-5}$
Unfractured crystalline rock	$3 \times 10^{-14} \sim 2 \times 10^{-10}$

Source: https://www.enviro.wiki/index.php?title=File:Newell-Article_1-Table1r.jpg, accessed on February 12, 2020




Conversion Factors for Permeability and Hydraulic Conductivity Units

	Permeability, k*		
	cm²	ft²	darcy
cm²	1	1.08×10^{-3}	1.01×10^{8}
ft²	9.29×10^{2}	1	9.42×10^{10}
darcy	9.87×10^{-9}	1.06×10^{-11}	1
m/s	1.02×10^{-3}	1.10×10^{-6}	1.04×10^{5}
ft/s	3.11×10^{-4}	3.35×10^{-7}	3.15×10^{4}
U.S. gal/day/ft²	5.42×10^{-10}	5.83×10^{-13}	5.49×10^{-2}

	Hydraulic conductivity, K		
	m/s	ft/s	U.S. gal/day/ft²
cm²	9.80×10^{2}	3.22×10^{2}	1.85×10^{9}
ft²	9.11×10^{5}	2.99×10^{6}	1.71×10^{12}
darcy	9.66×10^{-6}	3.17×10^{-5}	1.82×10^{1}
m/s	1	3.28	2.12×10^{6}
ft/s	3.05×10^{-1}	1	6.46×10^{5}
U.S. gal/day/ft²	4.72×10^{-7}	1.55×10^{-6}	1

*To obtain k in ft², multiply k in cm² by 1.08×10^{-3}.

Fig. 11.8 Range of values of hydraulic conductivity and permeability & conversion factors. (Source: Freeze and Cherry 1979)

11.2.4 *Physical Properties of Soil*

As groundwater moves through soils and rocks, its movement is affected by the physical properties of the soil. Such properties may be different in different types of soil. As a porous mixture of rocks or mineral particles, soil is classified by the size of the solid mass or particles contained.

The mixture of solid mass of soil includes boulder, cobbles/pebbles, gravel, sand, silt, clay, and even colloids. Boulders are large pieces of rock with the size larger than 25–30 cm. If such rock breaks into small fragments in the size ranging between 15 and 25 cm, they are called cobbles or pebbles. Smaller rock fragments in the range between 5 mm and 15 cm are called gravel. From about 5 mm down to 0.05 mm, the rock particles are called sand (USDA 1938). Sand is also divided into coarse sand (5 to 3 mm) and fine sand (<1 mm). Rock particles from about 0.005 down to 0.002 mm are then called silt. The smallest soil particles are called clay (with diameters of less than 0.002 mm). This reaches the size range of mineral particles.

Soil classification is based on relative proportions of sand, silt, or clay in a soil with its name following the primary constituent particle size or a combination of the most abundant particles sizes (e.g. sandy clay or silty clay). Another term, loam, is used to describe a roughly equal concentration of sand, silt, and clay. In the U.S., 12 soil texture classifications are used by the U.S. Department of Agriculture. Figure 11.9 shows this classification as so-called the soil texture triangle diagram.

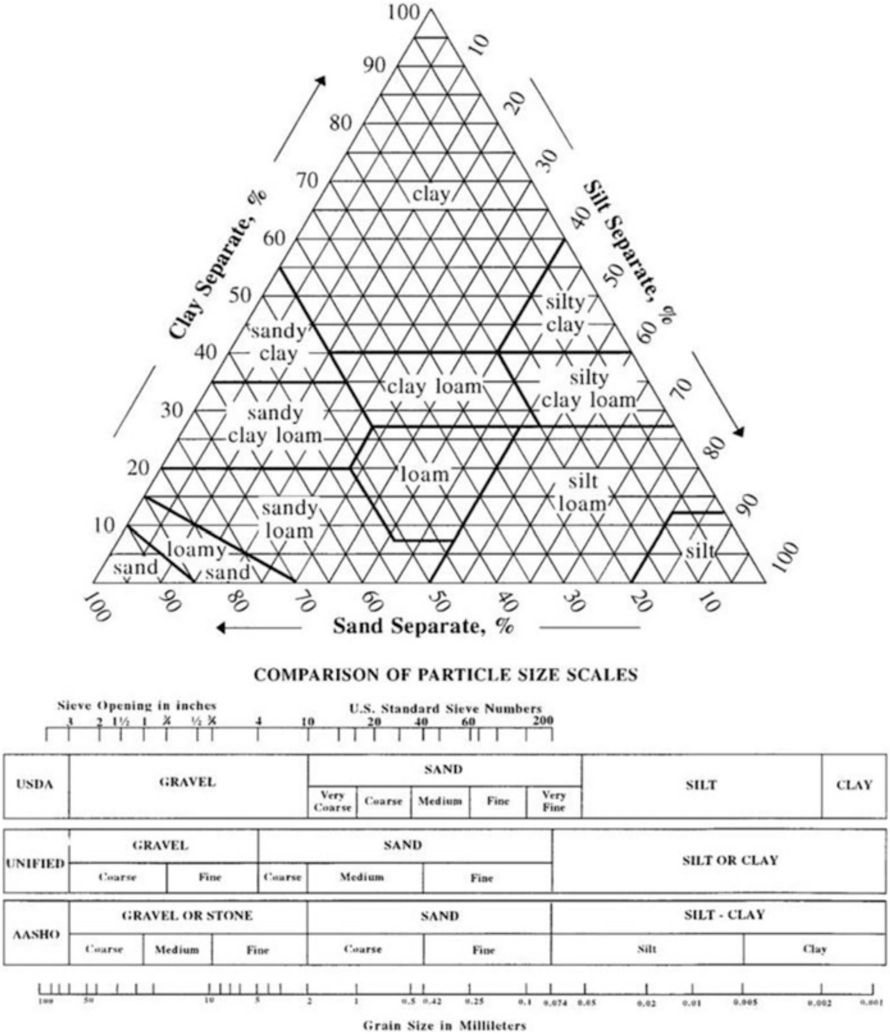


Fig. 11.9 Soil texture triangle (source: Wikimedia Commons 1993)

The exact scale of particle size distribution used for soil classifications may vary in different countries.

The values of the permeability of soil in gravel, sand, silty sand, silt, and clay are provided in Fig. 11.9.

As groundwater moves through the void spaces in a porous medium, description of the flow requires quantification of the levels of void in the medium that contributes to the movement of groundwater. For these purposes, the terms, porosity and effective porosity, are defined. Porosity is the relative volume of voids in the soil compared to the total soil volume (including the void and solid volume; the void can be occupied by water or air) as shown,

$$Porosity\ (\varepsilon) = \frac{V_{void}}{V_T} = \frac{V_T - V_S}{V_T} = \frac{V_w + V_a}{V_T} \qquad (11.10)$$

where, ε is the porosity, V_{void} is the volume of void, V_T is the total volume, V_S is the volume of the solid portion, V_w is the volume of water, and V_a is the volume of the air.

Among the void spaces, some are not accessible by water and do not actually contribute to groundwater movement. Such void spaces includes the very small size pores (e.g., in a clay medium). This means using porosity defined as above is not appropriate to support the description of groundwater movements. This leads to the definition of effective porosity. Effective porosity is the total connected void volume actually contributing to groundwater flow divided by the total bulk volume.

$$Effective\ porosity\ \left(\varepsilon_f\right) = \frac{V_{void} - V_{ivoid}}{V_T} = \frac{V_{cvoid}}{V_T} \qquad (11.11)$$

where, ε_f is the effective porosity, V_{ivoid} is the volume of void which is isolated from the groundwater flow, and V_{cvoid} is the volume of the connected void allowing water flow. Therefore, effective porosity is always smaller than total porosity (i.e. $\varepsilon_f < \varepsilon$).

Porosity of a porous medium depends on the degree of packing of the solids (called grains), their shape, size distributions, and arrangements. A medium with non-uniform grain size distribution will have a smaller porosity than one with uniform ones as small grains will fit into the openings left between the grains of larger size. A medium with uniform grain size has high porosity as the packing efficiency of the medium is low. An example of this is clay. Clay has relatively uniform grain size while the size of the grains is small. Therefore, clay has high porosity (~42%) but its effective porosity is much lower (~6%). This is because the sizes of the pores within the clay medium are very small making water transmission in clay very difficult. Effective porosity can have large influence on hydraulic conductivity of a medium.

The typical values of porosity and effective porosity of aquifer materials are listed in Table 11.3. As expected, while effective porosity is always smaller than porosity, the medium with smaller grain sizes show larger differences between the two quantities. Also rocks in general have lower porosities than soils and poorly sorted rock deposits have lower porosities than well-sorted ones.

Example 11.3: Determination of Groundwater Travel Time

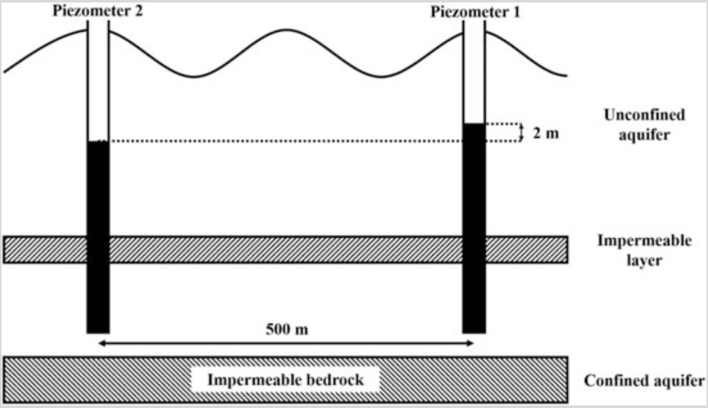
For the confined aquifer depicted in the following figure, determine the time required for groundwater to move from the location of the monitoring well to the location of drinking well. The distance between two wells is 500 meter. The difference in hydraulic head between the two well locations is measured to be 2 meter. Assume the confined aquifer is a sandstone aquifer.

(continued)

Table 11.3 Typical values of porosity and effective porosity of aquifer materials

	Total Porosity (mean)	Effective Porosity (mean)
Unconsolidated sedimentary deposits		
Gravel	0.25 ~ 0.44 (0.31)	0.13 ~ 0.44 (0.25)
Coarse sand	0.31 ~ 0.46 (0.39)	0.18 ~ 0.43 (0.34)
Medium sand	(0.40)	0.16 ~ 0.46 (0.30)
Fine sand	0.25 ~ 0.53 (0.40)	0.01 ~ 0.46 (0.28)
Silt, loess	0.35 ~ 0.50 (0.36)	0.01 ~ 0.39 (0.20)
Clay	0.40 ~ 0.70 (0.47)	0.01 ~ 0.18 (0.06)
Sedimentary and crystalline rocks		
Karst and reef limestone	0.05 ~ 0.50 (0.30)	0.00 ~ 0.36 (0.14)
Limestone, dolomite	0.00 ~ 0.20	0.01 ~ 0.24
Sandstone	0.14 ~ 0.49 (0.34)	0.02 ~ 0.41 (0.24)
Siltstone	0.21 ~ 0.41	0.01 ~ 0.33 (0.12)
Basalt	0.05 ~ 0.50	–
Fractured crystalline rock	0.00 ~ 0.10	–
Weathered granite	0.34 ~ 0.57	–
Unfractured crystalline rock	0.00 ~ 0.05	–
Tuff	0.04 ~ 0.49	0.02 ~ 0.47 (0.21)

Example 11.3 (continued)



Solutions:

For a sandstone aquifer, the average value of the effective porosity is 0.24 (Table 11.3) and the hydraulic conductivity varies between 3×10^{-10} and 6×10^{-6} (m/s) (Table 11.2). To give a conservative estimate of water movement, let's assume the highest hydraulic conductivity value (6×10^{-6} m/s)

(continued)

Example 11.3 (continued)

The average linear velocity of groundwater in the aquifer $= -\frac{K}{\varepsilon_f}\frac{\Delta h}{\Delta L} = -\frac{6\times10^{-6}\left(\frac{m}{s}\right)}{0.24}\frac{-2(m)}{500\ (m)} = 10^{-7}\left(\frac{m}{s}\right)$

Groundwater travel time $= \frac{500(m)}{10^{-7}\left(\frac{m}{s}\right)} = 5\times10^{9}\ (s) = 158.6$ year

If we assume the lowest value of the hydraulic conductivity, 3×10^{-10} (m/s), the resulting travel time will be much longer (by a factor of 20,000), i.e., 3.17×10^{6} (year). This example indicates the uncertainty in the estimation of groundwater travel time and the importance of charactering hydraulic conductivity in an aquifer system.

Another parameter of importance related to the concept of porosity is moisture content. Moisture content (θ) is the fraction of the volume occupied by water in the soil and is defined as,

$$\theta = \frac{V_W}{V_T} \tag{11.12}$$

where, V_T is the total volume of a soil or rock and V_W is the volume of the water within the given soil unit.

Another quantity of interest in describing water flow in porous medium is the bulk density of soil. Note that the total mass of solid in a porous medium is determined as the product of the density and the volume of solid. The resulting mass should also be the same per bulk volume basis. Therefore,

$$\rho_s V_s = \rho_b V_b \rightarrow \rho_b = \frac{\rho_s V_s}{V_b} = \frac{\rho_s(V_b - V_v)}{V_b} = (1-\varepsilon)\rho_s \tag{11.14}$$

where ε is the porosity and ρ_s *and* ρ_b are the density of the solid phase and the bulk volume of the soil, respectively. V_b, V_s, *and* V_v are the volume of the soil bulk mass, the solid, and the void, respectively.

The value of bulk density of soil is typically 2.65 g/cm^3 for most sands and soils.

11.2.5 Hydraulic Head Mapping Using Field Measurements

To describe the movement of groundwater in a region, the distributions of hydraulic head in the region must be known. Such distributions can be obtained in two ways, i.e., by field measurements using piezometers or by solving mathematical models called groundwater flow equation.

If a large number of piezometers are installed throughout the three dimensional hydrogeologic system, hydraulic heads of the system can be obtained and contoured on a two-dimensional map. Each individual data point represents a measure of the hydraulic head at the given location in the aquifer. Lines can then be constructed by connecting the points of equal hydraulic heads. These are called equipotential lines. Equipotential lines with incremental changes (decrease) in hydraulic head can be drawn on a map. The resulting x-y plane map is referred to as a potentiometric map. Figure 11.10 shows an example of a potentiometric map. This particular map is for an area near Milwaukee, Wisconsin in the U.S.

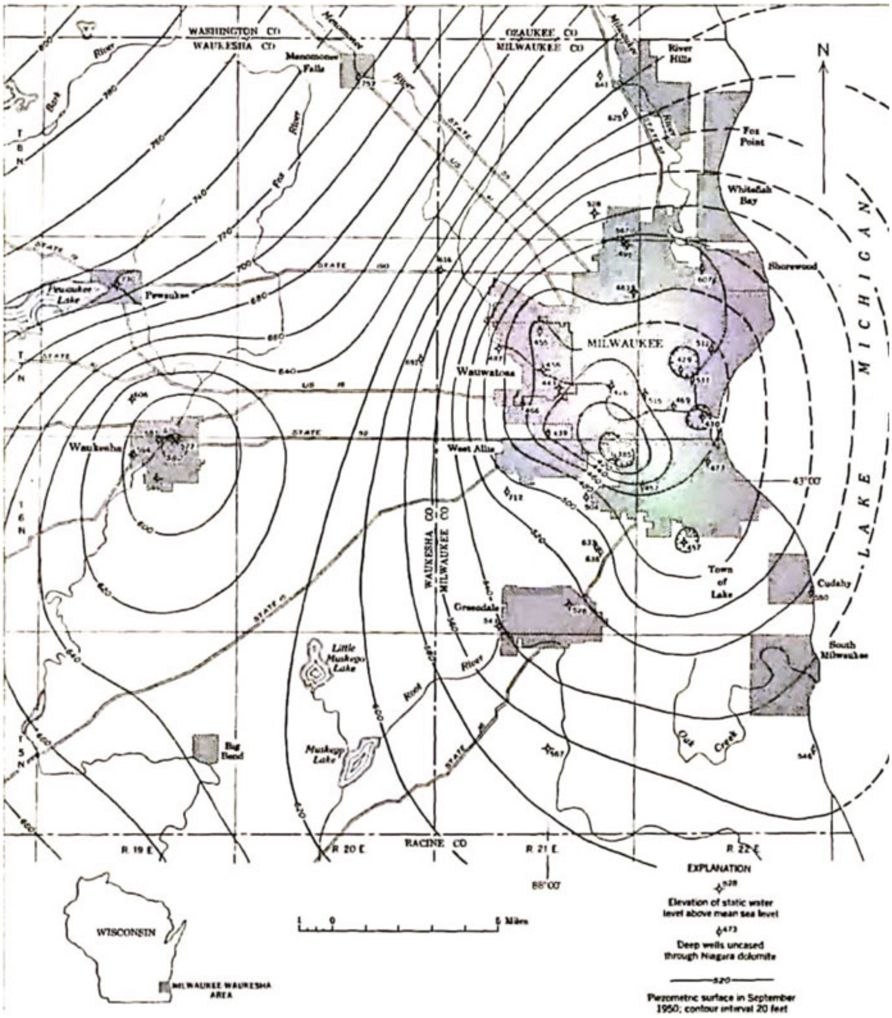


Fig. 11.10 An example of potentiometric map in Milwaukee, Wisconsin, USA

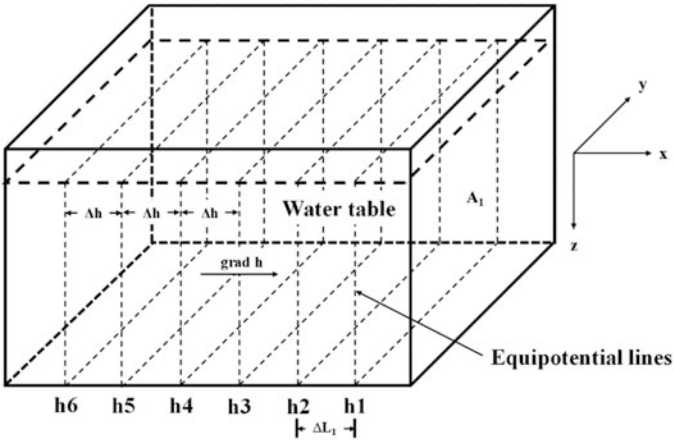


Fig. 11.11 Cross section and horizontal projection of lines connecting points of equal head

The potentiometric map is a projection of vertical equipotential planes onto the horizontal plane as depicted in Fig. 11.11. The surface depicted in the x-y plane in the figure is a potentiometric surface representing the projection of vertical equipotentials on the x-y plane. In this case, it is assumed that the hydraulic head variation in the vertical direction can be ignored and that the flow in the aquifer moves horizontally.

On the potentiometric map, the head losses between the adjacent equipotential lines are the same. When no gains or losses occur in the total flow, the volumetric flow rate passing through one equipotential line is the same as what passes through the other equipotential lines downgradient. This means (following the same definitions in Eq. 11.5) that

$$Q_1 = Q_2 \rightarrow K_1 A_1 \frac{\Delta h}{\Delta L_1} = K_2 A_2 \frac{\Delta h}{\Delta L_2} \tag{11.15}$$

where the subscript number represents different locations shown in the figure. As Δh is the same between the equipotential lines, the following relationship is valid (by assuming a uniformly thick aquifer, $A_1 = A_2$).

$$\frac{K_1}{K_2} = \frac{\Delta L_1}{\Delta L_2} \tag{11.16}$$

Therefore, the ratio of the hydraulic conductivities equals the ratios of the distance between the equipotential lines.

11.2.6 Estimating Hydraulic Head Distributions Using Groundwater Flow Equation

An alternative to field measurements is to use mathematical models based on the fundamental laws of physics of groundwater. Such an approach requires solving the equation for the groundwater flow in the region under consideration.

The equation for groundwater flow is based on a mass-balance in a control volume. The net change in the mass of groundwater (i.e., accumulation or loss of material) in a control volume is equal to the change in the mass of groundwater with time in the control volume. This is captured in the following equation.

$$-\left[\frac{\partial q_x}{\partial x}+\frac{\partial q_y}{\partial y}+\frac{\partial q_z}{\partial z}\right]\rho_w dxdydz=(\alpha\rho_w g+\varepsilon\beta\rho_w g)\rho_w dxdydz\frac{\partial h}{\partial t} \tag{11.17}$$

where, q_x, q_y, q_z are flow per unit cross-sectional area in x, y, and z direction, respectively, ρ_w is the fluid density, and $(\alpha\rho_w g + \varepsilon\beta\rho_w g)$ is called the specific storage, S_s. Here ρ_w is the density of the water (M/L^3), g is the acceleration of gravity (L/T^2), α is the compressibility of the aquifer (1/(M/LT2), ε is the porosity (L^3/L^3), and β is the compressibility of the water (1/(M/LT^2)) (here, M is of mass, L is of length, and T is of time).

The specific storage (S_s) is the amount of water per unit volume of a saturated formation that is stored or expelled from storage per unit change in hydraulic head due to the compressibility of the porous medium. This is also called elastic storage coefficient. This implies that when the head in a confining unit changes, water will either be stored or expelled. The specific storage has the dimension of 1/L.

By using $q_i=-K_i\frac{\partial h}{\partial i}$ $(i = x,\ y,\ or\ z)$, Eq. 11.17 yields the main equation of groundwater flow in three dimensions as follows.

$$\frac{\partial}{\partial x}\left(K_x\frac{\partial h}{\partial x}\right)+\frac{\partial}{\partial y}\left(K_y\frac{\partial h}{\partial y}\right)+\frac{\partial}{\partial z}\left(K_z\frac{\partial h}{\partial z}\right)=S_s\frac{\partial h}{\partial t} \tag{11.18}$$

The Eq. 11.18 is valid for a confined aquifer and can also be valid for an unconfined aquifer if the term in the right-hand side is set to be zero (i.e., if no water is stored or expelled when the head changes).

If the hydraulic conductivity in the aquifer is assumed a constant (i.e., does not change with location or direction), the equation can be modified as,

$$K\left(\frac{\partial^2 h}{\partial x}+\frac{\partial^2 h}{\partial y}+\frac{\partial^2 h}{\partial z}\right)=S_s\frac{\partial h}{\partial t} \tag{11.19}$$

If the flow is at steady state, then there is no change in head with time and the equation becomes,

$$\frac{\partial^2 h}{\partial x} + \frac{\partial^2 h}{\partial y} + \frac{\partial^2 h}{\partial z} = 0 \tag{11.20}$$

This equation represents the steady state flow of groundwater in a homogeneous and isotropic aquifer. Here, a homogeneous aquifer means the hydraulic conductivity in the system remains constant, independent of position and an isotropic aquifer indicates that the hydraulic conductivity is constant in different directions, regardless of the direction of measurement in the aquifer system (these terms are explained further in the next section).

If the flow is not steady state, the volume of water that a permeable unit absorbs or expels from storage (per unit surface area per unit change in hydraulic head) can be defined as the storage coefficient, or storativity (S). The storage coefficient is a dimensionless quantity. For a confined aquifer, the storativity is the product of the specific storage (S_s) and the aquifer thickness (b): $S = bS_s$. The value of specific storage is very small, generally ~0.0003 m^{-1} (0.0001 $\mathrm{ft.}^{-1}$) or less.

Using this definition of storativity, the Eq. 11.19 (for a confined aquifer) becomes,

$$\frac{\partial^2 h}{\partial x} + \frac{\partial^2 h}{\partial y} + \frac{\partial^2 h}{\partial z} = \frac{S_s}{K}\frac{\partial h}{\partial t} = \frac{S}{Kb}\frac{\partial h}{\partial t} = \frac{S}{T}\frac{\partial h}{\partial t} \tag{11.21}$$

where, T is defined as transmissivity (L^2/T) as the product of the hydraulic conductivity and the thickness of the aquifer ($T = Kb$). Transmissivity is a measure of the amount of water that can be transmitted horizontally through a unit width of the aquifer under a unit hydraulic gradient.

The equation for groundwater flow is solved by specifying the initial and boundary conditions for the given aquifer. If the hydraulic conductivity in the aquifer is a constant and the boundaries can be described with algebraic equations, then the equation can be solved analytically (i.e., by using an analytical solution). If the hydraulic conductivity in the aquifer is not a constant, then a numerical solution approach is needed. Results are the distributions of hydraulic heads in the aquifer.

11.2.7 Homogeneity and Isotropy of Aquifer

The homogeneity or isotropy of aquifer is defined depending upon how the hydraulic conductivity K of an aquifer system changes in different positions or directions.

If the hydraulic conductivity within a geologic formation remains constant, independent of position, the formation is called homogeneous. If the hydraulic conductivity within a geologic formation varies at different positions, the formation is called heterogeneous. This is illustrated in Fig. 11.12. A good example of heterogeneity is a layered rock system. In the layered system, the individual beds

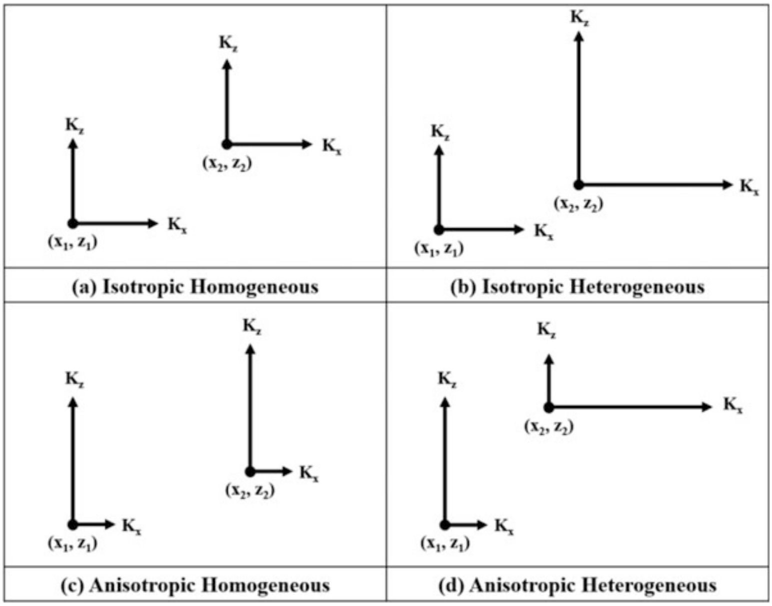


Fig. 11.12 Depiction of homogeneity, heterogeneity, isotropy, and anisotropy of a geologic formation

making up the formation can be homogeneous but the entire system as combination of layers is heterogeneous.

If the hydraulic conductivity remains constant in different directions at a point (being independent of the direction of measurement), the formation is called isotropic at that point. If the hydraulic conductivity changes with the change in the direction of measurement at a point, the formation is called anisotropic at that point. This is again illustrated in Fig. 11.12. The anisotropy of an aquifer is caused by rock structures and the orientation of rock-forming minerals.

In the case of an isotropic aquifer, the groundwater moves in a direction perpendicular to the equipotential lines, crossing the equipotential lines at right angles. If anisotropic, the flow crosses the equipotential lines at an angle dictated by the degree of anisotropy. Discussions in this chapter assume that the aquifer system under consideration is isotropic.

Consider a layered system of rock formations as shown in Fig. 11.13 as an example. Assume that each layer is homogeneous and isotropic with the values of hydraulic conductivity of each i_{th} layer as K_1, K_2, . . .and K_n.

If we assume that the groundwater flows vertically, i.e., perpendicular to the layering, the Darcy velocity of water entering the top layer must be equal to that leaving the bottom layer. With Δh_i representing the head loss across the i^{th} layer, the total head loss in the system is the sum of the head losses in each layer, i.e. $\Delta h = \Delta h_1 + \Delta h_2 + \ldots + \Delta h_n$.

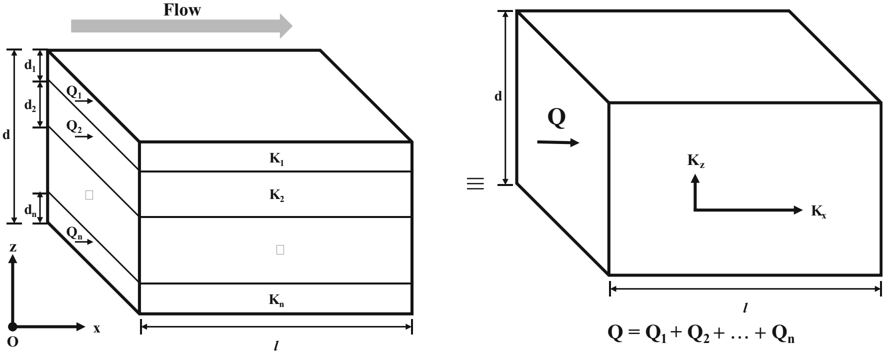


Fig. 11.13 An example of a layered rock system

According to Darcy's law,

$$v = K_1 \frac{\Delta h_1}{d_1} = K_2 \frac{\Delta h_2}{d_2} = \ldots = K_n \frac{\Delta h_n}{d_n} = K_z \frac{\Delta h}{d} \tag{11.22}$$

where K_z is an equivalent vertical hydraulic conductivity for the layered system, v is the Darcy velocity, and d_i is the depth of each i_{th} layer. Solving the relationship for K_z and replacing Δh_i with vd_i/K_i,

$$\begin{aligned} K_z &= \frac{vd}{\Delta h} = \frac{vd}{\Delta h_1 + \Delta h_2 + \ldots + \Delta h_n} \\ &= \frac{vd}{vd_1/K_1 + vd_2/K_2 + \ldots + vd_n/K_n} \end{aligned} \tag{11.23}$$

Therefore,

$$K_z = \frac{d}{\sum\limits_{i=1}^{n} d_i/K_i} \tag{11.24}$$

This K_z is the equivalent vertical hydraulic conductivity of the layered system. Note this is the harmonic mean (inverse average) of the hydraulic conductivities of each layer.

If we assume that the groundwater flows horizontally, parallel to the layering, then the total flow through the entire layered system is the sum of the flows in each layer. Then the flow through the system, due to the head loss Δh over a horizontal distance l (assuming a unit thickness for the flow area), is given by

$$\begin{aligned} Q &= Q_1 + Q_2 + \ldots + Q_n = K_1 d_1 \frac{\Delta h}{l} + K_2 d_2 \frac{\Delta h}{l} + \ldots + K_n d_n \frac{\Delta h}{l} \\ &= K_x d \frac{\Delta h}{l} \end{aligned} \tag{11.25}$$

where, K_x is an equivalent horizontal hydraulic conductivity of the layered system.

Then,

$$v = K_x \frac{\Delta h}{l} = \sum_{i=1}^{n} \frac{K_i d_i}{d} \frac{\Delta h}{l} \tag{11.26}$$

This leads to,

$$K_x = \sum_{i=1}^{n} \frac{K_i d_i}{d} \tag{11.27}$$

This shows that the equivalent horizontal hydraulic conductivity of a layered rock system is the arithmetic mean of the hydraulic conductivities of each layer.

For all possible sets of values of K_1, K_2, ...,K_n, the equivalent horizontal hydraulic conductivity of a layered rock system is always greater than the equivalent vertical hydraulic conductivity. An example of this is given below. As shown the example (Example 11.4), the horizontal flow in a layered rock system is dominated by the most permeable rock units. For a vertical flow, the least permeable units play the dominate role in controlling the flow in the system.

Example 11.4: Hydraulic Conductivity

Consider a 100-m thick layered rock system as a sequence of interbedded sandstone and shale. About 70% of the layer is sandstone. The sandstone has a horizontal and vertical hydraulic conductivity of about 10^{-3} cm/s while the shale has a horizontal and vertical hydraulic conductivity of about 1.9×10^{-10} cm/s.

Find the equivalent horizontal and vertical hydraulic conductivity of the layered system.

<u>*Solution:*</u>

The equivalent horizontal hydraulic conductivity of the layered system, K_x, is,

$$K_x = \sum_{i=1}^{n} \frac{K_i d_i}{d} = \frac{10^{-3} \frac{cm}{s} \cdot 70\ m + 1.9 \times 10^{-10} \frac{cm}{s} \cdot 30m}{100m} = 7.0 \times 10^{-4} \frac{cm}{s}$$

(continued)

Example 11.4 (continued)

The equivalent vertical conductivity of the layered system, K_z, is,

$$K_z = \frac{d}{\sum_{i=1}^{n} d_i/K_i} = \frac{100m}{\left(70m/10^{-3}\frac{cm}{s}\right) + \left(30m/1.9 \times 10^{-10}\frac{cm}{s}\right)}$$
$$= 6.3 \times 10^{-10}\frac{cm}{s}$$

In this case, the horizontal flow is six orders of magnitude faster than the vertical flow.

11.2.8 Flow Lines and Flow Nets

We saw an example of a potentiometric map in Fig. 11.10 where equipotential lines of a groundwater system are given. As the groundwater moves in a direction perpendicular to equipotential lines, the lines that are perpendicular to the equipotential lines (in the direction of the maximum potential gradient) can also be drawn. These lines are called flowlines. A flow line is an imaginary line of groundwater movement in an aquifer. It is also an imaginary line that traces the path of a particle in the groundwater in an aquifer. The resulting set of intersecting equipotential lines and flowlines as a map of contour lines is known as a flow net.

A flow net is a graphical representation of two-dimensional groundwater flow through an aquifer under steady state. Flow nets are useful to determine various quantities of interest including the travel time of groundwater, the total volumetric flow, and the distributions of transmissivity, in the case of isotropic medium. Figure 11.14 shows an example of a flow net showing a network of multiple equipotential lines along with the associated flow lines.

A flow net is composed of a number of stream tubes where each stream tube is an area between two adjacent flow lines. Each stream tube is also composed of a

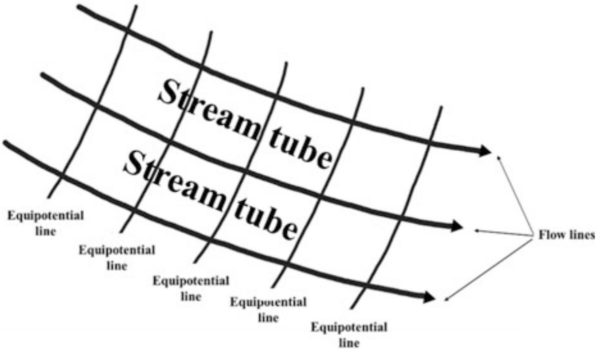


Fig. 11.14 An example of flow net and stream tubes

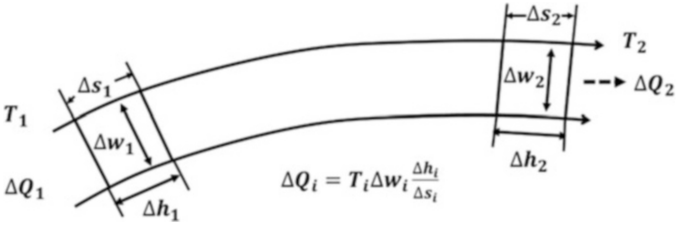


Fig. 11.15 Use of a stream tube to illustrate the calculation of transmissivity

number of "square (or near-squares)" figures as shown in Fig. 11.14. The total number of "(near) squares" in a stream tube represents total number of pressure drops while the groundwater moves over the distance represented by the flow line. The flow rate is constant along a stream tube (as discussed in Sect. 11.2.7). Moreover, if the flow lines are equally spaced, the flow of groundwater through each streamtube is the same.

To illustrate the use of the flow net/stream tube concept in the analysis of groundwater movement in an aquifer, consider a stream tube depicted in Fig. 11.15. The figure shows two-dimensional horizontal flow with no vertical components. Here, we also use the concept of transmissivity *T*, as discussed in association with Eq. 11.21 (note, $T = Kb$, where K = hydraulic conductivity, b = thickness of the aquifer).

11.2.8.1 Calculation of Transmissivity Distributions in a Stream Tube

Consider a stream tube shown in the following figure.

For the flow within the given stream tube, the flowrate within the first "square" (resulting in the first pressure drop Δh_1) can be written as,

$$\Delta Q_1 = T_1 \Delta w_1 \frac{\Delta h_1}{\Delta s_1} \tag{11.28}$$

where, Δw_1 is the width of the stream tube and Δs_1 is the distance between the equipotential lines represented in the square.

As$\Delta Q_1 = \Delta Q_2$ in the given stream tube,

$$T_1 \Delta w_1 \frac{\Delta h_1}{\Delta s_1} = T_2 \Delta w_2 \frac{\Delta h_2}{\Delta s_2} \tag{11.29}$$

If we use a uniform contour where the head loss in each square node, Δh_i 's are the same, then the transmissivity in the square node*, i*, can be given by

$$T_i = T_1 \frac{(\Delta s/\Delta w)_i}{(\Delta s/\Delta w)_1} \tag{11.30}$$

Fig. 11.16 An illustration of travel time calculation using a flow line

11.2.8.2 Calculation of Travel Time

Consider a flow line shown in a stream tube shown below (Fig. 11.16).

According to the Darcy's law,

$$(v_0)_i = \frac{T_i}{b}\frac{\Delta h_i}{\Delta s_i} \tag{11.31}$$

Then the average linear velocity of the groundwater on the flow line is,

$$v_i \equiv \frac{\Delta s_i}{\Delta t_i} = \left(\frac{v_0}{\varepsilon_f}\right)_i = \frac{T_i}{b\varepsilon_f}\frac{\Delta h_i}{\Delta s_i} \tag{11.32}$$

By solving this for Δt_i, we obtain $\Delta t_i = \frac{b\varepsilon_f}{T_i}\frac{(\Delta s_i)^2}{\Delta h_i}$. Therefore, the total travel time within the entire stream tube is

$$t = \sum_i \Delta t_i = \sum_i \frac{\varepsilon_f b}{T_i}\frac{(\Delta s_i)^2}{\Delta h_i} \tag{11.33}$$

11.2.8.3 Calculation of Flow in a Flow Net

The total flow of groundwater in a flow net (the volume of water flow per unit width of aquifer) can be determined as the sum of the flow rates in all stream tubes.

$$Q = \sum_j^N \Delta Q_j = \sum_j^N T_j \frac{\Delta h}{\Delta s_j}\Delta w_j = \Delta h \sum_j^N T_j \frac{\Delta w_j}{\Delta s_j} \tag{11.34}$$

where, N is the total number of connected stream tubes in the flow net, Δs is the distance for the equipotential drop, Δw is the width of the streamtube, and T_j is the transmissivity of each stream tube, j. If T_j is constant, the total flow is,

$$Q = T\Delta h \sum_j^N \frac{\Delta w_j}{\Delta s_j} \approx T\Delta h \cdot N \frac{\Delta w_j}{\Delta s_j} \tag{11.35}$$

In most groundwater conditions, the width of a stream tube is equal to the distance for equipotential drop ($\Delta w_j \cong \Delta s_j$). Then, the total flow can be calculated by a simple expression.

$$Q = T\Delta h \cdot N = T \cdot \frac{M}{H} \cdot N \tag{11.36}$$

where, M is the number of equipotential drops, H is the total head loss over the length of the streamlines, and N is the number of connected stream tubes in the flow net.

Example 11.5: Calculation of Travel Time in the Streamtube

For the potentiometric map shown in Fig. 11.10, sketch the flow line which originates at the circled 800 (ft) in the upper-left corner of the map. The properties of the underlying aquifer are:

Transmissivity T = 20,000 gpd/ft. (1 gallon = 0.1337 ft^3)

Thickness = 750 ft.

Effective porosity = 0.15

Determine the travel time of groundwater from the 800 ft. contour to reach the 600 ft. using the data in the table below (Δs_i of each interval i is also estimated as given). Note that unit conversion between ft. and m is not necessary in this example.

Interval (Δh)	Distance (miles)	Distance (ft) (1 mile = 5280 ft)
800–780	2.4	12,672
780–760	2.0	10,560
760–740	2.0	10,560
740–720	1.4	7392
720–700	1.2	6336
700–680	1.1	5808
680–660	1.0	5280
660–640	0.8	4224
640–620	0.7	3696
620–600	0.8	4224

Solution:

Using the equation $t = \sum_i \Delta t_i = \sum_i \frac{\varepsilon_f b}{T_i} \frac{(\Delta s_i)^2}{\Delta h_i}$ (from Eq. 11.33)

$$\mathrm{T} = 20000\,\frac{\mathrm{gpd}}{\mathrm{ft}}\left(\frac{0.1337 ft^3}{1 gal}\right) = 2673.6\,\frac{ft^3}{day - ft}$$

(continued)

Example 11.5 (continued)

$$t = \frac{\varepsilon_f b}{T\Delta h_i}\sum_i (\Delta s_i)^2$$

$$= \frac{0.15 \cdot 750ft}{2673.6\frac{ft^3}{day-ft} \cdot 20ft}$$

$$\times \left[12672^2 + 10560^2 + 10560^2 + 7392^2 + 6336^2 + 5808^2 + 5280^2 + 4224^2 + 3696^2 + 4224^2\right] ft^2$$

$$t = 1{,}239{,}930 \ days = 3{,}397 \ years$$

11.2.9 Groundwater Flow in Fractured Rock

As discussed in Chap. 10, igneous rocks like granite or basalt are one of the likely candidates of geological formations for nuclear waste repository. In these igneous rocks, presence of fractures are very much the characteristics of the rock body. These fractures will have significant influence on the movement of groundwater in the rock body. In contrast, fractures typically play little role in determining the flow regime in sedimentary rocks, another candidate for geological repositories. This is because sedimentary rocks often have high interconnected porosity and presence of fractures does not add much to the overall hydraulic conductivity of the system. Some sedimentary rocks like salt flow plastically performing self-sealing of fractures thus minimizing the effect of fractures.

The overall permeability of the igneous rock bodies is largely determined by the presence of fracture network. Fractures also provide preferred pathways for groundwater movement in these rocks as the fastest flowing water will move through the fractures rather than the voids in the bulk body of the porous rock.

How to analyze the flow of groundwater in fractured rocks depends on the characteristics of the fracture formations. If the distribution of the fractures in the rocks is uniformly dense and random on the scale of the model, the groundwater flow can be modeled by using so-called the continuum approach. In this case, the fractured media acts in a hydraulically similar manner to granular porous media. If the fractures are far apart and sparsely located, the groundwater flow is described in relation to individual fractures or fracture sets. In this case, the discontinuum (discrete) modeling approach is used.

The continuum approach is a simplified method in which groundwater movement in the fractured system is described by Darcy's law. There are two different classes of models in the continuum approach, equivalent porous medium model or dual-porosity model.

In the equivalent porous medium model, the fractured rock is represented by the average hydraulic and transport properties representing the effects of fractures on groundwater movement over the entire porous region of interest. The standard flow and transport models for porous medium are applied in this case.

The dual-porosity model notes the differences in mass transport between the fractures and the nonfractured rock matrix. The model assumes that mass transport is mainly through the combination of advection in the fractures and dispersion in the nonfractured matrix. The standard transport equations are applied to both regions but with the use of very different values of hydraulic conductivities, porosities, and transport parameters. The porosity assigned to the fracture system is much smaller than the one in the nonfractured matrix but the flow velocities are much higher in the fractures than in the matrix (Diodato 1994).

The discontinuum approach is a detailed complicated analysis for the fracture flow. An example is a discrete fracture modeling. In this case, only fracture provides paths for groundwater movement and the rock matrix is treated as impermeable to groundwater. In the approach, multiple fractures in a given geologic domain are treated as an interconnected network of fractures. The flow and transport along a single fracture are explicitly modeled by using the information on orientation and aperture of individual fractures. If the apertures are large, the flow may become turbulent which prevents the use of Darcy's law. Then the fluid mechanics principles embodied in the Navier-Stokes equations need to be employed in the analysis. The interactions between the contaminants and the rock matrix is incorporated in the contaminant transport model by describing dispersion into or out of the matrix through the use of a sink and source term. In this approach, characterizing individual discrete fractures is a major challenge.

An example of deriving the value of equivalent hydraulic conductivity for a fractured rock using the continuum approach is as follows.

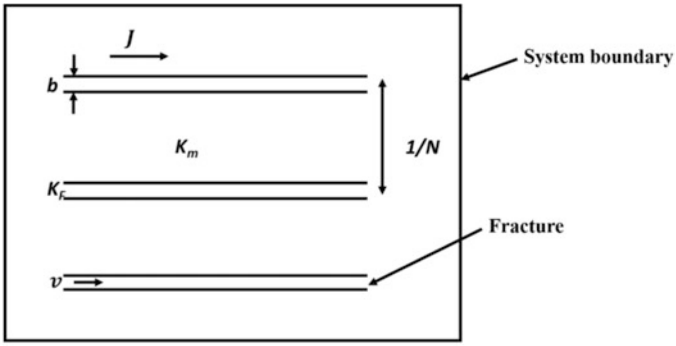
Assume a fractured rock system depicted in Fig. 11.17.

In this example, the fractures are uniformly spaced with each fracture having the aperture value, b. The number of fractures per unit distance across the face of the rock is denoted as N. The hydraulic conductivity of the porous rock is defined independent of the fractures and is denoted as K_m. The hydraulic conductivity of water in the fracture, denoted as K_F, is defined as,

$$K_F = \frac{\rho g b^2}{12\mu} \tag{11.37}$$

where, ρ is the bulk density, g is gravity, b is the aperture or opening size (L), and μ is the viscosity of water.

Recalling the discussion for the equivalent horizontal hydraulic conductivity in the layered rock system, the equivalent hydraulic conductivity for horizontal flow in the system, K_E, is given as,



$$\vec{v} = \frac{1}{12}\frac{\rho g b^2}{\mu}\vec{J} \equiv K_F \vec{J} \qquad K_F \equiv \frac{1}{12}\frac{\rho g b^2}{\mu}$$

Fig. 11.17 An example of fractured rock to illustrate the calculation of equivalent hydraulic conductivity

$$K_E = \frac{K_F b + K_m/N}{b + 1/N} = K_F b N \approx \frac{\rho g b^3 N}{12\mu} \qquad (11.38)$$

This relationship is sometimes called *the cubic law:* For a given hydraulic gradient, the flow through a fractured medium is proportional to the cube of the aperture of the fracture. The equivalent hydraulic conductivity of the system is also proportional to the hydraulic conductivity of water in the fracture with bN as proportionality constant.

Example 11.6: Equivalent Hydraulic Conductivity

For the rock system with fractures as shown in Fig. 11.17, determine the value of equivalent hydraulic conductivity for horizontal flow.

Given: $K_m = 10^{-11}$ (m/s), b = 10^{-5} (m). N = 1 (m^{-1})

<u>*Solution:*</u>

$$K_F = \frac{\rho g b^2}{12\mu} = \frac{0.998204\left(\frac{g}{cm^3}\right)\times 980.62\left(\frac{cm}{s^2}\right)\times\left(10^{-3}\right)^2\left(cm^2\right)}{12\times 0.01002\left(\frac{g}{cm-s}\right)} = 8.14 \times 10^{-3}$$

(cm/s) = 8.14×10^{-5} (m/s)

$$K_E = K_F b N = 8.14 \times 10^{-5}\, m/s \cdot 10^{-5}\, m \cdot 1 m^{-1} = 8.14 \times 10^{-10}\, [m/s]$$

This result indicates that the equivalent hydraulic conductivity of a fractured rock system is strongly affected by the size of fracture opening. As stated, for a given hydraulic gradient, flow of groundwater through a fractured medium is proportional to the cube of the fracture aperture.

Although the hydraulic conductivity of a fractured rock system can be measured by using lab tests on small size samples, the results are not valid as the samples are not representative. Rock would have to be very large to be representative for the purpose. At the same time, it is not possible to identify and characterize all of the discrete fractures in a large rock mass. This is due to natural variability in the system. Specifying the hydraulic conductivity of a fractured rock system remains an area of on-going research.

11.2.10 Groundwater Flow in the Unsaturated Zone

Most of the proposed HLW geologic repositories are located in the saturated rock system and discussions in the previous sections were focused on the behavior of groundwater in the saturated zone. One exception is the Yucca Mountain repository in the U.S. which is located in the unsaturated zone. Selection of Yucca Mountain as the repository site resulted in an increased in the studies of groundwater flow in the unsaturated zone. Understanding groundwater movement in the unsaturated zone is also important and necessary to describe infiltration of water from the surface into the repository at any repository sites.

11.2.10.1 Physical and Hydrological Properties of the Unsaturated Zone

The unsaturated zone is the soil region below the surface of the earth above the water table. The water table is the dividing boundary between the saturated and the unsaturated zone. On the water table, the water pressure in the pores of a porous medium is equal to the atmospheric pressure. Thus the pressure head is zero (i.e. $\psi = 0$, measured as gauge pressure) and the hydraulic head is equal to the elevation head ($h = z$).

The water in the unsaturated zone exists as soil moisture. This water as soil moisture is not sufficient to completely fill the voids and partially surrounds the soil grains. The remainder of the voids is taken up by air. The degree of (partial) water saturation in the soil is defined as moisture content. Moisture content varies depending upon the porosity of the soil, the distance from the water table, and the level of water precipitation at the surface. The moisture content is always less than the porosity of the porous medium (i.e. $\theta < \varepsilon$) in the unsaturated zone. .

Figure 11.18 shows the moisture content variations in different soils as a function of the distance above the water table. This curve is called *moisture retention curve*. The moisture retention curve is the characteristic curve of the soil controlled by capillary rise and pore size distribution. For the soil with relatively fine particles, the moisture content usually decreases rather abruptly and significantly above capillary fringe. This abrupt behavior is observed in the figure for clay and fine-grained sand. The minimum value of moisture content observed has a special name, the specific retention. Specific retention is the water-holding capacity of a porous soil.

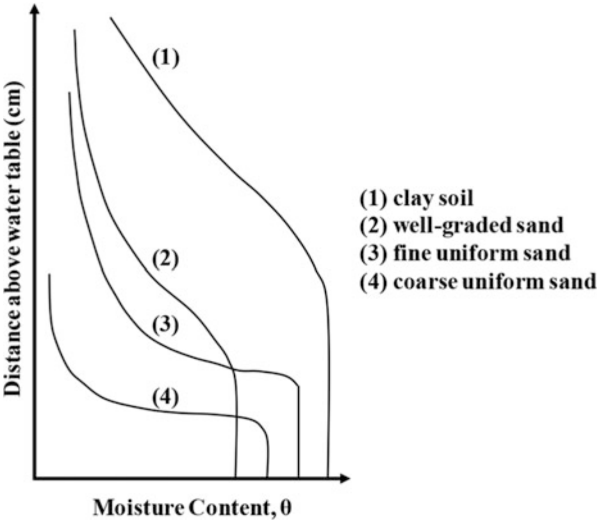


Fig. 11.18 Moisture retention curve

Capillary rise, also called capillary fringe, is a region where the water saturation in the pores are maintained above the water table due to surface tension-induced capillary rise. This is also called tension saturated zone and shown in Fig. 11.19a. How high will the capillary fringe rise depends on the permeability of the soil. The capillary fringe will be high for well-graded fine-particle soil (e.g., clay soil) but low for a coarse-grained large particle soil (e.g., coarse sand) as noted in Fig. 11.18. The pressure heads within the capillary fringe are still less than the atmospheric pressure.

In the unsaturated zone, the pressure head is always negative (i.e. $\psi < 0$) and is also called the tension head or suction head. Therefore, the hydraulic head is the elevation head minus the pressure head. Figure 11.19 shows, as examples, changes in the moisture content (Fig. 11.19b) and the pressure head (Fig. 11.19d) away from the water table in the unsaturated zone. The resulting changes in hydraulic heads are also shown in the figure (Fig. 11.19e).

How to measure the pressure head in the unsaturated zone is illustrated in Fig. 11.19c. In the unsaturated zone, piezometers are no longer useful as there will be no water level rise in the pipe due to the negative water pressure. To measure the negative pressure in this case, a device based on the concept of suction is used. Such device is called a tensiometer. As shown in the figure, water is already filled inside a tensiometer with closure at the top and with an air-tight porous membrane at the bottom. When the tensiometer is inserted into the soil at the desired depth, the air-tight porous membrane allows the passage of water from the tensiometer to the soil due to the presence of the negative pressure in the soil. Then the vacuum created at the top of the airtight tube inside the tensiometer after the removal of water from the tube provides a measure of the pressure head in the soil. Therefore, the tensiometer effectively takes advantage of the different degrees of suction in the soil for pressure head measurement.

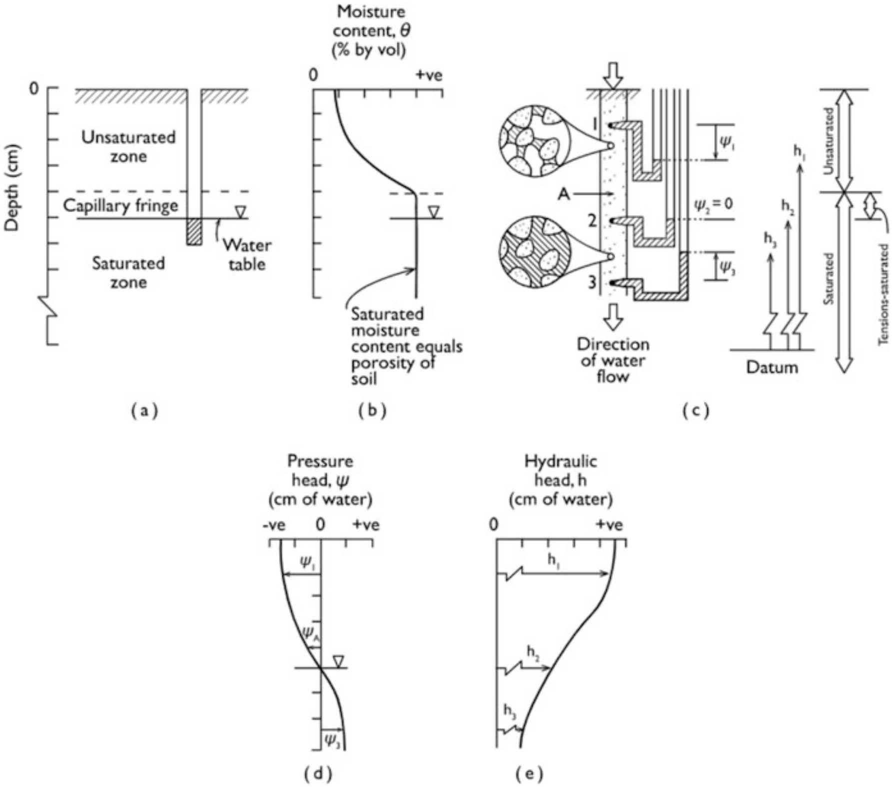


Fig. 11.19 Groundwater conditions in the unsaturated zone. (Source: Freeze and Cherry 1979)

The hydraulic conductivity, in the unsaturated zone, varies as a function of the pressure head and the moisture content. Also, the pressure head varies depending upon the distance from the water table. The variations in the negative pressure head and the moisture content with distance above the water table depend on the characteristics of the soil material (see Fig. 11.20).

There is another variable affecting the description of groundwater flow in the unsaturated zone. This is the wetting and drying cycle of the soil. Depending upon whether the moisture content is the result of wetting or drying in the soil, the pattern of moisture content variation in the unsaturated zone is different. Under the process of wetting, movement of water occupying the pore spaces is limited due to the presence of air entrapped. The entrapped air remains in the soil pores for some time after the soil has been wetted. However, under the process of drying, the blockage effect by the entrapped air disappears resulting in higher value of moisture content. This effect is called the hysteresis effect.

These variations in moisture content under wetting or drying also affect the observed pressure head and the resulting hydraulic conductivity behaviors. These

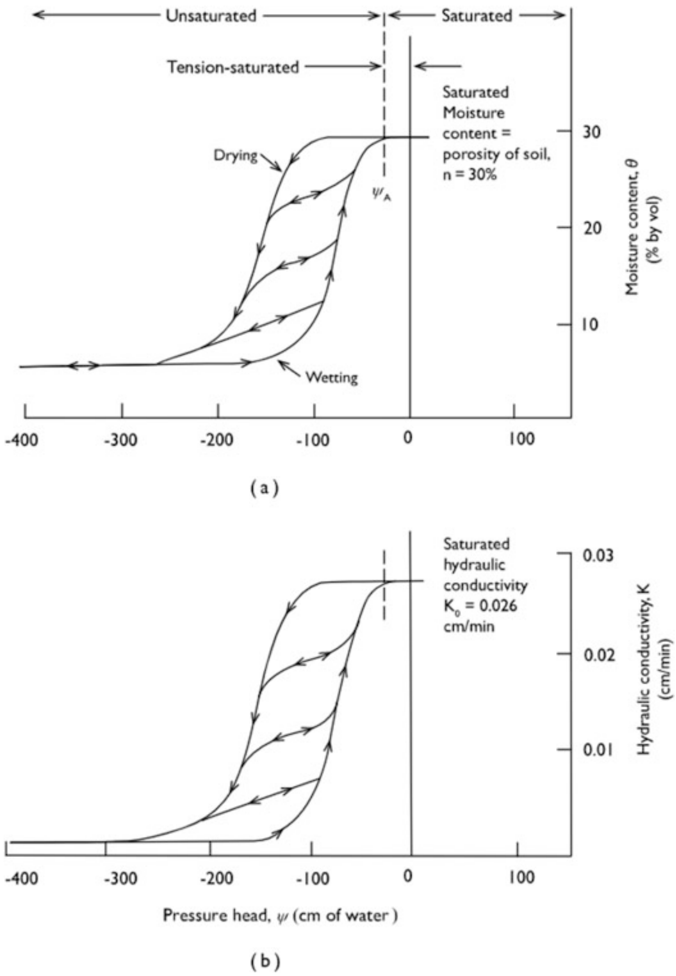


Fig. 11.20 Characteristic curves relating hydraulic conductivity and moisture content to pressure head. (Source: Freeze and Cherry 1979)

variant behaviors are also captured in Fig. 11.20. The internal lines in the figure show the course to be followed if the soil were only partially wetted and dried, or vice versa. As shown in the figure, moisture content, pressure head, and hydraulic conductivity are all interrelated in the soil. The relationships are nonlinear and complex.

The figure also shows that the moisture content θ and hydraulic conductivity are at the maximum at the water table and decreases as the distance from the water table increases. The pattern of decrease is dependent on pressure head ψ and also whether the process is under wetting or drying. Both the moisture content and hydraulic conductivity are higher under the drying cycle. In contrast to the unsaturated zone, the hydraulic conductivity in the saturated zone has a constant value, not a function of the pressure head ψ ..

The fact that pressure head, moisture content, and hydraulic conductivity all vary depending on the soil type and the distance from the water table make the description of groundwater flow in the unsaturated zone challenging.

11.2.10.2 Modeling Groundwater Flow in the Unsaturated Zone

Typically, groundwater in the unsaturated zone moves downward vertically. Once the water reaches the saturated zone, it moves mostly horizontally. Movement of groundwater both in the unsaturated zone and the saturated zone is described by the Darcy's law (as long as the flow is laminar): Driven by hydraulic gradient in a linear fashion with hydraulic conductivity as the proportionality constant.

Unlike the saturated zone, water movement in the unsaturated zone occurs only through water-filled spaces of the pores and the fraction of the pore spaces filled with water is highly variable. This presents challenge in describing groundwater flow in the unsaturated zone. Unlike in the saturated zone, hydraulic conductivity is not a constant but varies as a function of pressure head or moisture content. The following equation represents Darcy's law for the unsaturated flow.

$$v_0 = -K(\psi)\frac{dh}{dz} \tag{11.39}$$

Difficulties in characterizing the moisture content and the hydraulic conductivity in the unsaturated zone were noted in the discussions for Fig. 11.20. While the gradient of pressure head (i.e., the suction) dictates direction of water movement, the pressure head distribution (ψ) is highly variable in the unsaturated zone. Also, although the distribution of pressure heads can be readily measured, the effect of hysteresis in representing $K(\psi)$ is quite large. As an alternative to using the hydraulic conductivity as a function of pressure head, (ψ), the hydraulic conductivity can be represented as a function of moisture content, i.e., $K(\theta)$:

$$v_0 = -K(\theta)\frac{dh}{dz} \tag{11.40}$$

This reduces the hysteric effect as $K(\theta)$ is less affected by hysteresis than the $K(\psi)$ function. Problem in this case is the difficulty in measuring moisture content.

11.2.10.3 Steady Infiltration Case

While the flow of groundwater in the unsaturated zone is highly variable, let us be reminded that the eventual goal of groundwater analysis is to project potential human health impact from the migration of radionuclides. The most important health

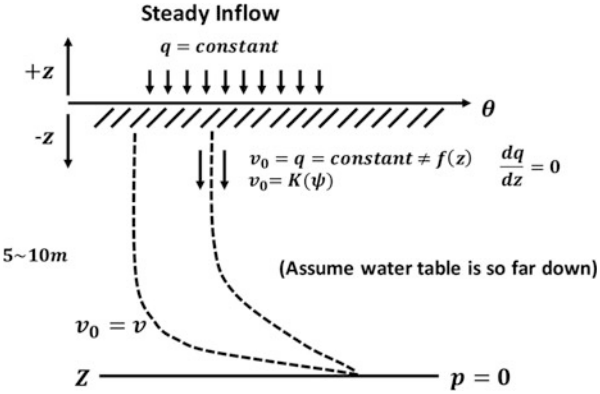


Fig. 11.21 Depiction of the steady infiltration case

impact of concern in this regard is cancer. As cancer is an outcome of a long-term cumulative process, the purpose of describing unsaturated flow for nuclear waste disposal is not so much on its time-dependent behavior but on the average behavior. Therefore, capturing only the long-term average behavior of unsaturated flow may be appropriate for the purpose of the analysis.

In this regard, the case of steady infiltration is considered to model unsaturated zone flow as a conservative case. The steady infiltration case assumes that groundwater flows vertically downward in the unsaturated zone with steady infiltration flow. This is captured in Fig. 11.21.

Then Darcy's law states,

$$v_0 = -K(\psi)\frac{dh}{dz} = -K(\psi)\left(1 - \frac{d\psi}{dz}\right) \tag{11.41}$$

where $h = z - \psi$.

Since the infiltration is steady, we can assume that moisture content also remains constant. Thus the pressure head remains also a constant (i.e., $d\psi/dz = 0$). Then Eq. (11.41) becomes,

$$v_0 = -K(\psi) = -K(\theta) \tag{11.42}$$

In this case, the average rate of movement of groundwater through the unsaturated soil (i.e., the average linear velocity) becomes:

$$v = v_0/\theta = -K(\theta)/\theta \tag{11.43}$$

Where, θ is moisture content of the soil medium. This rate can be used to determine the time of travel for unsaturated flow to reach the water table. The resulting travel time can be considered a time delay (due to the presence of the unsaturated zone) for the infiltrating water to reach the saturated zone.

If the time-dependent description of groundwater flow in the unsaturated zone is necessary, use of the following Richards equation is necessary. The equation represents the combination of the Darcy equation with the continuity equation.

$$\frac{\partial \theta}{\partial t} = -\frac{\partial}{\partial x}\left(K\frac{\partial \psi}{\partial x}\right) - \frac{\partial}{\partial y}\left(K\frac{\partial \psi}{\partial y}\right) - \frac{\partial}{\partial z}\left(K\frac{\partial \psi}{\partial z}\right) + \frac{\partial K}{\partial z} \tag{11.44}$$

11.2.10.4 Approximate Approaches to Quantify Hydraulic Conductivity and Moisture Content

Given the difficulty in describing hydraulic conductivity (K) and moisture content (s) as functions of pressure head in the unsaturated zone, approximate approaches have been suggested. These approaches use more readily determined parameters such as porosity and grain size to characterize K and s in a soil. One of the most widely used approaches is explained below (Meyer 1993).

Soil Moisture Contents

The volumetric water content (moisture content, s) of the unsaturated zone is the product of the saturated water content and the saturation ratio of the unsaturated zone. The saturated water content is the water content when the soil material is saturated hence is equal to the total porosity of the soil material. The saturation ratio is the ratio of the moisture content to the saturated water content. When the medium is saturated, the saturation ratio equals unity. Under unsaturated infiltration conditions, the saturation ratio, s, is a function of the infiltration rate, the saturated hydraulic conductivity, and the texture of the soil. It can be estimated by using the following equation.

$$s = \theta_r + (1 - \theta_r)\left(\frac{p}{K_{sat}}\right)^{0.25} \tag{11.45}$$

where θ_r is the residual water content, p is the infiltration rate (m/year), and K_{sat} is the saturated hydraulic conductivity (m/year).

Hydraulic Conductivity

The hydraulic conductivity in the unsaturated zone can be approximated by the following equation:

Table 11.4 Parameters of hypothetical disposal facility materials. (Source: Meyer 1993)

Material type	Residual water content, θ_r	Saturated water content, θ_s	α, van Genuchten parameter	n, van Genuchten parameter	K_{sat}, (cm/s)
Top soil	0.10	0.47	0.0440	1.523	1.0e-4
Upper gravelly sand	0.02	0.32	0.1008	2.922	1.0e-2
Pea gravel	0.03	0.26	4.6950	2.572	1.0e+0
Lower gravelly sand	0.02	0.34	0.1008	2.922	1.0e-2
Clay	0.0001	0.36	0.0016	1.203	1.0e-7
Sand	0.045	0.37	0.0683	2.080	3.0e-2
Gravel	0.014	0.51	3.5366	2.661	1.85e+0
Concrete	0.08	0.40	0.0063	1.080	1.0e-8
Undisturbed clay sand	0.21	0.30	0.0035	3.0	1.4e-7

$$K(\theta) = K(H) = K_{sat} H^{0.5} \left[1 - \left(1 - H^{\frac{1}{m}}\right)^m\right]^2 \tag{11.47}$$

where, $H = residual\ saturation = \frac{\theta - \theta_r}{\theta_s - \theta_r}$, $\theta_s =$ saturated water content, $m = 1 - \frac{1}{n}$, and $n =$ van Genuchten parameter.

The relevant parameters for this equation for various soil material types are given in Table 11.4.

If the hydraulic conductivity is represented as a function of pressure head, the expression is given as,

$$K(\psi) = K_{sat} \frac{\left[1 - (\alpha\psi)^{n-1} \cdot [1 + (\alpha\psi)^n]^{-m}\right]^2}{[1 + (\alpha\psi)^n]^{0.5m}} \tag{11.48}$$

where, α is another van Genuchten parameter as listed in Table 11.4.

11.3 Modeling Transport of Radionuclides in Groundwater

Understanding the movement of groundwater as discussed in previous sections provides the basis for the description of radionuclide migration in the groundwater systems. Additional processes that control the behaviors of contaminants in the groundwater in connection with the interactions with the porous medium are discussed in this section.

11.3.1 Drivers of Contaminant Transport in Groundwater

The radionuclides that are dissolved in the groundwater will be carried away along with the flowing groundwater in the porous medium. This process is called advection (advective transport). Advection refers to the process by which contaminants are transported by the bulk motion of groundwater. If the contaminants are nonreactive, their bulk mass moves at a rate equal to the average linear velocity of the groundwater by advection. The Darcy's law introduced in the previous section describes this bulk motion of the groundwater movement.

However, the bulk mas movement does not fully describe the transport of contaminant in the groundwater. This is because the dissolved contaminant species in the groundwater tend to spread out from the path of bulk movement of the flow system. The spreading of dissolved contaminants while moving in a porous medium is called dispersion, or hydrodynamic dispersion. This phenomena of dispersion cover a much larger volume of the subsurface space than would be expected from the bulk movement.

Most often, the contaminant species are reactive. They will react with other dissolved contaminant species in the water or have interactions with the solid surfaces of the porous medium. These interactions could lead to removal of the radionuclides from the groundwater or slowing down of their movements. Radioactive decay will also remove the radionuclides in the groundwater. Therefore, transport of radionuclides in the groundwater is subject to advection, dispersion, chemical/physical reactions, and radioactive decay.

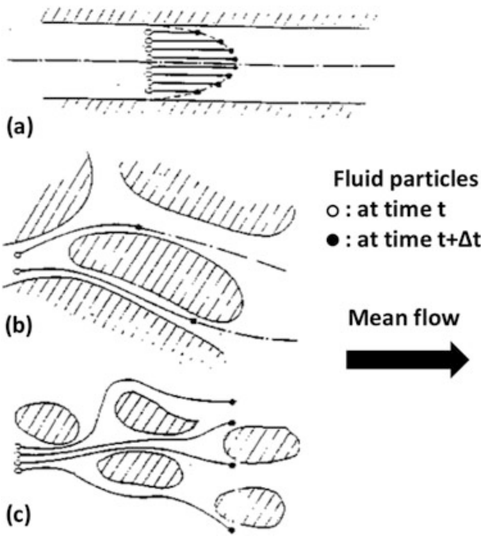
11.3.2 The Concept of Hydrodynamic Dispersion

Hydrodynamic dispersion, i.e., the spreading of contaminants in groundwater, is caused by mainly two processes, i.e., molecular diffusion and mechanical mixing (see Fig. 11.22).

Molecular diffusion occurs whenever a concentration gradient of a species is present. The dissolved radionuclides in water will disperse from the bulk mass (higher concentration region), spreading toward the surrounding (lower concentration) regions. This movement is due to the thermal-kinetic energy of the contaminant particles.

Mechanical mixing of contaminants is caused by the variations in groundwater velocity or by the turbulence in the flow. In a channel of groundwater movement, there are deviations in the rate of water movements due to the drag exerted on the fluid by the roughness of the pore surfaces. The drag slows down the water near the

Fig. 11.22 Causes of mechanical mixing in contaminant transport in groundwater



solid surface while the water in the center of a pore space moves faster. Also, as groundwater moves in a channel through three-dimensional tortuous paths with variations in the size of the pores, turbulence occurs. The results of these variations and turbulence is mixing of contaminants. Heterogeneities of solid particles in the medium add additional mixing effect. These mixing effects cause dispersion of contaminants in the flow medium.

Dispersion of dissolved contaminants causes the particles of contaminants to move at varying rates. Some particles will move faster and arrive at a point (e.g., a well) earlier than the arrival of the bulk mass. Some will move slower and arrive at the point much later. This is the characteristic feature of hydrodynamic dispersion of contaminants.

Figure 11.23 shows the dispersion of a nonreactive tracer in a packed column of porous medium under the condition of a steady state flow of groundwater. The changes in the concentration of the tracer in the column is represented by the ratio C/C_0, in the figure (where C is the time and spatially varying concentration of the tracer in the column and C_0 is the concentration initially introduced into the column). Prior to the introduction of the tracer, the tracer concentration in the column is zero. Once the tracer is introduced (at $t = t_0$), its concentration spreads in the column. This spreading causes some of the tracer molecules to move faster or slower than the average linear velocity.

The hydrodynamic dispersion of the dissolved species occurs both in the direction of bulk flow and in the direction normal to the flow path. The dispersion that occurs along the direction of bulk flow is called longitudinal dispersion. The dispersion in the direction perpendicular to the flow is called transverse dispersion. Figure 11.24 shows the spreading of contaminant plume due to longitudinal and transverse dispersion.

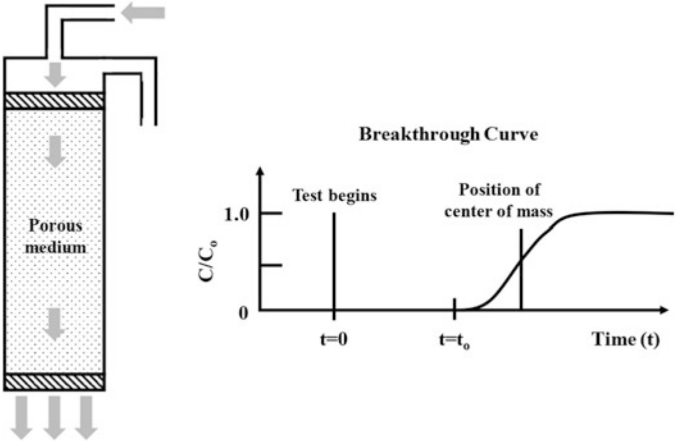
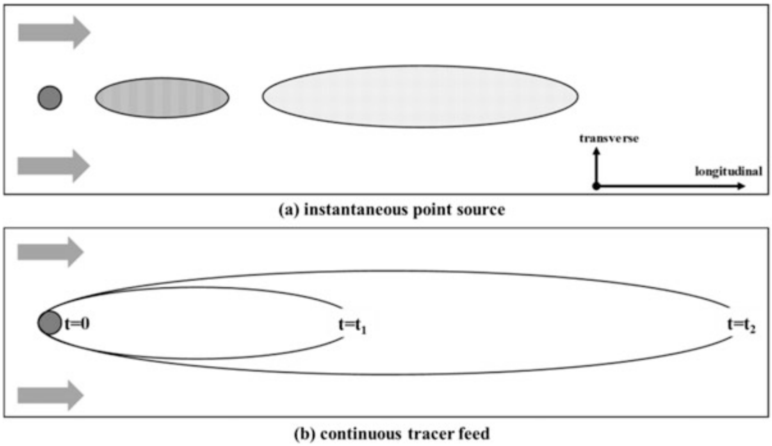


Fig. 11.23 Illustration of tracer dispersion in a column of porous medium



(a) instantaneous point source

(b) continuous tracer feed

Fig. 11.24 Spreading of a tracer in a two-dimensional uniform flow field in an isotropic porous medium

11.3.3 Modeling Contaminant Transport in Groundwater: No Chemical Reactions Involved

Development of a model to describe contaminant transport in a porous medium is based on examining mass balance within a control volume (a fixed region in space where a specified mass of a species crosses the boundary of the region). The following equation captures the mass balance within a control volume.

$$\begin{aligned}&[\textit{Net rate of change of mass of radionuclide}, i]\\&\quad = [\textit{Changes between influx\&outflux}] + [\textit{Gain from precursor decay}]\\&\qquad - [\textit{Loss due to decay of radionuclide}, i]\end{aligned} \tag{11.49}$$

Mathematical representation of this mass balance in 1-dimensional formulation gives the following equation (in the direction of increasing x, i.e., the direction of bulk groundwater movement).

$$\frac{\partial C_i \varepsilon_f}{\partial t} dx = -\frac{\partial q_i}{\partial x} + \lambda_{i-1} C_{i-1} \varepsilon_f dx - \lambda_i C_i \varepsilon_f dx \tag{11.50}$$

where

$C_i = C_i(x, t) =$ the liquid phase concentration of radionuclide, i [M/L^3],
$C_{i-1} = C_{i-1}(x, t) =$ the liquid phase concentration of the precursor nuclide of C_i,
$q_i = q_i(x, t) =$ the mass flux of radionuclide, i [M/L^2-T] as the flux of contaminant into and out of a fixed control volume within the flow of domain, and.
$\varepsilon_f =$ effective porosity.

This equation assumes that the porous medium is in the saturated zone. As C_i represents mass per volume of water, it is necessary to multiply the equation by effective porosity (the volume of pore water divided by the total volume), the porosity that actually contribute to water movement. Then the resulting concentration of radionuclides is defined for the total volume.

The flux of contaminant into and out of a fixed control volume results from the combined effect of advection and dispersion in the system. The flux due to advection is given as:

$$q_i^a = C_i \varepsilon_f v \tag{11.51}$$

The flux due to hydrodynamic dispersion is described by an expression analogous to Fick's first law of diffusion: The mass of fluid under diffusion is proportional to the concentration gradient given as following:

$$q_i^h = -D_{hi} \frac{\partial C_i \varepsilon_f}{\partial t} \tag{11.52}$$

where D_{hi} is coefficient of hydrodynamic dispersion. The negative sign indicates that the movement is from the areas of higher concentration to those of lower concentration.

The advective flux and dispersive flux are combined to represent the total flux as,

$$\frac{\partial q_i}{\partial x} = \frac{\partial\left(q_i^a + q_i^h\right)}{\partial x} = \frac{\partial}{\partial x}\left(C_i\varepsilon_f v - D_{hi}\frac{\partial C_i\varepsilon_f}{\partial t}\right)$$
$$= \frac{\partial}{\partial x}\left(C_i\varepsilon_f v - D_{hi}\varepsilon_f\frac{\partial C_i}{\partial t}\right) \tag{11.53}$$

Substituting Eq. 11.53 into Eq. 11.50,

$$\frac{\partial C_i\varepsilon_f}{\partial t} = -\frac{\partial q_i}{\partial x} + \lambda_{i-1}C_{i-1}\varepsilon_f - \lambda_i C_i\varepsilon_f$$
$$= -\frac{\partial}{\partial x}\left(C_i\varepsilon_f v - D_{hi}\varepsilon_f\frac{\partial C_i}{\partial t}\right) + \lambda_{i-1}C_{i-1}\varepsilon_f - \lambda_i C_i\varepsilon_f$$
$$\frac{\partial C_i\varepsilon_f}{\partial t} = \frac{\partial}{\partial x}D_{hi}\varepsilon_f\frac{\partial C_i}{\partial t} - \frac{\partial}{\partial x}C_i\varepsilon_f v + \lambda_{i-1}C_{i-1}\varepsilon_f - \lambda_i C_i\varepsilon_f \tag{11.54}$$

In homogeneous medium, the above equation becomes,

$$\frac{\partial C_i}{\partial t} = \frac{\partial}{\partial x}D_{hi}\frac{\partial C_i}{\partial t} - v\frac{\partial C_i}{\partial x} + \lambda_{i-1}C_{i-1} - \lambda_i C_i \tag{11.55}$$

If the disintegration of a precursor as a source for a radionuclide is ignored (which is not common), the equation becomes,

$$\frac{\partial C_i}{\partial t} = \frac{\partial}{\partial x}D_{hi}\frac{\partial C_i}{\partial t} - v\frac{\partial C_i}{\partial x} - \lambda_i C_i$$

This is the advection-dispersion equation to describe the transport of nonreactive radionuclides in groundwater in the saturated system.

11.3.3.1 Coefficient of Hydrodynamic Dispersion

As the process of molecular diffusion and mechanical mixing both contribute to hydrodynamic dispersion, the coefficient of hydrodynamic dispersion is expressed as the combination of both effects.

$$D_h = \alpha v + D^* \tag{11.56}$$

Where, D_h is the coefficient of hydrodynamic dispersion, α is the dynamic dispersivity [L], D^* is the coefficient of molecular diffusion [L^2/T], and v is the average linear velocity [L/T]. The term αv represents the effect of mechanical mixing.

Table 11.5 Example of diffusion coefficients of ions in free water [Samson et al. 2003]

Cation	D^* (10^{-9} m^2/s)	Anion	D (10^{-9} m^2/s)
Na^+	1.334	OH^-	5.273
K^+	1.957	Cl^-	2.032
Mg^{2+}	0.706	SO_4^{2-}	1.065
Ca^{2+}	0.792		

11.3.3.2 Molecular Diffusion Coefficient

The (molecular) diffusion of radionuclides results in a net flux of the substance regardless of the direction of flow represented by Fick's law. The diffusion coefficient, as a property of the dissolved species, characterizes the movement of the substance through unit cross sectional area of a region per unit of time when the concentration gradient is unity. It is typically on the order of 10^{-9} to 10^{-11} m^2/s (Table 11.5). Normally in transport of contaminant in groundwater, the effect of molecular diffusion is very small compared to that of mechanical mixing. However, if groundwater moves very slowly, the effect of molecular diffusion could become significant (this is represented in Fig. 11.27 where relative importance of molecular diffusion and mechanical mixing in contaminant transport in groundwater is shown).

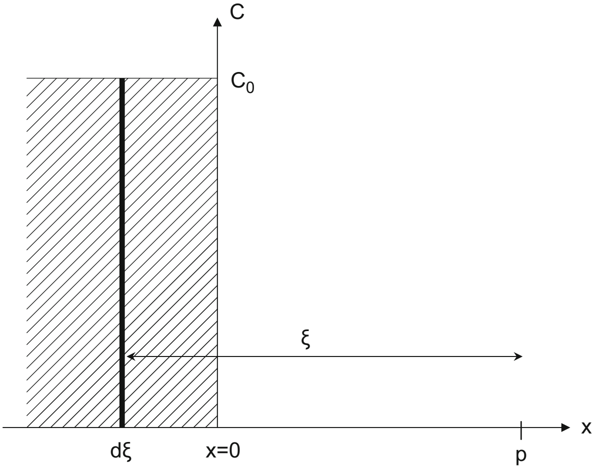
11.3.3.3 The Behavior of Hydrodynamic Dispersion of Contaminant

The process of hydrodynamic dispersion is an important part of advancing the contaminant plume in groundwater. Figure 11.24 shows an example of hydrodynamic dispersion of a tracer in a two-dimensional flow field. In Fig. 11.24a, the tracer is introduced instantaneously as a point source into the flow regime. As the tracer spreads away, albeit the total mass in the flow regime remains the same, the mass occupies an increasingly larger volume. A similar behavior of dispersion is also observed in the case of continuous release of the tracer in the source where the source region is always included in the contaminant plume (Fig. 11.24b).

As observed, the dispersive transport of contaminant in groundwater occurs both in the direction of flow and the direction normal to the flow with different degrees of dispersion: The dispersion is much stronger in the direction of flow than in the directions normal to the flow line. As the flow of groundwater in an aquifer is mostly horizontal, i.e., defined on a x-y plane, two coefficients can be defined: the longitudinal dispersion coefficient and the transverse dispersion coefficient. In case the dispersion is considered in three dimensional directions, the vertical dispersion is also included. In this case, transverse dispersion in the horizontal plane is generally much larger than the vertical dispersion. Overall, the longitudinal dispersion is much larger than both the transverse and vertical dispersion (up to 100 times larger).

The coefficient of longitudinal and transverse dispersion, respectively, is represented in the following equations.

Fig. 11.27 A schematic of geometry showing the effect of a continuous source in an infinite column (in x ≤ 0) on a concentration front



$$D_{hL} = \alpha_L v + D^*, D_{hT} = \alpha_T v + D^* \tag{11.57}$$

where

D_{hL} = The coefficient of longitudinal hydrodynamic dispersion
D_{hT} = The coefficient of transverse hydrodynamic dispersion
α_L = The longitudinal dynamic dispersivity
α_T = The transverse dynamic dispersivity
v = The average linear velocity of groundwater
D^* = Molecular diffusion coefficient

The coefficient of longitudinal and transverse hydrodynamic dispersion can be estimated based on the empirical observation of the data as

$$D_{hL} = \frac{\sigma_L^2}{2t}, D_{hT} = \frac{\sigma_T^2}{2t} \tag{11.58}$$

where, t is time, σ_L^2 is the variance in longitudinal spreading of the contaminant plume, and σ_T^2 is the variance in transverse spreading of the contaminant plume.

11.3.3.4 Dispersivity (Dynamic Dispersivity)

Dynamic dispersivity, or simply dispersivity, is a property of porous rocks as a medium for groundwater movement. It captures the degree of mechanical mixing of the contaminant with respect to the changes in the flow velocity due to movements through tortuous complex pore structures. It also reflects the heterogeneities of the system in hydraulic conductivity and permeability.

Table 11.6 The Data to Represent Longitudinal Dispersivity (for Eq. 11.59)

Type of rock	c (The characteristic parameter)	The scaling exponent
Unconsolidated sediments	0.20	0.44
Sandstones	0.01	0.92
Carbonates	0.80	0.40
Basalts	0.15	0.61
Granites	0.21	0.51

Dispersivity can be determined by field tests through measurements of contamination levels using a tracer in an aquifer system. Values of the longitudinal dispersivity, α_L, normally range between 0.1 and 30 m (Schulze-Makuch 2005). Challenge in determining the values of dispersivity is that the dispersivity increases as the scale of the hydrological system under investigation increases. This is because the spreading of dissolved contaminants in the groundwater tends to increase with the distance of travel. This is also in part due to the increase in the degree of heterogeneities in the system with increase in the travel distance.

While acknowledging the difficulty in its determination, the longitudinal dispersivity α_L (m) can be estimated by using empirical relationships. The following scaling relationship is a result of extensive review of published data.

$$\alpha_L = c(L)^m \tag{11.59}$$

where, c is a parameter characteristic of a geological medium, m is the scaling exponent, and L is the flow distance (m). The mean value of the scaling exponent is ~0.5. The value of c depends on the heterogeneity of rocks. Table 11.6 gives the values of c and m for several rock types. The upper bound for L to use the relationship is ~10 km for most rocks except for granites (100 m in this case).

Values of the transverse dispersivity, α_T, are found to be in the range between 0.0002 m and 0.2 m (Zech et al. 2019). In general, the values are within the range of 1% to 10% of the value of the longitudinal dispersivity α_L. Unlike the longitudinal dispersivity, no apparent scale effects can be noted with the transverse dispersivity.

The following relationship is also suggested to determine the value of transverse dispersivity, α_T (m), as a function of hydraulic conductivity of the medium (Carey et al. 2018),

$$\alpha_T = 0.08(K)^{-0.16} \tag{11.60}$$

where, K is hydraulic conductivity (m/s) of a medium and α_T is in millimeter.

11.3.3.5 Relative Importance of Molecular Diffusion and Mechanical Mixing

To understand the role of the underlying physical mechanisms in hydrodynamic dispersion, comparison of the contributions by molecular diffusion and mechanical

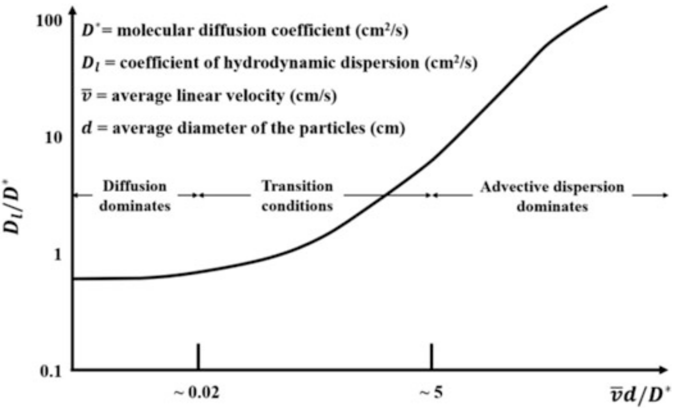


Fig. 11.25 Importance of diffusion vs. advection as a function of Peclet number

mixing is considered. Figure 11.25 shows this comparison based on the spreading of solutes migrating through a saturated sand columns. A dimensionless parameter known as the Peclet number, $v_x\, d/D^*$ is defined as the basis of this comparison, where v_x is the average velocity of the solute movement, d is the average diameter of the solid particles as the characteristic dimension of the porous medium, and D^* is the molecular diffusion coefficient. The y-axis of the figure represents the ratio of longitudinal dispersion, D_L, to molecular diffusion, D^*.

At very low flow velocities, the figure shows that molecular diffusion has larger influence than mechanical mixing on the overall hydrodynamic dispersion. In this case, the ratio of D_L/D^* is at a constant value of about 0.7. With increase in the velocity, the molecular diffusion effect decreases and the effect of mechanical mixing becomes more significant. Between a Peclet number of about 0.4 to 5, molecular diffusion and mechanical mixing have about the same influences. As the velocity increases further, the effect of mechanical mixing becomes dominant.

According to the 10CFR 60, U.S. NRC's regulations, there is a specified travel time requirement for high level nuclear waste disposal: The movement of groundwater to the accessible environment (at 5 km from the site boundary) should take longer than 1000 years. Therefore, by regulation, the maximum velocity of groundwater movement for any candidate sites for HLW disposal should be less than 5 m/year. Considering that the characteristic dimension of the porous medium (i.e., permeability) typically range between 0.01 and 0.001 m and that the molecular diffusion coefficient ranges from 10^{-4} to 10^{-5} cm^2/s, the ratio of $v_x\, d/D^*$ ranges between 10^{-2} and 1. This indicates that molecular diffusion is important and cannot be ignored in describing the migration of radionuclides in groundwater at a HLW geological repository.

11.3.4 Analytical Solutions of Contaminant Transport Equation

The most general treatment of the advection-dispersion equation (Eq. 11.55) requires using numerical solutions approach to describe the movements of radionuclides in groundwater. Simplified approaches are also available by using analytical solutions of the equation by assuming D_{hi} is a constant. Two examples of analytical solutions are given below.

11.3.4.1 Case 1: Pulse Injection of a Contaminant into an Infinite, Homogeneous Column of Porous Material

Assume a steady flow of water through an infinite, homogeneous column of porous material. Suppose that at time $t = 0$ a very thin slug of M kg of a non-reactive contaminant is introduced per cross sectional area (m^2) into the column at $x = 0$. The average linear velocity in the column is v. The initial concentration of the contaminant is zero in the column. The contaminant is stable and no decay or chemical reactions are involved. The influence of transverse dispersion can be ignored.

The equation for the system can be written as

$$\frac{\partial C}{\partial t} = D_h \frac{\partial^2 C}{\partial x^2} - v \frac{\partial C}{\partial x} \tag{11.61}$$

where D_h is the longitudinal dispersion coefficient and v is the average linear velocity.

For a moment, let's ignore the advection term and only examine the dispersion term,

$$\frac{\partial C}{\partial t} = D_h \frac{\partial^2 C}{\partial x^2}, \quad \left(\text{Equivalent to Fick's 2nd Law}\right) \tag{11.62}$$

$$C(x = \pm\infty, t) = 0$$

The solution of the equation takes the form of the Gaussian distribution,

$$C(x,t) = \frac{A}{\sigma\sqrt{2\pi}} e^{-\frac{1}{2}\left(\frac{x-\mu}{\sigma}\right)^2} \tag{11.63}$$

By noting that, $\mu = 0$ and $\sigma = \sqrt{2D_h t}$ (thus $D_h = \sigma^2/2t$) for a diffusional process,

$$C(x,t) = A \cdot \frac{1}{t^{1/2}} e^{-\frac{x^2}{4D_h t}} \tag{11.64}$$

Now, the total amount of the substance diffusing in the entire medium should be equal to M (kg/m^2),

$$M = \int_{-\infty}^{\infty} \varepsilon C(x,t)dx = \int_{-\infty}^{\infty} \varepsilon \cdot \frac{A}{t^{1/2}} e^{-\frac{x^2}{4D_h t}} dx \tag{11.65}$$

Let $\frac{x^2}{4D_h t} = \frac{y^2}{2} \rightarrow dx = \sqrt{2D_h t}\,dy$: $M = \int_{-\infty}^{\infty} \varepsilon \cdot \frac{A}{t^{\frac{1}{2}}} e^{-\frac{y^2}{2}} \sqrt{2D_h t}\,dy = \varepsilon A\sqrt{2D_h} \int_{-\infty}^{\infty} e^{-\frac{y^2}{2}} dy = \varepsilon A\sqrt{4\pi D_{h.}}$

Therefore,

$$A = \frac{M}{\varepsilon\sqrt{4\pi D_h}} = \frac{M/\varepsilon}{\sqrt{4\pi D_h}} \tag{11.66}$$

$$C(x,t) = \frac{M/\varepsilon}{\sqrt{4\pi D_h t}} e^{-\frac{x^2}{4D_h t}} \tag{11.67}$$

Now, consider the addition of the advective transport term in Eq. 11.62.

$$\frac{\partial C}{\partial t} = D_h \frac{\partial^2 C}{\partial x^2} - v\frac{\partial C}{\partial x}, C(x = \pm\infty, t) = 0 \tag{11.68}$$

The effect of adding the advection term on the solution of the equation is to move the center of mass of the contaminant plume. In other words, the center of mass will no longer be at $x = 0$ but move according to the average linear velocity of the groundwater, i.e. $\mu = vt$.

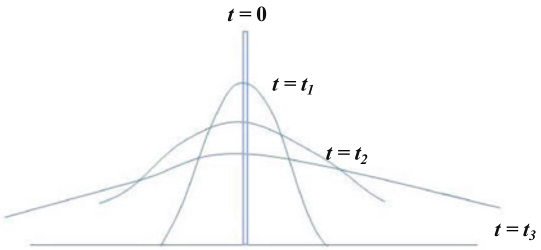
Then the solution representing the effect of having the advective transport term becomes,

$$C(x,t) = \frac{M/\varepsilon}{\sqrt{4\pi D_h t}} e^{-\frac{(x-vt)^2}{4D_h t}} \tag{11.69}$$

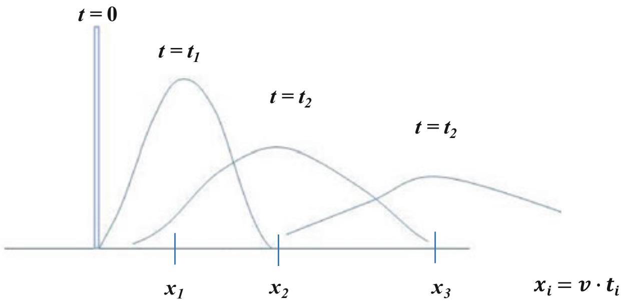
If the contaminant is a radionuclide with its decay constant λ, the solution including the effect of radioactive decay becomes,

$$C(x,t) = \frac{M/\varepsilon}{\sqrt{4\pi D_h t}} e^{-\lambda t} e^{-\frac{(x-vt)^2}{4D_h t}} \tag{11.70}$$

In general, radioactive decay results in irreversible decline in the activity of a radionuclide and is important for contaminant attenuation if the half-life of the radionuclide is comparable to or less than the residence time of the plume in the aquifer system.



(a) Solution of dispersion equation – the Gaussian distribution at x = 0



(b) Solution of advection and dispersion equation–moving the Gaussian distribution with time

Fig. 11.26 Gaussian distribution as the solution of advection and dispersion equation (see example 11.7)

The spreading of contaminant during its movement in the groundwater follows the pattern of the Gaussian distribution (whether the effect of advection of plume is considered or not) as illustrated in Fig. 11.26. The peak of the Gaussian distribution travels at the same rate as the average linear velocity of the water, and the width of the spreading increases proportionally with the square root of the travel time (this is the characteristics of the diffusion process).

Example 11.7: Graphical Solution of Advection-Dispersion Equation

The time-dependent solution of the dispersion equation (i.e., ignoring the effect of advection), $\frac{\partial C}{\partial t} = D_h \frac{\partial^2 C}{\partial x^2}$, will take the form of the Gaussian distribution with the increasing degree of plume spreading with time as shown in Fig. 11.26a. With increase in time, the peak concentration decreases and the concentration curve becomes flatter.

If the advection term is included (i.e., $\frac{\partial C}{\partial t} = D_h \frac{\partial^2 C}{\partial x^2} - v \frac{\partial C}{\partial x}$), the time-dependent solution will reflect the displacement of the center-of-mass of the plume while keeping the same exact shape of the Gaussian distribution as the solution of the dispersion equation (Fig. 11.26b). The center-of-mass movement follows the rate of movement dictated by the average linear velocity.

The solution of the advection and dispersion equation can be expanded in two-dimensional representation, as shown below (by using $\sigma_x = \sqrt{2D_{hx}t}$ and $\sigma_y = \sqrt{2D_{hy}t}$).

$$\begin{aligned} C(x, y, t) &= \frac{C_0}{\sigma_x\sqrt{2\pi}\cdot\sigma_y\sqrt{2\pi}} e^{-\frac{1}{2}\left(\frac{x-\mu_x}{\sigma_x}\right)^2} e^{-\frac{1}{2}\left(\frac{y-\mu_y}{\sigma_y}\right)^2} \\ &= \frac{C_0}{4\pi\sqrt{D_{hx}D_y}} e^{-\left(\frac{(x-v_x t)^2}{4D_{hx}t}+\frac{y^2}{4D_{hy}t}\right)} \end{aligned} \tag{11.71}$$

$$C(x, y, t) = \frac{C_0}{4\pi\sqrt{D_L D_T}} e^{-\left(\frac{(x-v_x t)^2}{4D_L t}+\frac{y^2}{4D_T t}\right)} \tag{11.72}$$

where, D_L and D_T represent the coefficient of hydrodynamic dispersion for the movement in the longitudinal and transverse direction, respectively.

This solution represents the case of a slug injection into a two-dimensional uniform flow field where the center of y-directional movement is at $y = 0$.

11.3.4.2 Case 2: Movement of Concentration Front in an Infinite Column from a Continuous Source (Steady State Flow)

Consider a presence of a semi-infinite (source) region of a dissolved non-reactive contaminant at concentration C_0. This region is next to a semi-infinite region of porous medium with pure water (no contamination) by being separated with a sharp interface between the regions at $x = 0$. At time $t = 0$, the interface separating the regions breaches and the dissolved contaminant starts moving into the porous medium region of pure water. The water movement is steady and moves at average linear velocity of v.

For one-dimensional case, the system equation is

$$\frac{\partial C}{\partial t} = D_h \frac{\partial^2 C}{\partial x^2} - v\frac{\partial C}{\partial x} \tag{11.73}$$

with the following initial and boundary conditions:

$C(x, 0) = C_0 - \;\infty\; < x < 0$
$C(x, 0) = 0 \;\; 0 < x < \infty$
$C(-\infty, t) = C_0$
$C(\infty, t) = 0$

The solution from the point pulse injection case (Case 1) can be utilized here to obtain the solution to the problem. In this (Case 2) problem, the contaminant source region can considered a combination of an infinite number of line sources in the region $x \leq 0$ (Fig. 11.27). Each corresponding solution from the line sources can be superposed on a location in $x > 0$ to represent the impact of having the source region continuously releasing the contaminant at the same concentration level, i.e., as the extended source distribution composed of an infinite number of line sources.

In the figure, the diffusing substance in a line element of width $d\xi$ is the dissolved contaminant with source strength $C_0 d\xi$.

Then, following the solution from the Case 1 problem, the concentration at point p at distance ξ from the element (at time t) is,

$$C(x = p, t) = \frac{C_0 d\xi}{\sqrt{4\pi D_h t}} e^{-\frac{\xi^2}{4D_h t}} \tag{11.74}$$

From the existence of infinite line sources, the complete solution is given by summing over the successive elements, $d\xi$, by (where the distance ξ ranges from x to ∞).

$$C(x, t) = \frac{C_0}{\sqrt{4\pi D_h t}} \int_x^{\infty} e^{-\frac{\xi^2}{4D_h t}} \, d\xi \tag{11.75}$$

Let, $\eta = \frac{\xi}{\sqrt{4D_h t}} \to d\xi = \sqrt{4D_h t}\, d\eta$ and ξ ranges from x to ∞; η ranges from $\frac{x}{\sqrt{4D_h t}}$ to ∞. Then,

$$\begin{aligned} C(x, t) &= \frac{C_0}{\sqrt{4\pi D_h t}} \sqrt{4D_h t} \int_{\frac{x}{\sqrt{4D_h t}}}^{\infty} e^{-\eta^2} \, d\eta = \frac{C_0}{\sqrt{\pi}} \int_{\frac{x}{\sqrt{4D_h t}}}^{\infty} e^{-\eta^2} \, d\eta \\ &= \frac{C_0}{\sqrt{\pi}} \left\{ \int_0^{\infty} e^{-\eta^2} \, d\eta - \int_0^{\frac{x}{\sqrt{4D_h t}}} e^{-\eta^2} \, d\eta \right\} \end{aligned} \tag{11.76}$$

Here, we use the definition of error functions: $erf(z) = \frac{2}{\sqrt{\pi}} \int_0^z e^{-\eta^2} \, d\eta$. Note $\int_0^z e^{-\eta^2} \, d\eta = \frac{\sqrt{\pi}}{2} erf(z)$ and $erf(\infty) = 1$. Then,

$$\begin{aligned} C(x, t) &= \frac{C_0}{\sqrt{\pi}} \left\{ \frac{\sqrt{\pi}}{2} erf(\infty) - \frac{\sqrt{\pi}}{2} erf\left(\frac{x}{\sqrt{4D_h t}} \right) \right\} \\ &= \frac{C_0}{\sqrt{\pi}} \left\{ \frac{\sqrt{\pi}}{2} - \frac{\sqrt{\pi}}{2} erf\left(\frac{x}{\sqrt{4D_h t}} \right) \right\} = \frac{C_0}{2} \left\{ 1 - erf\left(\frac{x}{\sqrt{4D_h t}} \right) \right\} \\ &= \frac{C_0}{2} erfc\left(\frac{x}{\sqrt{4D_h t}} \right) \end{aligned} \tag{11.77}$$

where, complementary error function, $erfc(z) = 1 - erf(z)$.

If we include the effect of advective movement,

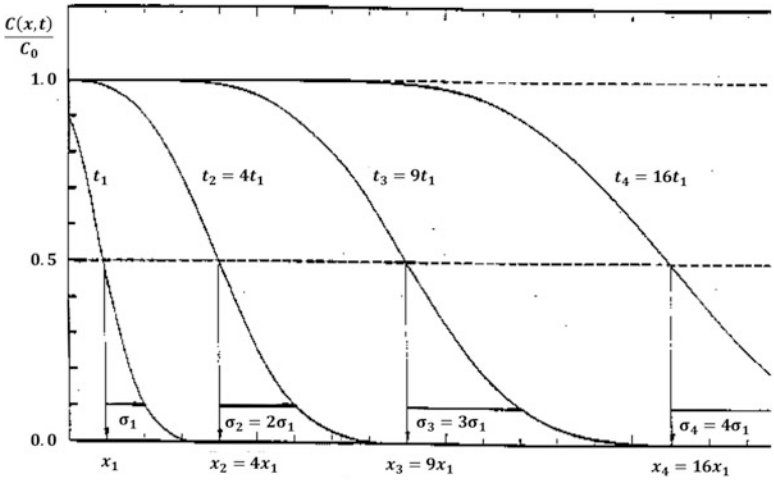


Fig. 11.28 Movement of concentration front in an infinite column (Steady one-dimensional flow)

$$C(x,t) = \frac{C_0}{2} erfc\left(\frac{x - vt}{\sqrt{4D_h t}}\right) \tag{11.78}$$

Graphical representation of this solution is shown in Fig. 11.28. The mid-point of the transitional zone (i.e., where $C(x,t) = 0.5C_0$) moves downstream at the average linear velocity, and the width of the zone increases with the square root of the travel time.

11.3.5 Modeling Contaminant Transport in Groundwater with Chemical Reactions

The discussions in the previous section assumed no chemical interactions between the contaminant and the porous medium. In reality, dissolved radionuclides in groundwater interact with solid surfaces of the porous medium during their transport. These interactions cause the contaminant to be distributed between the aqueous phase and the solid phase (soil surfaces). This interactions also slow down the migration of contaminant, causing its retardation relative to the groundwater movement within the aquifer. Therefore, the mass balance relationship needs to be reestablished including both the solid phase and the liquid phase.

Assuming reversible chemical reactions between the two phases, the equation for each phase can be written as follows.

- The material balance in the aqueous phase:

$$\varepsilon\frac{\partial C_i}{\partial t} = \varepsilon D_h\frac{\partial^2 C_i}{\partial x^2} - \varepsilon v\frac{\partial C_i}{\partial x} - \varepsilon k_1 C_i + (1-\varepsilon)\rho_s k_2 s_i - \lambda_i \varepsilon C_i \tag{11.79}$$

- The material balance in the solid (immobile) phase:

$$(1-\varepsilon)\rho_s\frac{\partial s_i}{\partial t} = \varepsilon k_1 C_i - (1-\varepsilon)\rho_s k_2 s_i - \lambda_i(1-\varepsilon)\rho_s s_i \tag{11.80}$$

where, $C_i(x,t)$ = average concentration of radionuclide i, in the aqueous phase (mass/volume of water)

$s_i(x,t)$ = the mass fraction in the solid phase (mass/mass of solid)
v = the average linear velocity
k_1 = the first order rate constant for sorption
k_2 = the first order rate constant for desorption
ε = the porosity (it is assumed that the effective porosity is equal to the total porosity, $\varepsilon = \varepsilon_f$)

Here,

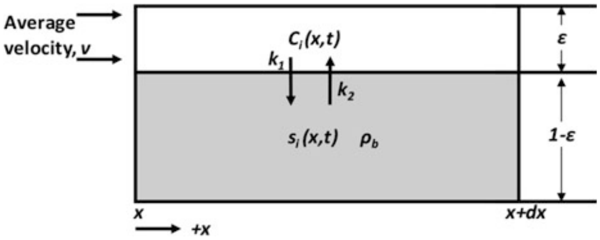
$\varepsilon k_1 C_i$ = the amount of mass removed from the aqueous phase due to chemical reaction and sorbed onto the solid phase. The rate of chemical reaction is given by k_1, in $\left[\frac{V_{water}}{V_{total}}\cdot sec^{-1}\cdot\frac{mass}{V_{water}}\right]$. $(1-\varepsilon)\rho_s k_2 s_i$ = the amount of mass released into the aqueous phase due to chemical reaction from the solid phase. The rate of chemical reaction is given by k_2, in $\left[sec^{-1}\cdot\frac{mass}{mass}\cdot\frac{V_{total}-V_{water}}{V_{total}}\frac{mass}{V_{solid}}\right]$.

Please note that the terms, $\varepsilon k_1 C_i$ and $(1-\varepsilon)\rho_s k_2 s_i$, each represent the amount of material movement between the aqueous phase and solid phase due to chemical reactions.

These two equations can be solved simultaneously to obtain the solutions for C_i and s_i.

As an alternative, we note that the solutions of these equations can be much simplified by making an assumption. The assumption is that the sorption and desorption reactions between the aqueous and the solid phases occur instantaneously and that local equilibrium is established immediately. Figure 11.29 shows this situation. The amount of mass removed from the aqueous phase due to chemical

Fig. 11.29 Sorption in a porous medium



reaction and sorbed onto the solid phase is equal to the amount of mass released into the aqueous phase due to chemical reactions from the solid phase.

Therefore,

$$\varepsilon k_1 C_i = (1-\varepsilon)\rho_s k_2 s_i \tag{11.81}$$

The equation can be rearranged as follows.

$$\frac{s_i}{C_i} = \frac{\varepsilon k_1}{(1-\varepsilon)\rho_s k_2} = \frac{\varepsilon k_1}{\rho_b k_2} = \frac{\theta k_1}{\rho_b k_2} \qquad (\varepsilon = \theta, \text{if saturated}) \tag{11.82}$$

This ratio is defined as distribution coefficient, K_d as follows,

$$K_d = \frac{k_1 \theta}{k_2 \rho_b} \tag{11.83}$$

where, k_1 = the forward kinetic rate (aqueous to solid)

k_2 = the backward kinetic rate
θ = the saturated water content (i.e., = ε)
ρ_b = the bulk density

Then, by using the definition of K_d, the solid phase concentration is represented by the aqueous phase concentration as follows. This relationship is also called the linear adsorption isotherm.

$$s_i(x,t) = K_{d,i} C_i(x,t) \tag{11.84}$$

where $K_{d,\ i}$ is the distribution coefficient of species *i* (ml/g). Therefore, distribution coefficient is the concentration of the species per gram of solid phase divided by its concentration in the liquid phase at equilibrium.

The unit of distribution coefficient is in volume per mass as shown below.

$$\frac{s_i(mg/g)}{C_i(mg/ml)} = \frac{\varepsilon k_1}{(1-\varepsilon)\rho_s k_2} = \frac{\varepsilon k_1}{\rho_b k_2} = \frac{\theta k_1}{\rho_b k_2} \equiv K_d(ml/g) \tag{11.85}$$

Now, summing Eqs. 11.79 and 11.80 gives,

$$\varepsilon \frac{\partial C_i}{\partial t} + (1-\varepsilon)\rho_s \frac{\partial s_i}{\partial t} = \varepsilon D_h \frac{\partial^2 C_i}{\partial x^2} - \varepsilon v \frac{\partial C_i}{\partial x} - \lambda_i \varepsilon C_i - \lambda_i (1-\varepsilon)\rho_s s_i \tag{11.86}$$

By using Eq. 11.84, all of the solid phase concentration, s_i can be substituted by C_i in Eq. 11.86 to give,

$$\left[1+\frac{1-\varepsilon}{\varepsilon}\rho_s K_{d,i}\right]\frac{\partial C_i}{\partial t}=D_h\frac{\partial^2 C_i}{\partial x^2}-v\frac{\partial C_i}{\partial x}-\left[1+\frac{1-\varepsilon}{\varepsilon}\rho_s K_{d,i}\right]\lambda_i C_i \tag{11.87}$$

Dividing all of the terms in Eq. 11.87 by $\left[1+\frac{1-\varepsilon}{\varepsilon}\rho_s K_{d,i}\right]$ gives

$$\frac{\partial C_i}{\partial t}=\frac{D_h}{\left[1+\frac{1-\varepsilon}{\varepsilon}\rho_s K_{d,i}\right]}\frac{\partial^2 C_i}{\partial x^2}-\frac{v}{\left[1+\frac{1-\varepsilon}{\varepsilon}\rho_s K_{d,i}\right]}\frac{\partial C_i}{\partial x}-\lambda_i C_i \tag{11.88}$$

Let R_i= retardation factor $= 1+\frac{1-\varepsilon}{\varepsilon}\rho_s K_{d,i} = 1+\frac{\rho_d K_{d,i}}{\varepsilon}$. If the system is not completely saturated, $R_i = 1+\frac{\rho_b K_{d,i}}{\theta}$ where θ = moisture content. Then Eq. 11.88 is simplified to

$$\frac{\partial C_i}{\partial t}=\frac{D_h}{R_i}\frac{\partial^2 C_i}{\partial x^2}-\frac{v}{R_i}\frac{\partial C_i}{\partial x}-\lambda_i C_i \tag{11.89}$$

This equation is identical to the advection dispersion equation for the nonreactive contaminant case except the terms, $\frac{D_h}{R_i}$ and $\frac{v}{R_i}$. If these two terms are viewed as the effective dispersion coefficient ($D_{h,\ eff}$) and the effective average linear velocity (v_{eff}), respectively, then Eq. 11.89 is exactly the same as Eq. 11.55.

This observation can be explained as: In the case of chemically reactive contaminant, the values of average linear velocity and the coefficient of hydrodynamic dispersion are effectively reduced from the case of non-reactive contaminant case. The magnitude of this reduction is represented by the retardation factor: The retardation factor represents the effect of the chemical reactions involved in the system which effectively slows down the movement of contaminants.

11.3.6 Use of $\mathbf{K_d}$ *for Modeling Sorption in Contaminant Transport*

The chemical reactions between the dissolved contaminant and the solid surfaces of the porous medium covers a variety of interactive processes. These reactions are generally referred to ‘sorption’. As discussed in Sect. 4.3.5., the term ‘sorption’ can mean a simple mechanical adherence of ions to a solid surface or chemical incorporation of ions into the mineral structure.

Sorption is generally not permanent because the first ions sorbed can be displaced by others and dissolve back into the solution as the groundwater continues to move. For each contaminant ion, its motion can be visualized as taking place in a series of steps of sorption and desorption. Then the net effect of such interactions is *retardation* of its movement. With such retardation, the contaminant movement lags behind the flow of groundwater. The degree of such slowing down of a contaminant depends on the sorptive properties of the contaminant and the properties of the solid surface.

Example 11.8: Radionuclide Travel Time in Groundwater

Assume that a horizontal aquifer with a permeable clay formation has groundwater movement at Darcy velocity of 0.1 m/year.

(a) Calculate the hydraulic gradient in the aquifer.
(b) if plutonium is dissolved in the groundwater, estimate how long it would take for the dissolved Pu in the groundwater to move a distance of 5 km. Assume the K_d value of Pu in the system is 2000 ml/g.

Given: K (clay hydraulic conductivity) $= 10^{-6}$ cm/s
ε, porosity of the clay formation $= 0.1$
ρ_b, bulk density of the clay formation $= 2.5$ g/cm^3
Assume the effective porosity is equal to the porosity of the system.

Solution:

(a) Calculate the hydraulic gradient in the aquifer.

From Darcy's law, $v = -k\frac{dh}{dl}$, or
$dh/dL = -\ v/K = -0.1$ (m/year)/10^{-6} (cm/s) $= -0.317$

(b) Estimate how long it would take for the dissolved Pu in the groundwater to move a distance of 5 km.

- Without sorption: $v = v_0/\varepsilon_0 = 0.1$ (m/year)/0.1 $= 1$ m/year in this no sorption case, the travel time for Pu to reach the destination is 5000 years.
- With sorption, the effective velocity accounting for the retardation of Pu movement becomes,
- $v_{eff} = v/R \Rightarrow R = 1 + 2.5$ (g/cm^3) / 0.1 * 2000 (ml/g) $= 50{,}001$
- $v_{eff} = 1/50001 = 2.0 \times 10^{-5}$ (m/year)
- travel time $= 5000$ (m)/2.0×10^{-5} (m/year) $= 2.5 \times 10^{8}$ (year)

Thus with the sorption of Pu in the solid surfaces, the travel time for Pu to reach the destination increases from 5000 years to 250 million years. This difference in the estimated Pu travel time shows the importance of K_d, i.e., considering the effect of sorption. It should be noted that the value used for K_d should be carefully selected as the K_d value has large implications on potential outcome of Pu migration.

11.3.6.1 Sorption Isotherm Approaches

If the sorption process takes place rapidly in comparison with velocity of groundwater movement, it can be assumed that the dissolved species will reach an equilibrium condition with the sorbed phase. To represent the amount of solute sorbed onto the surface of solids in this case, different equilibrium sorption isotherm approaches have been suggested. Here an isotherm refers to a curve giving the functional relationship between a surface and the interacting species in a constant-temperature adsorption process. These approaches include the linear sorption isotherm, the Freundlich isotherm, and the Langmuir isotherm. The assumptions behind the isotherms approaches are: (1) the reactions are reversible; (2) equilibrium is quickly reached, and; (3) the reactions are independent of variations in local water chemistry, the rock water ratio, or the species concentration.

As explained earlier (Eq. 11.84), the linear sorption isotherm is given as,

$$S = K_d C \tag{11.90}$$

where

C = the concentration of the solute in solution in equilibrium (mg/ml)
S = the mass of the solute sorbed onto the solid surface per unit mass (mg/g)
K_d = the distribution coefficient (ml/g)

Based on this approach, the retardation factor in contaminant movement becomes,

$$R = 1 + \frac{\rho_b K_d}{\varepsilon} \tag{11.91}$$

The linear sorption isotherm is very useful in its use due to simplicity but has major limitations. According to the given relationship, the amount of solute that can be sorbed onto the solid is not constrained and continues to rise with increase in the aqueous concentration. In reality, the isotherm behavior is expected to be nonlinear as the concentration of the dissolved species increase in water.

An alternative nonlinear approach is the Freundlich isotherm represented in the following (with the use of n as exponent).

$$S = K_d C^n \tag{11.92}$$

In this case, the amount of solute that can be sorbed onto the solid can be constrained with the use of value of n to be less than 1. Based on this approach, the corresponding retardation factor becomes,

$$R = 1 + \frac{\rho_b K_d n C^{n-1}}{\varepsilon} \tag{11.93}$$

Another nonlinear and more intuitive approach is the Langmuir isotherm. This approach notes that a solid surface possesses a finite number of sorption sites. Once all the sorption sites are filled, the surface will no longer be available to sorb the solute from the solution. The following equation reflects this.

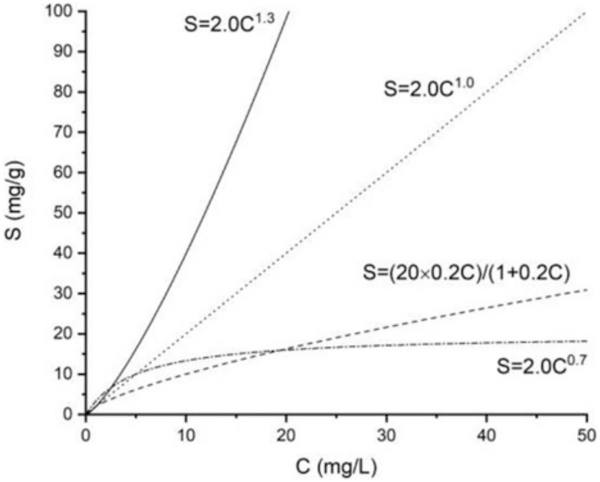


Fig. 11.30 Comparisons of different isotherm approach

$$S = \frac{Q_0 K_d C}{1 + K_d C} \tag{11.94}$$

where Q_0 is maximum sorptive capacity for the surface (mg/g).

Based on the Langmuir isotherm approach, the retardation factor becomes,

$$R = 1 + \frac{\rho_b}{\varepsilon}\left[\frac{K_d Q_0}{(1 + K_d C)^2}\right] \tag{11.95}$$

The relationships between the concentration in the aqueous phase and the mass in the solid phase are represented by using these different isotherm approaches in Fig. 11.30. The Langmuir isotherm always show non-linear saturating behavior whereas the Freundlich isotherm can give sublinear or supra linear behavior depending on the choice of value for *n*.

11.3.6.2 Measurement of K_d

The values of K_d for elements in soils can be measured typically through batch adsorption experiments (EPA 1999a, b, c). This experiment is performed by combining a solution with a known concentration of stable element with a known mass of the soil type under consideration. The solution and the soil mass are mixed and allowed for sorption to reach an equilibrium (typically for 1 to 7 days). The concentration of the element in the equilibrated solution is then measured and the difference between this measured concentration and the original solution concentration is used for the calculation of K_d as follows.

$$K_d = \frac{V_w(C_i - C_{as})}{M_s C_{as}} \tag{11.96}$$

where, V_w is volume of water, C_i is initial concentration of an aqueous contaminant, C_{as} is bulk concentration of the contaminant in the solution remaining after sorption, and M_s is mass of soil used in the experiment. The term $(C_i - C_{as})$ represents what is sorbed onto the soil.

Other methods for K_d measurement in the soil also include in-situ batch method, laboratory flow-through method, field monitoring method, and K_{oc} method (EPA 1999a).

Typical values of K_d measured for various elements are listed in Table 11.7. These values are the geometric means of the measured data in sand, silt, clay, and organic soil. Here the geometric mean is based on the use of lognormal distribution for the measure K_d values. In this case, the geometric mean (μ_g) is defined as,

$$\mu_g = \exp\left(\mu_{\ln(K_d)}\right) \tag{11.97}$$

Therefore, the geometric mean is the natural exponential of the mean of the log-transformed values of K_d.

It can be noted that elements that become cations when dissolved (e.g., Cs^+ and Fe^{2+}) tend to have higher K_d values. These cations are more strongly sorbed than the anions (e.g., I^-, HCO_3^-, and TcO_4^-) because most mineral surfaces in ordinary rocks are negatively charged. Iodine has a very low K_d values in all types of soil as it behaves mostly as iodide (I^-) in the environment. While the K_d values of heavy metals are shown to be high, the interactions of their ionic species in the porous medium are complex and estimating their K_d values is difficult due to the presence of multiple valences (see Sect. 4.1.2).

11.3.6.3 Other Considerations in the Use of K_d

If the concentration of a contaminant is limited by the solubility limit, use of the K_d approach to represent aqueous concentration of a species would not be valid. If the radionuclides are present mostly in colloidal forms, the K_d approach is also not recommended as the colloids provide an extra phase for the radionuclide speciation in the flowing groundwater. As typical applications of the K_d approach assume constant values of K_d for a given species, cautions must be exercised. If the variations in the chemistry conditions are expected to cause significant changes in K_d, the value of K_d should be conservatively selected or perhaps using a stochastic approach to reflect the effect of K_d value variations should be considered.

In the case of fractured flow modeling, the sorption can only occur at points where rock and water come into contact. In this case, using the so-called surface-related K_d approach can be considered. Then, a surface-related distribution coefficient, K_a, is defined as,

Table 11.7 Values of the distribution coefficients (K_d) (in ml/g as geometric mean) of different elements in various soils

	Sand		Silt		Clay		Organic	
Element	K_d (ml/g)	s.d.[1]	K_d (ml/g)	s.d.[1]	K_d (ml/g)	s.d.[1]	K_d (ml/g)	s.d.[1]
Actinium	450		1500		2400		5400	
Silver	90	1.8	120	1.1	180	0.4	15,000	0.9
Americium	1900	2.6	9600	1.4	8400	2.6	112,000	1.7
Beryllium	250		800		1300		3000	
Bismuth	100		450		600		1500	
Bromine	15		50		75		180	
Carbon	5	0.8	20		1		70	
Calcium	5		30		50		90	
Cadmium	80	1.5	40	1.6	560	0.9	800	2.3
Cerium	500	1.6	8100	1.5	20,000	0.5	3300	
Curium	4000	2.4	18,000	0.7	6000		6000	
Cobalt	60	2.8	1300	1.3	550	1.8	1000	1.5
Chromium	70	2.1	30	2.9	1500		270	2.7
Cesium	280	2.5	4600	1.3	1900	1.6	270	3.6
Iron	220	2.6	800	0.7	165	1.6	600	
Hafnium	450		1500		2400		5400	
Holmium	250		800		1300		3000	
Iodine	1	2.2	5	2.0	1	1.5	25	2.0
Potassium	15		55		75		200	
Manganese	50	1.4	750	2.6	180	2.0	150	
Molybdenum	10	1.1	125		90	1.2	25	0.5
Niobium	160		550		900		2000	
Nickel	400	1.5	300		650	0.7	1100	0.9
Neptunium	5	1.7	25	1.2	55	3.8	1200	0.4
Phosphorous	5		25		35		90	
Protactinium	550		1800		2700		6600	
Lead	270	2.3	16,000	1.4	550		22,000	0.5
Palladium	55		180		270		670	
Polonium	150	1.6	400	1.3	3000		7300	
Plutonium	550	1.7	1200	1.2	5100	2.1	1900	2.6
Radium	500	3.2	36,000	3.1	9100	1.3	2400	
Rubidium	55		180		270		670	
Rhenium	10		40		60		150	
Ruthenium	55	1.4	1000		800		66,000	0.3
Antimony	45		150		250		550	
Selenium	150	0.4	500		740	0.5	1800	0.5
Silicon	35		110		180		400	
Samarium	245		800		1300		3000	
Tin	130		450		670		1600	
Strontium	15	1.6	20	1.7	110	2.0	150	1.8
Tantalum	220		900		1200		3300	

(continued)

Table 11.7 (continued)

	Sand		Silt		Clay		Organic	
Element	K_d (ml/g)	s.d.[1]	K_d (ml/g)	s.d.[1]	K_d (ml/g)	s.d.[1]	K_d (ml/g)	s.d.[1]
Technetium	0.1	1.8	0.1	1.1	1	0.06	1	1.8
Tellurium	125		500		720		1900	
Thorium	3200	2.1	3300		5800	2.6	89,000	4.6
Uranium	35	3.2	15	3.3	1600	2.9	410	2.5
Yttrium	170		720		1000		2600	
Zinc	200	2.6	1300	2.4	2400	1.4	1600	1.6
Zirconium	600		2200		3300		7300	

Source: Thibault et al. (1990)

[1] Standard deviation of the natural logarithms of the observed values

$$K_a = K_d \rho_b / A \tag{11.98}$$

where A is the specific fracture surface area (L^2/L^3) and ρ_b is the bulk density of the rock (M/L^3).

In reality, the flow velocity in the fracture may be sufficiently high preventing the establishment of sorption equilibria. Under such conditions, the K_d approach is not applicable.

11.3.7 General Analytical Solutions for Contaminant Transport Equation

Consider a case of unidirectional transport of a single dissolved radionuclide with three-dimensional dispersion in a saturated homogeneous and isotropic aquifer.

In this case, the advective transport of the radionuclide is only in the x-direction but the dispersion takes place in x,y,z directions. If we assume an infinite aquifer, with thickness h, parallel to the x-y plane, the space and time-dependent radionuclide concentration in the aquifer is represented by the following equation.

$$\frac{\partial C}{\partial t} = \frac{D_{hx}}{R}\frac{\partial^2 C}{\partial x^2} + \frac{D_{hy}}{R}\frac{\partial^2 C}{\partial y^2} + \frac{D_{hz}}{R}\frac{\partial^2 C}{\partial z^2} - \frac{v}{R}\frac{\partial C}{\partial x} - \lambda C \tag{11.99}$$

where, D_{hx}, D_{hy}, D_{hz} are the coefficient of hydrodynamic dispersion in the x, y, and z directions respectively, given as

$$D_{hx} = \propto_L v + D^*, D_{hy} = \propto_T v + D^*, D_{hz} = \propto_v v + D^* \tag{11.100}$$

where, $\propto_L$, $\propto_T$, $\propto_v$ are the longitudinal, transverse, and vertical dispersivity, respectively and D^* is the diffusion coefficient.

The aquifer is bounded at $z = 0$ and h with no flow of radionuclides vertically upward or downward at the boundaries. There is no presence of the radionuclides far away from the source region where the radionuclide is present. Thus the boundary conditions are,

$$\left.\frac{\partial C(x,y,z,t)}{\partial z}\right|_{z=0} = \left.\frac{\partial C(x,y,z,t)}{\partial z}\right|_{z=h} = 0$$

$$C(x = \pm\infty, y, z, t) = 0$$

$$C(x, y = \pm\infty, z, t) = 0 \tag{11.101}$$

Given these assumptions of the initial and boundary conditions, the solution to the Eq. (11.99) can be obtained as an analytical solution. This solution, though, depends on the configuration of the source of release (e.g., point, line, area). Examples of these analytical solutions are described below (Till and Meyer 1983). These solutions are for the case of radionuclide being released instantaneously at $t = 0$ and are based on the use of the so-called Green's functions X_i, Y_i, *and* Zi . These Green's functions are the response of the system from the introduction of a unit source into the system, as defined in Table 11.8.

- Point Source: Source at $x = y = 0$ and $z = z_s$, where $0 \leq z_s \leq h$,

(a) Aquifer with finite depth

$$C(x,y,z,t) = \frac{1}{\varepsilon_f R} X_1(x,t) Y_1(y,t) Z_1(z,t) \tag{11.102}$$

(b) An infinitely deep aquifer (i.e. h → ∞): Replace $Z_1(z,t)$ by $Z_2(z,t)$.

- Vertical Line Source ($0 \leq z \leq h$) at $x = y = 0$

Table 11.8 Green's functions used in the solutions for aquifer concentrations

$X_1(x,t) = \frac{1}{\sqrt{4\pi D_{hx} t/R}}\, exp\left[-\frac{(x - vt/R)^2}{4D_{hx}t/R} - \lambda t\right]$
$X_2(x,t) = \frac{1}{4a}\left\{erf\left[\frac{x + a - vt/R}{\sqrt{4D_{hx}t/R}}\right] - erf\left[\frac{x - a - vt/R}{\sqrt{4D_{hx}t/R}}\right]\right\} exp\left[-\lambda t\right]$
$Y_1(y,t) = \frac{1}{\sqrt{4\pi D_{hy} t/R}}\, exp\left[-\frac{y^2}{4D_{hy}t/R}\right]$
$Y_2(y,t) = \frac{1}{4b}\left\{erf\left[\frac{b + y}{\sqrt{\frac{4D_{hy}t}{R}}}\right] + erf\left[\frac{b - y}{\sqrt{\frac{4D_{hy}t}{R}}}\right]\right\}$
$Z_1(z,t) = \frac{1}{h}\left\{1 + 2\sum_{m=1}^{\infty} \exp\left[-\frac{m^2\pi^2 D_{hz} t}{h^2 R}\right] \cos\left(\frac{m\pi z_s}{h}\right) \cos\left(\frac{m\pi z}{h}\right)\right\}$
$Z_2(z,t) = \frac{1}{\sqrt{4\pi D_{hz} t/R}}\left\{exp\left[-\frac{(z - z_s)^2}{4D_{hz}t/R}\right] + exp\left[-\frac{(z + z_s)^2}{4D_{hz}t/R}\right]\right\}$

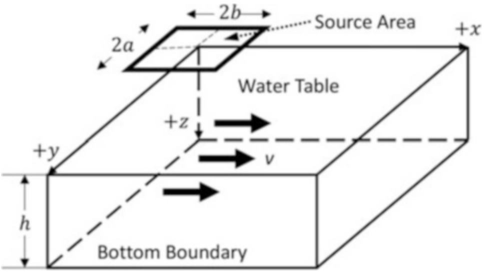


Fig. 11.31 Schematic of groundwater dispersion from a horizontal area source

$$C(x,y,z,t) = \frac{1}{\varepsilon_f R} X_1(x,t) Y_1(y,t) \frac{1}{h} \tag{11.103}$$

- Horizontal Area Source of length $l = 2a$ and width $w = 2b$ centered at (0,0,0) in an aquifer of constant depth h, (see Fig. 11.31)

$$C(x,y,z,t) = \frac{1}{\varepsilon_f R} X_2(x,t) Y_2(y,t) \frac{1}{h} \tag{11.104}$$

- Vertical Area Source $(-b < y < b,\ 0 < z < h)$ at x = 0

$$C(x,y,z,t) = \frac{1}{\varepsilon_f R} X_1(x,t) Y_2(y,t) \frac{1}{h} \tag{11.105}$$

These solutions of the instantaneous release cases can be also used to describe the situation of continuous release. In that case, the outcome of the release from each time point needs to be added together to represent the cumulative effect of the continuous releases. This can be done by using the concept of convolution integral as follows.

$$C(x,y,z,t) = \int_0^t d\tau\, \widehat{C}(x,y,z,t-\tau)\, Q(\tau) \tag{11.106}$$

where, $\widehat{C}(x,y,z,t-\tau)$ is the solution at time $(t-\tau)$ for an instantaneous release at $(t-\tau) = 0$, and $Q(\tau)$ is the source release rate at τ.

Analytical solutions to the advection and dispersion equation are the simplified representation of the space- and time-varying concentrations of contaminants under the specified assumptions. For example, the specified assumptions can be about the initial presence (or non-presence) of contaminants (initial conditions) or the distributions in the overall domain of interest (boundary conditions). Specifying the applicable initial and the boundary conditions allows a unique solution to the equation.

Applicability of the analytical solution approach in a given problem depends on the applicability of the simplifying assumptions employed to describe hydrogeological characteristics at the site including the homogeneity and isotropy of the porous medium, a unidirectional velocity field, and a simple flow domain geometry.

Due to the simplicity in its approach, analytical solutions may not provide accurate results for the space-time dependent concentration profiles of the contaminant under investigation. Nevertheless, they are useful for screening analysis or for testing and benchmarking the numerical solution results from using computer models. Also, when the available data do not warrant the use of a complex detailed model, analytical solutions are as good as the results from the use of complex computational models.

If the site under consideration is complex and the conceptual model for the sites' hydrogeology cannot warrant the use of simplifying assumptions, use of computer models based on space-time dependent analysis should be pursued. In this case, numerical solution methods such as finite difference method, finite element method, or method of characteristics are employed to solve the advection and dispersion equation. The approach allows the incorporation of the site's hydrogeology or geometry or the heterogeneities of the system in a more realistic way. Such an approach comes with complexity and increased cost in the analysis requiring extensive use of computational resources. The efforts to prepare the input data to represent site-specific features of the parameters should also be made.

Example 11.9: Contaminant Groundwater Concentration at a Well

Assume that one curie of a radionuclide is spilled out of a storage system through a highly permeable unsaturated zone resulting in a contamination of an unconfined aquifer. The resulting initial contamination can be assumed an area source covering a square area (50 m × 50 m) located on the surface. From the locations of radionuclide leakage, the groundwater is moving toward a drinking water well (x = 200 m, y = 0 m) with an average linear velocity of 1.5 m/day. Assume that the radionuclide's half-life is very long compared to the travel time to the well. The aquifer has a depth of 50 m with an effective porosity of 0.2. The longitudinal and transverse dispersivities ($\propto_L$ and $\propto_T$) are 20, 10 m, respectively. The well is screened over the entire depth of the aquifer. The retardation factor of the radioactive pollutant in the medium is 20.

Determine the concentration of the radionuclide at the location of the well.

Solution:

The source is a horizontal area type and, at the receptor location, water is collected over the total depth of the well. Eq. 11.104 applies to this example (the release is assumed to be instantaneous):

$$C(x,y,z,t) = \frac{1}{\varepsilon_f R} X_2(x,t) Y_2(y,t) \frac{1}{h},$$

where

(continued)

Example 11.9 (continued)

$$X_2(x,t) = \frac{1}{4a}\left\{ erf\left[\frac{x+a-vt/R}{\sqrt{4D_{hx}t/R}}\right] - erf\left[\frac{x-a-vt/R}{\sqrt{4D_{hx}t/R}}\right]\right\} exp\left[-\lambda t\right]$$

$$Y_2(y,t) = \frac{1}{4b}\left\{ erf\left[\frac{b+y}{\sqrt{4D_{hy}t/R}}\right] + erf\left[\frac{b-y}{\sqrt{4D_{hy}t/R}}\right]\right\}$$

Given:

$x = 200\ m$
$y = 0\ m$
$a = b = \frac{50}{2} = 25\ m$ (see Fig. 11.31)
$h = 50\ m$
$R = 20$
$\varepsilon_f = 0.2$
$v = 1.5\ m/day$

The coefficients of hydrodynamic dispersion for a unidirectional transport from Eq. 11.100, assuming the effect of molecular diffusion is negligible, become:

$D_{hx} = \propto_L v = 20 \times 1.5 = 30\ m^2/day$
$D_{hy} = \propto_T v = 10 \times 1.5 = 15\ m^2/day$

If half-life is very long, it can be assumed that effect of radioactive decay is negligible for $X_2(x, t)$

(i.e. $exp[-\lambda t] \sim 1$)

Plugging in the above values for Eq. 11.104 to get concentration as a function of time, we get:

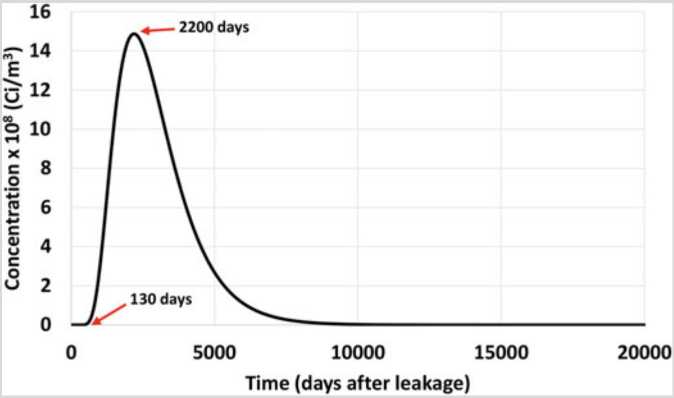
$$\begin{aligned} C(x,y,z,t) &= \frac{1}{\varepsilon_f R} X_2(x,t) Y_2(y,t) \frac{1}{h} \\ &= \frac{1}{0.2 \cdot 20} \cdot \frac{1}{4(25)} \\ &\quad \times \left\{ erf\left[\frac{200+25-\frac{1.5t}{20}}{\sqrt{4\cdot\frac{30t}{20}}}\right] - erf\left[\frac{200-25-\frac{1.5t}{20}}{\sqrt{4\cdot\frac{30t}{20}}}\right]\right\} \cdot \frac{1}{4(25)} \\ &\quad \times \left\{ erf\left[\frac{25+0}{\sqrt{4\cdot\frac{15t}{20}}}\right] + erf\left[\frac{25-0}{\sqrt{4\cdot\frac{15t}{20}}}\right]\right\} \cdot \frac{1}{50} \end{aligned}$$

A plot of the solution is shown below as a function of time.

(continued)

Example 11.9 (continued)

(Note that **y-axis is the concentration multiplied by 10^8**, because concentration is very small, meaning the actual concentration is the value in the figure divided by 10^8)



The results indicate that the contaminant plume arrives at around 130 days at the drinking well but the peak arrives about 6 years after the initial spill at the concentration of ~0.15 µCi/m^3 (~0.0055 Bq/ml).

11.4 Effects of Geochemistry on the Migration of Radionuclide in Groundwater

Through interactions with the components of the soils and rocks in the porous medium, migration of chemically reactive radionuclides in groundwater is subject to the effect of local chemistry. Precipitation and dissolution reaction will limit the concentration of some of the radionuclides in the solution. Aqueous complexation and oxidation/reduction reactions also affect solubility of species while controlling their distributions between the water and the solid phase. Such distributions will also be affected by other surface related phenomena. These chemistry effects are discussed below mainly regarding the changes in the mobility and solubility of radionuclides in groundwater.

11.4.1 Effect of Chemistry in Near-Field and Far-Field

As contaminants migrate away from their release points, they will encounter progressive changes in the conditions of the environment (e.g., the changes in rock composition) along with variations in the chemical species dissolved in groundwater. These changes affect the prevailing pH, oxidation potential (Eh), temperature, and ligand concentrations, which in turn control the mobility of radionuclides along with their solubility in the groundwater.

Most natural waters are near neutral with the pH ranging between 5 and 9 but the redox potential changes widely between −300 to +500 millivolts. Natural waters also contain a variety of ligands as agents of chemical reactions. Most commonly encountered ligands in natural waters include bicarbonate/carbonate (HCO_3^-/CO_3^{2-}), hydroxide (OH^-), Cl^-, SO_4^{2-}, and humic substances. Significant alterations in solubility and sorption characteristics of the radionuclides are possible within the conditions of natural waters.

The effects of geochemistry on radionuclide migration are also different depending on the characteristics of the surrounding settings of. These settings are classified into the near-field and the far-field region of a geological repository.

Near-field is a zone including the engineered barrier systems and the immediate surrounding rocks extending to the excavated repository. The region may extend for some meters or tens of meters depending on the disturbances created by the presence of nuclear waste and the degree of time dependent changes in the chemical and environmental conditions. The chemical reactions in this zone may not be in equilibrium due to possible presence of steep gradient in pH and/or Eh in the region. These chemical reactions along with groundwater infiltration directly affect material degradation (corrosion of waste packages) and radionuclides release characteristics. These reactions are also affected by the presence of heat and radiation from nuclear waste. The resulting radionuclides release due to the degradation of engineered barriers constitute the source term for the repository safety assessment.

Far-field is the natural geological system away from the nuclear waste repository. Compared to the near-field, the far-field is undisturbed, in a relatively ‘steady state’ with regard to chemistry, temperature, and hydrology. Any changes taking place are subject to normal rates of slow geological evolution. The far-field controls the infiltration of groundwater into the near-field and, as a long-distance migration route, determines the eventual fate and transport of radionuclides.

In both the near-field and the far-field, solubility and sorption are very important in controlling the fate and transport of radionuclides ultimately affecting safety performance of geologic repository. The solubility of radionuclides controls the leaching from the nuclear waste forms into the surrounding water and their concentration in groundwater in both near-field and far-field. The sorption characteristics will control the rate of radionuclide migration in groundwater in both the near-field and the far-field.

11.4.2 Solubility

Solubility, as the maximum Far-field is the natural geological system away from the nuclear waste repository. Concentration of a chemical species dissolved in water at equilibrium, reflects the extent to which the reactant (radionuclide in solid) or products (ions in solution) are favored in a dissolution-precipitation reaction. The radionuclides of major importance in nuclear waste can be in two groups in terms of solubility in groundwater:

1. Those radionuclides that would react with the water to form compounds that are less soluble than the waste matrix from which they came.

These include Np, Pu, and Tc. Under the slightly alkaline and reducing conditions (typical of most groundwater), they become very insoluble oxides (NpO_2, PuO_2, Tc_3O_4) through precipitation reaction. Therefore, if they are liberated from the waste, their concentrations in groundwater will be effectively constrained by their low solubility.

2. Nuclides with no solubility constraints (Sr, Cs, I, and Ra)

The nuclides of Sr, Cs, I and Ra have no defined 'solubility' or their solubility is very high (or non-limiting). Therefore, when these radionuclides dissolve in groundwater, their concentrations will be a function of the dissolution rate of nuclear waste forms and the flow rate of the groundwater in the system, not being controlled by solubility.

Numerical values of solubility can be estimated by using thermodynamic data or by laboratory experiments under repository conditions. The values given in Table 11.9 are examples for the elements of interest in repository safety assessment. The values are given for four different combinations of Eh and pH. The maximum permissible concentrations (MPCs) are also listed for comparison purposes. The MPC represents the level of concentration in water as the maximum allowable level through drinking water consumption not to violate dose limits for a member of the public.

Table 11.9 Comparison of solubility of major nuclides: concentration (mol/L) of each nuclides in equilibrium with its most insoluble compound under the specified Eh and pH at 25 °C and 1 atm

	Reducing Conditions		Oxidizing Conditions	
	Eh = −0.2 V		Eh = +0.2 V	
Nuclide	pH 9	pH 6	pH 9	pH 6
Se	$>1 \times 10^{-5}$	$>1 \times 10^{-5}$	$>1 \times 10^{-5}$	$>1 \times 10^{-5}$
Sn	8×10^{-10}	8×10^{-10}	8×10^{-10}	8×10^{-10}
Sr	7×10^{-6}	~ 1	7×10^{-6}	~ 1
Cs	~ 1	~ 1	~ 1	~ 1
I	$\sim 2 \times 10^{-3}$	$\sim 2 \times 10^{-3}$	$\sim 2 \times 10^{-3}$	$\sim 2 \times 10^{-3}$
Tc	10^{-15}	$>1 \times 10^{-5}$	$>1 \times 10^{-5}$	$>1 \times 10^{-5}$
U	4×10^{-9}	4×10^{-11}	$>4 \times 10^{-6}$	$>4 \times 10^{-6}$
Np	4×10^{-10}	4×10^{-10}	4×10^{-8}	4×10^{-7}
Pu	4×10^{-11}	4×10^{-10}	4×10^{-11}	4×10^{-9}
Am	4×10^{-14}	4×10^{-11}	4×10^{-14}	4×10^{-11}
Ra	4×10^{-9}	4×10^{-7}	4×10^{-9}	4×10^{-7}
Pb	5×10^{-7}	5×10^{-6}	5×10^{-7}	5×10^{-6}

Table 11.10 Species of Actinides in Aqueous Solution

	Oxidation State				
Nuclide	III	IV	V	VI	VII
Tc		TcO_2			TcO_4^-
Th		Th^{4+}, ThO_2			
U	U3+	U^{4+}, UO_2, UOH^{3+}	UO_2^+	UO_2^{2+}, $UO_2(OH)_2$, $UO_2(OH)_3^-$	
Np	Np^{3+}	Np^{4+}, NpO_2, $Np(OH)_4$ $Np(OH)^{3+}$ $Np(OH)_2OH$	NpO_2^+, NpO_2OH	NpO_2^{2+}, $NpO_2(OH)^+$, $NpO_2(OH)_2$	
Pu	Pu^{3+}, $PuOH^{2+}$	Pu^{4+}, PuO_2	PuO_2^+	PuO_2^{2+}, $PuO_2(OH)_2$	
Am	Am^{3+}, $AmOH^{2+}$, $Am(OH)_2^+$	AmO_2	AmO_2^+		
Cm	Cm^{3+}	CmO_2			

The first column, i.e., the reducing conditions at pH 9, represents the most likely Eh-pH combination expected in repository water (slightly alkaline and slightly reducing condition). The solubility values are in general low with Sn, Pu, Am while some elements show high solubility (Se, Sr, Cs, I). Also, from the table, it is noted that the solubility of certain elements (Tc, Np, U) changes rather significantly between the oxidizing and reducing conditions. These large variations are associated with the Eh condition changes from oxidizing to reducing, resulting in the soluble species being precipitated as insoluble oxide. This is discussed further below. Notable changes in solubility are also observed between pH 9 and pH 6 for Sr, Am, and Ra.

Table 11.10 shows the available oxidation state of select elements of interest in aqueous solutions. Please be reminded that the electrons of the actinides are located in the 5f, 6d, and 7 s subshells as discussed in Sect. 4.1.2. Compared with the lanthanides whose electrons are in the 4f subshells, these electrons extend father from the nucleus and are more loosely bound than the ones in the filled subshells (with binding energies of a few eV). Therefore, many actinides can easily loose the electrons in the 6d or 7 s subshells exhibiting multiple oxidation states.

Possible oxidation states of these actinides (also shown in Fig. 8.5) are summarized below for the likely observable states in natural waters.

Th ($\underline{\mathbf{4}}$), U (3, **4**, $\underline{\mathbf{6}}$), Np (3, **4**, $\underline{\mathbf{5}}$, 6), Pu (**3**, $\underline{\mathbf{4}}$, 5, 6), Am ($\underline{\mathbf{3}}$,4), Cm ($\underline{\mathbf{3}}$,4)

Here the common oxidation states in natural waters are bolded and the most prevalent one in the repository groundwater is underlined. The most stable oxidation state is +6 for U, +5 for Np, +4 for Pu and Th, and + 3 for Am and Cm. Other stable

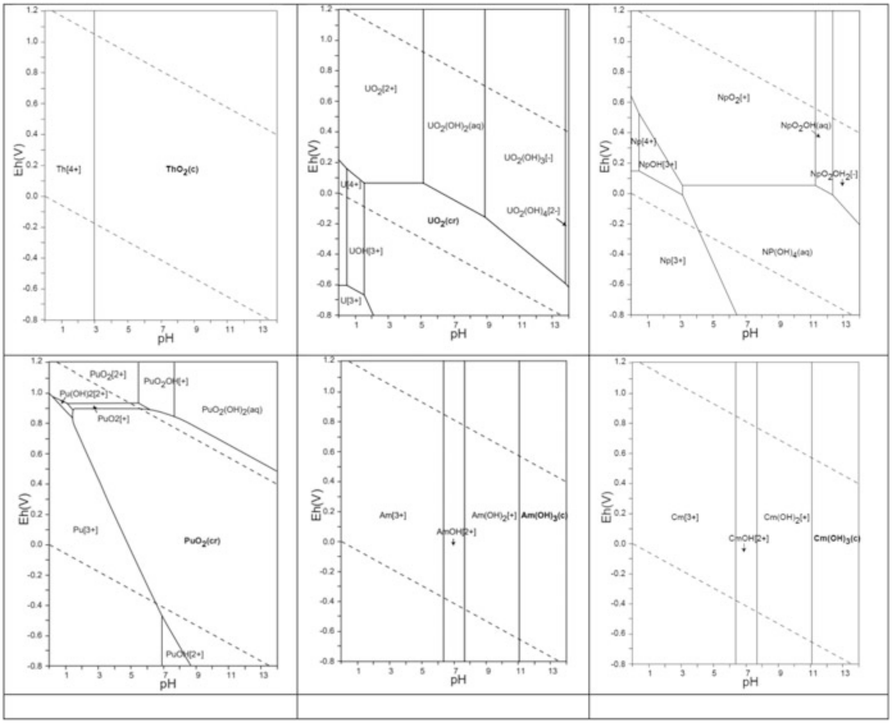


Fig. 11.32 Eh-pH Diagram of the Actinides (Takeno 2005)

oxidation states include +4 for U and Np, and + 3 for Pu. Am and Cm are mostly likely in the +3 state in natural water.

The chemical forms that are associated with different oxidation states are shown in Table 11.10. Figure 11.32 also shows the dominant aqueous species associated with different oxidation states under the given Eh-pH conditions. The Eh-pH diagrams of the actinides in Fig. 11.32 assume that the total concentration of elements is low at 10^{-10} mole/kg at 298.15 K and 10^5 Pa. The expression used in the diagrams for the solid includes “c” or “cr” (crystalline), or “s” (solid). The Eh-pH diagram of iodine and technetium, two other major elements of interest in repository safety assessment, are also shown for comparison in Fig. 11.33.

As to the Eh-dependent changes in solubility (Table 11.9), neptunium, for example, exists as NpO_2^+ ($Np^{(V)}$) under oxidizing conditions which is soluble. Under reducing conditions, neptunium is reduced to $Np^{(IV}$ and becomes insoluble NpO_2. The same also occurs with uranium. Uranium is soluble as UO_2^{2+} or $UO_2(OH)_2$ ($U^{(VI)}$) in the oxidizing environment but becomes reduced to $U^{(IV)}$ as UO_2 under reducing conditions with much reduced solubility. Uranium as $U^{(IV)}$ can exist in the form of UOH^{3+}but only under highly acidic conditions. While somewhat similar behavior is observed with plutonium, the solubility of plutonium is generally low. In the case of Am, the variation of solubility between the oxidizing and

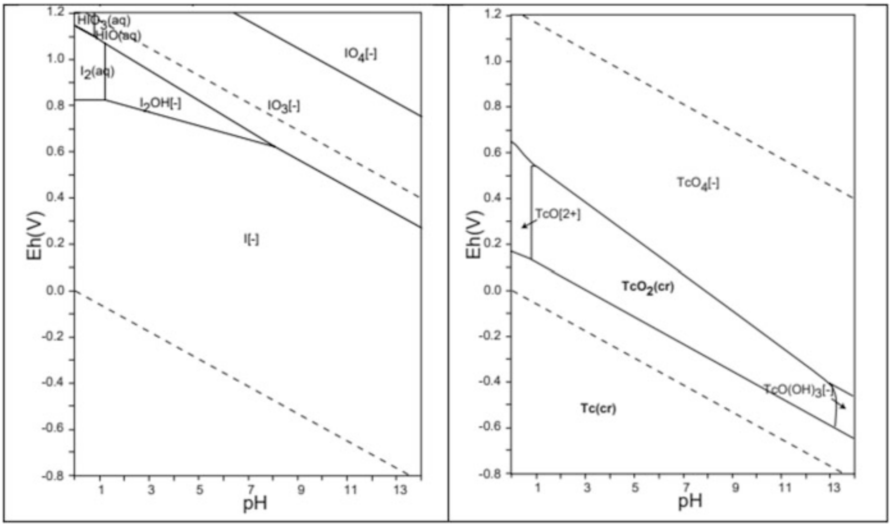


Fig. 11.33 Eh-pH Diagram of Iodine and Technetium (Takeno 2005)

reducing conditions is not observed. This is because Am is always present in trivalent form in natural water (as the other oxidation states are not really stable in aqueous solution). Americium in the trivalent form has in general low solubility.

Another observation from Table 11.9 is that the solubility also depends on the pH in the system. In fact, most of the element with low solubility show pH dependent solubility characteristic: In general, solubility decreases with pH increase. This can be related to the increase in c*arbonate* concentrations from *the conversion of* CO_2. *Increase in the presence of carbonate ligands raises the chance of complex formations and lowers the solubility of the actinides.* Increase in pH can also be associated with higher concentration of hydroxide ions (OH^-) increasing the formation of hydroxide complexes which may lead to lower solubility. This variations in solubility with pH change are also noted with other ionic species such as strontium and radium as shown in Table 11.9. An exception is noted for uranium in Table 11.9, i.e., solubility increases with pH increase. This behavior would be due to the conversion of UO_2 ($U^{(IV)}$) to $UO_2(OH)_2$ ($U^{(VI)}$) under the reducing conditions as captured in Fig. 11.32.

Another element of interest in this discussion is technetium (transition metal). Technetium exists predominantly as highly soluble pertechnate ion TcO_4^- ($Tc^{(VII)}$) under the oxidizing conditions. Then, under the reducing conditions, the pertechnate is reduced to $Tc^{(IV)}$ and is precipitated as insoluble TcO_2 (see Fig. 11.33).

There are elements whose solubility is always very high as shown in Table 11.9. These elements, such as iodine or cesium, always exists as ions (I^- or Cs^+, respectively) under both oxidizing and reducing conditions and is soluble in water (see also Fig. 11.33). In general, in a polar substance like water, a substance existing as ionic species is soluble.

In general, the concentration of the actinides in natural waters are generally low, in the order of 10^{-6} molar or lower (Runde 2000). In the case of typical groundwater conditions of Yucca Mountain, plutonium is likely to be in the IV oxidation state thus having low solubility. In contrast, neptunium is likely to be in the V oxidation state which is far more soluble than the IV oxidation state. In this case, neptunium becomes a major nuclide of concern in safety assessment. However, if the salinity of the groundwater is high such as in a repository in salt rock (such as at the WIPP site in New Mexico), plutonium favors the VI oxidation state which renders Pu to be much more soluble than in the IV state. Therefore, plutonium would be a major nuclide of concern at WIPP.

Increase in solubility beyond the solubility limit can also occur. Such cases are through formation of complexes with other constituents in the groundwater or through colloid formation. Sources of mobile colloids in groundwater include dispersed subsurface soils, dissolved secondary mineral phases, and precipitated constituents of groundwater.

In summary, the solubility of certain elements, such as the actinides and technetium, are very sensitive to Eh and pH. If these elements remain in the engineered barrier systems of a geological repository, they are in the low oxidation states (III or IV) as the system is typically reducing. Accordingly, the actinides and technetium will be maintained as insoluble oxides. Corrosion of metallic containers or presence of microbial activities also contribute to the system to be reducing. The conditions may change, however, in the far field, away from the repository. If the groundwater in the far field is slightly more oxidizing or more acidic, these radionuclides may become soluble and become part of groundwater migration.

Recently, the possibility of using microorganisms to provide the reducing geochemical environment is being considered. Microorganisms were also found to be capable of selective removal of certain radionuclides. If such approach can become technically mature, microorganism-based control of local geochemistry may provide another natural barrier against the migration of radionuclides in geological disposal of nuclear waste.

11.4.3 Distribution Coefficients (K_d)

The value of K_d of an element depends on its chemical properties subject to the local chemistry conditions. Key chemistry conditions affecting K_d are the pH and Eh of the solution, ionic strength, ionic radius, presence of ions that compete for the sorption sites, extent and nature of the solid surfaces providing sorption sites, and presence of ligands that can form stable complexes with the dissolved ions. The chemical reactions that could control the K_d values of elements include aqueous complexation, dissolution-precipitation, oxidation-reduction, acid-base reaction, and ion-exchanges.

The pH of water plays a particularly prominent role. The increase in pH raises the chance of complex formation through the promotion of the presence of ligands (e.g.,

Table 11.11 The estimated values of K_d (ml/g) of Pu (EPA 1999b)

	Clay Content (wt. %)								
	0–30			31–50			51–70		
	Soluble Carbonate (meg/l)			Soluble Carbonate (meg/l)			Soluble Carbonate (meg/l)		
Kd	0.1–2	3–4	5–6	0.1–2	3–4	5–6	0.1–2	3–4	5–6
Minimum	5	80	130	380	1440	2010	620	1860	2440
Maximum	420	470	520	1560	2130	2700	1980	2550	3130

carbonate and hydroxide ions). As the resulting aqueous complexes are negatively charged and, as soil surfaces are negatively charged, the complexes tend to have lower sorption on soil surfaces. Therefore, under high pH, K_d of radionuclides is expected to be lower in general. The pH also affects a number of aqueous and solid phase properties which directly control sorption. These pH dependent properties include surface charge of solid surfaces, competition for sorption sites, and cation-exchange capacity (CEC) of soil. For example, pH increase causes the solid surfaces to become increasingly more negatively charged with the buildup of aqueous complexes increases. This causes reduction in K_d. Presence of various ionic species, through pH dependent species dissolution, would compete for sorption sites leading to reduction in K_d. The CEC, referring to the amount of positive charge that can be retained on soil surfaces per mass of soil, increases with pH and results in the increase in K_d. Higher clay contents or organic matter presence in soil also increase sorption by providing additional sorption sites (see Table 11.11 for the example of plutonium).

The sorption behaviors of actinides are strongly affected by the oxidation state related changes, the effect of dissolved carbonates, and pH. By changing the effective charges, redox related aqueous complexation also changes the preference of actinides to form complexes. The effective charge of actinides varies with the changes in the oxidation state (Runde 2000). This change of effective charge of the central actinide ion at different oxidation states is shown in the following (*An* stands for actinides):

Effective charge: +4 with An^{4+} ($An^{(IV)}$); +3.3 with $AnO2^{2+}$ ($An^{(VI)}$); +3 with An^{3+} ($An^{(III)}$); +2.3 with $AnO2^{+}$ ($An^{(V)}$)

According to these observations, *An*(IV) with the highest effective positive charge is expected to form strong bond to the anionic ligands by electrostatic interactions. The results will be the production of the most stable complexes and also the most stable precipitates. *An*(V) or *An*(III), with the lowest effective charge, is expected to form weaker complexes and less stable precipitates. Accordingly, with *An*(IV), high degree of sorption and lowest solubility is expected while low sorption and high solubility is expected with *An*(V) or *An*(III).

In general, this behavior applies to plutonium, neptunium, and uranium. As Pu (IV) is the most stable and common Pu species in natural water, Pu(IV) is expected to have limited mobility with low solubility in groundwater. In terms of specifics, two most significant factors in describing the sorption of plutonium are dissolved carbonate concentrations and the clay content of the soil (EPA 1999b). The effect of pH was found insignificant with plutonium. The following table shows the changes in the K_d value of Pu with variations in soluble carbonate and clay content (Table 11.11).

In the case of Np, the +5 state (with the weakest effective charge) is the most stable and common oxidation state. Accordingly, neptunium is expected to be the most soluble and transportable actinide in groundwater. However, if reduction from the +5 state to +4 state under the reducing conditions occurs, such change would increase sorption and also solubility. Sorption of neptunium is found to be strongly dependent on pH and carbonate concentrations.

For uranium, important factors influencing its sorption behavior include oxidation state, pH, dissolved carbonate concentrations and the clay/organic matter content. Importance of these factors are captured in Table 11.12 where distinctively different values of K_d are noted between U(IV) and U(VI). In general, the K_d value is highest at neutral pH and increases with higher presence of fine matters (clay/organic matter/hydrous-oxide). A very significant changes in the K_d value of uranium are observed with the changes in the fine matter content.

Americium exists mostly in the +3 oxidation state in natural waters as uncomplexed Am^{3+} in moderately to highly acidic conditions and as aqueous carbonate complex $AmCO_3^+$ in near neutral to alkaline conditions. In both cases, americium exhibits high sorption behavior.

Oxidation/reduction reactions also influence the sorption capacity of solid surfaces or affect aqueous complexation. For example, ferrous mineral oxides provide strong sorption sites for metals, but, when the mineral oxides dissolve due to the changes in the redox condition, the sorption potential of the surface is reduced.

The radionuclides in cationic form such as cesium or strontium generally exhibit high sorption behavior. Their sorption occurs through cation exchange, strongly controlled by the CEC of the soil. Presence of competing ionic species is also an important controlling factor. Experiments conducted with cesium and strontium show that their sorption capacity decreases with increasing salinity of groundwater due to the presence of the sodium ions in the solution. If salt is chosen as host rock of a repository, high content of sodium in the groundwater would limit the sorption of many of the radionuclides (including the actinides) in the paths of migration.

Table 11.12 The estimated values of Kd (ml/g) of U (overall), U(IV) and U(VI) (EPA 1999c)

Material	pH								
	≥9			5–9			≤5		
Fine matters (%)[a]	<10	10–30	>30	<10	10–30	>30	<10	10–30	>30
U (overall)	0	5	50	0	50	500	0	5	50
U(IV)	200	500	1000	100	250	500	20	30	50
U(VI)	0	1	2	1	2	5	2	5	20

[a] Fine matters (%) = sum of the percentage of clay, organic matter, and hydrous-oxide in soil

Table 11.13 The ranges of Kd values for important radionuclides in rock materials being considered for repository siting (McKinley and Scholtis 1992) (The values in the parenthesis are considered suitably conservative)

	Granite	Basalt	Tuff	Clay/Shale	Salt
Se	0.4–20 (5)	0.4–20 (5)	0.4–20 (5)	0.4–20 (5)	2–100 (20
Sn	10–500 (100)	10–500 (100)	20–500 (100)	20–500 (100)	1–100 (10)
Sr	1–200 (20)	5–200 (20)	2–1000 (20)	5–500 (20)	0–10 (1)
Cs	10–1000 (100)	10–1000 (100)	6–1000 (50)	20–20,000 (100)	0–200 (1)
I	0	0	0	0	0
Tc	0–4 (0.4)	0–10 (0.4)	0–10 (0.4)	0–2 (0.4)	0–2 (0.4)
U	1–50 (5)	2–100 (5)	0.4–20 (4)	5–500 (20)	1–6 (2)
Np	1–50 (10)	1–50 (10)	1–50 (10)	1–40 (10)	1–30 (5)
Pu	1–500 (20)	10–500 (50)	5–500 (20)	50–2000 (100)	1–1000 (20)
Am	50–5000 (300)	6–5000 (50)	30–5000 (100)	20–5000 (80)	20–500 (100)
Ra	5–500 (50)	5–500 (50)	5–500 (50)	5–500 (50)	0.4–50 (5)
Pb	1–20 (5)	2–50 (5)	2–50 (5)	2–50 (5)	0.4–10 (2)

The radionuclides present in predominantly anionic forms (e.g., I^-, Cl^-, TcO_4^-, SeO_4^{2-}) show low sorption behavior, resulting in low K_d values. Their movements are insensitive to migration field composition. This is shown in Table 11.7 where typical values of K_d in different types of soils are given. As discussed in Sect. 11.3.6, iodine, one of the most significant nuclides affecting the long term performance of a repository, has very low values of K_d. In the case of Tc, +7 state in anionic forms (TcO_4^-) under the oxidizing conditions imply negligible effect of sorption. When Tc is reduced to Tc(IV), however, the product is an insoluble oxide which is essentially immobile.

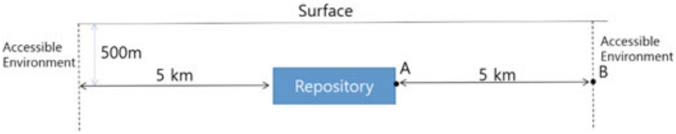
Typical values of K_d for major elements of interest are summarized in Table 11.13 for major host rocks of geological repository. The values given in the parenthesis are the representative but conservative estimates. The high value in each range is for the most common repository conditions (Eh: -0.2 to 0.3 V, pH 7 to 9) while the low figure is an estimate of the minimum value under less favorable conditions of Eh, pH, or complexing. The numbers for salt are given for the case of sorption on ordinary rock material due to the movement of high-salinity groundwater from a salt repository.

11.5 Conclusion

Migration of dissolved radionuclides in groundwater provides a connection between the engineered barrier systems of a geological repository and the biosphere. This chapter described how groundwater transport and contaminant migration in the porous rock medium can be described, subject to the natural site conditions of hydrology, geology, and geochemistry. Use of models was emphasized in the discussions for quantitative modeling of radionuclide migration in groundwater. Importance of chemical reactions in describing the migration of radionuclides in groundwater was also discussed, highlighting the role of sorption and solubility of elements. In particular, as heavy elements often have multiple valences and feature sharply contrasting solubilities and sorption characteristics in different chemistry environments, the importance of the redox potential (Eh) of the system was noted. In this regard, presence of chemically reducing conditions is emphasized as desirable to provide favorable natural barrier conditions.

Homework

Problem 11.1: The accident at Three Mile Island Unit 2 in March 1979 occurred approximately 4 months[2]. A site with the following characteristics has been proposed for the first nuclear waste repository in your country. The repository is to be located in a granite rock body at a depth of 500m. The rock is situated in a regional groundwater basin in which the hydraulic gradient has a magnitude of 2×10^{-3} in the horizontal direction. The effective porosity of the rock is 2%. The hydraulic conductivity is 10^{-6} m/s.



(a) The depth from the surface to the water level in a piezometer located at point A (in the figure) is 250 m. Estimate the groundwater pressure (in N/m2) at point B.
(b) U.S. NRC's 10CFR Part 60 requires a minimum groundwater travel time of 1000 years from the proposed outer boundary of the repository to the "accessible environment". Would the proposed site meet this requirement?

Problem 11.2: Your team of environmental scientists has monitored the concentration of a pollutant in an aquifer. One set of data was taken on April 1, 1990. The second set was taken a year later. The pollutant, a chloride ion, is expected to flow at the same average velocity as the groundwater.

The data shown in the following represent the concentration of the chloride ion at various positions in the aquifer. Use these data to estimate:

(a) The average linear velocity of the groundwater,
(b) The date when the chloride ion was introduced into the water,
(c) The location of the discharge, and
(d) The hydrodynamic dispersivity

On April 1, 1990

x(m)	0	10	20	30	40	50	60	70	80	90	100
Concentration	0.0	0.01	0.05	0.13	0.31	0.60	0.99	1.35	1.53	1.45	1.14
x(m)		110	120	130	140	150	160	170	180	190	200
Concentration		0.75	0.41	0.19	0.07	0.02	0.01	0.00	0.00	0.00	0.00

On April 1, 1991

x(m)	300	310	320	330	340	350	360	370	380	390	400
Concentration	0.00	0.00	0.01	0.01	0.03	0.05	0.09	0.15	0.23	0.33	0.45
x(m)		410	420	430	440	450	460	470	480	490	500
Concentration		0.58	0.71	0.81	0.87	0.88	0.84	0.75	0.64	0.50	0.38
x(m)		510	520	530	540	550	560	570	580	590	600
Concentration		0.26	0.17	0.11	0.06	0.03	0.02	0.01	0.00	0.00	0.00

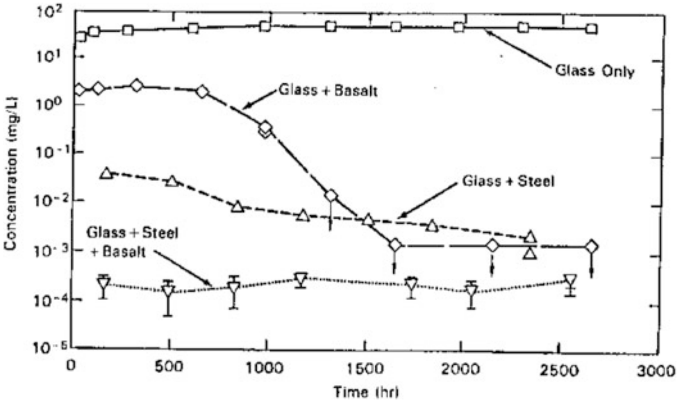
Problem 11.3: The following Table 11.6 shows the examples of K_d values of various radionuclides in groundwater. By using the same problem given in Example 11.7 and the given values of K_d for the radionuclides, estimated the expected travel time to the Accessible Environment (i.e., 5 km distance). Determine also how many half-lives of each radionuclide are needed to reach the Accessible Environment. Discuss the implications of the results.

Table: The K_d values of selected radionuclides

Species	K_d
^{226}Ra	500
^{99}Tc	20 (1–100)
^{129}I	1 (0–1)
^{135}Cs	700
^{239}Pu	400
^{234}U	10,000
^{237}Np	100

Problem 11.4: Explain which chemical reactions affect the solubility and sorption characteristics of actinides in the aqueous system. Describe why such effects occur.

Problem 11.5: The following figure show the approximate results of standard leaching experiments using a technetium-doped borosilicate glass under different conditions. As seen in the figure, the amount of leaching is the highest when only the glass samples are in the water. The other experiments were performed by replacing about the half of the glass samples with basalt rock or steel bars or both. Explain why the results of technetium leaching change in different conditions.



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Chapter 12
Performance Assessment of Geological Repository

Abstract Final disposal of nuclear waste requires the projection of the long-term behavior of the geological repository, through performance assessment, to demonstrate regulatory compliance with safety standards. Performance assessment considers natural as well as human initiated disruptions to describe the state of nuclear waste isolation and impacts of the failures of isolation on humans and the environment. This chapter describes how performance assessment is conceptualized and conducted and how the results are interpreted.

Keywords Performance assessment · Waste isolation failure scenarios · Uncertainty · Monte Carlo method · Regulatory compliance

For the demonstration of regulatory compliance with safety standards, disposal of nuclear waste in a geological repository requires the projection of the potential long-term impacts of placing nuclear wastes in the facility on humans and the environment. Such projection is termed performance assessment, i.e., performance assessment of geologic repository. This chapter discusses how performance assessment is conceptualized and conducted and how the results should be interpreted.

12.1 Definition of Performance Assessment

Performance assessment (i.e., repository safety performance assessment) is an integrated safety assessment of a geological nuclear waste repository. The assessment is mainly to examine whether the system meets the safety standards set by the regulatory authority. In this sense, performance of a repository means the measure of safety in nuclear waste disposal. Typically such safety measure refers to potential releases of radioactivity from a repository or potential radiation dose to the affected individuals or impacts on the environment over specified time periods. The time periods covered by performance assessment are at least 10,000 years or up to a

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https://doi.org/10.1007/978-94-024-2106-4_12

million years given the long-term presence of radioactive materials in nuclear waste (see also Sect. 2.4.2).

As performance assessment is to determine regulatory compliance of a system for the future, i.e., protection of the public and the environment in the long-term future, such compliance demonstration cannot be based on experimental results but requires the use of predictive computational models. These models are the tools for the projections of the future conditions and evolution of the geological repository system reflecting the best available knowledge of today. In fact, it is very difficult, if not impossible, to use models to predict the future as models do not represent the reality.

This implies that the goal of performance assessment is not to predict the future but to provide the basis for the judgment on regulatory compliance. The compliance is with respect to performance requirements in the future for geological disposal of nuclear waste. Through performance assessment, a reasonable assurance must be provided that the geological repository will remain safe during the specified time periods of interest (NCRP 2005).

Performance assessment for a geologic repository is sometimes called "Total System Performance Assessment (TSPA)" as the assessment involves descriptions of the total repository system. The description is to project consequences of nuclear waste disposal based on integrative understanding of the system behavior. For the purpose, models are set up to represent the evolution of physical phenomena that may ultimately lead to exposure of humans to radiation from the materials released from the waste. The questions addressed in performance assessment include: How and under what circumstances are radionuclides released from a repository? How likely are such releases? What could be the consequences of such releases to humans and the environment?

12.1.1 Meaning of Performance in Performance Assessment

While performance of a geological repository serves as a regulatory measure of safety in nuclear waste disposal, the required performance varies from country to country. Please refer to Sect. 2.4.2 where an overview of the considerations for the required performance is provided.

Most countries adopt annual dose limits as the primary performance requirement. Lifetime cancer risk is sometimes used. As discussed in Sect. 2.4.2, other performance requirements used include the limits on cumulative or annual release of radionuclides, groundwater travel time, or requirements on the containment period for waste packages. Examples of performance requirements for the geological repositories in the U.S. and Finland are as follows.

U.S. Nuclear Regulatory Commission (NRC 2009): ". . .reasonable expectation that the reasonably maximally exposed individual receives no more than the following annual dose from releases from the undisturbed Yucca Mountain disposal system: (1) 0.15 mSv for 10,000 years following disposal; and (2) 1.0 mSv after 10,000 years up to 1,000,000 years, but within the period of geologic stability".

Radiation and Nuclear Safety Authority of Finland (STUK 2018): “The disposal of nuclear waste shall be so designed that the radiation impacts arising as a consequence of expected evolution: a) the annual dose to the most exposed individuals remains below the value of 0.1 mSv; and b) the average annual doses to other individuals remain insignificantly low.” . . . “. . .constraints for radioactive releases to the living environment (average release of radioactive substances per annum) . . . are as follows: 0.03 GBq/year for long-lived, alpha-emitting radium, thorium, protactinium, plutonium. . .”.

In terms of setting up the dose limits, common approaches specify the limit at a fraction of 1 mSv/year which corresponds to the ICRP’s recommended value for annual public dose limit (this was also discussed in Example 2.8). Typically, the dose limits are to protect members of critical group among the potentially exposed population (as discussed in Sect. 2.4.2.2). Also the time periods covered in performance assessment vary among the countries, typically ranging from 10,000 years to 1000,000 years (see also the discussions in Sect. 2.4.2.3 and Table 2.1).

Along with the use of individual dose limits, upper limit on population dose is also suggested as a measure of public health impact. Population dose refers to the total combined dose of each individual among the affected population.

One caveat in using population dose is that however small the individual dose is, if the numbers are added over a large number of people, the population dose becomes a large value. Such value is often translated into cancer risk by using linear-no-threshold dose response model. This translation is, however, not recommended as the resulting estimate of cancer incidence gives misinterpretation of the potential public health impact. This practice ignores the very low possibility of cancer occurrence at very low dose level and was warned against by the International Council on Radiation Protection (ICRP) as quoted below (ICRP 2007):

> The aggregation of very low individual doses over extended time periods is inappropriate, and in particular, the calculation of the number of cancer deaths based on collective effective dose from trivial individual doses should be avoided.

In the case of U.S. it is suggested that calculations of population dose include only those individuals who receive annual committed effective dose equivalents in excess of 0.01 mSv (1 mrem) (NCRP 1994). Annual doses to individuals below this level are regarded as *de minimis* not contributing to cancer risk.

12.1.2 Model Development for Performance Assessment

Mathematical and conceptual models are relied upon in performance assessment to describe all the natural processes and component behaviors with respect to isolation and release of radionuclides from nuclear wastes. Therefore, models to describe the behavior of various components of the total repository system are needed. As the disposal system is located in deep geological formations, the models describe the movements of groundwater in geological formations and the related interactions with the surroundings.

The sequence of modeling in performance assessment follows the description of groundwater infiltration, radionuclide releases from the waste packages, interactions and movements in the near-field, transport in the far-field (geosphere), transport in the biosphere and through related exposure pathways, and the resulting impacts on humans (and the environment). Here geosphere refers to all of the non-living parts of underground system (i.e., rocks, minerals, and groundwater). Biosphere refers to the collection of all ecosystem where life exists.

Development of performance assessment models begins with conceptual model. A conceptual model is a conceptualization of a repository system or a specific subcomponents of a repository at a specific site, and becomes a basis for developing quantitative performance assessment. For a repository system, such conceptualization is based on the interpretation of site's climatic and hydrogeological data, the scenarios selected in relation to the release of radionuclides, and the potential outcome of the releases through the contamination of the groundwater or the ecosystem. Many pieces of data and information collected from site characterization support the development of a conceptual model.

Based on this, a system level computer model is developed to support the concrete description of the conceptual model. The system level computer model is to actually perform the calculations, i.e., to analyze isolation/release of nuclear waste and their consequences. As geological repository is composed of various components, the system level model must include the description of all of the components as sub-models. These components include the waste form, the waste package, the geological repository, and the portion of the geosphere and the biosphere surrounding the geological repository connecting the affected humans (and the biological species).

Understanding the processes involved with the performance of a repository in details demands the handling of huge collection of data and information that cannot be fully contained in the models. It is therefore necessary to leave out some details as every piece of detailed information is not needed in the computer model. Through progressive simplification, only the information relevant to be the primary driver for the process under consideration is selected and used. This process is called model abstraction (DOE 1998). Model abstraction is a necessary process for the preparation of a system level model. This abstraction process is repeated for all of the major processes involved in each sub-model. The final models are generally the most compact or abstracted models of all while capturing the essential features of the site and the key processes affecting the performance of the repository. The models should also be efficient enough to handle the required computational tasks. The overall structure of model abstraction is shown in Fig. 12.1. Based on the use of detailed mechanistic models, key features of the processes are extracted providing the basis of subsystem models. The subsystem models are then combined to describe the total system.

The outcome of model abstraction is an integrated total system model. The total system model includes various sub-models to perform calculations for the selected

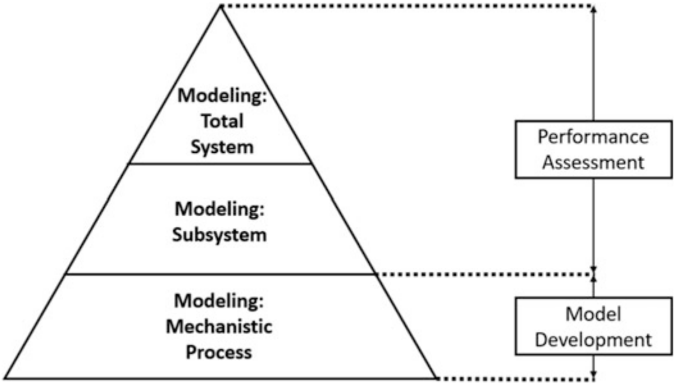

Fig. 12.1 Model abstraction for repository performance assessment

scenarios of radionuclides release. For the base case scenario of groundwater contamination, the sub-models include the models for source term, near-field transport, far-field transport, and exposure pathways in the biosphere. These sub-models are in a smaller scale also integrative models. For example, the source term model describes the interactions between the infiltrating water and the multiple components of the engineered barrier system.

If the scenario of human induced events, such as inadvertent intrusions, are to be considered, a separate set of models may need to be employed to describe the scenarios involved with such incidents. The scenarios other than the groundwater infiltration scenario are described in Sect. 12.4.2.

12.1.2.1 Models for Infiltration Analysis

Starting from precipitation, infiltration analysis follows the movements of water from the surface into the repository system. The analysis is based on describing surface water balance: The water that is not lost by surface run-off, evaporation, or transpiration by the plants enters the unsaturated zone flow system. Therefore, the model captures water movements through the unsaturated zone until it enters the waste disposal system. Changes in the site conditions over time are also captured in the analysis.

12.1.2.2 Models for Engineered Barrier Analysis

Engineered barrier analysis is to describe the behavior of nuclear waste packages and to determine the time of their failure. Using the models for various mechanisms of corrosion, the analysis examines the degradation of nuclear waste package materials.

Interactions of the waste packages with infiltrating groundwater and impacts of any early failure of defective container are described in the analysis.

12.1.2.3 Models for Source Term Analysis

Source term analysis is to describe the release of radionuclides from the waste forms into the near-field rocks over time. Therefore, the analysis considers the outcome of water infiltration and engineered barrier degradation with respect to the release of radionuclides. To support the analysis, various parameters and their evolutions over time are also described. Depending upon the level of rigor required, these parameters may include time history of container-wall and fuel-rod temperature, pH, Eh, chloride concentration, and internal container water. Source term analysis is the central part of performance assessment as the results drive the outcome of the repository performance assessment.

12.1.2.4 Models for Groundwater Flow and Radionuclide Transport

Based on the results from the source term analysis, the analysis of groundwater flow and radionuclide transport examines the movement of radionuclides through the near-field and the far-field. The models describe hydrologic processes in the porous medium under the influence of geological and geochemical features. If fractures are present in the rock system, description of fracture flow needs to be included in the models.

12.1.2.5 Models for Dose Analysis

The final phase of repository performance assessment is to analyze the impacts of radionuclides migration on the biosphere. This is also called biosphere analysis in a more general term. As discussed in Sect. 2.4.2.6, the focus of the analysis is on humans. This assumes that if the impacts on humans are acceptable then the consequences on other biota are also acceptable.

By taking the results from the analysis of groundwater movement and radionuclide transport (i.e., the concentration of radionuclides in groundwater at the receptor locations), the dose to the exposed individuals is estimated. This analysis requires the understanding of the behaviors of the affected humans in terms of potential radiation exposure from the usage of the contaminated groundwater. The usage could include drinking of contaminated groundwater or consumption of contaminated foodstuff through the food chains. Transfer of radionuclides to animals, animal products (i.e. milks), vegetables, crops, and fruits are also described to determine the total amount of radionuclide intake into the human body. The distribution of radionuclides and their energy deposition in human body are modeled in the analysis to assess the resulting dose impact. It should be noted that the affected individuals in the analysis are hypothetically constructed people to represent potential human radiation exposure in the future.

This biosphere analysis is subject to large uncertainty. The uncertainty arises not only due to individualistic human behaviors but also from the unknown-ness of the ecosystem in the long-term future. The conditions of the surface environment as the setting for human activities will also be subject to variation. This issue of making assumptions as part of the analysis is further discussed in Sect. 12.2.3.

12.2 Steps in Performance Assessment

Performance assessment is an iterative process. Major steps involved in a performance assessment include: (1) development of scenarios, (2) development of models, (3) performing integrated consequence analysis, and (4) examination and interpretation of results and uncertainties. The results of the analysis may require changes in the design of the system and the steps of performance assessment are followed again per the design changes made. This iterative process helps to build confidence in regulatory compliance.

12.2.1 Scenario Development

To project time-dependent evolution of a geological repository with respect to the behavior of nuclear waste packages and radionuclide release, possible scenarios must be developed. Such scenarios become the basis of evaluating the consequences relevant to the performance of the repository.

Development of scenarios requires an understanding of how the environmental, geological, and hydrological characteristics of the site interact and affect the isolation of nuclear waste in the repository. This exercise of scenario development is based on characterizing features, events, and processes (FEPs) at a site leading to the release and migration of radionuclides from nuclear waste.

Here, features refer to the characteristics of a site with respect to meteorology, geology, or hydrology including annual precipitation rate, rock types, depth of water, presence of fractures, etc. Events refer to occurrences of specific events that affect isolation of nuclear waste in the repository such as earthquakes, volcanic eruptions, glaciations, climatic fluctuations, or human initiated disruptions (e.g., drilling or mining). Processes are on-going phenomena that have gradual, continuous interactions with the repository system affecting isolation of nuclear waste. These processes include soil erosion, water infiltration, temperature changes, materials degradation, etc. Therefore, detailed understanding of FEPs that affect the integrity of nuclear waste isolation over the course of time provides the basis of developing scenarios covered under performance assessment.

If all possible features, events, and processes are combined, the number of scenarios can be quite large. Therefore, screening of scenarios is necessary. For the screening, various events and processes conceivable at the candidate site are examined and classified based on their physical reasonableness, probability, and potential consequences. They are then further screened to select the potentially significant FEPs based on importance criteria. In the case of U.S. Department of Energy, the following importance criteria are used for FEP screening (DOE 2002):

(i) FEP has at least 1 chance in 10,000 of occurring over 10,000 years.
(ii) Exclusion of FEP would significantly change radiological exposure or radionuclide release.

The IAEA have suggested about 60 scenarios as potentially relevant to performance assessment (IAEA 1981; IAEA 1983). There is also a more recent effort to establish the International FEP Database to support achieving comprehensiveness of performance assessment by covering 134 FEPs (NEA 2000).

12.2.2 Performing Integrated Analysis for Repository Performance

Once the scenarios are selected, the models for the subsystems of the geological repository are connected and interactively used to describe the overall behavior of the system and potential consequences of scenario evolutions with respect to the performance requirements. The overall scheme of the integrated performance assessment within the U.S. regulatory framework is represented in a diagram shown in Fig. 12.2.

As implied in the discussions of Sect. 12.1, results of performance assessment cannot be treated as deterministic one. The calculated results of performance assessment, i.e., the dose to an individual or the cumulative releases of radionuclides to the accessible environment, are the projections based on agreed sets of assumptions for a particular scenario selected. Also, these results are within the bounds of models but not as actual measures of future consequences. Therefore, these results represent a sort of “probable’ behaviors of a repository system under the defined conditions (DOE 1998).

In this regard, to answer the questions asked in performance assessment (“What can happen?”, “How likely is it to happen?”, and “What are the consequences?”), the analysis needs to be organized employing the concept of risk and addressing the issue of uncertainty. Accordingly, performance assessment uses a framework of probabilistic analysis.

To quantify risk and uncertainties as repository system performance within a probabilistic framework, the analysis seeks to mimic the natural behavior of the system that is inherently uncertain or variable. Therefore, in the analysis, the models are run repeatedly for the given scenario using many combinations of parameter

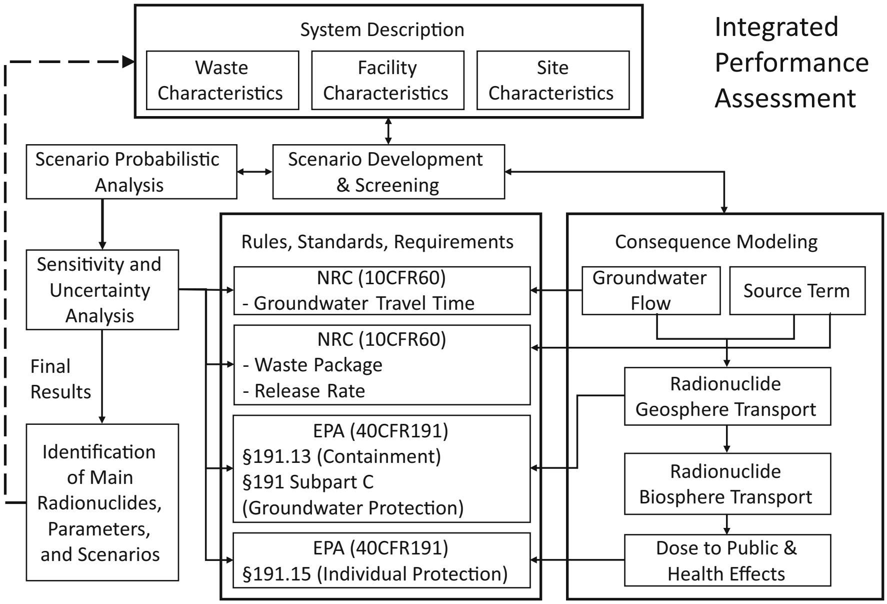

Fig. 12.2 Overall scheme of integrated performance assessment. (Gallegos 1991)

values within the expected bounds as an attempt to reflect the range of behaviors or the values of parameters in the absence of perfect or complete knowledge. Therefore, each run is one realization of the input parameters from the possible ranges of parameter values and the result provides some finite possibility of capturing the performance. When the results from multiple calculations are combined, the outcome is likely to capture the consequences as probabilistic performance measures.

12.2.3 *Evaluation of Uncertainty in Models and Parameters*

Uncertainties naturally arise in performance assessment. The uncertainties are due to the variable nature of the processes and phenomena and the limited nature of scientific knowledge in understanding those processes and phenomena. This limitations in scientific knowledge is also reflected in the use of mathematical/computer models to project the performance of a repository into the future. The models and available data used including the need for data extrapolation are all sources of uncertainty.

Uncertainties in this analysis can be either epistemic or aleatory. Epistemic uncertainty comes from inadequacy of knowledge. Aleatory uncertainty comes from natural variability. Both are always present in characterizing uncertainties in performance assessment. Epistemic uncertainty is manifested in the use of models and the scenarios for future state of things while aleatory uncertainty mostly resides with the use of parameters and their values used in the models. These uncertainties are represented in performance assessment as model uncertainty and parameter uncertainty. While model uncertainty represents epistemic uncertainty more closely, epistemic and aleatory uncertainty are present in both model uncertainty and parameter uncertainty.

12.2.3.1 Model Uncertainty

Model is a conceptual approach used to assess system behavior. The system behavior under consideration is complex and evolves over time. In performance assessment, mathematical equations are used to capture the behavior of time-evolving complex repository system to represent the concepts or phenomena involved. Therefore, model uncertainty refers to the uncertainty arising from abstracting a real system and its evolution in the future.

Uncertainty in future state of things includes the uncertainty in site conditions affecting processes and events at a site. The possibility of ice age or global warming is an example. Changes in the nature and society are also part of this uncertainty including the uncertainty in social developments and human institutions. As medical science and technology advance toward curing cancer and other human diseases, the background societal risk level may also change by shifting the desired level of performance outcome. Capturing all of these uncertainties may be beyond the realm of science.

While we may not fully capture the uncertainty in future state of things, this type of uncertainty is addressed through relevant policy positions as part of government decision making. For example, in the case of the Yucca Mountain project, U.S. DOE uses the societal conditions of today as the basis for judging the safety in the future (DOE 2008). Therefore, performance assessment is conducted based on the state of current knowledge: The populations would remain at their present locations with similar behaviors and human institutions remain intact in the future. This assumes that protecting the public at the current state of things provides adequate basis for the protection of the public in the future. The future climatic conditions may be estimated based on what is currently known about the past and the present with additional considerations of the impacts of human activities on the climate in the future. As a conservative approach, the U.S. DOE assumes that the current climate is the driest it will ever be at Yucca Mountain.

Limitations in using models to represent complex system behaviors are an important part of model uncertainty. This uncertainty is embedded in the process of conceptual model development, in setting up the supporting mathematical models and equations, and in developing and executing computer codes to solve the equations. Uncertainty in conceptual model development arises from the presence of alternative ways in handling issues such as spatial/temporal variability, heterogeneity, coupled processes, and system dimensionality. Uncertainties in mathematical and computational models are due to the use of simplifying assumptions or from the implementation uncertainty. Simplifying assumptions include linearization of equations, process decoupling, limited use of spatial dimensions to represent the process, using steady state assumption for a transient phenomenon, and algebraic or programming mistakes. Implementation uncertainty includes uncertainty due to discretization or numerical convergence. The initial and boundary conditions used to support the mathematical model formulations also constitute the model uncertainty.

Characterization of model uncertainty is challenging. Part of the challenge comes from the difficulty in identifying all likely processes and events, in evaluating the plausibility of the selected models, and in assessing the accuracy of model predictions.

Approaches suggested to address model uncertainty are either model-focused or prediction-focused. The distinction depends on whether the attention is directed toward the plausibility of the model hypotheses or the accuracy of its predictions. Using the prediction-focused approach is not possible in performance assessment as truth in the predictions cannot be known. The model-focuses approach expresses the likely processes and events as a set of alternative scenarios and conceptual models. Each conceptual model is supported by the corresponding computational models with a specified set of input variables. The plausibility of each conceptual model is then expressed by using probability, i.e., the likelihood of the model to represent the reality. The assessed probabilities are used as weighting to combine the results of the models and uncertainties from the alternative models.

An example of this approach is shown in Fig. 12.3. In the figure, the probabilities, P_{Ai} and P_{Bi} represent the relative likelihood of the selected conceptual model Ai or Bi under two different scenarios of A and B to represent the process under consideration. The approach allows integrative use of alternative models to determine the final outcome while reflecting model uncertainty. Apparent challenge in the

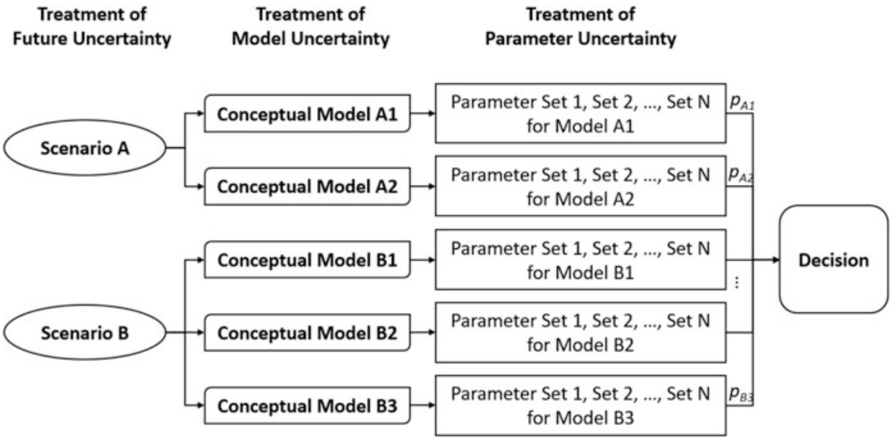

Fig. 12.3 An example of framework to analyze uncertainty in performance assessment (P_{Ai} and P_{Bi} represent the relative likelihood of the selected conceptual model Ai or Bi under two different scenarios of A and B to represent the process under consideration)

approach is the difficulty in quantifying the relative likelihood of alternative conceptual models in different scenarios.

Reducing model uncertainty in the development of models is also a part of an effort to address model uncertainty. In this regard, an important effort is model verification and validation. Model verification is the process of confirming that a model is correctly implemented with respect to the conceptual model and the solution to the model. Therefore, verification checks whether the conceptual model is properly implemented in the computational model setup and in the use of input parameters to run the computational model. Verification is not concerned whether the conceptual model is correct or the results from the computational model is accurate. Model validation is the process of checking the accuracy of the model's representation of the real system (such as using experimental data). Therefore, validation is concerned about the correctness of the conceptual model as well as the accuracy of the results from the use of the model. Ideal validation requires a comparison between model predictions and observations of the real system over the temporal and spatial scales relevant to the analysis (NRC 1991). While recognizing the challenge in performing such comparison, validation of performance assessment models is pursued by using laboratory experiments, field test data, and natural analogue observations as the basis for such comparison in the use of specific component models. Use of natural analogues for model validation is discussed in Sect. 12.5.

12.2.3.2 Parameter Uncertainty

Parameter uncertainty refers to the uncertainty from the use of the data and parameters in the models, i.e., the uncertainty in determining parameter values used in a model. It includes the uncertainty associated with inherent randomness of natural phenomena and the intrinsic heterogeneity of natural systems, the uncertainty in capturing natural phenomena or characteristics by using laboratory and field measurements, and the uncertainty in translating the measurement data into the input parameter values used in a model. The uncertainty associated with laboratory and field measurements includes measurement errors due to instrument error, reading error, and biases in the data from insufficient observations. Applicability of the laboratory measured data to field situation is also a source of parameter uncertainty.

Efforts can be made to reduce parameter uncertainty by obtaining additional data or information about the parameters used in the model. Such additional data or information can be obtained from the literature, through laboratory analysis or field studies, from the soft data collected based on qualitative observations, or by examining correlation between the variables.

In comparison to model uncertainty, analysis of parameter uncertainty is widely exercised. This is because uncertainty in input parameters can often be quantified. Methods for parameter uncertainty analysis include the Monte Carol method, the probability-distribution approach, the bounding approach, sensitivity analysis, analytical methods, differential methods, or expert judgment (these are not mutually exclusive). While the Monte Carlo method is the most popular approach, other methods are also applicable depending on the type or complexity of the mathematical models used. To quantify input parameter uncertainty in support of the probability-distribution approach, the bounding approach and expert judgment are often used when not enough information is available.

12.2.3.3 Monte Carlo Method

The Monte Carlo method is most often used for parameter uncertainty analysis in combination with the probability-distribution approach in performance assessment. The name Monte Carlo method comes from the physicists working on nuclear weapon projects at Los Alamos National Laboratory in the 1940s. At that time they noted that use of randomness and the repetitive nature of the process in numerical simulations are analogous to the activities conducted at a casino for gambling. So they termed the method in reference to the Monte Carlo Casino in Monaco. Monte-Carlo methods involve selecting input values from each input parameter probability distribution (this is called sampling) and repeatedly running a simulation using the combinations of the sampled input data. Thus by repeatedly running the simulation model using various sets of randomly selected input data, Monte Carlo methods provide a way of propagating the uncertainties in all of the selected inputs variables through repeated executions of simulation models. An output distribution is produced by collecting and evaluating the values of the output parameter of interest from the repeated simulations.

Monte Carlo simulation proceeds as following (see Fig. 12.4):

(i) Specify a probability distribution for each of the selected input parameters as random variable.
(ii) For each specified probability distribution of an input parameter, construct a corresponding cumulative distribution function. In this way, all of the values of the random variable have one-to-one match with the cumulative probability (which ranges between 0 and 1).
(iii) Generate a random number which ranges between 0 and 1 by using a random number generator. This random number is used to randomly select (i.e., sample) the value of an input parameter from the cumulative distribution function (Fig. 12.5 captures this process). Practically pseudo-random generators are used for random number generation by using a "random seed (a starting value)" as an input.
(iv) By repeating step (iii) for each of the selected input parameters, one sample from each input distribution is selected. By continuing this for all input parameters which are considered random variable, a complete set of input values is constructed.
(v) By repeating step (iii) and (iv), many complete sets of input values can be constructed.

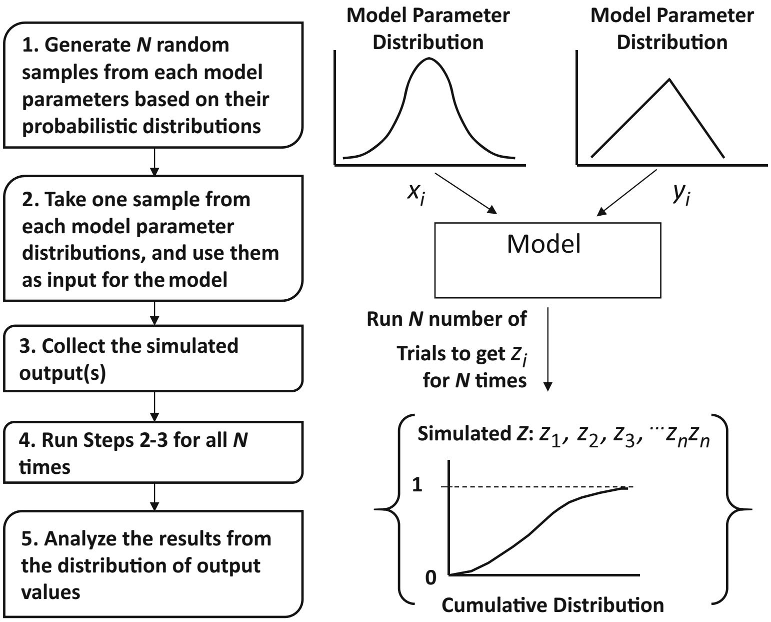

Fig. 12.4 Application of Monte Carlo analysis to a model

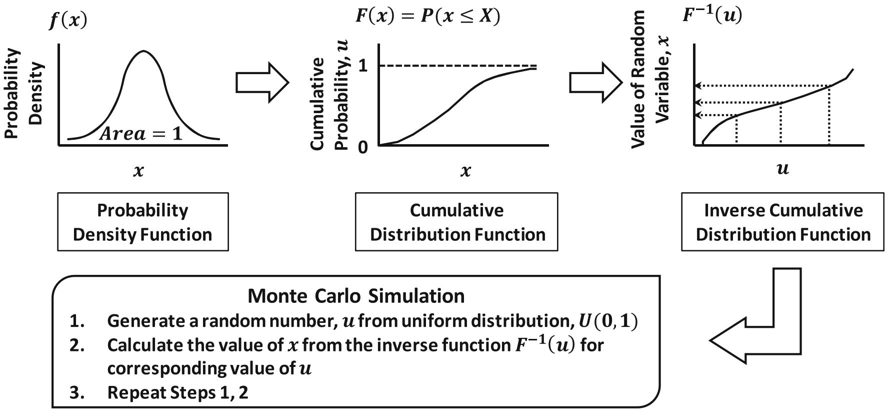

Fig. 12.5 Use of random numbers to represent a probability distribution in Monte Carlo analysis

(vi) Each set of input values is entered into the simulation model and the model is executed. Therefore, although the sample values of the input parameters are probabilistically generated, execution of the model is deterministic for a given set of input parameter values.

(vii) Repeat steps (iii) through (vi) until the specified number of sample realization (or model iterations) is completed while saving the results of each simulation.

Thus for the output parameter of interest, a set of samples of model output results is obtained.

(viii) The results of outputs are summarized by using various statistics such as mean, variance, and various percentile values.

Results from the Monte Carlo analysis are used to produce a distribution of model calculations which represents the range of uncertainty in the simulation results. The results are compared with respect to the performance criteria set up in the regulatory requirements to examine how uncertainties in the results should be interpreted. If the results as distribution are well below or greatly exceed the specified criteria, then judgment on the performance of the system can be made with confidence, i.e., performance is satisfactory or unacceptable, respectively. If the results exceed the criteria with probability slightly higher than what is acceptable, either changes in the design to improve the performance or refinement in the analysis may be considered to reduce uncertainty. The latter requires sensitivity analysis to identify those parameters which contribute the most to the uncertainty in the results. The sensitivity analysis compares the variations in input parameters with variations in the resulting output values to identify the key parameters of importance. Based on the results, new data collection effort can be made for the identified key parameters for uncertainty reduction.

Repeated sampling of value for an input parameter can be done by using simple random sampling or Latin hypercube sampling (LHS). LHS is a form of stratified (or constrained) sampling where the sampling process is guided by design rather than being purely random. For example, in a standard LHS, the sampling space (i.e., the input value range) for an input parameter is first divided up into m equi-probable intervals. Then a single value is sampled at random once within each of these intervals. By repeating the process m times, then the entire input space of that input parameter is sampled. By doing the same for all of the inputs, i.e., by selecting one value at random from each of the inputs from the m sample values for each input, m complete sets of input values are generated for model execution. The constrained sampling of LHS allows efficient mapping out of the input value space making the process of estimating output distributions very efficient. The method is only random in the pairing of values of the input parameters. Thus creating any spurious correlation in the pairing of input values is to be avoided. To remove spurious input correlations in the pairing, a method of restricted pairing is used for the implementation of LHS (Iman et al. 1980).

12.2.4 General Framework of for Uncertainty Analysis

A general structure of uncertainty analysis to consider both model uncertainty (including the uncertainty in future conditions) and parameter uncertainty in a probabilistic framework has been suggested (NRC 1993). This framework is shown in Fig. 12.5.

In this structure, uncertainty in future state of things are captured by using different scenarios as particular conceptualizations of the future of a repository site. Each different scenario can be assigned a probability as an indication of the relative frequency of the scenario. Within each scenario of the future, model uncertainty is addressed by postulating alternative conceptual models of the behavior of the repository system (or subsystem). Each alternative conceptual model describes the behaviors of the repository system (or subsystem) by using a particular set of mathematical models. The likelihood of different models being appropriate to describe the system behavior can be specified by using an assigned probability. Within each model, parameter uncertainty analysis can be performed by using Monte Carol method, i.e., running the model by using alternative sets of input parameter values selected from respective probability distributions for each input.

The results of the analysis may be combined by using the assigned probabilities as relative weights to the scenarios and the alternative conceptual models. Therefore, relative confidence levels are assigned as probabilities to each of the scenarios and the models used.

An alternative way of addressing model uncertainty is the use of a logic tree (EPRI 1992). A logic tree is a sequential conceptualization of events leading to a consequence. The approach allows conceptual representation of uncertain models and scenarios in a systematic way. For example, uncertainties in the states of nature and in the occurrences of future events leading into a consequence can be addressed by specifying probability to different conceptualization at each event's occurrence. This approach is shown in Fig. 12.6 for the description of the cases of water infiltration, radionuclide release to groundwater and migration reaching a biosphere location toward human exposure. The figure reflects a particular case where the chance of water infiltration to be high or low is less than 10%, the chance of the

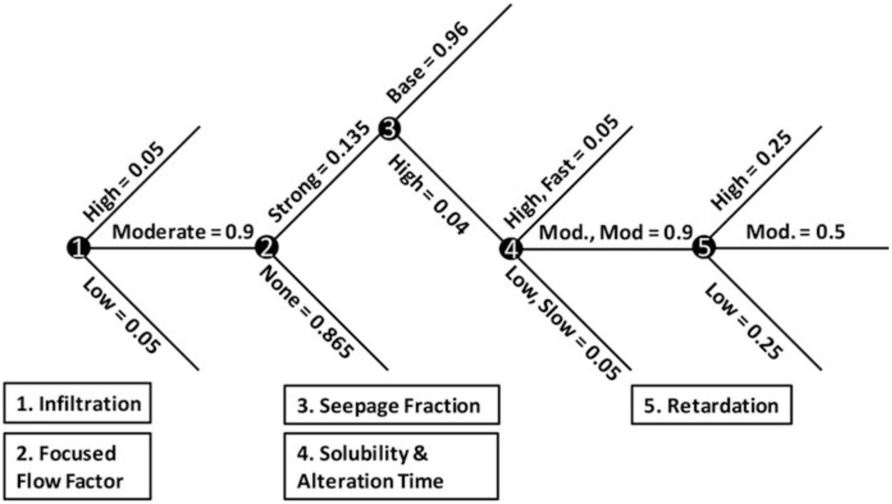

Fig. 12.6 An example of logic tree method [EPRI IMARC-7] to describe radionuclide release and migration from nuclear waste. (Source: EPRI 1996)

infiltrating water hitting the waste containers is 13.5% (focused flow factor), the chance of the contaminated groundwater to pass through the porous medium is about 96% (seepage fraction), the solubility of radionuclides and the alternation time of waste forms are predominantly moderate, and the retardation factor of radionuclides are mostly set at moderate values.

Uncertainty in performance assessment cannot be totally removed or completely characterized. This is due to the nature of long-term future predictions, along with practical constraints in time and financial resource, or simply due to the incomplete nature of knowledge. However, the goal of uncertainty analysis in performance assessment is not to quantify the uncertainty itself but to support decisions. As long as uncertainty analysis helps to make decisions with insights to the development of a geologic repository, the effort is meaningful (a carefully conducted uncertainty analysis will add confidence to the decision at stake). Such decision could be in relation to design improvement, compliance demonstration, or risk communication for a geological repository.

12.3 A Simplified Performance Assessment

The example given in this section is a very crude simplified case of performance assessment based on the disposal of HLW or spent fuel from LWR at the Yucca Mountain geological repository. The example describes the release of radionuclides from waste form, transport of radionuclides in the unsaturated zone and the saturated zone, and radiation dose to a member of the public by using a set of simplified equations.

12.3.1 Source Term Model

The source term model is to describe the release of radionuclides from different waste forms after the failure of waste packages. The waste forms considered in this example include spent fuel, vitrified glass, ceramics, and metallic waste. These models show the waste forms having different degradation mechanism with varying rates of radionuclide releases.

12.3.1.1 Spent Fuel (SF) Waste Form

The rate of dissolution of spent fuel can be represented as (CNWRA 2007):

$$r_{\mathrm{SNF}} = r_0 e^{-34300/\mathrm{RT}} \left[\frac{mg}{m^2 - \mathrm{day}}\right] \tag{12.1}$$

where r_{SNF} is the dissolution rate of SF [mg/m^2−day]; r_0 is the reference dissolution rate [mg/m^2−day] (the recommended value is 323,184.8 as mean); R is the gas constant, 8.314 [J/mol/K]; T is the temperature of the waste package [K].

The dissolution rate of spent fuel is multiplied by the effective specific surface area (a_{SNF}) [m^2/kg] (the recommended value is 0.22) to determine the fractional release rate from spent fuel. The effective specific surface area is a function of spent fuel particle radius, spent fuel density and the volume fraction of the waste form contacted by water.

Then, the rate of release per year from spent fuel is,

$$r_{\text{SNF\_frac}} = r_{\text{SNF}} \times a_{\text{SNF}} \times \frac{365}{1000000} \left[yr^{-1}\right] \tag{12.2}$$

Therefore, the rate of mass release from spent fuel in spent fuel waste is given as:

$$\text{release}_{\text{SNF}} = m_{\text{SNF}} \times r_{\text{SNF\_frac}} \left[\frac{\text{kg}}{\text{year}}\right] \tag{12.3}$$

where m_{SNF} [kg] is the mass of spent fuel (excluding the cladding).

12.3.1.2 Glass Waste Form

The rate of dissolution of glass is given by (CNWRA 2007):

$$r_{\text{glass}} = k_0 10^{\eta \cdot pH} e^{-E_{ag}/RT} \left[\frac{g}{m^2 - \text{day}}\right] \tag{12.4}$$

where: r_{glass} is the dissolution rate of glass [g/m^2−day]; k_0 is the intrinsic dissolution rate of glass [g/m^2−day] (the recommended values are 8.41×10^3 and 2.82×10^1 for the acidic and alkaline conditions, respectively) (Bechtel SAIC 2004); pH is the pH value of the water in the waste package; η is pH dependence coefficient (−0.49 and 0.49 for the acidic and alkaline conditions, respectively); E_{ag} is effective activation energy [J/mol] (31,000 and 69,000 for the acidic and alkaline conditions, respectively); R is the gas constant 8.314 [J/mol/K]; T is the temperature of the waste package [K].

The release rate per year from glass becomes,

$$r_{\text{glass\_frac}} = r_{\text{glass}} \times a_{\text{glass}} \times \frac{365}{1000} \left[\text{year}^{-1}\right] \tag{12.5}$$

where, a_{glass} is the effective specific surface area [m^2/kg] represented by

$$a_{\text{glass}}(\text{t}) = f_{\text{exposure}} \cdot \left(2.7 \times 10^{-3}\right) \cdot \left(m_{o,\text{glass}} - \sum m_t\right) \left[\frac{m^2}{kg}\right] \tag{12.6}$$

In the equation, f_{exposure} is the exposure factor to account for the uncertainty in accessibility of water due to the cracks in the glass (the recommended value is 4); $m_{o,\text{glass}}$ is the initial mass loading of glass (kg); $\sum m_t$ is the total released glass mass (kg) before time t. Then the rate of release from glass for a mass loading of m_{glass} [kg] glass waste is,

$$\text{release}_{\text{glass}} = m_{\text{glass}} \times r_{\text{glass\_frac}} \left[\frac{\text{kg}}{\text{year}}\right] \tag{12.7}$$

12.3.1.3 Ceramic Waste Form

The rate of ceramic waste dissolution is represented by the following two relationships for the alkaline or acidic condition (ANL 2006):

$$r_{\text{CWF\_alkaline}} = 1.4 \times 10^4 \cdot 10^{0.64pH} \cdot e^{-83000/\text{RT}} \left[\frac{g}{m^2 - \text{day}}\right] \tag{12.8}$$

and

$$r_{\text{CWF\_acid}} = 1.26 \times 10^{11} \cdot 10^{-0.36pH} \cdot e^{-72000/\text{RT}} \left[\frac{g}{m^2 - \text{day}}\right] \tag{12.9}$$

The dissolution rate is multiplied by an effective specific surface area (a_{CWF}) [m^2/kg] to estimate the fractional release rate from the waste form.

$$r_{\text{CWF\_frac}} = r_{\text{CWF}} \times a_{\text{CWF}} \times \frac{365}{1000} \left[\text{year}^{-1}\right] \tag{12.10}$$

Then the rate of release from ceramic for a mass loading of m_{CWF} [kg] ceramic waste is:

$$\text{release}_{\text{CWF}} = m_{\text{CWF}} \times r_{\text{CWF\_frac}} \left[\frac{\text{kg}}{\text{year}}\right] \tag{12.11}$$

12.3.1.4 Metallic Waste Form

The rate of release from metallic waste can be estimated as follows (Bauer and Morris 2007),

$$r_{\text{metal}} = A \cdot ln\left(1 + \frac{B}{A}t\right)/365 \left[\frac{g}{m^2 - \text{day}}\right] \tag{12.12}$$

where,

$$A = e^{7.9812+2.3938\times10^{-4}[Cl^{-}]-1.2273pH}\ \left[\frac{g}{m^2}\right]$$

$$B = e^{-0.54624-0.69046pH+\left(0.019665+5.8415\times10^{-6}[Cl^{-}]\right)T}\ \left[\frac{g}{m^2-\text{day.}}\right]$$

for given T (temperature in °C), pH(pH value of the bulk solution), $[Cl^{-}]$(the halide or chloride ion concentration in ppm or mg/L), and t (time in days)

The release rate of metallic waste is multiplied by an effective specific surface area (a_{metal}) [m^2/kg] (with the recommended value of 4.42×10^{-3}) to estimate the fractional release rate (ANL 2000).

$$r_{\text{metal\_frac}} = r_{\text{metal}} \times a_{\text{metal}} \times \frac{365}{1000}\ \left[\text{year}^{-1}\right] \tag{12.13}$$

Then the rate of release from the metallic waste form for a mass loading of m_{metal} [kg] metal waste is:

$$\text{release}_{\text{metal}} = m_{\text{metal}} \times r_{\text{metal\_frac}}\ \left[\frac{\text{kg}}{\text{year}}\right] \tag{12.14}$$

12.3.1.5 Release from Waste Form to a Waste Package

When the water enters the waste package after its failure, the overall mass balance model for the nuclide inventory in the water in a failed waste package is (CNWRA 2007):

$$\frac{dm_i}{dt} = w_{li}(t) - w_{ci}(t) - m_i\lambda_i + m_{i-1}\lambda_{i-1} \tag{12.15}$$

where:

m_i = the amount *of* i^{th} *ra*dionuclide in the waste package water at time t [mol];
λ_i = the decay constant of i^{th} radionuclide [year^{-1}];
m_{i-1} = the amount of $(i\text{-}1)^{th}$ radionuclide in the waste package water at time t [mol];
λ_{i-1} = the decay constant of $(i\text{-}1)^{th}$ radionuclide [year^{-1}];
$w_{li}(t)$ = the rate of transfer from the spent fuel into the waste package water through leaching of SF [mol/year] (function of flow rate of water, composition of the water and the element solubility in the water);
$w_{ci}(t)$ = the rate of advective transfer out of the waste package [mol/year] $=C_i(t)\ q_{out}(t)$ where $C_i(t)$ is the concentration of i^{th} radionuclide in the waste package water [mol/m^3] (mass m_i divided by volume of water in the waste package) and $q_{out}(t)$ is the water flow leaving the waste package at time t [m^3/year].

The estimated release rate of each radionuclide from the engineered barrier system becomes the source term, designated as $q_c(t)$ [Bq/year or Ci/year] represented by

$$q_c(t) = c \cdot \frac{m_i}{V_{wp}} \cdot q_{\text{out}}(t) \tag{12.16}$$

Here, $q_c(t)$ is determined by dividing m_i by the volume of water available in the WP (V_{wp}) and then multiplying the water flow leaving the waste package (q_{out}) by a conversion factor c (in Bq/mol or Ci/mol).

12.3.2 Unsaturated Zone Transport

The unsaturated zone is assumed to be a homogeneous, isotropic, porous media with constant, unidirectional flow in the vertical (downward) direction. The contaminant transit time in the unsaturated zone can be assumed as:

$$T_a = \frac{X_u}{U_u} R_u \text{ [year]} \tag{12.17}$$

where

X_u= the distance from the base of the source volume to the top of the aquifer [m]
R_u= the retardation factor of the radionuclide in the unsaturated zone
U_u= the unsaturated zone average pore velocity [m/year] $= \frac{P}{\varepsilon_{f_u}}$
P=net water percolation rate [m/year]
ε_{f_u} = effective porosity in the unsaturated zone

The release rate from unsaturated zone to an aquifer (saturated zone) is:

$$q_a(t) = e^{-\lambda_i T_a} q_c(t) \left[\frac{Bq}{yr} or\ Ci/\text{year}\right] \tag{12.18}$$

12.3.3 Saturated Zone Transport

In this example, the transport of contaminant in the saturated zone is represented by the following advection-dispersion mass balance equation (Codell and Duguid 1983):

$$\frac{\partial C}{\partial t} + U\frac{\partial C}{\partial x} = D_x \frac{\partial^2 C}{\partial x^2} + D_y \frac{\partial^2 C}{\partial y^2} + D_z \frac{\partial^2 C}{\partial z^2} - \lambda C \tag{12.19}$$

The solution of this equation depends on how the release takes place from the source. Please note that different types of solutions can be obtained for various source configurations as described in Sect. 11.3.7.

In the case of instantaneous release of mass, q (assuming the surface and initial concentrations are zero everywhere in the domain), the solution to this equation in an infinitely deep aquifer for a point source becomes:

$$C(x, y, t) = \frac{2q}{\varepsilon_f R} \frac{1}{\sqrt{4\pi D_x t/R}} \frac{1}{\sqrt{4\pi D_y t/R}} \frac{1}{\sqrt{4\pi D_z t/R}}$$

$$exp\left[-\frac{(x - Ut/R)^2}{4D_x t/R} - \lambda t\right] \cdot exp\left[-\frac{y^2}{4D_x t/R}\right] exp\left[-\frac{z^2}{4D_z t/R}\right] \left[\frac{Ci}{m^3}\right] \tag{12.20}$$

where: q is the released mass; D_x = the coefficient of longitudinal hydrodynamic dispersion [m^2/year]; D_y = the coefficient of transverse hydrodynamic dispersion [m^2/year]; D_z = the coefficient of vertical hydrodynamic dispersion [m^2/year]; U = the average linear velocity of groundwater [m/year]; R = the retardation factor.

The above equation for instantaneous release can also be utilized to describe the case of continuous release as follows:

$$C_{\text{longterm}}(x, y, t) = \int_0^t C(x, y, t - \tau) q(\tau) d\tau \tag{12.21}$$

12.3.4 Calculation of Human Dose

To determine the impact on humans, the first effort needed is identifying the groups of people who are potentially most significantly affected by the presence of the contaminated groundwater. Local communities who consume the water from an aquifer affected by the release of radionuclides from the geological repository are the likely group. For the identified group, behavioral analysis is made to characterize the patterns of their exposure to radiation considering various exposure pathways. If the radionuclides in the groundwater reaches the location of wells, the water from the well can be pumped out and directly consumed as drinking water by humans. The water can also be used for irrigation of crops and vegetables. The water can be used to water stock animals as well which become the source of contamination of milk or meat products. The radionuclides can be inhaled by humans if they become resuspended as fine particles and picked up by the wind. The groundwater could also reach surface water and be consumed though surface water usage by the local community.

Therefore, the radiation exposure routes include ingestion of food and water, inhalation of contaminated air, and direct external exposure. Typically, ingestion of food is the dominant one among them in terms of dose to humans from groundwater contamination. A simplistic way to estimate the dose to humans from various exposure pathways is to assess the dose from drinking contaminated groundwater and use the ratio of drinking water dose to total dose from all exposure pathways. The drinking water dose (effective dose) can be calculated as follows:

$$D_{\mathrm{DW}} = U_{\mathrm{water}} \times \sum_i C_i D_{cf,i} e^{-\lambda_i T_p} \left[\frac{\mu Sv}{\mathrm{year}}\right] \tag{12.22}$$

where, D_{DW} = dose from drinking water [μSv/year]; U_{water} = a usage factor that specifies the drinking water intake rate for an individual under consideration [m^3/year]; C_i = the concentration of nuclide i in groundwater [Bq/m^3]; $D_{cf,i}$ = the dose factor of radionuclide i from ingestion [μSv /Bq]; λ_i = the decay constant [year^{-1}]; T_p = the average transit time required for nuclides to reach the point of exposure [year].

A study by the U.S. National Academy of Science estimated the ratio of drinking water dose to total dose for a select group of radionuclides. The estimation was based on an individual dose resulting from a unit concentration of a radionuclide reaching the environment of the biosphere from a geological repository. The results are listed in Table 12.1. It can been that the ratio is about 20% for ^{238}U and ^{239}Pu and is as high as 30% for ^{93}Zr but also is very low for ^{14}C and ^{79}Se (less than 0.1%).

Table 12.1 Dose conversion factors and ratio of drinking water dose to total dose for selected radionuclides

Radionuclide	Dose (effective dose) conversion factor (Sv/Bq)*	Ratio of drinking water dose to total dose**
^{14}C	5.8×10^{-10}	1.22×10^{-4}
^{79}Se	2.9×10^{-9}	4.27×10^{-4}
^{93}Zr	1.1×10^{-9}	3.0×10^{-1}
^{99}Tc	6.4×10^{-10}	1.0×10^{-2}
^{126}Sn	4.7×10^{-9}	1.2×10^{-2}
^{129}I	1.1×10^{-7}	7.07×10^{-2}
^{135}Cs	2.0×10^{-9}	2.67×10^{-2}
^{210}Pb	6.9×10^{-7}	5.08×10^{-2}
^{226}Ra	2.8×10^{-7}	1.07×10^{-1}
^{230}Th	2.1×10^{-7}	6.91×10^{-2}
^{238}U	4.5×10^{-8}	2.15×10^{-1}
^{237}Np	1.1×10^{-7}	8.41×10^{-2}
^{239}Pu	2.5×10^{-7}	1.94×10^{-1}
^{241}Am	2.0×10^{-7}	4.3×10^{-2}

aFrom ICRP 119 report (2013) Annex F. Effective dose coefficients for ingestion of radionuclides for members of the public

bbased on an individual dose resulting from a unit concentration of a radionuclide reaching the environment of the biosphere from a geological repository) (NAS, p.249, Table 9.1, 1983)

The dose conversion factors of these radionuclides needed for the calculation in Eq. 12.22 are also given in Table 12.1 for the case of ingestion by an adult (ICRP 2013). The dose conversion factor captures the relationship between the intake of radioactivity into human body and the resulting dose to the whole body, as a numerical value. The dose conversion factor is specific to the route of exposure such as ingestion, inhalation, or direct exposure. In the case of ingestion, the dose conversion factor translates the absorption and distribution of radionuclides in the body upon entry (by mouth) to the resulting effective dose in the body after examining their energy deposition in the target organs.

Example 12.1: Estimation of Dose to Humans from Spent Fuel Assume a hypothetical geological repository for spent fuel disposal is located at the bottom of the unsaturated zone at a site. The top of the aquifer is 400 m below the surface. 1000 years after spent fuel emplacement, an unusually severe rain storm (53 cm of rain fall) came at the site. About 10% of the rainwater uniformly infiltrated into the mountain rocks in 2 hours from the storm. The water slug happened to hit one of the waste packages that had previously failed due to corrosion. This resulted in 33.3 m^3 of water slug entering the waste package. The waste package is emplaced vertically with a length of 4 meters and contained spent fuels with 1 kg of ^{135}Cs (this assumes about 7 spent fuel assemblies in the waste package). The release occurred uniformly throughout the length of the package and the plume enters into an aquifer underlying the repository.

Away from the location of the failed spent fuel package, there is a farm with a well. The distance between the location of the waste package and the well is 5 km. Groundwater flows directly from the location of waste package to the well. A farmer living in the farm draws all of his drinking water from the well. The volume of water pumped out from the well every year for his use is 1.35 m^3.

Assume:

- All of ^{135}Cs inventory in the spent fuel is released to the groundwater (only ^{135}Cs was released from the waste package from the incident).
- The velocity that water flows in the unsaturated zone is the same as the rate water was uptaken due to rain and travels as a slug through the repository to the aquifer (no dispersion of radionuclides or loss of water along the way).

Data given:

- Effective porosity in all rock is 0.1.
- Density of the rock in the aquifer is 2.65 g/cm^3.

(continued)

Example 12.1 (continued)

- The aquifer hydraulic conductivity is 10^{-3} cm/sec.
- The change in water level from wells at the repository to the wells at the accessible environment is 4 m.
- The aquifer is 50 m deep.
- The hydraulic dispersion coefficient in the aquifer is 10^{-4} cm^2/s.
- K_d for cesium is 100 ml/g.
- The concentration of ^{135}Cs in groundwater is not limited by solubility.
- The dose conversion factor for ^{135}Cs for the ingestion pathway is 2.0×10^{-9} Sv/Bq (Table 12.1).
- The dose from drinking water is 2.67% of the total dose (Table 12.1).

a) How long will it take for the water slug from the rain to reach the aquifer?
b) What is the rate of release of ^{135}Cs from spent fuel?
c) What is the concentration of ^{135}Cs in the slug coming out of the waste package?
d) When does the peak concentration reach the location of the well at the farm?
e) What is the concentration of ^{135}Cs in the drinking water consumed by the farmer?
f) Determine the annual dose to the farmer from drinking the contaminated water during the year of ^{135}Cs peak arrival.
g) Repeat b) through f) if the waste is in glass within the waste package.

Solutions:

a) **How long will it take for the water slug from the rain to reach the aquifer?**

From the problem, 10% of rainfall (53 cm) is uniformly soaked into mountain rocks. From Chap. 11,

$$v_{\text{unsat}} = \frac{v_{0,\text{unsat}}}{\varepsilon_f} = \left(\frac{53cm \cdot 0.1}{2h}\right)/0.1 = 26.5\ cm/h = 2320\ m/\text{year}$$

Assuming constant velocity towards the aquifer,

$$t_{\text{aquifer}} = \frac{h}{v_{\text{unsat}}} = \frac{400\ m}{2320\ m/\text{year}} = 0.172\ \text{year} = \boxed{63\ \text{days}}$$

b) **What is the rate of release of Cs-135 from spent fuel?**

Rate of release from spent fuel can be found using Eqs. 12.1~12.3:

(continued)

Example 12.1 (continued)

$$r_{\text{SNF}} = r_0 e^{-\frac{34300}{RT}} \left[\frac{mg}{m^2 - \text{day}}\right]$$

$$r_{\text{SNF\_frac}} = r_{\text{SNF}} \times a_{\text{SNF}} \times \frac{365}{1000000} \left[\text{year}^{-1}\right]$$

$$\text{release}_{\text{SNF}} = m_{\text{SNF}} \times r_{\text{SNF}_{\text{frac}}} \left[\frac{kg}{yr}\right]$$

$$= m_{\text{SNF}} \times r_0 e^{-\frac{34300}{RT}} \times a_{\text{SNF}} \times \frac{365}{1000000} \left[\frac{\text{kg}}{\text{year}}\right]$$

Assuming $r_0 = 323184.8$ (from recommended value for mean), $T = 290$ K (for underground temperature, which may vary depending on the location), $a_{\text{SNF}} = 0.22$ m^2/kg (Yucca Mountain value assumption),

$$\text{release}_{\text{SNF}} = 1\ \text{kg} \cdot 323184.8 \cdot e^{-\frac{34300}{8.314 \cdot 290}} \cdot 0.22 \cdot \frac{365}{1000000}$$

$$= 1.721 \times 10^{-5} \text{kg/year} = \boxed{0.0172 g/\text{year}}$$

c) **What is the concentration of ^{135}Cs in the slug coming out of the waste package?**

Volume of slug per waste package is given: 33.3 m^3. Half-life of ^{135}Cs is 2,300,000 years, so even in 1000 years after waste emplacement, the mass of ^{135}Cs is nearly the same. Since release occurred uniformly throughout the length of package (given),

$$\text{Concentration} = \frac{\lambda N}{V} = \frac{\left(\frac{\ln 2}{2,300,000 \cdot 365 \cdot 24 \cdot 3600\ s}\right)\left(0.0172 g \cdot \frac{1 mol}{135 g} \cdot \frac{6.022 \times 10^{23}}{1 mol}\right)}{33.3\ m^3}$$

$$= 2.20 \times 10^5 \frac{Bq}{m^3} = \boxed{5.94 \times 10^{-7} Ci/m^3}$$

d) **When does the peak concentration reach the location of the well at the farm?**

For the given example problem, peak concentration reaches the location of the well when the slug reaches the well. Using Darcy's law, $v_0 = k\frac{\Delta h}{\Delta l}$

$$v = \frac{v_0}{\varepsilon_f} = \frac{k\frac{\Delta h}{\Delta l}}{\varepsilon_f} = \frac{\left(10^{-5}\ \frac{m}{s}\right)\left(\frac{4\ m}{5000\ m}\right)}{0.1} = 8 \times 10^{-8}\ m/s$$

(continued)

Example 12.1 (continued)

$$\text{Time to reach the well at the farm, } t = \frac{\text{distance}}{\text{velocity}} = \frac{5000\ m}{8 \times 10^{-8}\ m/s}$$
$$= 6.25 \times 10^{10} s = \boxed{1982 \text{ year}}$$

e) **What is the concentration of ^{135}Cs in the drinking water consumed by the farmer?**

From Eq. 11.91 (Sect. 11.3.6), retardation factor can be found using $= 1 + \frac{\rho_b K_d}{\varepsilon}$.

$$R = 1 + \frac{(2.65)(100)}{0.1} = 2651$$

Assuming source will behave like a vertical line source (h = 4 m) (Sect. 11.3.7), the peak concentration can be found by taking the exponential terms of the Green's functions as ~1:

$C(x, y, z, t) = \frac{1}{\varepsilon_f R} X_1(x, t) Y_1(y, t) \frac{1}{h}$ for unit source (Eq. 11.103) and $D_x = D_y = 10^{-4}$ cm^2/s (given)

$$\rightarrow C = \left. \frac{Q}{\varepsilon_f Rh \cdot \sqrt{\frac{4\pi D_x t}{R}} \cdot \sqrt{\frac{4\pi D_y t}{R}}} \right|_{t=6.25\times10^{10} s}$$
$$= \frac{5.94 \times 10^{-7} \frac{Ci}{m^3} \cdot 33.34 m^3}{0.1 \cdot 2651 \cdot 4\sqrt{\frac{4\pi(10^{-4})(6.25\times10^{10})}{2651}} \cdot \sqrt{\frac{4\pi(10^{-4})(6.25\times10^{10})}{2651}}} \boxed{C = 6.31 \times 10^{-13} Ci/m^3}$$

(Note that this is a result of very conservative assumptions for the simplification of the calculation)

f) **Determine the total annual dose to the farmer from drinking the contaminated water during the year of ^{135}Cs peak arrival.**

Farmer gets 1.35 m^3 of water from well per year as drinking water.
Then, the total intake is

$$6.31 \times 10^{-13}\ Ci/m^3 \times 1.35\ m^3/\text{year} = 8.52 \times 10^{-13}\ Ci/\text{year}$$
$$= 0.0315\ [Bq/\text{year}]$$

Using the dose conversion factor of 2.0x10^{-9} (Sv/Bq),

(continued)

Example 12.1 (continued)

$$H_E = 0.0315\ Bq/\text{year} \times 2.0 \times 10^{-9}\left(\frac{Sv}{Bq}\right) = 6.3 \times 10^{-11}\ Sv$$
$$= \boxed{6.3 \times 10^{-5}\ \mu Sv \text{ per year}}$$

from drinking water. This corresponds to 2.67% of the total dose (given).

The total dose $H_E = \frac{6.3\times10^{-5}\mu Sv}{0.0267} = \boxed{0.0024\ \mu Sv \text{ per year}}$ from **all exposure pathways.**

If we further assume that there are 20,000 waste packages at the repository containing the same amount of ^{135}Cs and they all fail simultaneously, the outcome corresponds to ~ 48 μSv/year or 4.8×10^{-5} Sv/year This is not a real case but gives a quick simplified sketch of an outcome of the hypothetical incident.

g) **Repeat (b) through (f) if the waste is in glass within the waste package. Assume mass of the glass waste form is 50 kg, and the mass loss of glass is so small that it is negligible when finding $a_{glass}(t)$.**

Every step is the same except for (b).

b) for glass waste form,

Assuming acidic condition (assume pH = 5.5) and using Eqs. 12.4~12.7,

$$r_{\text{glass}} = k_0 10^{\eta \cdot pH} e^{-E_{ag}/RT} = 8.41 \times 10^3 \cdot 10^{-0.49\cdot5.5} \cdot e^{-\frac{31000}{8.314\cdot290}}$$
$$= 4.425 \times 10^{-5}\left[\frac{g}{m^2 - \text{day}}\right]$$

Assuming mass loss is so small that $m_{o,\ \text{glass}} - \sum m_t \approx m_{o,\ \text{glass}}$,

$$a_{\text{glass}}(t) = f_{\text{exposure}} \cdot \left(2.7 \times 10^{-3}\right) \cdot \left(m_{o,\text{glass}} - \sum m_t\right)$$
$$= 4 \cdot \left(2.7 \times 10^{-3}\right) \cdot (50 - 0) = 0.54\left[\frac{m^2}{kg}\right]$$

$$r_{\text{glass\_frac}} = r_{\text{glass}} \times a_{\text{glass}} \times \frac{365}{1000} = 4.425 \times 10^{-5} \times 0.54 \times \frac{365}{1000}$$
$$= 8.72 \times 10^{-6}\left[yr^{-1}\right]$$

$$\text{release}_{\text{glass}} = m_{\text{glass}} \times r_{\text{glass\_frac}} = 50 \cdot 8.72 \times 10^{-6} = 4.36 \times 10^{-4}\left[\frac{kg}{yr}\right]$$

Assuming 1 kg of Cs-135 is mixed evenly to form 50 kg of glass waste form,

(continued)

Example 12.1 (continued)

$$\text{release}_{Cs} = \frac{1\text{ kg}}{50\text{ kg glass}} \cdot 4.36 \times 10^{-4} \frac{\text{kg glass}}{\text{year}} = 8.72 \times 10^{-6}\text{kg/year}$$

$$= 0.00872\text{ g/year}$$

c) for glass waste form,

$$\text{Concentration} = \frac{\lambda \text{N}}{\text{V}} = \frac{\left(\frac{ln\,2}{2,300,000 \cdot 365 \cdot 24 \cdot 3600\ s}\right)\left(0.00872g \cdot \frac{1\text{mol}}{135g} \cdot \frac{6.022\times 10^{23}}{1\text{mol}}\right)}{33.3m^3}$$

$$= 1.115 \times 10^4 \frac{Bq}{m^3} = \boxed{3.01 \times 10^{-7} Ci/m^3}$$

d) for glass waste form,

Time to reach the well at the farm is the same: $\boxed{1982\text{ year}}$

e) for glass waste form,

$$C = \left.\frac{Q}{\varepsilon_f Rh \cdot \sqrt{\frac{4\pi D_x t}{R}} \cdot \sqrt{\frac{4\pi D_y t}{R}}}\right|_{t=6.25\times 10^{10} s}$$

$$= \frac{3.01 \times 10^{-7} \frac{Ci}{m^3} \cdot 33.34m^3}{0.1 \cdot 2651 \cdot 4\sqrt{\frac{4\pi(10^{-4})(6.25\times 10^{10})}{2651}} \cdot \sqrt{\frac{4\pi(10^{-4})(6.25\times 10^{10})}{2651}}} \quad \boxed{C = 3.20 \times 10^{-13} Ci/m^3}$$

f) for glass waste form,

$$H_E = 3.20 \times 10^{-13}\left(\frac{Ci}{m^3}\right) \times 1.35\left(\frac{m^3}{\text{year}}\right) \times 3.7 \times 10^{10}\left(\frac{Bq}{Ci}\right) \times 2.0$$
$$\times 10^{-9}\left(\frac{Sv}{Bq}\right)$$
$$= 3.2 \times 10^{-11}\ Sv = \boxed{3.2 \times 10^{-5}\ \mu Sv \text{ per year}}$$

from drinking water

Total dose = $H_E = 3.2 \times 10^{-5}\ \mu Sv \div 0.0267 = \boxed{0.0012\ \mu Sv \text{ per year}}$ from **all exposure pathways.**

Again, if we further assume that there are 20,000 waste packages at the repository containing the same amount of ^{135}Cs in the glass and they all fail simultaneously, the outcome corresponds to about ~24 μSv/year.

12.4 Results of Performance Assessment: Examples

12.4.1 *Comparison of Sites in the U.S. Through Generic Performance Assessment*

A preliminary performance assessment was performed as part of a national study on the isolation of nuclear waste for potential repository sites in different rock types in the U.S. (National Research Council 1983). The study was based on conceptual repository designs in basalt, salt, tuff, and granite. The basalt repository was at a site in Hanford, Washington. The location for the salt repository was in Permian basin in Texas or Paradox basin in Utah and Colorado. The granite repository was a generic case with no specific site under consideration. The tuff repository was assumed at the Nevada Test site but the results for this site is not included in this discussion as more detailed analysis is described in the next section.

The study assumed the disposal of vitrified HLW at each site from reprocessing of spent fuel equivalent to the fuel mass of 100,000 ton of uranium. Key hydrologic features of each site considered as input data are the distance from the repository to the affected biosphere and the groundwater velocity. The values of K_d in different host rocks for the radionuclides considered are listed in Table 11.13 Table 12.2 shows the summary of the results of the performance assessment. The results assume solubility-limited release of radionuclides. The peak dose to humans was highest at the salt repository (~10^{-5} Sv/year) compared to 10^{-7} or 10^{-9} Sv/year at the granite and basalt repository, respectively. Because there is no groundwater in salt, the analysis made an ad-hoc assumption that a major diversion of an aquifer in surrounding non-salt strata resulted in the contaminated flow from a salt repository. The results show that the estimated doses are lower than 10^{-4} Sv/year, a likely example of generic performance goal. If the release of radionuclides is assumed to occur congruently with the dissolution of the waste form, the results could be higher by two or three orders of magnitude. Among various radionuclides contributing to human dose, ^{237}Np was found to be one of the top contributors at all repository sites. This agrees with the expectation due to the long-half life and relatively mobile

Table 12.2 Observations from generic analysis of performance assessment for different repositories

	Basalt	Granite	Salt
Site	Hanford, WA	Generic	Permian basin (Texas) and Paradox basin (Utah, Colorado)
Distance to biosphere (km)	14	10	100
Water travel time	1.5×10^4	10^5	10^5
Maximum dose to humans (Sv/y)	3×10^{-9}	~10^{-7}	$3\text{x}10^{-5}$
Top dose contributing nuclides	^{237}Np, ^{14}C, ^{135}Cs, ^{79}Se, ^{210}Pb, ^{129}I, ^{99}Tc,	^{14}C, ^{237}Np, ^{135}Cs, ^{79}Se	^{135}Cs, ^{237}Np, ^{14}C, ^{99}Tc

and soluble nature of ^{237}Np (as discussed in Sect. 11.4). Other important radionuclides included ^{14}C, ^{135}Cs, ^{79}Se, ^{210}Pb, ^{129}I, and ^{99}Tc. No detailed uncertainty analysis was made in the study. Due to the preliminary nature of the analyses, however, the values of the estimated doses need to be interpreted with caution as conservative examples.

12.4.2 Results of Performance Assessment for the Yucca Mountain Repository

Two separate (but related) performance assessments have been conducted for the development of the Yucca Mountain repository by the U.S. Department of Energy (DOE). The first one, 1998 Viability Assessment (DOE 1998), is to determine the suitability of the Yucca Mountain site. The second one, 2008 Final Environmental Impact Assessment, is part of U.S. DOE's license application to US NRC for repository construction (DOE 2008). These analyses estimated potential human health impacts of the repository into the future by selecting a group of individuals whose living conditions and lifestyle may represent the reasonably maximally exposed individual (RMEI). A group of subsidence farmers living near water wells located at about 18 km downgradient from the repository was selected for this purpose. The performance was examined as radiological dose to these RMEI for the 10,000 and 1000,000 year post-closure periods.

12.4.2.1 Scenarios Analyzed

The principal scenario used for the performance assessment is groundwater induced human radiation exposure. Rainwater at the site moves down through the unsaturated zone into the repository and results in degradation of nuclear waste packages. The resulting releases of radionuclides are carried by the groundwater downward through the unsaturated zone and on through the saturated zone to locations where human exposure could occur.

A baseline assumption for the level of water precipitation at the site in the future is that future climates are similar to current conditions with possibility of being wetter. Water in the unsaturated zone generally moves downward in the rock matrix and through fractures. The rock at Yucca Mountain is tuff with varying degrees of fractures. The overall unsaturated flow system is very heterogeneous as the water flowing in the fractures moves much more rapidly than the water moving through the rock matrix.

A potential design feature of the Yucca Mountain repository is to maintain the temperature of the system to be high above the water boiling point using the decay heat of nuclear waste. This is possible as the repository is located with the unsaturated zone. If the loading of nuclear waste packages in the tunnel drifts is made to keep the waste packages to remain very close to each other, the resulting high heat density in the repository tunnels causes the temperature of the repository rocks to be high enough to drive off moisture or the water infiltrating into the repository. This effectively delays infiltration of water into the repository until the significant portion of the decay heat dissipates away due to radioactive decay. If the loading of nuclear waste is not dense enough and the temperature of the repository remains relatively low, then the infiltrating groundwater moves to reach the repository walls within first few hundred years and begins to drip into the waste packages in a few places. Over time, the number and locations of water infiltration vary depending on the changing climate conditions, natural rock conditions, or chemical alterations of the engineered materials. The chemistry in the repository tunnels is continually changing because of the interactions among the incoming water, circulating gas, and engineered materials (e.g., concrete or metals) in the tunnel drifts.

The nuclear waste emplaced in the repository will be enclosed in a double-barrier waste package with two different materials (i.e., stainless steel or carbon steel for the canister and Alloy 22 for the overpack). Corrosion of the waste packages will be influenced by the changing thermal, hydrologic, and chemical conditions in the repository. Water eventually penetrates a waste package through localized corrosion-induced failures and contacts the spent nuclear fuel. The water first contacts the cladding and with the breach of cladding, the fuel pellets are exposed to water. When the fuel pellets are contacted by water, dissolution of mobile species in the fuel rod voids takes place first. This is followed by the corrosion of grain boundaries and UO_2 matrix dissolution. Dissolution of UO_2 matrix will be strongly dependent on the oxygen-to-uranium ratio, the extent of burnup, and irradiation history. The reaction rates of individual nuclides will vary depending upon their chemical properties. Once the waste form begins to alter, it may take about 1000 years for the spent fuel waste to completely degrade (Bechtel SAIC 2004).

The dissolved radionuclides are carried away in a flowing water to move out of the waste package through a pit or any opening in the package into the waste emplacement tunnel. After escaping from the waste package, the radionuclides can move through materials on the floor of the tunnel. Some radionuclides may stick on the floor or may move in the water as dissolved ions or colloidal particles after being attached to them. The radionuclides in the water move downward beneath the repository with some water moving rapidly in fractures and some much more slowly in the rock matrix. Depending upon the degree of sorption, radionuclides may experience retardation in the movement with groundwater. When the water reaches the water table, it flows horizontally in a generally southerly direction toward the Amargosa Valley. The radionuclides gradually become more dispersed and the corresponding concentration continues to decrease.

Once the radionuclides reach the location of water wells, the contaminated groundwater can be pumped out and cause radiation exposure to humans through various exposure pathways. These exposure pathways include ingestion of food and water, inhalation of suspended particles in the air, and direct external exposure.

Other scenarios relevant to human radiation exposure at the site were also considered. These included earthquake, volcanic activity or human intrusion.

Earthquakes during the post-closure periods affect the repository, primarily through ground motions, which cause rockfall into the repository tunnel. The frequency of earthquake at Yucca Mountain is rather high. The probability of volcanic activity at Yucca Mountain as a repository-disturbing event was estimated to be extremely low according to the past records in the area. Nonetheless, formation of a small cinder cone by a dike that flowed up through the repository tunnels due to an explosive eruption was considered. A borehole drilled directly through the repository was also considered as an example of human intrusion. Through such drilling, the contents of waste package could be released to the groundwater. The probability of such event occurring was estimated and assumed to be absolute (i.e., always to occur with a finite probability).

12.4.2.2 Results from Site Viability Assessment

The first DOE performance assessment for the Yucca Mountain repository (DOE 1998) was mainly to examine the suitability of the site while the design of the repository was still in progress. The waste package design was based on using carbon steel canisters and Alloy 22 overpack. Loading of spent fuel or HLW was based on so-called low-temperature operating mode. Analyses were made for the 10,000 and 1000,000 year post-closure periods using both the base case scenario of groundwater-induced release of radionuclides and the case of disruptive events scenarios, i.e., volcanic activity, earthquakes, and human intrusions.

The Base Case – Groundwater Induced Release

For the 10,000 year post-closure periods, the estimated maximum dose to humans through groundwater contamination was about 4×10^{-7} Sv/year (0.04 mrem/year), occurring at 10,000 years. The major factors controlling the dose results were waste package failure times and water infiltration rate. Presence of unsaturated zone between the repository and the water table (spanning ~300 m) caused delays in the transport of radionuclides into the saturated zone for about 300 years. Transport of radionuclides in the saturated zone caused dilution of concentration from plume dispersion. The degree of dilution before getting to the receptor well location was about by a factor of 4 (as the median value) or 10 (as the mean value). Within the 10,000-year period, only nonsorbing radionuclides such as ^{99}Tc and ^{129}I reached the biosphere while most other radionuclides did not reach the biosphere due to retardation in their movement. The top dose contributors (see Fig. 12.7) were ^{99}Tc (38% of the peak dose rate), ^{129}I (31%), and ^{14}C (4%).

In the results for the 1000,000 year post-closure periods, the estimated peak dose was about 3×10^{-3} Sy/year (300 mrem/year) arriving at around 320,000 years. The key controlling factors were again water infiltration and the waste package failure history. The cladding failure rate was also found to be important to explain the radionuclide release behaviors. The unsaturated zone was found to be of little

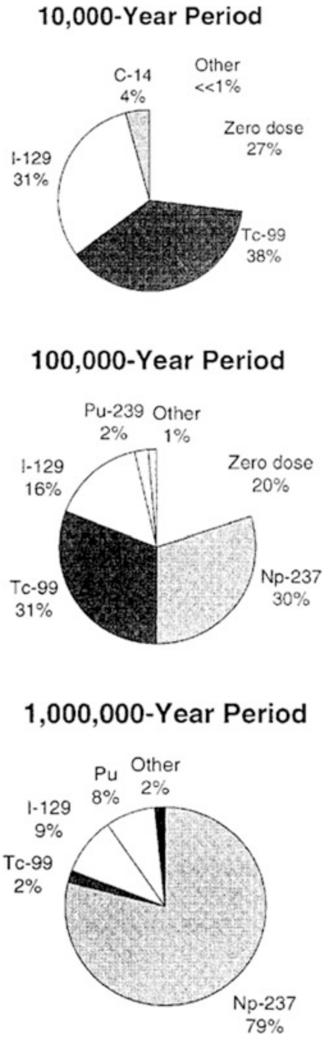

Fig. 12.7 Yucca Mountain repository viability assessment results as the average contributions of major radionuclide to the peak dose rate at three different post-closure periods (10,000 years; 100,000 years; and 1000,000 years). (DOE 1998)

consequence in affecting repository performance during the very long post-closure periods while the saturated zone transport was found to have a major effect on repository performance through plume dilution. ^{237}Np (79%) was the top contributor to the peak dose followed by ^{129}I (9%), ^{239}Pu and ^{242}Pu (8%), and ^{99}Tc (2%) (Fig. 12.7).

The release of ^{237}Np was mainly limited by the slow rate of cladding failures while the solubility-limited release played some role. With ^{129}I and ^{99}Tc, the release was mainly controlled by waste package failure rates. Their transport in the saturated zone was not limited by solubility and sorption didn't have much retardation effect on them either. The release of Pu was also solubility-limited and mainly dependent on cladding failures. The effect of solubility and sorption was also significant in the transport of Pu in groundwater.

The results also showed early rise in the projected dose which reflects the impact of early failures of waste packages. After this initial peak, waste canisters were expected to remain intact during 100 ~ 1000 years of post-closure period (the repository temperature remains relatively high during this period). Beyond 1000 years up to 10,000 years, waste canisters and overpack began to corrode with water infiltration into the repository. Minor releases of radionuclides began and high solubility nonsorbing nuclides resulted in the dose impact at the receptor location. The peak dose did not arrive during this period. Beyond 10,000 years and unto one million years, waste canisters and overpack might no longer be effective in isolating nuclear waste and groundwater continued to come in through the repository. All of the mobile radionuclides migrated to the accessible environment with groundwater. Their movements were controlled by solubility and sorption. The nuclides with long half-life survived and reached the biosphere resulting in human dose.

Disruptive Event Scenarios

The disruptive event scenarios include volcanic activity, earthquakes, and human intrusions. In the case of volcanic activity, three different sub-scenarios were considered: (1) direct release from volcanic eruption dispersing radioactively contaminated ash on the ground, (2) enhanced source term due to liquid magma intersecting the repository tunnels leading to failures of waste packages, (3) indirect effect as the liquid magma from the igneous activity not contacting the waste but changing the flow of groundwater (this affected groundwater related repository performance). The probability of volcanic event intersecting the repository area at Yucca Mountain was estimated at 1.5×10^{-8} per year with 90% confidence interval of 5.4×10^{-10} to 4.9×10^{-8} (CRWMS M&O 1996). In the case of direct release, the estimated dose was lower than the peak dose rate from the base case of groundwater infiltration by approximately a factor of two million. In the case of enhanced source term, the peak dose rate was estimated to be less than half of the peak dose in the base case. The case of indirect effect on groundwater flow was found to have no significant impact on the dose rate.

In terms of event probability, the estimation indicated that there is less than one in 1000 chance of igneous activity at Yucca Mountain over the 10,000 year period. Also, for the one million year period, there was less than 10% chance of igneous activity. The chance of direct release of radionuclides happening among the volcanic activities was estimated at less than 6% for both time periods. About 60% of the time, volcanic eruptions would cause an enhanced release of radionuclides from waste packages from liquid magma activities over the 10,000 years period. This chance slightly would increase up to 70% over the one million year period, according to the estimation.

Regarding earthquakes, their potential impacts on the repository were analyzed by examining the vibratory ground motions and displacements causing rockfall and disruptions to the waste packages (CRWMS M&O 1998). Results indicated that seismic activities had almost no impact on repository performance during the period of one million years. This was because the damages to waste packages were not severe enough to cause radionuclide releases. The probability of rockfall causing a waste package to breach was essentially zero over 10,000 years. Over one million years, rockfall was expected to cause about 30% of the waste packages to breach in the repository. However, the overall probability of waste package failure did not change when these breaches were added to the estimated rate of corrosion related failures. Other impacts such as changes in site hydrologic properties and alterations in groundwater flow patterns were found to have insignificant influence on groundwater related consequences.

Human intrusion through drilling assumed the penetration of a waste package by a drill hole. In this case, the drilling was assumed to take place as a deterministic event at some point in time during the post-closure periods. The resulting dose impact was found to vary (either smaller or larger than the base case dose depending upon the timing of drilling) but the overall consequence was within the range of variability of the base case results.

12.4.2.3 Results from the Final Environmental Impact Analysis for the Yucca Mountain Repository

As part of final environmental impact analysis, US DOE conducted a second round of performance assessment for Yucca Mountain (DOE 2002). This analysis reflected a number of key design changes compared to the 1998 Viability Assessment. Nuclear waste package design used the stainless steel for the canister instead of carbon steel. Loading of spent fuel and HLW to the repository was based on so-called high temperature operating mode. Therefore, the repository was assumed to be under hot conditions to drive off any infiltrating water preventing water from directly contacting waste packages. Titanium alloy drip shield was added to the design of nuclear waste package. The results showed enhanced performance of the repository in comparison to the results from the 1998 Viability Assessment.

The Base Case: Groundwater-Induced Impacts

In this 2002 analysis, possible effects of climate changes and seismic events were combined into the base case analysis of groundwater related waste package failures. Unlike the 1998 analysis, seismic events were incorporated into the base case analysis.

The results (Table 12.3) showed that for the first 10,000 years after repository closure, the mean peak dose was 2×10^{-10} Sv/year (0.00002 mrem/year) with the 95th percentile peak dose at 1×10^{-9} Sv/year (0.0001 mrem/year). These much

Table 12.3 Projected peak dose from groundwater release at the location of reasonably maximally exposed individuals during 10,000 or 1000,000 post-closure years (DOE 2002)

	Mean		95th-percentile	
Time Period	Peak dose (mrem/year)	Time of peak (year)	Peak dose (mrem/year)	Time of peak (year)
10,000 post-closure years	2×10^{-5}	4900	1×10^{-4}	4900
1000,000 post-closure years	150	480,000	620	410,000

lower results compared to the 1998 Viability Assessment were mainly due to improved durability of waste packages (i.e., from the use of drip shield and stainless steel canister). The waste packages were expected to remain intact significantly longer than 10,000 years. The challenge in this approach was to verify the long-term corrosion resistance of the waste packages when new materials such as titanium, alloy 22, and stainless steel were incorporated in the design. The radionuclides that were estimated to contribute the most to individual dose (the RMEI mean dose at 18 km) in 10,000 years were ^{99}Tc (77%), ^{14}C (16%), and ^{129}I (7%).

For the period between 10,000 year and one million years, the estimated mean and the 95th percentile individual annual peak dose (the RMEI mean dose at 18 km) was 1.5×10^{-3} Sv/year (150 mrem/year) and 6.2×10^{-3} Sv/year (620 mrem/year), respectively. The radionuclides contributed the most to the dose were ^{237}Np (79% of the peak dose), ^{129}I (9%), ^{239}Pu and ^{242}Pu (8%), and ^{99}Tc (2%). Arrival of multiple peaks were observed at 200,000 years or later after the closure of repository closure (this was caused bythe transitions in climate states) (Fig. 12.8).

Depending on the time frames covered by the analysis, different parameters contributed significantly to the overall uncertainty of the results. These are shown in Table 12.4. When the drip shields are intact and no water is dripping on the waste package (between 125,000 and 250,000 post-closure years), degradation of Alloy-22 overpack was governed by the humid air corrosion. After 250,000 years, most waste packages were estimated to have failed. At that point, the water infiltration scenario became the most important source of uncertainty. One of key issues was how to describe water flows through fractures leading into episodic infiltration contacting the waste packages. Also the variations in water infiltration due to climate changes were found to be an important source of uncertainty.

For the scenarios of disruptive events, i.e., volcanic activity and human intrusion, the results were not significantly different from those of the 1998 analysis. For the volcanic activity scenario, the maximum impact was estimated to occur at 300 years after the repository closure with an annual dose to a RMEI at less than 1×10^{-6} Sv/year (0.1 mrem/year). Human intrusion by drilling was assumed to occur 30,000 years after the closure of the repository with the expectation of enough degradation of waste packages (the scenario assumed no detection of radioactive object penetration by the driller). The resulting dose as mean was less than 2×10^{-8} Sv/year (0.002 mrem/year) occurring around 100,000 years after repository closure. This is at less than one-tenth of the radiological dose from a disruptive volcanic

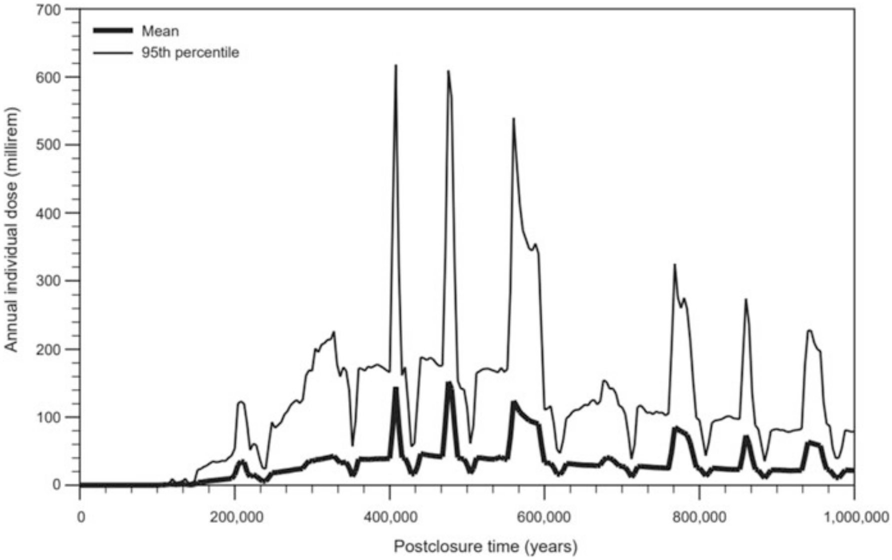

Fig. 12.8 Results of 2008 DOE total system performance assessment (TSPA): mean and 95th percentile annual individual dose during one million years after repository closure at the reasonably maximally exposed individual (RMEI) location (source: DOE 2002)

Table 12.4 Key parameters contributing to the uncertainty of performance assessment at different post-closure time (source: DOE 2002)

Time after closure	Two most important parameters
125,000 years	General humid air corrosion rate of Alloy-22 outer lid
	General humid air corrosion rate of Alloy-22 inner lid
250,000 years	General humid air corrosion rate of Alloy-22 outer lid
	General humid air corrosion rate of Alloy-22 inner lid
500,000 years	Episodic infiltration of water through fractured rock
	General humid air corrosion rate of Alloy-22 outer lid
1000,000 years	Episodic infiltration of water through fractured rock
	Infiltration rates (low, medium, and high)

event. Results indicated that the overall impact of disruptive events was lower than that of the base case.

12.4.3 Results of Performance Assessment from Select Countries

As a comparison to the results from U.S. geological repository, example results of performance assessment from a select group of other countries are shown in

Table 12.5 Examples of performance assessment results for geologic repositories in select countries

Country	Results of Performance Assessment	Site
Belgium	Maximum exposure (at 200,000 year time) 10 to 30 times lower than typical internationally accepted dose limits for geological disposal facilities of 0.1 to 0.3 mSv per year. (Chapman and McCombie 2016)	Boom clay
Canada	Peak dose ~900 times lower than the interim dose acceptance criterion of 0.3 mSv per year and occurs at ~100,000 years after closure. (Hunt et al. 2014)	Hypothetical crystalline rock
Finland	Max exposed group; peak dose ~10^{-5} Sv/year at ~3000 years after closure (POSIVA 2012)	Olkiluoto
France	0.019 mSv/year at 330,000 years. (ANDRA 2005)	Oxfordian limestone
Germany	10^{-5} Sv/a period of ~300,000 to 360,000 years	Konrad mine
	Several million years, same order of magnitude or lower occur exposures due to long-lived actinides and their daughter products (Brennecke 2004)	
Japan	~10^{-5} mSv/year at almost one million years. (JNC 2000)	Crystalline rocks, sedimentary rocks
Korea	0.01 mSv/year *at ~ 10,000 years* (Kim et al. 2001)	Plutonic rocks
Sweden	10^{-8} risk within 100,000 years. (Strömberg 2016)	Forsmark site
Switzerland	5.3×10^{-5} mSv/year at 1.0×10^{6} year (NAGRA 2002)	Opalinus clay
Taiwan	The sum of the peak annual doses is 4.08×10^{-4} μSv /year (TPC 2017)	Granite formation

Table 12.5. Some of the results are genetic without specifying a site for the repository. The majority of assessments have resulted in peak doses to the maximally exposed individual as significantly less than the 0.3 mSv/year dose level recommended by the IAEA. These projected doses are typically well below the average annual dose to an individual from the natural activity occurring in each respective country and indicate that the regulatory limits of performance assessment can be met with enough margin of safety (See Table 2.1 for national regulatory approaches to safety in nuclear waste disposal). These results indicate the feasibility of meeting the goal of nuclear safety in performance assessment under the variations in the natural repository conditions.

12.5 Natural Analogues

Natural analogues refer to the materials, artifacts, or processes which may be used to produce data or insights that are unobtainable by other means in support of repository performance assessment. Data available from natural analogues could be utilized to overcome the challenge associated with using data collected over relatively short periods of time with a relatively limited number of testing conditions.

12.5.1 Need for Natural Analogues

It is not feasible to perform long-term, real-time experiments on the behavior of materials for isolation and containment of nuclear waste prior to implementing nuclear-waste disposal. As an alternative, natural analogues, representing a study of uncontrolled “natural experiments” using nature or human artifacts of ancient origin thus, help to shed light on projecting the performance of the geological repository or its subsystems. With long-term history embedded, natural analogues may resemble what is expected to occur in a geological repository in the future.

Natural analogue studies have been made on ancient man-made artifacts (using copper, iron, concrete), uranium ore deposits, natural fission reactors, or marine sediments and various geological settings. Therefore, the models and databases needed to predict the behavior of waste forms, waste packages, or radionuclide movements in groundwater (glass, cement, metallic waste containers, clay backfill materials, and radionuclides in the near-field or the far-field) may be tested by using natural analogue studies. These natural analogues may also help public communication efforts as they provide some basis for scientific understanding of the processes involved in a visible way.

12.5.2 Natural Analogues for Waste Forms, Metallic Containers, and Backfills

12.5.2.1 Glass

Long-term stability of glass from natural origins has been known to the scientific community. Due to the differences in the composition and the exposed conditions, natural glasses cannot be used to quantitatively assess the leaching characteristics of vitrified HLW (IAEA 2005). Nevertheless, they can be useful to explain the mechanisms of glass corrosion and the formation of secondary alteration products as part of the explanations of long-term behavior of nuclear waste glass. Massive glass slabs found in Bet She’arim in Israel, an archaeological artifact, are an example of natural glass as a source of information for long-term behavior of vitrified HLW.

12.5.2.2 Cementitious Materials

Natural cement minerals exist in geological formations showing their long-term stability. The most comprehensively studied one is the Maqarin natural analogue in Jordan. The information from the Maqarin cement system has been used to test and evaluate hydrogeochemical computer code to support nuclear waste disposal in cementitious environment. There are also a number of archaeological analogues that show long-term durability of concrete or cement. These are mostly the remains of the

Roman constructions dating back to the third century BC. Studies of these archaeological analogues may help the predictive analysis of concrete system in nuclear waste disposal.

12.5.2.3 Metallic Containers

Archaeological analogues of iron metal are available from the iron nails used by the Roman soldiers and from various museum collections in the world. Studies of these artifacts provided the estimation of uniform corrosion rates of iron or iron alloys as ranging between 0.1 and 10 μm/year (Johnson and Francis 1980). Numerous archaeological analogues of copper are also available from the collection of early weaponry and tool fabrication (smelting/casting) dating back to 6000 BC. Studies on these archaeological artifacts provided the estimate of the uniform corrosion rates of copper or cooper alloys, ranging between 0.025 and 1.27 μm/year (DOE 2008).

12.5.2.4 Bentonite Backfill

A mountain range in central Cyprus, called the Troodos Ophiolite complex, contains a large mass of bentonite (hundreds of tons) with the site condition resembling what would be expected in a geological repository. Therefore, the site provides a natural analogue for the study of bentonite clay as waste backfill. Results from the studies at the site indicated very limited reactions between bentonite clay with alkaline (pH 10–12) groundwater over a long period (10^5 to 10^6 years). The finding supports the notion that any long-term reaction of bentonite backfill with alkaline groundwater in a geologic repository will be minimal.

12.5.3 Natural Analogues for Spent Fuel Disposal or Radionuclides Transport

A number of large uranium deposits exist in nature that could provide useful data for the dissolution and migration of uranium. Due to the differences in the experienced conditions, the information from these natural analogues may not be applicable to explaining the behavior of spent fuel in a geological repository. Nevertheless, studies of them provide useful qualitative understanding of UO_2 dissolution and alterations. Natural analogues for the study of radionuclide transport in groundwater are also available. These include Oklo mine (in Gabon), Cigar Lake (in Canada), Alligator Rivers (in Australia), Morro do Ferro (in Brazil), and Loch Lomond (in the U.K.). While the site conditions vary, these studies are very useful in testing and validating computer codes and databases to support the modeling of radionuclide transport.

12.5.3.1 Oklo Mine

Oklo is a site of uranium mine located in the West African country of Gabon with its mining operations begun in 1956. In 1972, a discovery was made by a French scientist performing mass spectroscopy that the isotope ratio $^{235}U/^{238}U$ in some parts of the mine was much lower than the normal. Subsequent investigations led to a conclusion that ^{235}U in the ore had been depleted in natural fission reactions. This meant presence of natural fission reactors at the site. Further studies found presence of fission products and concluded that the natural fission reactions took place intermittently, at a depth of about 3500 m, for more than half a million years, at a time about two billion years ago. Today Oklo is known as the only natural nuclear reactor in the world.

Studies at Oklo showed that the uranium ore at the site (uraninite) experienced little dissolution over time and that the actinides were retained locally with either low solubility along with sorption or retention in the uraninite crystal structure. Some of the mobile fission products (^{90}Sr, ^{137}Cs, ^{99}Tc) were found to have migrated with groundwater away from the reactor zone. The distance was within the predicted range based on the current state of knowledge.

As the information about the conditions of pH, Eh, and temperature changing over time at the site is not available, it is difficult to make meaningful geochemical predictions using the results from the Oklo study. It is also difficult to develop quantitative information on the dissolution of UO_2 as the uraninite at Oklo contains lower concentrations of fission products than what would be present in actual spent fuel. With low power density and low radiation and temperature effects, the fission products at Oklo were expected to have stayed mostly within the uraninite crystal. Nevertheless, Oklo remains an important natural analogue to test predictions of radionuclides movements.

Example 12.2: Geological Repository Analog Using the Oklo Natural Reactor The Oklo nuclear reactor is an often cited natural analogue for the containment of certain radionuclides in a natural setting.

a) Explain, from your understanding of geochemistry, how the very rich uranium ore deposits may have resulted in the first place.

Use the following information:

At the beginning of the earth, uranium was nearly uniformly distributed amongst rocks and soils and no significant rich deposits existed. The Oklo deposits are very rich uranium deposits that are located near the mouths of rivers that drain a large fraction of water in the western portion of Africa and they went critical approximately two billion years after the earth's inception.

(continued)

Example 12.2 (continued)

b) Does the natural analogue of the Oklo deposits have any positive or negative implications for the long-term geologic storage of spent fuel or high-level waste?

Answers:

a) Uranium may travel through groundwater extremely slowly. Nonetheless, uranium that are nearly uniformly distributed amongst the rocks and soils of western portion of Africa would be transported through groundwater to the mouths of the rivers. Not thousands or millions of years, but after ~two billion years, Oklo deposits finally may have become rich enough in uranium to go critical. This was possible because large fraction of water in the western portion of Africa would drain at near Oklo deposits. The resulting accumulation of uranium over billions of years could have resulted in high enough concentration levels.

b) The natural analogue of the Oklo deposits have positive implications in showing little migration of fission products away from the locations of fission and very low rate of dissolution of actinides. This may indicate the possibility of isolating and containing nuclear waste within natural geological barriers. The fact that it took over two billion years to accumulate the Oklo deposits may imply robustness of geological systems as natural barriers against migration of transuranic waste. There is also negative implication from the Oklo study. The general public may see the Oklo deposits showing the possibility of nuclear wastes being transported away through geological barriers as time passes. However, this requires complete dissolution of nuclear waste in groundwater which is virtually impossible due to low solubility characteristics of most actinides in the geologic disposal conditions.

12.5.3.2 Cigar Lake

Cigar Lake is the site of uranium mine located in northern Saskatchewan, Canada (the mining operation began in 2014). While the site is the location of the second largest uranium deposit in the world, the surface water in the lake on top of the uranium deposit shows no sign of uranium presence. It turns out that a clay mineral layer underneath a saturated sandstone deposit surrounds a very concentrated uranium oxide core. The clay layer provides the isolation of uranium deposit preventing the migration of uranium to the water body. Natural analogue studies at the site investigated the stability of the uraninite and the mechanisms of radionuclide retention in the ore body, including the study of the effect of colloids. The nuclides investigated include uranium plutonium, thorium, technetium, copper, nickel, barium, lead, strontium, zinc, molybdenum, chromium, and arsenic. The issues of near-field transport of radionuclides under the influence of radiation induced oxidation were also examined through hydrogeological modeling.

12.5.3.3 Alligator Rivers

The Koongarra uranium-ore deposits in the Alligator Rivers region of northern Australia serve as natural analogue for radionuclide transport modeling in groundwater (Golian et al. 1992). Geochemical and hydrogeological processes as part of the modeling work were examined for the various types of rocks at the site. The processes leading to the decomposition and leaching of the primary ore (uraninite and pitchblende) were also studied. Measurements made at the site include migration distances of nuclides, groundwater velocity, and sorption equilibria based on sequential extraction of rock samples for ^{238}U, ^{234}U, and ^{230}Th. Functional dependences of solubility and sorption on groundwater redox conditions and the effect of colloids were also studied. Through the studies, techniques for in-situ K_d measurements were improved and thermodynamic solubility and speciation predictions by hydrogeochemical computer codes were validated. Radiochemical database was also developed from these studies. Uranium mining activities have not taken place at the Koongarra unanium-ore deposits and the area is under protected as part of a national park since 2013.

12.5.3.4 Pocos de Caldas

A thorium deposit containing 20,000 Mg of Th located on the Pocos de Caldas plateau (known as a spar city), north of São Paulo in southeast Brazil, serves as natural analogue of far-field radionuclide transport.

The site, known as Morro do Ferro has been the location of extensive studies on transport and retardation of radionuclides under different redox conditions with consideration of the effect of colloids. Using the data collected at the site, hydrogeochemical computer models and related databases were validated with respect to the prediction of the solubility-limiting phase, the saturation concentration and the aqueous speciation of uranium, thorium, lead, strontium, nickel, manganese, aluminum, and zinc.

12.5.3.5 Other Natural Analogues

A number of other natural analogues are also available to support performance assessment for specific types of geological conditions. These include Loch Lomond in the U.K. for sedimentary rocks, Palmottu uranium deposits in Finland for crystalline rocks, and Mont Terri URL in Switzerland for clay rocks. At Loch Lomond, studies of migration of uranium, radium, iodine and bromine from the marine sediment layer into the neighboring freshwater layers have been conducted.

Natural analogues are also available to address other issues of long-term projection, such as impact of glaciation. Glaciated host rocks in Greenland were studied to better understand the hydrogeological and hydrogeochemical processes associated with future cold climate conditions and glacial cycles and their potential impact on long-term performance of deep geological repositories (Claesson et al. 2016).

For the issue of water infiltration through the unsaturated zone and its impact on waste packages, using various archaeologic artifacts in underground openings was considered. These archaeological artifacts support predictions of water infiltration and their effect on underground materials. Examples of these artifacts include the degree of preservation of Paleolithic cave paintings in southwestern Europe, murals and artifacts in Egyptian tombs, painted subterranean Buddhist temples in India and China, and painted underground churches in Cappadocia, Turkey.

12.5.4 Cautions in the Use of Natural Analogues

While natural analogue can be a very useful aide to performance assessment, cautions need to be exercised in the use of the data obtained from natural analogue studies. Natural experiments usually come with poor control of the conditions to which materials are exposed. Therefore, the conditions or processes experienced by the natural experiments imply large degree of uncertainty and may not reflect what would occur into the future in a geological repository.

To use natural analogue studies to support performance assessment, the effects of other processes which may have been involved in the geochemical system must be delineated to enable quantitative assessment of such effects. The physico-chemical conditions expected in a geological repository (temperature, pressure, concentration, pH, Eh, etc.) should not differ significantly from possible variations in the conditions represented in natural analogues. Also, the limitations in the use of the results of natural analogue studies to describe hydrogeological modeling of radionuclide transport should be fully acknowledged. The timescale of the processes involved in natural analogues needs to be well characterized to assess applicability for long-term projections as well.

12.6 Conclusion

One of the most important activities in nuclear waste disposal is to obtain license approval to build and operate geological repository. The approval is based on assuring safety in nuclear waste disposal through long-term protection of the public and the environment. In this respect, a performance assessment is a key milestone effort. It is to develop understanding of the level of safety associated with a proposed geological repository. While absolute assurance of regulatory compliance is not attainable, performance assessment attempts to provide the basis for the judgment on the expected safety level of the repository systems with reasonable assurance. Although performance assessment is quantitatively driven with numerical results as outcome, the substance of the results is qualitative in representing the level of safety of nuclear waste disposal. Performance assessment considers natural as well as human initiated disruptions to describe the state of nuclear waste isolation and potential release and migration of radionuclides in the environment over large spatial and temporal scales. Many scientific disciplines are involved in the development and execution of the models for performance assessment. This makes performance assessment an inherently uncertain projection. Nevertheless, performance assessment helps to identify the major sources of uncertainty and the need for the development of good conceptual models. The findings from performance assessment guide the effort in repository development in design and construction by helping to focus resources on the key issues of concern with most effect on repository performance. Performance assessment also provides a tool to evaluate specific design alternatives and strategies to achieve long-term isolation and containment of nuclear waste. By providing clearly defined subsystem performance data, performance assessment also provides transparency in safety demonstration and simplifies the necessary presentations and reviews.

Homework

Problem 12.1 List five key radionuclides of importance in performance assessment. Describe why they are the major dose contributing radionuclides.

Problem 12.2 Repeat the calculations of Example 12.1 for other radionuclides of importance in performance assessment such as ^{99}Tc, ^{129}I, ^{237}Np, and ^{239}Pu. Use the information in the table regarding the inventory of the radionuclides in the waste package, K_d, ad solubility. The data for dose conversion factors and the fraction of dose from drinking water to total dose can be found in Table 12.2. Compare the results of the calculations between using the nominal value and the high value of K_d.

	Inventory in WP (kg)	K_d (nominal) (ml/g)	K_d (high) (ml/g)	Solubility (mol/L)
Tc-99	2.6	0.4	10	1×10^{-5}
I-129	0.6	0–1	1	1×10^{-4}
Np-237	1.5	10–50	50	4×10^{-8}
Pu-239	16	20–500	500	4×10^{-11}

Problem 12.3 Assume a hypothetical situation where a spent fuel package containing 1 Ci (3.7×10^{10} Bq) of ^{99}Tc is buried in a concrete overpack within a saturated aquifer. Twenty years after the burial, the spent fuel package and the concrete overpack fail simultaneously, resulting in 10% of the ^{99}Tc in the package released to the groundwater in short periods of time. The package is 3 m long and the

release occurs uniformly throughout the length of the package. Away from the location of the buried spent fuel package, there is a well at a farm. The distance between the location of the waste package and the well is 5 km. Groundwater flows directly from the location of waste package to the well. A farmer living in the farm draws all of his drinking water from the well.

a) Calculate the time required for the peak of the ^{99}Tc plume to reach the well.
b) Calculate the concentration of ^{99}Tc peak arriving at the well.
c) Assume that the detection limit of ^{99}Tc is 0.1% of your estimated peak concentration, approximately how much time in advance of the peak could the release be detected?

- Given; Hydraulic conductivity of the aquifer is 10^{-2} m/sec
- Hydraulic head of the aquifer:
    - 1000 m at the location of the spent fuel package
    - 990 m at the location of the farm (groundwater flows directly from the location of spent fuel to the farm)
- K_d of ^{99}Tc in the aquifer is 5 ml/g.
- The effective porosity of the aquifer is 0.18.
- The bulk density of soil is 1.6 g/cm^3.
- Longitudinal dispersivity = 10 m
- Transverse dispersivity = 0.1 m
- Half-life of ^{99}Tc is 2.12×10^5 years

Problem 12.4 A site with the following characteristics has been proposed for a hypothetical nuclear waste repository in the United States. The repository is to be located in a granite rock body at a depth of 400 m. The rock is situated in a regional groundwater basin in which the hydraulic gradient at the repository depth is horizontal, with a magnitude of 2×10^{-3}. The effective porosity of the rock is 2%. The hydraulic conductivity is 10^{-6} m/sec. The repository occupies an area of 1.5 km × 1.5 km and extends 20 m in a vertical direction.

In this question, assume the followings:

1. The engineered barriers in the repository are completely ineffective, such that from the time that the repository is closed, all groundwater passing through the repository zone is contaminated with all of the radionuclides in the waste at concentrations corresponding to their solubility limits.
2. The hydrological properties of the repository zone are identical to those of the surrounding rock.
3. Longitudinal and transverse dispersion can both be neglected.
4. Assume that the initial inventory of isotope *i* remaining in the repository at time *t* after closure, $I_i(t)$, is given by:

$$I_i(t) = I_i(0)e^{-\lambda_i(t)} - \frac{v_0 A S_i \rho_w}{\lambda_i}\left(1 - e^{-\lambda_i t}\right)$$

where,

λ_i = decay constant of isotope i (sec^{-1})
v_0=Darcy velocity (m/sec)
A = cross sectional area of repository perpendicular to the direction of groundwater flow (m^2)
S_i = solubility of isotope i
ρ_w= water density

It is assumed that the isotope has no precursor. Use the data given in the following table.

Nuclide	Initial inventory in spent fuel (kg/1000MTHM)	Half-life (year)	Solubility (kg of element/kg water)	K_d (ml/gm)	mass fraction of element(*)
Sr-90	417	29	7×10^{-5}	15	1.0
I-129	177	1.6×10^{7}	0.5	0	1.0
Np-237	442	2.14×10^{6}	5×10^{-10}	40	1.0
Pu-239	5030	2.4×10^{4}	4×10^{-10}	300	0.647

aMass of the specific isotope/total mass of the element

a) Suppose that the repository is designed to contain 70,000 MTHM of spent fuel. If it is assumed that each radionuclide continues to be released into solution until the inventory in the repository has declined to 10^{-6} of its initial value, calculate the release duration for each of the four radionuclides.

b) Cumulative release limits into the accessible environment for individual radionuclides are established as in shown in the following table. Would the release limits set by EPA be met?

Element	EPA Release limits over 10,000 years (Ci/MTHM)
Sr	1000
I	100
Np	100
Pu	100

Problem 12.5 What are the limitations of natural analogues in their intended use in the study of nuclear waste disposal? Based on the observations at the major natural analogue sites, comment on the significance of geochemistry in understanding behaviors of radionuclides in natural environment.

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Chapter 13
Management of Low and Intermediate Level Waste

Abstract Low and intermediate level wastes (LILW) are generated from nuclear power plants, industrial processes, research laboratories, and hospitals. Although low in radioactivity contents in comparison to high level waste, LILW, with widespread presence of its generators in various sectors of society, demands foresight and careful planning and coordination for its safe management and disposal. This chapter describes an overview of LILW management approaches including their characterization, classification, treatment, packaging, and disposal. The issue of mixed hazardous and radioactive waste is also discussed.

Keywords LILW generation · LILW characterization · LILW classification · LILW treatment and processing · LILW disposal

Low level waste (LLW) and low and intermediate level waste (ILW) refer to two types of nuclear waste generated from nuclear power plants (NPPs), industrial processes, research laboratories or hospitals. The International Atomic Energy Agency (IAEA) defines LLW as being above clearance levels (10 μSv/year or 1 mrem/year), with limited amounts of long lived radionuclides. These wastes require isolation and containment for periods of up to a few 100 years and are suitable for disposal in engineered near surface facilities. IAEA goes on to define intermediate level waste (ILW), by its content of long lived radionuclides, which requires a greater degree of containment and isolation than provided by near surface disposal (IAEA 2009a). However, ILW needs no, or only a limited provision, for heat dissipation during storage and disposal.

The nuclear waste classification system in the U.S. uses the term LLW as it includes both LLW and ILW as generally defined by the IAEA (i.e. LLW = LILW in U.S.). In this chapter, the term low and intermediate level waste (LILW) will be used to refer to LLW of the U.S. for consistency (except for the case of using LLW as part of legal designation). Also in this chapter, although the term "radioactive waste" is more commonly used to refer to LLW and LILW, the term "nuclear waste" is still used for consistency with the rest of the book. Whenever the waste discussed is or

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potentially would be classified as an intermediate level waste, the term ILW will be used.

LILW must be carefully managed from generation to final disposal. Sources of LILW have to be carefully controlled to prevent contamination including necessary waste segregation. The waste needs to be treated to reduce volume and for conditioning of materials in preparation for shipment and disposal. Final disposal requires long-term planning and careful engineering practices.

13.1 Brief History of Low and Intermediate Level Waste Management in the U.S.

LILW management in the U.S. (NRC 2007) began with the Manhattan project. During the Manhattan project days (1943–1946), the LILW generated was handled in a variety of ways, including open pit burning, shallow land burial, incineration, dilution/dispersion, and storage. Selection of management techniques at that time were mainly driven by economics and security considerations with environmental concerns at a lower priority. With the beginning of the cold war after WWII, LILW was generated from the production of nuclear weapons and the related R&D activities.

At that time, disposal of LILW was managed by the U.S. Atomic Energy Commission (AEC) and was based on two methods, ocean dumping or shallow land burial. Ocean dumping assumed that dilution in ocean water along with radioactive decay would render the radiation levels of waste to be innocuous. The practice was used between 1946 and 1970 (see also Sect. 10.1) by dropping a 55-gallon steel drums filled with LILW in about 180 m (600 ft) waters. The wastes were mixed in the drum with cement or concrete to ensure sinking and to withstand the pressure in deep-sea. The disposal sites included the Pacific (between 1946 and 1970 disposing 56,261 containers with 14,981 Ci at +34 sites), the Atlantic (between 1951–1967 disposing 34,203 containers with 79,483 Ci at +24 sites), and the Gulf of Mexico (<1959 disposing 79 containers with <25 Ci at 2 sites). The ocean dumping practice phased out due to expense along with the concern over the contamination of ocean (see also the discussion in Sect. 10.1.1). At that time, the reported cost of land disposal ($5.15 per drum) was much cheaper than ocean dumping ($48.75 per drum). Shallow land burial started at AEC facilities (such as Oak Ridge National Laboratory, Los Alamos National Laboratory, the Hanford site, the Savannah River site, and Idaho National Engineering Laboratory) and then continued for the disposal of defense LILW and TRU wastes.

In the 1950s, the majority of LILW generation was limited to federal government related activities. Then, non-defense applications of radioactive materials started in the late 1950s including the operation of commercial nuclear power plants. This development resulted in a sharp increase in the generation of LILW from the commercial sector creating the need for commercial operation of LILW disposal sites. Initially, the commercial disposal of LILW was made at federal sites in Idaho and Oak Ridge. Then, in 1962, AEC authorized the operation of commercial LILW disposal facilities under pressure from states and the private sector. These facilities were authorized to accept only non-defense waste. The nation's first commercial LILW disposal site opened in 1962 in Beatty, Nevada (1962–1992) as a federally licensed facility. This was followed by the Maxey Flats facility in Kentucky (1963–1977), the West Valley facility in New York (1963–1975), the Richland facility in Washington (1965 – still operating), and the Sheffield facility in Illinois (1967–1978). In 1971, the Barnwell, South Carolina site started accepting waste as the sixth LILW disposal facility in the U.S.

Beatty, Nevada was a very dry site and had no major hydrogeological issues at the site. However, mismanagement and poor record keeping of the facility by the operating personnel was a key concern in the early part of the facility operation. Illegal sales of radioactively contaminated tools, lab equipment, and generators were commonly exercised at the site. After several incidents of leaking radioactivity from the shipping trucks, the disposal facility's operating license was suspended from 1976 till 1979. The site reopened in 1979 and operated until its closure in 1992 through an agreement between the Governor of Nevada and the Rocky Mountain Compact Board. The total volume and activity accepted at the facility were 1.3×10^5 m^3 (4.7 million cubic feet) and 1.7×10^{17} Bq (4.7 million Ci).

Maxey Flats, KY and West Valley, NY are two sites that were prematurely closed due to off-site contamination resulting from the release of radioactive materials from facility operations. During the operation of the Maxey Flats site in Kentucky (1963–1977), about 1.3×10^5 m^3 (4.7 million cubic feet) of LLW waste was disposed containing approximately 8.9×10^{16} Bq (2.4 million curies) of radioactivity. It was a site with low permeability soil, under a humid continental climate, having a complex subsurface geology. With lack of careful site characterization, the site was interspersed with perched water tables, with a water table at 84 m from the surface. When the trenches were built, there were hydraulic connections between the trenches. There was also continual infiltration of water through the trench caps which accumulated inside the trenches. The wastes were not properly segregated, resulting in tritium contaminated liquid waste and unstabilized solid waste being co-disposed. In addition, waste packaging and emplacement were not done uniformly. Accumulation of water also resulted in leaching of radionuclides from the waste at the site. When evaporators were employed to address water accumulation, their operation led to the release of radioactive materials offsite. In addition, the subsidence of trenches was noted and attributed to problems in trench construction. The site was closed in 1977 by the state, requiring on-going custodial care.

The West Valley site in New York operated from 1963 through 1975. It was a site in a cool moist climate with highly compacted glacial till with very low permeability. There were perched water table zones dispersed throughout the site with the water table located at approximately 30 m from the surface. About 6.8×10^4 m^3 (2.4 million cubic feet) of LLW was disposed at the site with approximately 2.7×10^{16} Bq (0.74 million curies) of radioactivity inventory. The wastes were packaged in steel drums along with cardboard and wooden boxes with minimal requirements for waste form. No segregation of waste was exercised and backfilling before trench closure was inadequate. While the trenches were being filled, water accumulated due to precipitation. Even after the closure of trenches, water infiltration continued into the trenches due to poor trench cover design and inadequate drainage. Due to the very impermeable nature of the soil, the water continued to accumulate inside the trench leading to a phenomenon called the bathtub effect. This resulted in an eventual surface discharge of contaminated trench water. Eventually, this overflow of trench water led to site closure in 1975.

The Sheffield, Illinois, a silt-clay-sand sediment site opened since 1967 and operated under the shared licensing responsibility between the State of Illinois and AEC. In 1975, the operator requested license renewal to US NRC to increase the capacity and lifetime of the facility. During the licensing review, the need for more site-specific geology and hydrogeology study was raised along with the discovery of tritium leakage and migration. These developments led to closure of one of the operating trenches. With the capacity expansion application pending and no space in other trenches, disposal operation ceased in 1978 after accepting 8.7×10^4 m^3 (3.1 million cubic feet) of LILW (containing 4.1×10^{16} Bq (1.1 million Ci)) through operation until 1977 at the site (INEL 1994).

As noted, the operations of the first generation commercial LILW sites had various problems of radioactivity release exacerbated by a lack of proper regulatory guidance and a poor safety culture. Better regulatory oversight was needed to improve the safety practices at these sites. In fact, at the time of beginning of commercial LILW disposal, there were no licensing criteria specific to the disposal of LILW. The general AEC regulations of 10 CFR 20.302 (a), 10 CFR 20.302 (b), and 10 CFR 20.304 were the only applicable licensing rules.

With the abolishment of AEC in 1974, the newly created NRC regulated LILW by using a collection of generic regulations specified in 10CFR Part 20, 10CFR part 30, 10 CFR Part 40, and 10 CFR Part 70. The approach still lacked comprehensive regulatory requirements though. Part 20, 30, 40, 70 of 10 CFR represents "Standards for Protection Against Radiation", "Rules of General Applicability to Domestic Licensing of Byproduct Material", "Domestic Licensing of Source Material", and "Domestic Licensing of Special Nuclear Material, respectively." Effort to develop a comprehensive regulatory framework for commercial LILW disposal began in 1978 under the direction of the Congress. LLW classification scheme was developed at that time along with the methods for the assessment of impacts of different LILW disposal concepts. These along with the proposed performance objectives and technical criteria became a draft regulation designated as 10 CFR Part 61. The final 10 CFR Part 61 rule was issued in December 1982 and became effective as of December 1983. Since then, 10 CFR Part 61 has provided comprehensive regulatory oversight over the management and disposal of LILW in the U.S.

Such developments were happening in parallel to support and improve the operation of existing LILW facilities in the U.S such as the facilities in Richland, Washington and Barnwell, South Carolina. The Barnwell, SC facility opened in 1971 with a 10,000,000 m^3 capacity and is still in operation. The site is expected to accept LLW until 2038. Currently, the site is operated by EnergySolutions and receives Class A, B, and C LLW from the Atlantic compact states (Connecticut, New Jersey, and South Carolina). The Richland, WA facility, currently operated by U.S. Ecology, has a 125,000 m^3 capacity and accepts Class A, B, and C LLW from the Northwest (Washington, Oregon, Idaho, Montana, Utah, Wyoming, Hawaii, and Arkansas) and Rocky Mountain (Colorado, New Mexico, and Nevada) compact states.

A key milestone in the effort to develop commercial LILW facilities in the U.S. was the enactment of LLW Policy Act in 1980. The Act begins with s statement that "Each state is responsible for providing for the availability of capacity either within or outside the state for disposal of LILW generated within its borders (excluding defense or other federal wastes)." The Act allowed the states to join and form regional compacts and authorized exclusion of LILW generated outside a compact after January 1, 1986 from the use of the regional disposal facilities. Each compact was to designate one active site as the compact's LILW disposal facility. After experiencing many difficulties, challenges, and confusion, an Amendment of the Act was announced in 1985. The Amendment's Act allowed existing LILW disposal facilities to remain available to waste generators until 1993 and provided incentives for new facility development. The earlier discussion on available LILW facilities identified which groups of states (compacts) can dispose of their waste in specific disposal facilities, and which disposal facilities are available to out of compact states. The Amendment also stated that DOE is responsible for disposal of commercial GTCC LLW which is to be disposed of at a HLW geologic repository.

Despite the efforts made by individual states, and the compacts (groups of states), none of the states or compacts successfully developed new LILW disposal facilities under the LLWPAA framework. In 1989 the U.S. government's Office of Technology Assessment (OTA) reviewed the LLWPAA process and found that some states enacted bans to legally restrict shallow land disposal despite the Federal regulations finding this disposal method technically sound. Other issues cited in the report were the rising costs of LILW disposal (at the time of the study, it had tripled in 20 years) and the management of mixed wastes (combined radioactive and hazardous waste).

Since the LLWPA, only two new disposal facilities have been licensed to accept LLW and both were accomplished outside of the LLWPAA framework. The first was the Clive Utah site which was originally a DOE uranium mill tailings disposal site, then in 1987 it was licensed, by the Utah Department of Environmental Quality, to accept NORM waste (uranium mill tailings). In 1991 the State of Utah approved a license for the site to dispose of class A LLW. The facility is operated by the Envirocare Company as a private facility. The site can accept waste from all 50 states (except those in the Rocky Mountain and Northeast Interstate Compact). Currently, the site disposes of 98% of the U.S. Class A waste volume, but does not accept sealed sources or biological tissue waste. The site is also licensed to dispose of mixed waste.

The second new LILW disposal facility is the Andrews, Texas site, opened in 2012 by Waste Control Specialists (currently owned by Lehman & Company), and accepts Class A, B, and C waste from the Texas compact (Texas and Vermont) generators. The Andrews site can also accept waste from the 34 U.S. states that do not have access to a disposal facility, up to 30% of the site's licensed capacity. These out-of-compact generators, must submit an import petition to the Texas Compact Commission and receive waste acceptance approval prior to shipping. Both facilities are currently under operation along with Richland, Washington and Barnwell, South Carolina facilities.

Currently, 10 CFR Part 61 addresses 84 regulatory issues related to land disposal of LILW including waste classification, waste characteristics, disposal facility licensing requirements, disposal concepts and performance objectives (protection of the public, stability of disposal, etc.), environmental monitoring, and financial assurances. The performance objectives of a disposal facility are to be met during the 1000 years of post-closure period. There is now also a new 10CFR61 rule pending (NUREG-2175) in the U.S. reflecting the need for supplement existing rules by providing detailed guidance on the inadvertent intruder analysis and using a graded level of effort for disposal facility performance assessment covering beyond 1000 years (up to 10,000 years for protective assurance and beyond 10,000 years for long-lived waste). Additionally, the U.S.NRC periodically publishes NUREGs (U.S. Nuclear Regulatory Commission Regulations) which include reports or brochures on regulatory decisions, research results, results of incident investigations, and technical and administrative information, and BTPs (Branch Technical Positions) as guidance documents for use by generators, processors and state regulators for safe and effective low level waste management.

13.2 Generation of Low and Intermediate Level Waste

Considering the world wide generation of nuclear waste from nuclear power plants, Table 13.1 provides some perspective on the quantities of waste volumes generated and the radioactive content of those wastes.

Sources of LILW include the nuclear fuel cycle, such as nuclear power plants, uranium reprocessing plants, fuel fabrication plants, and uranium enrichment facilities. Non-fuel cycle industries also produce LILW and are referred to as institutional waste generators. They include industry (R&D companies, manufacturers, nondestructive-testing operations, uranium mines, radiopharmaceutical manufacturers, etc.), government (i.e., local, state, and federal entities), universities, and hospitals. Between 2009 and 2018, the U.S. reported 732 licensees under

Table 13.1 Typical generation ratios, IAEA summary International nuclear power plants

Waste type	Volume (%)	Radioactive content (%)
High-level waste	0.06	9%
Intermediate-level waste	1.63	3
Low-level waste	69	2

Source: IAEA (2018)

Table 13.2 Average annual generation from 2009 to 2018

	Volume		Activity	
Generator type	meters^3 (ft^3)	%	Bq (Ci)	%
Academic	5280 (186,452)	61%	$3.922 \times 10^{+12}$ (106)	80%
Government	127,966 (4,519,094)		$3.811 \times 10^{+13}$ (1030)	
Industry	269,793 (9,527,653)		$2.072 \times 10^{+12}$ (55,987)	
Medical	95 (3336)		$2.738 \times 10^{+12}$ (74)	
Undefined (likely non-nuclear fuel cycle)	21,164 (747,411)		$2.640 \times 10^{+13}$ (713,573)	
Utility	268,814 (9,493,063)	39%	$7.067 \times 10^{+12}$ (191,002)	20%

Data retrieved from DOE (2018)

government regulation generating LLW (DOE 2018). The U.S. Department of Energy's radioactive waste Manifest Information Management System (MIMS) website data from 2009 to 2018 reports the volumes and activity by generator group as presented in Table 13.2.

13.2.1 Low and Intermediate Level Waste from Nuclear Power Plants

The vast majority of radioactivity in the LILW from NPPs comes from the waste processing systems that remove the radioactive contaminants from water and air effluents. Such processing is performed for the reactor coolants and any radioactive material streams from the reactor coolant system or through the connected auxiliary and support systems.

A small fraction ($\leq 0.01\%$) of nuclear fuels may exhibit cladding defects during normal NPP operation, leading to the release of fission products and actinides (IAEA 2009b). The fuel cladding and reactor structural materials under neutron irradiation go through activation and the resulting activated materials, through corrosion processes, become a constituent of the primary coolant. Also, stable corrosion products can be activated when transported through the core where they also become a constituent of the primary coolant. The primary coolant itself goes through neutron activation and becomes radioactive.

The radiation level elevates as these radioactive materials build up in the reactor coolant system. To control reactor coolant radioactivity concentrations and the build-up of radiation fields inside the plant, a small fraction of reactor coolant is continually diverted and purified. The media (primarily ion exchange resins and filter media) used for purification of the reactor coolant after the service period become LILW. These spent purification media are generally much higher in radioactivity concentrations than typical dry active waste.

The radioactive materials also move into various auxiliary and support systems connected to the coolant system. Circulation of water through these various systems leads to leakage of radioactive materials from system components such as valves, pumps, sampling locations, etc. These leaks ultimately enter floor drains and become part of a liquid stream requiring processing and result in liquid radioactive waste. Other sources of liquid radioactive waste in NPPs can include laundry wastes, laboratory drains, or floor drains. Special systems are in place to collect and process these radioactively contaminated coolant or liquid leakages for decontamination. These operations also result in the generation of secondary wastes in the form of LILW. Other solid materials (floor coverings, mop heads, etc.) which come into contact with radioactivity from these leaks also become LILW.

Gaseous wastes such as gases coming from plant systems, building ventilation air, or in the case of BWRs, gases ejected from the turbine condenser as part of system design are also treated before release to the atmosphere resulting in the production of LILW (typically HEPA (high efficiency particulate air) filters and activated carbon). Release of radioactive materials from the effluents, into the environment is strictly controlled to keep the radiation dose to members of the public in unrestricted areas “as low as reasonably achievable (ALARA)”.

These media or process waste or the contaminated products are LILW and can be generally grouped in two categories: “Wet waste” and “Dry waste”. Wet wastes include ion-exchange resins, filters, filter sludges, and evaporator concentrates (as explained in Sect. 13.3). Dry radioactive wastes are mostly called dry active waste (DAW). It mainly consists of contaminated trash, filters from ventilation systems, and scrap metals or components. Dry wastes also include activated hardware from the reactor internals.

Ion exchange resins are typically synthetic organic polymers used for cleanup of radioactive liquid streams. The resins include small porous bead resins or powdered resins, the latter is normally deposited on a filter as an overcoat. Bead resins are typically used for the deep bed demineralizers. Filters are often used in the form of cartridge filters or HEPA filters for the removal of particulate matter. Cartridge filters are assemblies of filters to provide mechanical filtration and are made of pleated cotton, fiberglass, and epoxy-impregnated paper and can be overcoated with powdered resins. HEPA filters are typically made of fiberglass. Filter sludges are the leftover waste materials after filtration treatment of radioactive liquid streams. Evaporator concentrates, sometimes called evaporator bottoms, are the residues after the processing of liquid solutions through evaporation, the resulting concentrates contains high levels of dissolved solids. There are other miscellaneous wastes such as sediments collected in the bottom of sumps, vessels, and tanks in nuclear power plants. There are also various used oil and miscellaneous liquids generated as waste from changing the lubrication oil for pumps and motors.

DAW includes a wide range of radioactively contaminated materials such as plastic bags and sheeting, paper coveralls, paper towels, plastic booties, failed parts and components, contaminated clothing items, etc. DAW often carries very low levels of contamination. The IAEA defines very low level waste (VLLW) as waste that does not necessarily meet the criteria of clearance, but does not need a high level of isolation and containment. It is therefore, suitable for disposal in a near surface hazardous waste disposal facility. According to the IAEA, more than 98% of DAW volume is classified as very low or low level waste, with a majority of the remaining volume being intermediate level waste. This is shown in Table 13.1 (see also Table 6.1 for comparison with the case of the U.S.). In terms of total radioactivity, approximately 98% of the radioactivity is associated with intermediate and high level waste (IAEA 2018).

Key characteristics of these wet and dry wastes in terms of sources and typical radiation dose rates along with typical treatment methods and the containers used for disposal are summarized in Tables 13.3 and 13.4 (EPRI 1996). As seen in the table, the radiation dose rates from the wet wastes vary widely and demand careful planning in the handling of the materials. The dose rates from the dry wastes is low in general with the exception of activated hardware which poses very high levels of dose requiring extreme care in handling. Comparison of the dose rates are also shown in Figs. 13.1 and 13.2 for wet and dry LILW, respectively. Various treatment or processing methods are applied to the wet and dry waste. These methods are discussed in Sect. 13.5.

13.2.2 Low and Intermediate Level Waste from Other Nuclear Fuel Cycle Facilities

LILW are produced from the treatment of gaseous and liquid effluents from the operation of nuclear fuel cycle facilities (such as for enrichment, conversion, fuel fabrication, and reprocessing) along with radioactively contaminated solid wastes as described in Sect. 6.4. Maintenance and servicing operations in these facilities also produce radioactively contaminated waste. In the case of the front-end fuel cycle, the contamination is mainly from uranium and is LILW. In the case of reprocessing, a variety of fission products and actinides are part of the contamination, generating a combination of both LLW and ILW. The types of materials that comprise the radioactive waste from these facilities are not much different from the LILW from nuclear power plant operation.

13.2.3 Low and Intermediate Level Waste from Industrial and Institutional Activities

LILWs are generated from industrial organizations that supply or use radioactive materials and institutions such as universities, hospitals, clinics, biomedical research

Table 13.3 Characteristics of wet low and intermediate level waste (LILW) from nuclear power plants (EPRI 1996)

	Bead resins	Powdered resins	Cartridge filters (mostly from PWR)	Evaporator concentrates	Miscellaneous sediments	Oils/liquids
Sources	Deep bed demineralizers	Filter demineralizers	In-plant filter system/portable underwater vacuum systems	Evaporators	Sumps, tanks	Sumps, equipment drains, lube oil leakage
Purpose	Ion exchange of dissolved contaminants	Ion exchange of dissolved contaminants	Filtration of suspended solids	Concentrate dissolved liquids	–	–
Side benefit	Some filtration of suspended solids	Filtration of suspended solids	–	–	–	–
Waste processing method	Dewatering, possible solidification, blending	Dewatering, solidification, blending	Dewatering, encapsulation, absorbed, blending	Solidification, drying	Dewatering, drying, solidification	Solidification, incineration
Containers	Concrete container, HIC, steel liner, steel drum	Concrete container, HIC, steel liner, steel drum	Concrete container, HIC, steel liner, steel drum	Drums	HIC, steel liner, steel drums	Steel drums
Typical dose rates	0.1 ~ 3000 mSv/h (10 ~ 300,000 mrem/h)	0.1 ~ 3000 mSv/h (10 ~ 300,000 mrem/h)	1.0 ~ 800 mSv/h (100 ~ 80,000 mrem/h)	2.0 ~ 20 mSv/h (200 ~ 2000 mrem/h)	0.1 ~ 20 mSv/h (10 ~ 2000 mrem/h)	< 0.05 mSv/h (< 5 mrem/h)

Table 13.4 Characteristics of dry low and intermediate level waste (LILW) from nuclear power plants (EPRI 1996)

	Incinerable trash[a]	Non-incinerable trash[a]	Contaminated metals	Other non-incinerable materials	Activated hardware
Common materials	Polyethylene, polyurethane, cloth, paper, wood, etc.	Polyvinyl chloride, nylon reinforced plastic, rubber, filters, glass, etc.	Tools, pipes, valves, components, scaffolding	Spent grit blast waste, rubble, large quantities of contaminated dirt	Reactor components, fuel assembly hardware, control rod blades, etc.
Source	Various	Various	Various	Various	Reactor core components
Waste processing method	No processing, incineration, compaction, supercompaction	No processing Compaction, supercompaction	Decontamination, metal-melt, supercompaction	Supercompaction	Metal melt, segmentation, supercompaction
Containers	Drums, steel boxes, sea-land containers	Drums, steel boxes, sea-land containers	Drums, steel boxes, sea-land containers	Drums, steel boxes, sea-land containers	HIC, spent fuel trash liner
Typical dose rates	Generally <0.5 mSv/h (generally <50 mrem/h)	Generally <0.5 mSv/h (generally <50 mrem/h)	Generally <0.5 mSv/h (generally <50 mrem/h)	Generally <0.5 mSv/h (generally <5 mrem/h)	2.0 ~ 400 Sv/h (200 ~ 40,000 rem/h)

[a] Sorting wastes into the incinerable and non-incinerable categories, and their sub categories is a key component of effective DAW management

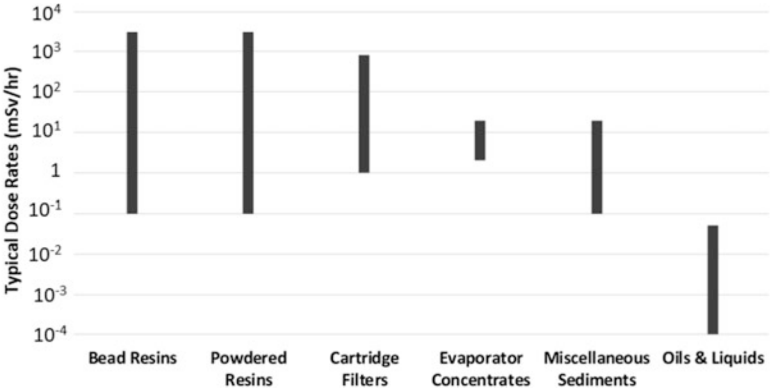


Fig. 13.1 Typical dose rate distribution from wet low and intermediate level waste (LILW)

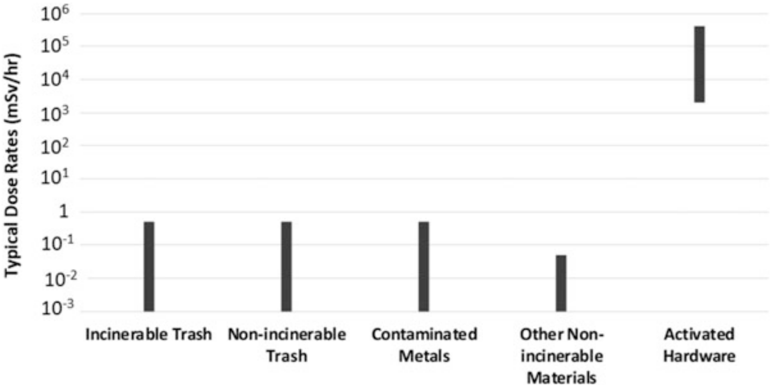


Fig. 13.2 Typical dose rate distribution from dry low and intermediate level waste (LILW)

laboratories, and other industrial entities where radioactive materials are used for research, clinical or manufacturing purposes.

Industrial uses of radiation sources or radioactive materials include, industrial radiography (using x-rays, or gamma rays from ^{192}Ir or ^{60}Co) for inspection of metal parts for flaws, using sealed sources (e.g., ^{137}Cs) for oil and gas exploration through gamma ray attenuation profiling of field materials. They also include gauging for aluminum or paper manufacturing (using beta particles from ^{85}Kr for continuous testing of the thickness of paper or aluminum), using gamma rays for sterilization, etc. When sealed sources from these activities, become disused sources (no longer used and no intention of using), they are accounted for and controlled as either LLW or ILW depending on radioactive contents (IAEA 2014).

Radiopharmaceuticals are used extensively in diagnostic procedures. A number of body organs can be visualized by introducing a radiopharmaceutical into a patient and imaging the resultant distributions of a tracer radionuclide using a gamma camera. The most widely used tracer radionuclide for medical diagnosis is technetium-99 m ($t_{1/2}$ = 6 h), followed by ^{125}I, ^{51}Cr, ^{75}Se, and ^{85}Sr. ^{99m}Tc is prepared by being extracted from a longer-lived isotope ^{99}Mo ($t_{1/2}$ = 66 h) for each procedures.

For the purpose of understanding the mechanism of reactions and biological processes in humans and animals, radionuclides such as ^{14}C ($t_{1/2}$ = 5730 years), ^{3}H ($t_{1/2}$ = 12.3 year), ^{32}P ($t_{1/2}$ = 14 days), ^{35}S ($t_{1/2}$ = 87 days), ^{125}I ($t_{1/2}$ = 59 days) are widely used for labelling/tracing compounds in the pharmaceutical and organic chemical industries. Radiation sources (x-ray machines or sealed sources of gamma emitting nuclides, such as ^{60}Co or ^{137}Cs) are also used in radiation therapy, to kill cancer cells.

LILW resulting from these procedures include sealed sources, liquid scintillation vials, absorbed aqueous and organic liquids, animal carcasses, other biological wastes, laundry waste, and other radiologically contaminated trash. Liquid scintillation vials refer to the left-over glass vials containing organic solvents (e.g., toluene or xylene) and liquid scintillator (e.g., zinc sulfide) used for the detection of radiation typically from beta (e.g., ^{3}H, ^{14}C) or alpha emitters through conversion of radiation energy to light energy by using the scintillation process.

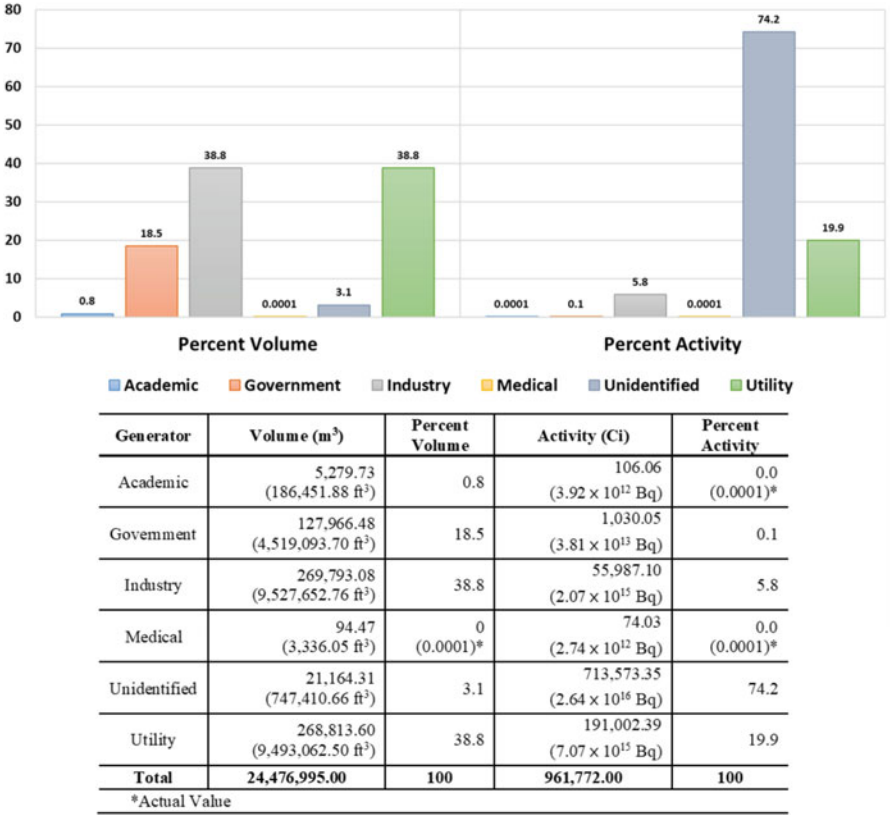
Generation of LILW from various commercial sectors in the U.S. in terms of volume and activity are shown in Fig. 13.3. The figures are based on LILW generation between 2009 and 2018 from academia, government, industry (non NPP industry), medical institutions, utility, and unidentified generators.

Note that some sealed sources do not qualify as LLW because of their remaining long term hazard. Also, sealed sources with high decay generation may not qualify as ILW if the LLW classification considers decay heat contents in the waste (this is the case in the Republic of Korea and IAEA's 2006 classification scheme (IAEA 2006a) included the consideration of decay heat generation).

13.3 Characterization of Low and Intermediate Level Waste

Efforts in the management of LILW must be commensurate with the level of hazard presented by the material. In other words, once the waste is generated, the requirements for the treatment, packaging, and disposal could vary depending upon the hazardousness of the waste. To understand the level of potential hazard associated with each type of generated waste, characterization of the waste is necessary.

Waste characterization is used to identify the radioactive composition (i.e., the nature and quantities of radionuclides and their concentration) of waste. Waste characterization is necessary not only to provide the necessary protective measures during handling and transportation but also to accurately assess the radionuclide inventory in a waste disposal facility. Thus, LILW characterization allows the determination of the total inventory of radionuclides at a disposal facility, as the sum of each radionuclide shipped to the facility which also allows for the projection



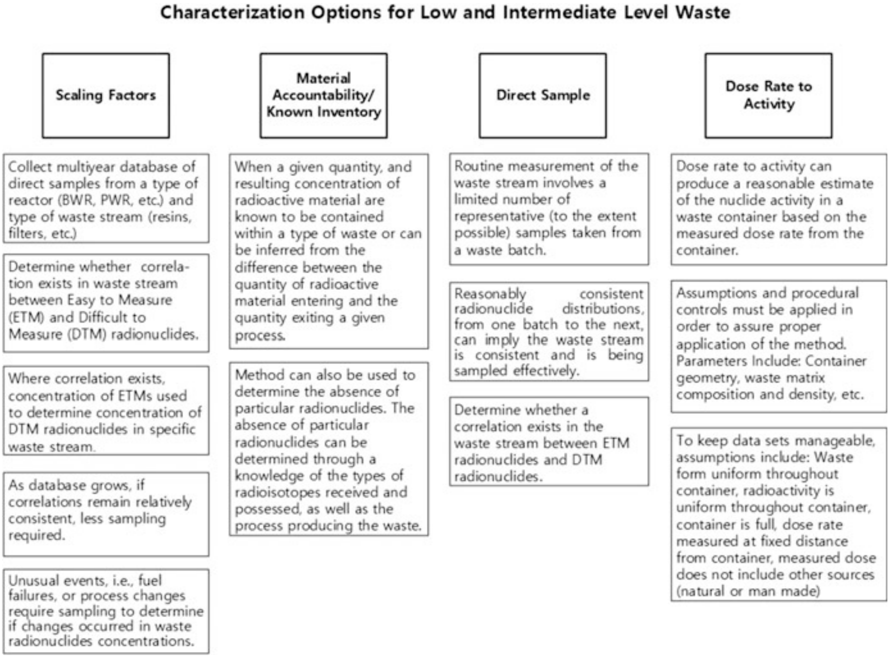
Generator	Volume (m^3)	Percent Volume	Activity (Ci)	Percent Activity
Academic	5,279.73 (186,451.88 ft^3)	0.8	106.06 (3.92 x 10^{12} Bq)	0.0 (0.0001)*
Government	127,966.48 (4,519,093.70 ft^3)	18.5	1,030.05 (3.81 x 10^{13} Bq)	0.1
Industry	269,793.08 (9,527,652.76 ft^3)	38.8	55,987.10 (2.07 x 10^{15} Bq)	5.8
Medical	94.47 (3,336.05 ft^3)	0 (0.0001)*	74.03 (2.74 x 10^{12} Bq)	0.0 (0.0001)*
Unidentified	21,164.31 (747,410.66 ft^3)	3.1	713,573.35 (2.64 x 10^{16} Bq)	74.2
Utility	268,813.60 (9,493,062.50 ft^3)	38.8	191,002.39 (7.07 x 10^{15} Bq)	19.9
Total	**24,476,995.00**	**100**	**961,772.00**	**100**

*Actual Value

Fig. 13.3 U.S. Percent LILW volume and curie content generated by each of the six categories of commercial generators (2009–2018)

of dose to the public from LILW disposal facilities. Along with identifying the nature and quantities of radionuclides contained in the waste, LILW characterization seeks information on the chemical and physical forms of waste for risk characterization (IAEA 2007). Thus, knowledge of waste generating processes and the effect of plant design and operational parameters are useful for the characterization of LILW generated at nuclear power plants.

LILW characterization is dictated by government regulations (NRC 1983). In the U.S. regulatory requirements come from 10CFR Part 61 and the wastes are classified, depending on nuclide concentrations, into class A; class B; class C and greater than class C (GTCC). The requirements include waste characteristics that intended to facilitate handling at the disposal site and provide protection of personnel at the disposal site. Examples of waste characteristics to be examined under the regulations are waste package, packaging for liquid and solid waste, and the waste content not being capable of exploding-detonating or contain toxic gases, etc. Sampling activities and radiation measurements are performed as part of the characterization as



Characterization Options for Low and Intermediate Level Waste

Scaling Factors

Collect multiyear database of direct samples from a type of reactor (BWR, PWR, etc.) and type of waste stream (resins, filters, etc.)

Determine whether correlation exists in waste stream between Easy to Measure (ETM) and Difficult to Measure (DTM) radionuclides.

Where correlation exists, concentration of ETMs used to determine concentration of DTM radionuclides in specific waste stream.

As database grows, if correlations remain relatively consistent, less sampling required.

Unusual events, i.e., fuel failures, or process changes require sampling to determine if changes occurred in waste radionuclides concentrations.

Material Accountability/ Known Inventory

When a given quantity, and resulting concentration of radioactive material are known to be contained within a type of waste or can be inferred from the difference between the quantity of radioactive material entering and the quantity exiting a given process.

Method can also be used to determine the absence of particular radionuclides. The absence of particular radionuclides can be determined through a knowledge of the types of radioisotopes received and possessed, as well as the process producing the waste.

Direct Sample

Routine measurement of the waste stream involves a limited number of representative (to the extent possible) samples taken from a waste batch.

Reasonably consistent radionuclide distributions, from one batch to the next, can imply the waste stream is consistent and is being sampled effectively.

Determine whether a correlation exists in the waste stream between ETM radionuclides and DTM radionuclides.

Dose Rate to Activity

Dose rate to activity can produce a reasonable estimate of the nuclide activity in a waste container based on the measured dose rate from the container.

Assumptions and procedural controls must be applied in order to assure proper application of the method. Parameters Include: Container geometry, waste matrix composition and density, etc.

To keep data sets manageable, assumptions include: Waste form uniform throughout container, radioactivity is uniform throughout container, container is full, dose rate measured at fixed distance from container, measured dose does not include other sources (natural or man made)

Fig. 13.4 Characterization options for low and intermediate level waste

explained below. Results of LILW characterization are summarized in a document for shipping called the waste manifest, and contains information on what is being transported. In general, LILW characterization in the U.S. is based on using one of three methods listed below (there is a fourth method identified in the guidance, “classification by source” which is not typically use at NPPs).

- Material accountability/Known inventory; in this case the inputs and outputs of a process are known (for example, sealed sources).
- Direct sample; where the actual sample results are used to quantify the activity in the waste based on the mass of the waste and the specific activity (Bq/kg or nanocuries/gram) of each radionuclide in the sample.
- Dose rate to activity; Contents of gamma emitting radionuclides are determined based on gamma scanning and the contents of non-gamma emitters are estimated based on so-called scaling factors. Use of scaling factors for this purpose is described in Sect. 13.2.3. The majority (>95%) of LILW is characterized using the dose rate to activity method.

The processes utilized to characterize LILW are captured in Fig. 13.4. The figure presents each process in general terms. Individual programs will vary.

13.3.1 Sampling for Waste Characterization

As noted in Fig. 13.3, LILW characterization through direct measurement or scaling factor development and maintenance is performed by extracting samples from LILW before performing any solidification or encapsulation treatment. Sampling typically uses crude techniques such as “dipping” (extracting a sample by using scoop-like device), grabbing, dry-product sampling, automatic sampling by using continuous transfer, or manual cutting, etc. As samples are a very small fraction of the total bulk waste (typically 1 g or ml for each sample), determination of the composition of LILW through sampling requires representativeness of the samples. Thus multiple samples may need to be collected. Representative sampling often requires significant sampling time (e.g., by mixing the waste or taking multiple samples) and could result in considerable radiation exposure to the plant personnel. The number of samples to be collected depends on homogeneity of the distribution of radionuclides in the waste. Samples need to be chemically preserved if volatile losses of radionuclides are expected (e.g., ^{14}C and ^{129}I).

13.3.2 Analysis of the Samples

The collected samples are analyzed to determine their contents. Analyzing the gamma emitting radionuclides contained in the waste can be readily performed. However, as many of the radionuclides in LILW do not emit gamma radiation, using complicated time-consuming radiochemical analyses is often needed on the samples to identify long lived radionuclides of importance to waste disposal. In the U.S., the list of nuclides to be analyzed for LLW classification under 10CFR Part 61 includes ^{3}H (β), ^{14}C (β), ^{55}Fe (*x*-ray), ^{59}Ni (*x*-ray), ^{63}Ni (β), ^{90}Sr (β), ^{94}Nb (β/γ), ^{99}Tc (β), ^{129}I (β/γ), ^{237}Np (α/β/γ), and the isotopes of Pu, Am, and Cm (α plus β or γ). These radionuclides emit alpha, beta, or low energy gamma rays and belong to the difficult-to-measure (DTM) (also called hard-to-measure (HTM)) nuclides group. For these HTM nuclides, a detailed analytical chemistry work is needed. The results of the analyses of wet waste samples are given in activity per unit mass or volume. However, for DAW samples (using swipes and surrogate filters), when the units of activity per sample are provided, only the ratios of the nuclides to the total are used in the dose rate to activity models.

13.3.3 Use of Scaling Factors

An alternative to sample analysis for quantifying the presence of the HTM radionuclides is to use scaling factors. A scaling factor is a multiplier to determine the concentration of HTM radionuclides in a waste by using the concentration value of "easy-to-measure" (ETM) radionuclides. Once developed for a specific waste stream (i.e., type), scaling factors can be used to determine the inventory of HTM radionuclides as long as the characteristics of the waste stream remain the same. ETM radionuclides are the ones that emit strong gamma radiation such as ^{60}Co or ^{137}Cs.

In the case of developing scaling factors for fission products and TRUs, the key ETM radionuclide used is ^{137}Cs whose release depends on fuel defects. For corrosion products, ^{60}Co is used as the key scaling nuclide. Sometimes when ^{144}Ce is found in the sample, it is also used as a scaling nuclide as ^{144}Ce is known to correlate very well to TRUs, such as ^{239}Pu. Such use of ^{144}Ce should be limited to the current core fuel cycle, though, due to its short half-life (284.4 days). Recently, with the very low rate of fuel failure/defects, using ^{137}Cs has become difficult due to its very low concentration in the coolant. In this case using ^{60}Co is exercised as the key scaling nuclide for all HTM nuclides.

Extensive efforts have also been undertaken in a number of countries, such as U. S., Sweden, France and Spain to develop industry-wide scaling factors for specific types of waste streams (IAEA 2009b). These efforts can be driven by the nuclear industry or specified by the government or disposal facility. In Sweden, the term correlation factor is used instead of scaling factor. As defined, the correlation factor approach requires a correlation between the activity content of a HTM nuclide and a key scaling nuclide.

The results of Sweden's efforts with comprehensive list of radionuclides are shown in Table 13.5. Although this is a nation-specific results, comparison with other countries effort show no significant differences (SKB 2007).

The scaling factors for TRUs (^{238}Pu, ^{240}Pu, ^{241}Am, ^{242}Cm, ^{244}Cm) using ^{239}Pu as the scaling nuclide are also shown in Table 13.6 for the U.S. PWRs and BWRs. For TRUs (i.e., ^{238}Pu, ^{239}Pu, ^{240}Pu, ^{241}Am, ^{242}Cm, ^{244}Cm), the scaling factors were found to be nearly constant with low dispersion in all industry data regardless of the waste streams.

While the use of scaling factors for ^{99}Tc and ^{129}I has continued over the years, developing scaling factors for these two radionuclides has been particularly challenging as their concentrations are often below the lower limit of detection (LLD). It has been reported that using scaling factors derived from LLD values would result in overestimation of the inventory of ^{99}Tc and ^{129}I at 100–1000 times higher than actual (EPRI 2015). As ^{99}Tc and ^{129}I are dominant contributors to the projected dose from LILW disposal, industry-wide efforts have been made to improve the determination of the scaling factors of ^{99}Tc and ^{129}I in LILW (NRC 2000). These efforts are based on using mass spectrometry analysis for accurate determination of ^{99}Tc and ^{129}I contents in the samples. From these efforts, more accurate scaling factors were developed (NRC 2000; EPRI 2015) as summarized in Table 13.7.

As indicated in the table, ^{99}Tc is scaled to the activated corrosion product ^{60}Co when ^{137}Cs/^{60}Co < 10 with the scaling factor of 1.3×10^{-6}. When ^{137}Cs/^{60}Co > 10, ^{99}Tc is scaled to the fission product ^{137}Cs using the scaling factor of 2.5×10^{-8}. The latest research shows that ^{129}I should be scaled to ^{137}Cs when present using the scaling factor of 2.0×10^{-7}. However, if ^{137}Cs is not present in the waste due to

Table 13.5 Correlation factors for LILW from Swedish BWRs and PWRs

Scaled (HTM) Nuclide	Correlation Factor	BWR	PWR	Scaled (HTM) Nuclide	Correlation Factor	BWR	PWR
H-3	Co-60	1×10^{-4}	1×10^{-3}	Ag-108	Co-60	6×10^{-5}	6×10^{-5}
Be-10	Co-60	6×10^{-10}	6×10^{-10}	Cd-113 m	Cs-137	6×10^{-4}	6×10^{-4}
C-14	**Co-60**	$\mathbf{3 \times 10^{-3}}$	$\mathbf{8 \times 10^{-2}}$	Sb-125	Co-60	1×10^{-1}	1×10^{-1}
Cl-36	**Co-60**	$\mathbf{6 \times 10^{-7}}$	$\mathbf{6 \times 10^{-7}}$	Sn-126	Cs-137	5×10^{-7}	5×10^{-7}
Fe-55	Co-60	1	1	**I-129**	**Cs-137**	$\mathbf{3 \times 10^{-6}}$	$\mathbf{3 \times 10^{-6}}$
Ni-59	**Co-60**	$\mathbf{1 \times 10^{-3}}$	$\mathbf{3 \times 10^{-2}}$	Ba-133	Co-60	1×10^{-5}	1×10^{-5}
Ni-63	**Co-60**	$\mathbf{8 \times 10^{-2}}$	**4**	Cs-134	Cs-137	1	1
Se-79	Cs-137	4×10^{-6}	4×10^{-6}	**Cs-135**	**Cs-137**	$\mathbf{1 \times 10^{-5}}$	$\mathbf{1 \times 10^{-5}}$
Mo-93	**Co-60**	$\mathbf{1 \times 10^{-6}}$	$\mathbf{1 \times 10^{-6}}$	Pm-147	Cs-137	9×10^{-1}	9×10^{-1}
Zr-93	Co-60	1×10^{-6}	1×10^{-6}	Sm-151	Cs-137	3×10^{-3}	3×10^{-3}
Nb-93	Co-60	1×10^{-3}	1×10^{-3}	Eu-152	Cs-137	7×10^{-5}	7×10^{-5}
Nb-94	Co-60	1×10^{-5}	1×10^{-5}	Eu-154	Cs-137	1×10^{-1}	1×10^{-1}
Tc-99	**Cs-137**	$\mathbf{9 \times 10^{-4}}$	$\mathbf{9 \times 10^{-4}}$	Eu-155	Cs-137	7×10^{-2}	7×10^{-2}
Pd-107	Cs-137	1×10^{-6}	1×10^{-6}	Ho-166 m	Co-60	4×10^{-6}	4×10^{-6}

Aimed to represent waste 1 year after discharge from the nuclear facility. Bold nuclides are updated as of 2007; Nonbold nuclides are from 1998

Table 13.6 Scaling factor for transuranics with ^{239}Pu for U.S. PWR and BWR (using all data without outlier removal)

	PWR	BWR
^{239}Pu/^{144}Ce	5.8×10^{-3}	9.6×10^{-3}
^{238}Pu/^{239}Pu	1.0	1.6
^{241}Pu/^{239}Pu*	1.08×10^{2}	1.06×10^{2}
^{241}Am/^{239}Pu[a]	5.5×10^{-1}	9.6×10^{-1}
^{242}Cm/^{239}Pu	1.5	1.1
^{244}Cm/^{239}Pu	5.4×10^{-1}	7.9×10^{-1}
Alpha>5 year/^{239}Pu	3.5	4.8

Source: SKB (2007)

[a]Highly time dependent analysis as ^{241}Pu decays to ^{241}Am in 14.4 year half-life, and ^{241}Am derived from ^{241}Pu

Source: EPRI (1996)

Table 13.7 Improved scaling factors for 99Tc and 129I based on mass spectroscopy analysis of samples

	Scaling factor	Condition to apply
^{99}Tc/^{137}Cs	2.50×10^{-8}	If ^{137}Cs/^{60}Co > 10
^{99}Tc/^{60}Co	1.30×10^{-6}	If ^{137}Cs/^{60}Co < 10
^{129}I/^{137}Cs	2.00×10^{-7}	If Cs is present in the waste
^{129}I/^{60}Co	3.20×10^{-8}	If Cs is not present in the waste

NRC (2000), EPRI (2015)

improved fuel integrity, ^{129}I should be scaled to ^{60}Co using the scaling factor 3.2×10^{-8} (EPRI 2015).

LILW characterization by sampling is done with two sampling schedules. The first one is to establish the baseline database for the calculation of the scaling factors. This initial sampling should be based on using multiple samples from each waste stream under consideration. The second one is to assure that the waste characteristics are not changed substantially or that the changes are tracked to produce new scaling factors. The changes in the waste characteristics can be caused by the change in fuel cladding performance, a change in materials or equipment in the system (e.g., steam generator replacement), or a change in operating chemistry. Updating the scaling factors from the second sampling can be done by simply replacing the old ones with the new ones, but in general new scaling factors are combined with the historical data for updating.

In the U.S., the standard sampling frequency suggested is once a year for class B and C wastes and biannually (every 2 years) for class A waste. Actual sampling at U.S. NPPs typically consists of: taking DAW swipes during outages (every 18 months), testing Class B and C filters annually, and sampling spent resins (A and B/C) when the resins are transferred from the tanks to the disposal containers. However, the sampling frequency may be increased or decreased based on consideration of the facility operations or the characteristics of waste stream or radionuclides.

LILW characterization must be confirmed periodically through direct measurements. If the radionuclide distribution remain consistent, increasing the length of time between the sampling campaigns can be considered by the power plant. If any process changes in a waste stream occur, potentially causing an upward classification of the waste, more frequent analysis of the waste should be performed.

13.3.4 Dose Rate Measurements

Along with sampling, dose rate measurements for gamma-emitting radionuclides can be used (EPRI 1996). The measurement allows LILW characterization based on the dose-to-curie method. The following steps are followed in the approach. First, the dose rate measurement is taken by using a detector at a specified distance and a geometrical position from the waste package. The resulting dose rate estimates (Sv/h or rem/h) are then converted to activity contents by using dose-to-activity calculations utilizing the following equation:

$$A = \frac{D}{\sum_{i=1}^{n} d_i f_i} \tag{13.1}$$

where A = total activity, D = dose rate measured at specified distance, d_i = radionuclide specific dose-to-activity conversion factor for a given waste package configuration, f_i = fractional abundance of radionuclide i.

The technique requires a priori knowledge of the waste composition and usually assumes uniform distribution of activity throughout the bulk waste. Such dose-to-activity calculations also assume that the measured dose rate is proportional to the activity in the container. In this calculation, the relative abundance of the radionuclides in the waste as gamma sources is needed which is obtained from the analysis of samples. The dose-to-activity conversion factors for radionuclides are determined from a separate shielding calculation using computer codes or relevant calculations using equations. These calculations are made by assuming a unit activity concentration of individual radionuclides in the waste with the specification of waste densities and geometries and container configurations. Multiple dose rate measurements in the same geometry from different locations on the container will improve the accuracy in estimating the activity through better averaging of any inhomogeneity.

The measured dose rates along with the data on relative abundance of gamma emitting radionuclides are converted to their activity concentrations. Then the results are used with the scaling factors to determine the activity concentrations of HTM nuclides in the waste package.

13.4 Classification of Low and Intermediate Level Waste

As discussed in Subsection 2.4.2.4, waste classification is needed to streamline the follow-on activities of waste management such as treatment, packaging, and disposal. Thus the classification scheme determines the specific protective measures to be taken in the waste management activities. While the scheme for classification varies in different countries, the classification scheme suggested by the IAEA provides a general guideline which is followed in many countries. The IAEA classification is based on the activity content and half-life of radionuclides in the waste as discussed in Sect. 6.4.5.

A review of nation-specific approaches to classification in different countries indicates that there are three broad generic approaches to classification: (1) the U.K./ France/Finland approach, (2) the Sweden/Belgium/Spain approach, (3) the U.S. approach.

13.4.1 The European Approaches to Waste Classification

The U.K./France/Finland approach is most closely aligned with the IAEA classification which is also recommended by the European Commission (EC) (LLW Repository 2016). The U.K. classification following activity concentration limits is shown in Table 13.8.

In Sweden, Belgium and Spain, the classification is primarily based on considering the dose rate of the waste package in handling and disposing of wastes at a disposal site, as laid out in facility-specific safety cases (LLW Repository 2016). Table 13.9 indicates that the Swedish classification scheme. Note specific disposal facilities are designated for the disposal of different classes of waste.

13.4.2 The U.S. Approaches to Waste Classification

As opposed to the waste classification approaches listed above, the U.S. waste classification is based primarily on the overall characteristics of the wastes as determined by their origin (Department of Energy (DOE) or commercial waste generators). Furthermore, in the U.S., a very different waste classification scheme is used by dividing LILW into different classes: class A, class B, class C, and greater than class C. The classification depends on the concentration of both short-lived and long-lived radionuclides in the waste. In comparison to the IAEA classification, class A represents short-lived low activity waste. This may corresponds to IAEA's EW (exempt waste), VSLW (very short lived waste), or VLLW (very low level waste). Class B represents more or less short-lived high activity waste that may corresponds to IAEA's LLW (low level waste)). Class C is for the long-lived high activity waste

Table 13.8 Low and intermediate level waste classification scheme in U.K (te = tonne)

Waste class	Activity limits	Description
ILW (intermediate level waste)	$>4 \times 10^0$ GBq/te α or ≥ 12 GBq/te, β/γ	Waste with radioactivity levels exceeding the upper boundaries for LLW but which do not require decay heat to be taken into account in the design of storage or disposal facilities.
LLW (low level waste)	$\leq 4 \times 10^0$ GBq/te α or ≤ 12 GBq/te, β/γ	Radioactive waste having a radioactive content not exceeding 4 GBq/te alpha activity or 12 GBq/te beta/gamma activity. LLW includes two sub-categories of very low level waste (VLLW).
VLLW (very low level waste) consists of two subcategories of LLW: LLW1 (low volume) and LLW2 (high volume)		
VLLW –subcategory of LLW1: Low volume (dust-bin disposal)	$< 4 \times 10^{-2}$ GBq/m^3	Waste which can be safely disposed of with municipal, commercial or industrial waste, each 0.1 m^3 of material containing less than 400 kBq β/γ activity or single items containing less than 40 kBq β/γ activity (note no α allowance and special requirements for waste containing C-14 and tritium).
VLLW –subcategory of LLW 2: Bulk disposals – High-volume VLLW	4×10^{-3} GBq/te	Radioactive waste with a maximum concentration of 4 MBq/te (0.004 GBq/te) of total activity, which can be disposed of to specified landfill sites. For waste containing tritium, the concentration limit for tritium is 40 MBq/te (0.04 GBq/te).
Exempt waste	$< 4 \times 10^{-1}$ Bq/g	Materials which are exempt from regulation. These can be disposed of with ordinary waste.

Source: Defra (2007)

and may overlaps both LLW and ILW (intermediate level waste). Greater Than Class C (GTCC) is ILW unless it is heat generating, in which case it is likely HLW. GTCC is long-lived very high activity waste except that the waste is not generated from the process of reprocessing. Examination of the U.S. LLW classification scheme is useful as the system is based on a quantitative analysis of the potential hazards involved.

Consideration of both long-lived and short-lived radionuclides in the U.S. classification scheme is related to the necessary use of institutional control, waste forms, and disposal methods to limit the potential long-term hazard, i.e., the dose to humans in the future. The required dose limits for the public are 0.25 mSv/year (25 mrem/year) to whole body, 0.75 mSv/year (75 mrem/year) to thyroid, 0.25 mSv/year (25 mrem/year) to any other organ of the body, and 5.0 mSv/incident (500 mrem/incident) in the case of an inadvertent intruder. The active institutional control was assumed for 100 years period after the closure of the facility.

Table 13.9 Low and intermediate level waste classification scheme in Sweden

Waste class	Disposal facility	Package dose rate	Half-life of significant radionuclides
Low and intermediate level long-lived waste (LILW-LL)	SFR disposal facility/ SFL repository (operation 2045)		Significant amounts of radionuclides with half-lives >31 years
Intermediate level short lived waste (ILW-SL)	Silo Concrete cylinder	< 500 mSv/h	Significant amount of radionuclides with half-lives <31 years
	BMA rock vault	< 100 mSv/h	
	BTF Two rock vaults	< 10 mSv/h	
Low level short-lived waste (LLW-SL)	BLA rock vault	< 2 mSv/h	Significant amounts of radionuclides with half-lives <31 years
Very low level, short-lived waste (VLLW-SL)	Shallow land burial at reactor site	< 0.5 mSv/h	Small amount of radionuclides with half-lives <31 years

Two potential scenarios of human exposure were considered as the basis of determining the dose to humans in the development of waste classes. These two scenarios are (1) an inadvertent intruder in direct contact with the disposed waste, and, (2) an individual's or population's radiation exposure from potential consumption or use of contaminated groundwater.

As a short-lived low activity waste, the class A waste is considered a low hazard level waste that poses little concern in terms of radiation exposure to humans either from direct contact or through inadvertent intrusion or through consumption or use of contaminated groundwater. Thus no requirement on the waste form or disposal site design is imposed except for limiting the liquids content and the need for segregation from class B&C if not stabilized. During the 100 year period of active institutional control, no human radiation exposure is expected from the presence of the disposal facility as intrusion into the facility is strictly prohibited. However, at the end of institutional control, an intruder may be exposed to radiation from the waste through various scenarios but the class A concentration limit controls the resulting dose to be less than the dose limit. The scenarios could involve building a house or raising crops at the site and consuming them.

The class B waste is a short-lived high activity waste and requires stabilization of the waste for 300 years to minimize the chance of direct contact with an inadvertent intruder before the decay of its contents. Stabilization of waste may be through waste immobilization or packaging in a structurally stable container, or use of any other stabilization measures at a disposal facility for the protection of an inadvertent intruder. It is assumed that at the end of the institutional control period (100 years), an inadvertent intruder digs into waste materials (that is other than natural soil) but terminates excavation work after 6 h recognizing that what's being

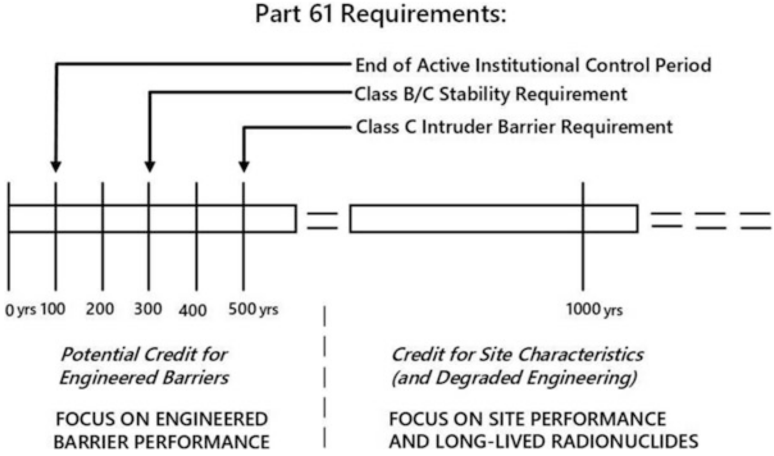


Fig. 13.5 Time lines associated with the use of institutional control, waste form, and intruder protection barrier in relation to waste classification

dug up is not a natural material. During the 6 h, he or she may be exposed to radiation through direct exposure. Nevertheless, the dose received is less than the limit to the intruder due to the concentration limit of the class B waste. This scenario assumes that an inadvertent intrusion involving the exposure to class B waste takes place between 100 and 300 years after the facility closure. After 300 years, the class B waste should not pose hazards due to the decay of the short-lived radionuclides.

The class C waste requires both stabilization and special site design (using intrusion barrier) to protect an inadvertent intruder beyond 300 years. In this case, an intrusion is assumed to take place between 100 and 500 years after facility closure. Safety against any intrusion between 100 and 300 years is provided by waste stabilization, through waste immobilization or waste packaging (e.g., using special canisters such as Modular Concrete Canisters (MCC)). This is the same approach used for class B waste, but an intrusion beyond 300 years is no longer protected by the waste stabilization requirement. In the case of beyond 300 years, the class C waste needs to be buried in a segregated manner (at least 5 m below surface) with an intruder barrier. The intruder barrier can be a concrete structure and is assumed to be effective for 500 years after facility closure. Thus, between 300 and 500 years, an intruder is prevented from contacting the waste due to the presence of the intruder barrier. However, after 500 years, it is assumed that the barrier no longer exists (due to concrete degradation), and an intruder can excavate the site and perhaps build a house and live at the site. In this case, after 500 years, a person's exposure to radiation can occur through construction of a house or various agricultural activities at the site. The class C waste concentration limit is set to limit the resulting radiation dose within the annual dose limits.

The time lines associated with the use of institutional control, waste form, and an intruder protection barrier relative to waste classification are summarized in Fig. 13.5. The concentration limits determined for different classes of LILW according to these scenarios and analyses are given in Table 13.10. The figure also indicates that for the required performance of the LILW disposal facility over 1000 years, site characteristics are important and considered for the protection of the public regardless of the failures of engineered systems.

The concentration limits used to classify LILW (classes A, B and C) in the U.S. are given in Table 13.10. It is recognized that a number of radionuclides likely to be of concern are not reflected in Part 61, however these radionuclides are typically identified and analyzed in individual disposal site license conditions.

The majority of NPP GTCC waste are activated metal wastes generated during the decommissioning process. The portions of the nuclear reactor vessel such as the core shroud and core support plate will be GTCC. The prevalent radionuclides that result in a GTCC designation in activated metals are ^{14}C, ^{54}Mn, ^{55}Fe, ^{59}Ni, ^{63}Ni, ^{94}Nb and ^{60}Co (DOE 2016). According to the 1985 LLW Policy Act Amendment, GTCC wastes are to be disposed of at a HLW geologic repository under the responsibility of U.S. DOE, although the waste was produced from commercial nuclear power generation.

The following figure (Fig. 13.6) is a good representation of waste types and waste class disposed annually in the in U.S. in terms of volume, based on EPRI data presented in 2006.

Example 13.1: Estimation of Dose Rate from LILW Drum
A 55 gallon (200 liter) drum containing ion-exchange resins from a PWR is labeled LILW. The dose rate measurement at 50 cm from the surface of the container gave a value of 1 mR/h. Based on the data of the resins at the sampling location in the plant, the waste is known to have the relative abundance of 93.6% of ^{137}Cs and 6.4% of ^{60}Co. The waste drum is also known to contain ^{3}H, ^{14}C, ^{99}Tc, and ^{129}I. By using the following dose-to-activity conversion factor and scaling factors, determine the class of this LLW according to the U.S. classification scheme.

Given:

Dose-to-activity conversion factors:

^{137}Cs: 2.228 (mR/h/mCi)
^{60}Co: 9.422 (mR/h/mCi)

Scaling factors:

^{3}H/^{60}Co: 1×10^{-3}
^{14}C/^{60}Co: 8×10^{-2}
^{99}Tc/^{137}Cs: 2.5×10^{-8}
^{129}I/137Cs: 2.0×10^{-7}

Solution:

By using Eq. (13.1),

(continued)

Table 13.10 Concentration limits for LLW classification under 10 CFR Part 61

Radionuclide	Long Lived Nuclides Concentration (Ci/m^3)
C-14	8
C-14 in activated metals	80
Ni-59 in activated metals	220
Nb-94	0.02
Tc-99	3
I-129	0.06
Alpha emitting transuranic nuclides with half-lives >5 years	100 nci/g
Pu-241	3500 nci/g
Cm-242	20,000 nci/g

[a] These concentration limits are class C limits and based on the intruder scenarios.

	Short lived nuclides concentration (Ci/m^3)		
Radionuclide	**Class A**	**Class B**	**Class C**
Total of all nuclides with half-lives <5 years	700		
H-3	40		
Co-60	700		
Ni-63	3.5	70	700
Ni-63 in activated metal	35	700	7000
Sr-90	0.04	150	7000
Cs-137	1	44	4600

For long-lived radionuclides:
(i) If the concentration does not exceed 0.1 times the value in the table, the waste is class A
(ii) If the concentration exceeds 0.1 times the value in the table but does not exceed the value in Table 13.10, the waste is class C
(iii) If the concentration exceeds the value in the table, the waste is not generally acceptable for near-surface disposal
(iv) For wastes containing mixtures of radionuclides listed in the Table 13.10, the total concentration shall be determined by the sum of fractions rule described shown in Example 13.2
For short-lived radionuclides,
(i) If the concentration does not exceed the value in Column 1, the waste is class A
(ii) If the concentration exceeds the value in Column 1, but does not exceed the value in Column 2, the waste is class B
(iii) If the concentration exceeds the value in Column 2, but does not exceed the value in Column 3, the waste is class C
(iv) If the concentration exceeds the value in Column 3, the waste is not generally acceptable for near-surface disposal
(v) For wastes containing mixtures of the nuclides listed in the table, the total concentration shall be determined by the sum of fractions rule
[a] (beyond class A concentration limits) These wastes shall be class B unless the concentrations of other nuclides (short-lives nuclides) in Table 13.10 determine the waste to the class C independent of these nuclides

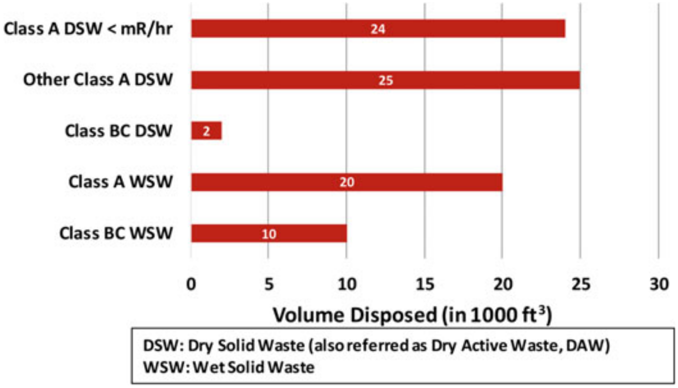


Fig. 13.6 Operating U.S. reactors: annual LILW disposed by waste class (from data: NEI 2006)

Example 13.1 (continued)

$$\frac{1\left(\frac{mR}{h}\right)}{0.064\times 9.422+0.936\times 2.228\left(\frac{\frac{mR}{h}}{mCi}\right)}=0.372\,mCi\left(1.38\times 10^{7}Bq\right).$$

Therefore, the waste contains,

$0.372\ \text{mCi}\times 0.936=0.348\ \text{mCi of }^{137}\text{Cs}$
$0.372\ \text{mCi}\times 0.064=0.024\ \text{mCi of }^{60}\text{Co}$

The contents of ^{3}H, ^{14}C, ^{99}Tc, ^{129}I are:

^{3}H: 0.024 mCi of $^{60}\text{Co}\times 1\text{x}10^{-3}=2.4\times 10^{-8}$ Ci
^{14}C: 0.024 mCi of $^{60}\text{Co}\times 8\text{x}10^{-2}=1.92\times 10^{-6}$ Ci
^{99}Tc: 0.348 mCi of $^{137}\text{Cs}\times 2.5\text{x}10^{-8}=8.7\times 10^{-12}$ Ci
^{129}I: 0.348 mCi of $^{137}\text{Cs}\times 2.0\text{x}10^{-7}=6.96\times 10^{-11}$ Ci

The concentration of the radionuclides in the waste (with the volume of waste at 200 L = 0.2 m^{3}):

The concentration of the radionuclides in the drum:

^{137}Cs: 3.48×10^{-4} Ci/0.2 (m3) = 1.74×10^{-3} (Ci/m^{3})
^{60}Co: 2.4×10^{-5} Ci/0.2 (m3) = 1.2×10^{-4} (Ci/m^{3})
^{3}H: 2.4×10^{-8} Ci/0.2 (m3) = 1.2×10^{-7} (Ci/m^{3})
^{14}C: 1.92×10^{-6} Ci/0.2 (m3) = 9.6×10^{-6} (Ci/m^{3})
^{99}Tc: $8.\times \text{x}10^{-12}$ Ci/0.2 (m3) = 4.4×10^{-11} (Ci/m^{3})
^{129}I: 6.96×10^{-11} Ci/0.2 (m3) = 3.5×10^{-10} (Ci/m^{3})

For short lived radionuclides (with respect to the Class A limit)

(continued)

Example 13.1 (continued)

$$\frac{1.2\times10^{-7}}{40}\left({}^{3}H\right)+\frac{1.2\times10^{-4}}{700}\times\left({}^{60}Co\right)+\frac{1.74\times10^{-3}}{1}\left({}^{137}Cs\right)=0.00174<1$$

For long-lived radionuclides (with respect to the Class A limit)

$$\frac{9.6\times10^{-6}}{0.8}\left({}^{14}C\right)+\frac{4.4\times10^{-11}}{0.3}\times\left({}^{99}Tc\right)+\frac{3.5\times10^{-10}}{0.006}\left({}^{129}I\right)=1.2\times10^{-5}<1$$

From the consideration of both short- and long-lived ones, the waste is determined to be Class A.

Example 13.2: Classification of Low and Intermediate Level Wastes

Using the U.S. classification scheme, determine the class of the LILW with the given nuclide concentrations.

^{129}I at 0.075 Ci/m^3 & ^{60}Co at 750 Ci/m^3

Sums of fractions rule can be used as following to find the total concentration for radionuclides a, b, …

If $\frac{concentration_a}{Limit_a}+\frac{concentration_b}{Limit_b}+\ldots>1$, then Class Limit is exceeded.

Solutions:

^{129}I is a long-lived nuclide while ^{60}Co is a short lived. So we examine each one separately.

- The limit for ^{129}I for Class C is 0.06 Ci/m^3: 0.075/0.06 = 1.25 > 1. As the Class C limit is exceeded. This is GTCC (greater than Class C).
- The limit for the ^{60}Co (short lived nuclides) for Class A is 700 Ci/m^3: 750/700 = 1.07 > 1. As the limit for Class A is limited, this is Class B.
- With the mixture of GTCC (^{129}I) and Class B (^{32}P), the more conservative class is used. So the waste is GTCC. In other words, the waste class is dictated by the most conservative table.

13.5 Treatment/Processing of Low and Intermediate Level Waste

LILWs are often treated for immobilization or volume reduction, then packaged for long-term isolation. Treatment of LILW refers to any processes applied to the waste before or during the packaging of the waste for disposal. For example, during the early periods of radioactive waste management in the 1950s and 1960s, "dilute and disperse" was exercised as a way of treating LILW. However, such practices were discontinued after the advancement in radiation biology indicating that there may not be a threshold level of dose for adverse effect of radiation. In other words, there was still a concern for the outcome of "dilute and disperse' as low level contamination could still potentially cause damage to humans.

The primary objectives of LILW treatment is to reduce the volume of waste to be disposed of and/or to condition the waste for long-term stabilization prior to final disposal. If possible, minimizing the production of LILW is also pursued. For example, the "delay and decay" process could prove useful to minimize the production of LLW if the radionuclides of concern are relatively short-lived. Hold-up storage is an example of "delay and decay" and has been used for short-lived noble gases at NPPs. Regarding waste minimization, pretreatment of waste is often needed prior to LLW treatment. Pretreatment includes segregation and decontamination. Segregation is to separate the waste that is non-radioactive from LLW. In the U.S., initial onsite segregation of DAW is conducted to separate the waste into: metal and nonmetal, or combustible and noncombustible. In addition, on-site segregation may be used to separate the waste that is non-radioactive material from LLW. The on-site segregation is to identify potentially clean waste based on the location of waste origin, i.e., waste from a walkway in a radiologically controlled area (clean) versus inside a radiologically contaminated area. In addition, potentially clean waste is also identified based on a contact dose rate of <2 mSv/h (< 200 mrem/h). However in the U.S., the final survey and disposal of these materials for free release is undertaken by an off-site processor. For U.S. NPPs, the majority of pretreatment/decontamination is contracted to offsite vendors. This segregation results in the separation of material that can be sent to landfill sites licensed to accept industrial wastes, or class A wastes being sent to a disposal facility that only accepts class A waste delivered in a transportation package (i.e., sealand container) but does not require a waste disposal container.

Decontamination is used to remove radioactive contamination from metal objects, using mechanical or chemical processes to render the item free of loose radioactive contamination. Typically, on-site decontamination at U.S. NPPs involves items such as hand tools or scaffolding, allowing these tools and equipment to be reused inside the radiologically controlled area. Any decontamination undertaken to reclassify a LLW item as non-radioactive would likely be done by an off-site vendor. Decontaminated waste is surveyed and can be released as non-radioactive if appropriate. Application of segregation or decontamination will depend on types of waste stream, materials and the related costs.

In terms of actual treatment, most often the approaches used involve the concept of "concentrate and contain", to achieve volume reduction and stabilization. Therefore through effluent treatment, the release of radioactive materials as effluents in the waste water and off-gases to the environment is minimized. Treatment could also be for stabilization as required by the regulations. For example, stabilization is performed by the process of dewatering resins to meet free liquid process requirements and placing them in a steel liner and then modular concrete canisters for disposal. Diatomaceous earth (small particles of silicon oxide) is added to liners with filter cartridges, in order to adsorb small amounts of water, to meet the free standing liquid requirement.

In a broad sense, LILW treatment processes can be accomplished via four approaches: (1) transfer technologies, (2) concentration technologies, (3) transformation technologies, and (4) conditioning technologies. Selection of a technology for treatment of LILW depends on the chemical and physical characteristics of the waste under consideration. It is important to recognize that, while the volume of waste may be reduced, the amount of radioactivity remains the same, i.e., the waste will be more concentrated in radioactivity contents as the volume is reduced.

13.5.1 Transfer Technologies

Transfer technologies refer to the processes that remove radioactive species from a waste stream and transfer them to another medium. Most of the liquid and gaseous radioactive waste treatment processes employed in nuclear power plants are using transfer technologies. Use of ion-exchange resins and filters as discussed in Sect. 13.2.1 are key examples of transfer technologies. At the end of applying transfer technologies, the radioactivity of the waste will be collected (as more concentrated) in the treatment media which become LILW. Besides ion-exchange and filtration, reverse osmosis, ultrafiltration, and chemical regeneration are also examples of transfer technologies. Decontamination of solid LILW such as metals is also an example of transfer technologies.

Ion-exchange removes dissolved radioactive species (like cesium) in the water through the ion exchanges of one ion for another, often with demineralizer resins. The removed radionuclides are retained in the resins. Filtration is used to remove particulates by using filters such as cartridge filters or HEPA filters for particulate matter or a charcoal filter for iodine removal. The radioactivity in cartridge filter waste is captured and retained in very small pores through filtration, impingement, and adsorption. Filters are placed upstream of ion-exchange treatment to improve the efficiency of ion-exchange resins.

Reverse osmosis is a special type of filtration based on the use of a semipermeable membrane. When a solution with two different concentration levels is contained in a chamber but divided by a semipermeable membrane, water will move from the higher concentration solution to the lower concentration solution thus equalizing the concentration on each side of the membrane. This is a natural process and is called osmosis. However, pressure can be used to push the higher concentration solution through the membrane. This is called reverse osmosis. Through reverse osmosis, radioactively contaminated water is pushed through a membrane under pressure. Then the dissolved ions and particles in the contaminated water are filtered out resulting in a purified water stream. Reverse osmosis has become common technology in the U.S. nuclear industry for purification of reactor coolant (e.g., in the condensate systems of BWRs) when large volumes of water require processing.

Ultrafiltration is similar to reverse-osmosis as pressure is used to push the contaminated stream through a semipermeable membrane. The main difference is the pore size. Ultrafiltration membranes have pore sizes ranging from 1 to 100 nm while reverse osmosis membranes are at about 0.1 nm. Ultrafiltration removes macromolecules and colloidal particles while reverse osmosis removes very fine particles and dissolved ions.

Chemical regeneration is used to re-transfer the collected contamination in ion-exchange resins that are not in the higher activity range (IAEA 2012a). Thus through chemical regeneration, the spent resins can be regenerated for reuse. However, U.S. utilities often choose to send the resins directly to disposal rather than regenerating them because of the additional cost of regeneration and the additional need to neutralize, treat, and dispose of the regeneration wastes. Chemical cleaning is used to recover membrane in reverse-osmosis or ultrafiltration systems. In both processes of regeneration and chemical cleaning, acid, alkaline, surfactant or detergent solutions are used.

Metal melting is a technique of volume reduction and decontamination of metallic waste. With metal melting, the radioactive elements are oxidized and largely transferred to a slag phase. Thus the metal is significantly decontaminated and the contaminants are concentrated in the slag. The decontaminated metal still contains residual radioactivity and may be blended into new steel for reuse under certain regulatory regimes (Germany for one). The extent to which metal melting is used depends on the country's regulatory requirements or alternative disposal options and the accessibility of the melting facility. For example, Sweden (SSM 2015), France (France 2014) and Canada (IAEA 2006c) use metal melting whereas the U.S. has mostly replaced the use of this technology with bulk burial at the class A disposal facility in Utah.

13.5.2 Concentration Technologies

Concentration technologies are to reduce the waste volume. They are widely used to process liquid and solid LILW. Major examples of these technologies are compaction or supercompaction for solids; evaporation for liquids, and; drying or dewatering for wet solids. The use of these technologies varies by country, depending on available disposal and technology development.

Compaction or supercompaction is a well-developed technology of applying hydraulic press for volume reduction (VR) of solid LILW waste. The VR factor is the ratio of initial volume to volume after treatment. Depending on the level of compaction force applied, the compactors are called low-force compactors or super compactors. Compactors apply a force of 2 to 400 tons (20–4000 kN), while supercompactors typically apply 1000 ton (10,000 kN) to 2200 ton (22,000 kN) of force (Jolley et al. 1986). Supercompaction is applied for waste that cannot be handled in lower force compactors. Typically, the VR factor achieved ranges between 3 and 12 (IAEA 1983), depending on the type of waste material being treated (the input density, theoretical density, and potential springback or re-expansion) and compaction force factors applied. Application of compaction or supercompaction depends on the level of liquid contained in the waste. In the case of supercompaction, the technique cannot be applied if free standing or absorbed liquid exists in the waste. Low force compactors are more typically located at the NPP site, whereas supercompactors are used at a vendor facility or disposal facility. Recently, with the availability of class A bulk disposal in Utah, compaction is not typically used by U.S. NPPs. In Canada, France, Sweden and other countries, compaction is widely used (ANL 2011). Typically, bulk wastes in the U.S. are disposed in Utah where the wastes are compacted in soil lifts at the disposal facility (EnergySolutions 2015).

Evaporation is used for liquid waste, in particular in the boric acid treatment system. Through evaporation, the water in the waste is boiled off and the steam is condensed leaving most of the radioactive material behind as the residue. The evaporator residue becomes LILW. A factor of 2–4 volume reduction is expected by applying an evaporator (IAEA 1983). However, because evaporator bottoms must often be solidified thus increasing the volume, this efficiency is largely lost.

Drying or dewatering is an important processing step for wet solid wastes (WSWs). Drying uses heat to remove liquid while dewatering uses pumping or gravitational flow to draw water from wet solids. Dewatering is widely applied to ion exchange resins, filters, filter sludges, and miscellaneous sediments. Drying is also applied to evaporator concentrates (bottoms) and is more volume efficient than cement solidification.

13.5.3 Transformation Technologies

Transformation technologies are used to concentrate radioactive waste through changes in the physical form. They include incineration, steam reforming, supercritical-water oxidation, catalytic extraction processing (also called molten-metal technology) and plasma arc technology.

Incineration burns the waste in a controlled manner. Incineration converts the waste into ash, flue gas and heat. It is mainly used to treat organic materials in the waste through decomposition. However, in a number of countries the low activity portion of their low level waste are incinerated. Depending on the country, this can be done at an offsite vendor location (France, CENTRACO facility) at the disposal facility (Spain, El Cabril for institutional waste only) or at the power company's

LILW storage facility (Canada, OPG Western Waste Management Facility) (OPG 2019). In the use of these technologies, the facility operation must comply with the effluent release limits.

In steam reforming, high temperature steam is used to breakdown organic materials in the waste through reactions in an oxygen deficient environment. Supercritical water oxidation uses the special properties of supercritical water for the destruction of hazardous materials in the waste. At supercritical condition (i.e., water at temperatures and pressures above the thermodynamic critical point), water becomes an agent of gas-phase free radical reactions and causes decomposition of the reacting materials. Steam reforming or supercritical water oxidation can be applied to organic wastes such as fire resistant sheeting and rubber gloves, and ion exchange resins.

Catalytic extraction processing (CEP) uses high temperature metal to break down hazardous and radioactive materials in the wastes. This results in benign forms of materials by breaking molecular bonds of the waste and reducing it to its constituent elements. The constituent elements are then recombined to make inorganic gases or metals that could be recycled through use in other industrial activities. In the U.S., the technology is used to treat ion exchange resins and non-metal filter cartridges. The VR factor of about 7 and 35 was achieved for ion exchange resins and non-metal filter cartridges, respectively (IAEA 2006b).

Plasma arc technology is based on creating high temperature plasma arc to treat waste for breakdown of organic components into simpler atoms/molecules. The remaining inorganic components are melted and cooled into a glassy slag for disposal.

13.5.4 Conditioning Technologies

Conditioning technologies are to stabilize or immobilize the waste for disposal. These technologies were described as part of waste form discussions in Sect. 9.4. As discussed in Sect. 9.4, encapsulation/solidification of waste using a good waste form significantly reduces the potential for the release of radionuclides to the environment.

Other than concrete, polymers, or glass, grouting is also used for LILW encapsulation. Grouting is defined as an injection of slurry cementitious materials under pressure into structures in order to fill and seal voids, cracks or other cavities in the system (EPA 1984). Bituminization is another approach to LILW encapsulation. It is generally used for the treatment of liquid wastes/sludges and involves drying of the waste before it is combined with the bitumen to form a conditioned product. The bituminization process, due to the flammable nature of the constituent materials, needs to be carefully controlled.

Stabilizing the waste by using durable, long-life containers is exercised in the U. S., as part of waste conditioning. Currently, most disposal facilities accepting class B and C wastes rely more heavily on concrete over packs/canisters by placing waste packages in them. In this case, the necessary stabilization or isolation of waste is mainly achieved by the use of concrete overpacks and canisters.

Examples of the usage of the conditioning techniques in the management of LLW and ILW in different countries are presented in Table 13.11.

In the U.S., a large fraction of commercial LILW is disposed of without solidification. This is in part due to economic penalties in the disposal rate structure for waste volume increase caused by solidification. In addition, U.S. NRC gives no credit to using chemically stable waste form in disposal facility performance assessments and emphasizes the structural stability of waste form. This removes the incentive for the use of chemically stable waste form for LILW. Currently, the U.S. industry practices waste stabilization using durable, long-life containers (high integrity containers (HICS)) along with reliance on concrete overpacks and canisters provided by the disposal facility.

High-integrity containers (HICs) are, in comparison to carbon steel drums and liners, more durable type of container for the disposal of low and intermediate level waste and thus for the disposal of long-lived high activity waste. HICs as a composite barrier can be made from corrosion resistant metal alloys, reinforced concrete, high density polyethylene (HDPE), or polymer-coated metals. One type of HIC widely used is a combination of both a HDPE and a concrete overpack. HICs are required to have a minimum lifetime of 300 years by the U.S. NRC regulations (NRC 1983).

Example 13.3

(a) The water contained in the reactor coolant system of the damaged unit at Three Mile Island was severely contaminated during the accident and had to be decontaminated before further work on the core could proceed. The principal radiological gamma hazard in the reactor water was ^{137}Cs.

At the time that decontamination began, the inventory of ^{137}Cs solution in the reactor coolant system was 4300 Ci. The volume of coolant was 345 m^3.

The decontamination was performed by means of a zeolite ion-exchanger with a throughput of 50 liters per hour. The nominal decontamination factor (DF) achieved in the ion-exchanger was 10,000 (DF = inlet concentration/exit concentration).

How long did it take to reduce the ^{137}Cs activity in solution to the target level of 0.1 μCi/ml?

(b) Suppose that the ^{137}Cs loading on the dewatered resin is 10 μCi/cm^3 at the time of packaging the waste. Assume further that there are no other radionuclides on the resin. The resin is to be immobilized within a concrete matrix. If the waste loading in a concrete matrix is 15% for the dewatered resin, what is the class of the solidified waste?

(continued)

Table 13.11 Summary of waste conditioning in five countries

Waste Type	Canada	France	Korea	Spain	Sweden	U.S.
Homogeneous, wet waste: Resins Filters Sludge Concentrates	Dewater[a] or Cement solidification[b]	Polymer solidification or Cement solidification	Resin drying or Dewatering Concentrates: Polymer solidification	Cement solidification	Cement solidification or Bituminization Or Dewater[e]	Pyrolysis or Dewatering or Drying and Concrete
Final disposal			Overpack[c]	Overpack[d]		Overpack[f,g]
Heterogeneous DAW/DSW: Combustible Compactable Non-combustible	Compaction and/or incineration or Metal melt	Compaction or incineration or Metal melt	Compaction or Vitrification	Compaction plus cement encapsulation or Cement stabilization	Compaction plus cement encapsulation or Incineration or Metal melt	Bulk disposal – Rare: Compaction Incineration Metal melt
Final disposal			Overpack[c]	Overpack[d]		

Source: Kim (2016)

[a] In Canada intermediate level wastes are not conditioned, except for dewatering resins at the NPP site

[b] CanDu reactor Active Liquid Waste Treatment System sludge – cemented in relatively small (pail size) quantities generated

[c] Final disposal container (concrete overpack)– all wastes

[d] Final disposal container – most wastes

[e] Low activity resins – are dewatered and placed in concrete tanks for disposal in the LLW-LL disposal cavern, (cavern called BTF)

[f] Barnwell cement overpack

[g] Modular concrete container

Example 13.3 (continued)

(c) Assume that a lifetime dose limit of 10^5 rads to the concrete matrix has been specified because of concern over the effects of radiation damage to the concrete. What class of waste should the resin be disposed of to meet the lifetime dose limit?

Note: The average gamma energy per ^{137}Cs decay = 0.276 MeV. Assume that the contribution from beta radiation can be ignored with respect to radiation damage.

Solutions:

(a) Initial concentration of ^{137}Cs in the coolant = 4300 Ci/345 m^3 = 12.4 μCi/ml

Target concentration = 0.1 μCi/ml

$\lambda = 0.693/(30 \text{ years}) = 0.023 \text{ year}^{-1}$

dC/dt = −λC – (total removal rate in ion exchange resins)/(total volume)

dC/dt = −λC – (50 L/h)(C)(8766 h/year)(1000 ml/L)/345 × 10^6 ml = −(λ + 1.27)C

ln(C(t)/C(0)) = −(λ + 1.27)t

C(t) = C(0)exp(−(λ + 1.27)t)

/12.4 = 8.06 × 10^{-3} = exp(−1.293 t)

t = 3.73 year

(b) The limits for class A, B, and C for ^{137}Cs are 1 Ci/m^3, 44 Ci/m^3, and 4600 Ci/m^3, respectively.

Given the waste loading of 15%, the volume of the solidified waste form for a 1 m3 of dewatered resin becomes 1/0.15 = 6.7 cm^3. The concentration of ^{137}Cs is then 10 Ci/6.7 m^3 = 1.49 Ci/cm^3. This belongs to Class B.

(c) To meet the limit 1 Ci/m^3 of class A, the concentration of the dewatered resin needs to be lowered by a factor of 10. Thus the volume of concrete to be added is 9 times the volume of resin,

As the waste is already class B, no further treatment is needed to be in the class B category.

Also, it is not necessary to consider class C as that is beyond the current concentration.

To examine whether the cumulative dose limit can be met as class A or class B waste:

(continued)

Example 13.3 (continued)

Let's determine the dose rate for the given the concentrations of either class A or class B waste. Then, use that number for the entire decay periods of 137Cs.

The ^{137}Cs loading on the resin is 10 μCi/cm^3 at the time of the discharge.

$$\frac{10\mu Ci}{cm3\,\frac{37000Bq}{\mu Ci}} = 3.7 \times 105\ \mathrm{Bq/cm^3}$$

The resulting dose rate from this loading is

3.7×10^5 Bq*0.275 MeV*1.6×10^{-6} (erg/MeV)*1 (rad/(100 erg/g)) = 1.63×10^{-3} (rad/s)(g/cm^3)

During the entire period of ^{137}Cs being radioactive, the total dose =

$$\begin{aligned}1.63 \times 10^{-3}\left(\frac{rad}{s}\right)\int_0^\infty e^{-\lambda t}dt\left(\frac{g}{cm3}\right) &= 1.63 \times 10^{-3}\left(\frac{rad}{s}\right)\frac{1}{\lambda}\\ &= 1.63 \times 10^{-3}\left(\frac{rad}{s}\right)\\ &\quad\times\frac{1}{7.3\times10^{-10}(sec^{-1})}\\ &= 2.23 \times 106\ \mathrm{rad} - (\mathrm{g/cm3})\end{aligned}$$

The density of ion exchange resin: 0.7 ~ 0.8 g/cm^3

Assuming 0.8 g/cm3, the total dose of the waste as class B is 2.23×10^6/0.8 = 2.8×10^6 rad.

This is beyond the dose limit of 10^5 rads. So the cumulative dose limit cannot be met as class B.

If the waste is solidified with concrete to be class A, the volume needs to be increased 9 times.

The density of dewatered resin in concrete waste form = ~1.6 g/cm^3.

The total dose then becomes,

$$\frac{2.23 \times 10^6\left(rad * \frac{g}{cm3}\right)}{1.6\left(\frac{g}{cm3}\right)} \times \frac{1}{9} = 1.5 \times 10^5\mathrm{rad}$$

This exceeds the lifetime dose limit. Therefore the cumulative dose limit cannot be met as class A waste either. The results indicate that the waste needs to be encapsulated in a different medium for disposal.

13.6 Packaging of Low and Intermediate Level Waste

After LILWs are processed based on whether the radioactive waste is liquid, wet, or dry solid wastes, the resulting waste products are placed in a shipping container/packaging. Selection of the package used depends on the type, volume, radioactivity levels of the radioactive material processed. The containers /packaging provide protective barriers against physical and chemical stresses during transportation, interim storage, and disposal. Waste containers used in the burial of LILW vary from country to country. Generally waste containers include carbon steel drums, steel liners and boxes. In the U.S. some LILWs are shipped in bulk in reusable intermodal containers. These wastes are disposed at facilities that accept Class A wastes such as in Utah. High integrity containers (HICs) are used when the waste form is required to meet isolation from the environment for 300 years. HIC's are typically placed in site specific concrete containers (i.e., Andrews, Texas, Barnwell, South Carolina, Envirocare, Utah disposal facility). The packaged LILW is placed in a shipping cask and shipped to a processor for treatment before disposal or directly to a disposal facility.

The shipping of LILW is per international transportation requirements set out by the IAEA and are used by countries to standardize LILW shipping requirements (IAEA 2012b). Therefore, LILW shipping between countries is standardized. Each country's LILW regulator typically incorporate those requirements with existing regulations for public protection. In the US, the Department of Transportation (DOT) and the Nuclear Regulatory Commission (NRC) have outlined their respective responsibilities for regulating the transportation of radioactive waste in a memorandum of understanding. The NRC regulates packaging and shipment of fissile material and of wastes containing radioactive materials greater than a Type A quantity (defined below). The DOT regulates the transport of all other radioactive wastes.

Implementation of the shipping requirements for LILW from NPPs is described below according to the U.S. regulations. The packaging includes the use of general design package for low specific activity (LSA) material or surface contaminated objects (SCO) (in the US, these are shipped as exclusive use shipment), Type A packages, and Type B packages.

Examples of general design packages for LSA and SCO material include steel drums, wooden boxes, metal trunks, etc. In the case of reusable shipping containers, the waste is packed into an inner container, which is directly disposed of with the wastes at the disposal site.

Type A packages are used for the transport of quantities of radioactive material below the Type A activity limits that is not LSA or SCO material. Type A packages are required to maintain integrity during normal transport conditions. Demonstration of the compliance with the requirements is through testing under free drop, stacking or compression, and puncture tests. Common examples of Type A packaging are steel drums and liners but fiberboard box or wooden box can be Type A packaging.

Table 13.12 Type A package activity content limits for selected radionuclides

Radionuclide	A_1 (in TBq[a]) (special form)	A_2 (in TBq[a]) (normal form)
^{3}H	4×10^{1}	4×10^{1}
^{14}C	4×10^{1}	3×10^{0}
^{32}P	5×10^{-1}	5×10^{-1}
^{60}Co	4×10^{-1}	4×10^{-1}
^{63}Ni	4×10^{1}	3×10^{1}
^{99}Mo (including decay products)	1×10^{0}	6×10^{-1}
^{99}Tc	4×10^{1}	9×10^{-1}
^{129}I	Unlimited	Unlimited
^{137}Cs (including decay products)	2×10^{0}	6×10^{-1}
^{192}Ir	1×10^{0}	6×10^{-1}
^{201}Pb	1×10^{0}	1×10^{0}
^{226}Ra (including decay products)	2×10^{-1}	3×10^{-3}
^{241}Am	1×10^{1}	1×10^{-3}
^{235}U	Unlimited	Unlimited
^{238}U	Unlimited	Unlimited
^{239}Pu	1×10^{1}	1×10^{-3}

Source: IAEA (2012b)

[a] 1 TBq = 10^{12} Bq

Type B packages are under the most stringent packaging requirements and used for the transport of highly radioactive material such as ion exchange resins, spent fuel, irradiated hardware, etc. Type B packages are certified by the NRC for normal conditions of transport and hypothetical accident conditions of transport.

The quantity of radioactive material in each package must be within the specified limits for the package (see Table 13.12). In the case of Type A package, the maximum activity value is the A_1 value for an indispersible solid material or a sealed source (these are called special form material) or A_2 for normal form (other than special form, LSA or SCO material). Most LILW shipped from NPP is in a Type A package under the limit of A_2. The values of A_1 and A_2 limits are given in Table 13.12 for a select group of radionuclides. For the case of mixture of radionuclides, the sum of the ratios of the activity content to the A_1 or A_2 limits values should be less than or equal to zero for the mixture to meet the Type A quantity limit.

Protection of the general public during transport is accomplished by applying public radiation dose limits. The limits applied to these packages are 2 mSv/h (200 mrem/h) at any point on external surface of the package and 0.1 mSv/h (10 mrem/hr) at 1 m from the surface.

While all countries shipping LILW follow the IAEA international transportation requirements, some countries process the wastes within a package. For example, France embeds its ion exchange resins in a polymer matrix, in a cement cylindrical container and is rated as an IP-2 container for transport (LLW Repository 2018). (IP-2 containers are containers designed and certified for radioactive contaminated cargoes for storage and transport by ship, barge, road, or rail). These packages are transported via truck or rail. Sweden cement solidifies their ILW resins in concrete molds (concrete boxes) which are placed in an ATB container (equivalent to an IP-2 container) for transport by sea (SKB 1996).

13.7 Disposal of Low and Intermediate Level Waste

Similar to HLW, LILW disposal demands high level efforts in site selection and facility development. For the screening of potential sites, information to be reviewed includes land use, transportation, geography and demography, ecology, meteorology, hydrogeology, geology and seismology. The socioeconomics of the population and regional historic, archaeologic or cultural landmarks should also be considered. These efforts can be followed by onsite visits, a preliminary survey, physical inspections, and a limited amount of sampling (soil, surface water). From these screening activities, candidate sites can be identified. As discussed in Chap. 10, such screening efforts may not amount to successful site selection unless the local government/public consent to the development. Therefore, the effort must be paralleled with activities of engaging with the public finding volunteer communities and developing appropriate compensation package. Upon obtaining consensus with the local government and the public, detailed site characterization can be performed. With such support from the local government and the public, a LILW disposal facility can be constructed, contingent upon licensing approval by the government for the performance assessment of the facility.

In the U.S., approximately 1.2×10^5 m^3 (4.25 million ft^3) and 5.0×10^{15} Bq (135,000 Ci) of LILW were disposed of at commercial facilities in 2019. The volume and radioactivity of waste disposed of vary from year to year based on the types and quantities of waste shipped. The following figure (Fig. 13.7) shows the variations in the volume and radioactivity of LILW disposed of at domestic LILW disposal facilities from 1986 through 2018. The volume of the disposed LILW steadily decreased over the years mainly due to the fee structure of LILW disposal which is primarily based on volume.

13.7.1 Disposal Methods for Low and Intermediate Level Waste

The design of a disposal facility depends on the selected disposal methods. The methods used for LILW disposal include shallow land burial, intermediate depth disposal, above-ground vault, below-ground vault, earth-mounted concrete bunker, and modular concrete canister. If not near-surface facility, shaft disposal or mined cavity concept is also used. As with the case of HLW disposal, long-term isolation of waste is pursued as the goal of LILW disposal. Thus the design of a LILW disposal facility should aim at minimizing contact of water with waste while maintaining the

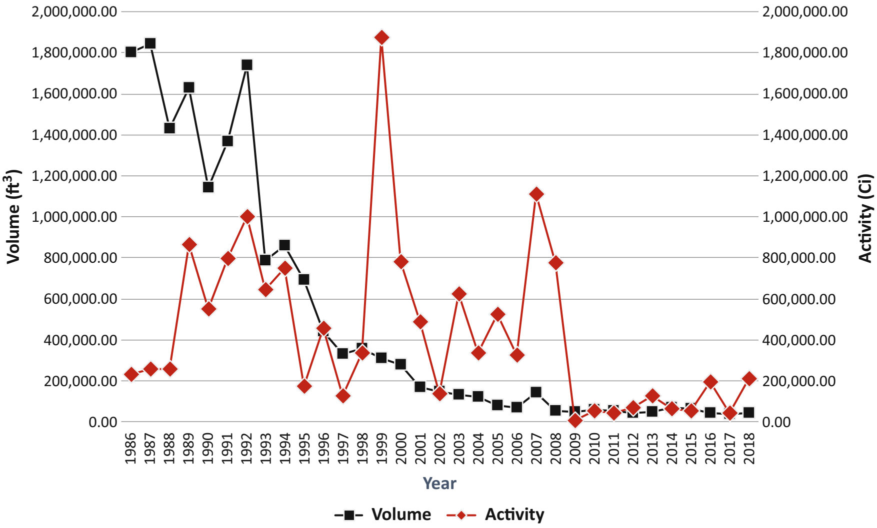


Fig. 13.7 Total annual volume and activity disposed at all commercial US disposal facilities except Envirocare

integrity of the engineered barrier(s). Avoiding the need for continued active maintenance, after site closure, is also desired.

Shallow land burial (SLB) is a landfill, from the surface to 30 m deep, with elaborate soil cover systems (IAEA 2009a). Waste disposal trenches are excavated and lined with gravel and plastic liners, and waste containers are placed inside the trench by stacking them in an orderly fashion. Once the trenches are filled, the wastes are covered by the backfill, compacted, and then enclosed by a cover system with layers of soil. Nearly all LILW in the U.S. has been disposed of through shallow land burial. Since 1995 the Barnwell, South Carolina disposal facility has used modular concrete canisters to place waste packages for enhanced waste isolation. Modular concrete canisters are also used in the more recently established (2012) Andrews, Texas site.

Intermediate-depth disposal is similar to shallow land burial. However, the disposal trenches are deeper and the cover system is thicker.

The above-ground vault (AGV) uses a concrete structure on the site as an above ground structure located permanently on the surface. Below-ground vault (BGV) is also a concrete structure but below ground. It is built in a trench. Once the facility is full, it is buried underground covered with a layer of clay and a concrete roof. These above and below ground vaults are backfilled with either gravel (Spain) or concrete or gravel (France) (SKB 2011). Earth-mounded concrete bunker (EMCB) is a trench lined with concrete as above ground structures. Therefore EMCB is similar to AGV but includes with modular concrete canisters as part of the system. Once the facility is full, it is covered with an earthen cover. With the modular concrete canister design, individual waste containers are placed in these concrete canisters, on a pad as above ground structures. International experiences show that EMCB is most widely adopted in a number of countries for LILW disposal. The sites using EMCB include the La Manche site in France, the L'Aube site in France, and the Drigg site in the U.K. The LLW disposal facility in Andrews, Texas, also uses modular concrete canisters but as a below grade facility.

Other different approaches used include shaft disposal or mined cavity concept. Shaft disposal, sometimes called the augered hole or borehole disposal, places LILW containers within boreholes. The mined cavity concept uses old near-surface underground mines for LILW disposal. Examples include Sweden's intermediate depth rock cavity disposal at Forsmark, Germany's deep mine disposal at the Konrad facility, the Richard and Bratrství repository in Czech Republic, and the Olkilluoto and Loviisa facilities in Finland.

In comparison to HLW disposal, less stringent requirement on waste isolation is imposed on LILW disposal. The required time period of waste isolation and institutional control is also comparatively shorter. This is because the activity and longevity of radionuclides contained in LILW is much less than those in HLW. The LILW disposal requirements in the U.S., the 10CFR61 requirements, ask for the demonstration of relevant safety performance for a period of 1000 year after the closure of a site. However, the performance assessment period may be increased to 10,000 post-closure years to assure protection of the public.

Construction of a typical LILW disposal facility requires consideration of the cover system design, the site drainage system design, the performance of concrete institutional barrier, and stability of waste forms and packages. In the disposal area, any void spaces must be minimized and backfilled once the disposal capacity, i.e., the limit on the site's maximum activity content, is reached. Surface cover dose rates should be below the unrestricted area limits (i.e., 5 mSv/year (500 mrem/year) and 0.02 mSv/year (2 mrem/h)). Boundaries and positions of the disposal units must be located and mapped by land survey and permanent trench markers must be set up.

From an engineering perspective, the life cycle of a typical near-surface disposal facility can be envisioned as: site selection and characterization (1–2 years), preoperational licensing (1–2 years), construction and active disposal operations (20–40 years), site closure and stabilization (1–2 years), and institutional control (100 years in the U.S., $\leq$ 300 years in France and Spain). Though technically these analyses and events (envision facility to active disposal operations) can be conducted in the time frames provided, there are often public acceptance issues that may significantly delay the actual development of the site.

13.7.2 Performance Assessment of Low and Intermediate Level Waste Disposal Facility

In conducting performance assessment for LILW disposal facilities, the principles and the methodology are basically the same as those discussed for a HLW geological

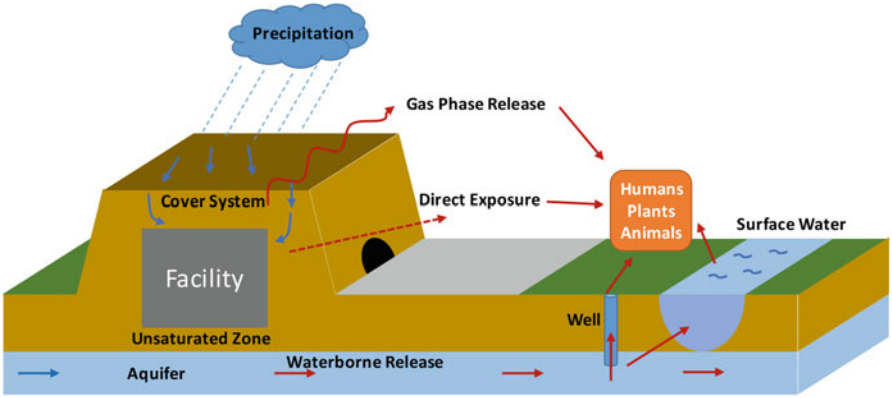


Fig. 13.8 Depiction of a hypothetical facility and the key areas for performance assessment in LILW

repository. The main objectives of a LILW performance assessment are to determine whether reasonable assurance of compliance with quantitative performance objectives can be demonstrated and to identify waste acceptance criteria related to quantities of wastes for disposal. A hypothetical depiction of a facility and the key areas for performance assessment are shown in Fig. 13.8.

The models involved in LILW facility performance assessment are similar to those of a HLW repository performance assessment. The models include source-term/near-field models, radionuclide transport models, and exposure/dose models. Source-term/near-field models track the degradation of the facility, waste contained therein, and the release of radionuclides into the surrounding subsoil. The radionuclide transport models track the movement of radionuclides from the subsoil to potential human exposure sites. The exposure/dose models track the uptake, exposure and dose equivalent that may result from exposure to any transported radionuclides.

While the methodology employed in these models are similar to those used in HLW repository performance assessment, the modeling for radionuclide release from waste needs to consider LILW specific waste forms. As currently practiced, LILWs are disposed in a wide variety of materials and forms, including metals, resins, filters, mixed trash, cement, or sorbent media. Modeling may also need to consider the use of soil or cement backfills in the waste packages. Also, volatilization of wastes through biodegradation of organic materials or hydrogen production from radiolysis needs to be examined along with the analysis of the impact of gaseous release of radionuclides such as ^{14}C and ^{3}H.

13.7.3 Cost of Low and Intermediate Level Waste Disposal

A number of countries have built and operated LILW disposal facilities. From these experiences, the cost of developing a facility can be estimated. However, as the facilities have to meet widely different domestic requirements regarding waste definition and acceptable waste types, generalizing the cost of disposal is difficult. In general, the undiscounted cost ranges from 0.02 to 0.17 US mills per kWh (NEA 1990). This cost can be considered a small part of the total cost of electricity generation from nuclear power. The cost associated with planning and licensing is found to be a significant portion of the total cost. The economy of scale was also important for the construction of near-surface disposal facility. The construction cost of near-surface facilities is in general lower than that of mined cavity facilities.

If multiple facilities are built adjacent to each other, the construction cost is lower due to cost sharing. Non-technical considerations such as socio-political factors, regulatory approaches, and taxes and insurance are also found very important along with the time value of money issue.

In terms of a LILW generator, the cost of LILW disposal is the payment to be made to the disposal facility operator. In the case of the U.S., such disposal cost has been on the rise since the commercial operation of LILW disposal facilities. In contrast, the volume of LILW disposed of at commercial facilities steadily decreased as part of the disposal cost reduction effort by the generators (see Fig. 13.7).

Table 13.13 provides a comparison of U.S. NPP LILW disposal cost at the four active disposal facilities in based on the data in 2018, 2015, 2012, 2010, and 2008. These cost data are reported in NUREG 1307 as the payment for sending the waste to each facility. The availability of data from the Texas and Utah facilities depends upon the operating history. Envirocare's Clive facility start accepting class A waste in 2001 and Andrews, Texas facility in 2012.

To understand how these fees translate into disposal costs, the differences in the charges used at each facilities need to be understood. For example, sending the waste to Utah facility requires payment of only the volume based charge along with non-compact tax. In the case of the Texas facility, surcharges are added depending on the curie content, ^{14}C activity, and the weight of waste. At the South Carolina facility, besides the base weight fee, surcharges are added through dose multiplier, millicurie surcharge, and ACC administration surcharge. At Richland, the fees include basic disposal fee, shipment charge, container charge, engineered concrete barrier charge, perpetual care charge, site availability charge, and business tax commissioner fee. Then surcharges are added per dose rate.

Table 13.13 Comparison of LLW disposal costs at U.S. disposal facilities (2008–2018) (based on NRC 2012 Appendix A)

NUREG-1307	Rev 17	Rev 16	Rev 15	Rev 14	Rev 13
Disposal of LLW at Texas facility[a.]					
Waste type	2018 charge ($/ft^3)	2015 charge ($/ft^3)	2012 No fees	2010 No fees	2008 No fees
Class A LLW – routine	$100	$100	–	–	–
Class A LLW – shielded	$180	$180	–	–	–
Class B and C LLW	$1000	$1000	–	–	–
Sources	$500	$500	–	–	–
Biological waste (untreated)	$350	$350	–	–	–
Disposal of class A LLW in Clive Utah facility					
	2018 charge ($/ft^3)	2016 charge ($/ft^3)	2012 Charge ($/ft^3)	2010 charge ($/ft^3)	2008 charge ($/ft^3)
Large components	$398	$379	$350	$300	–
Debris	$165	$157	$145	$125	–
Oversize debris	$188	$179	$165	$145	–
Resin/filters	$523	$498	$460	$400	–
Combustibles	$653	$622	$575	$500	–
Evaporator bottoms, $/gallon	$27	$25	$14	$12	–
Disposal rates for LLW at South Carolina Atlantic compact waste[b]					
	2018 charge ($/pound)	2016 charge ($/pound)	2012 Charge ($/pound)	2010 charge ($/pound)	2008 charge ($/pound)
Density range > 120lbs/ft^3	$8.169	$7.589	$7.516	$6.702	$6.191
Density range 45–60lbs/ft^3	$14.298	$13.283	$13.155	$11.730	$10.836
Dose multiplier based on container dose level 0–200 mR/h multiplier on weight rate	1.00	1.00	1.00	1.00	1.00
Container dose level 200 mR/h–1R/h multiplier on weight rate	1.08	1.08	1.08	1.08	1.08
Container dose level 10–25 R/h–multiplier on weight rate	1.37	1.37	1.37	1.37	1.37
Millicurie surcharge $/millicurie	$0.612	$0.569	$ 0.563	$0.502	$0.464
Max millicurie charge (4000,000) $/shipment	$244,843	$228,451	$225,083	$200,702	$185,600
Irradiated hardware $/shipment	$92,845	$86,253	$85,422	$76,169	$70,364
Disposal rates for LLW at Richland Washington facility[c]					
	Rev 17	Rev 16	Rev 15	Rev 14	Rev 13
Site Avaiability charge Annual charge per generator	2018 charge	2016 charge	2012 charge	2010 charge	2008 charge
20–40 ft^3 or 100–200 mR/h.	$2111	$1986	$1773	$1668	$1562
160–320 ft^3 or 800–3200 mR/h	$14,931	$14,045	$12,539	$11,801	$11,050

(continued)

Table 13.13 (continued)

NUREG-1307	Rev 17	Rev 16	Rev 15	Rev 14	Rev 13
Disposal of LLW at Texas facility[a.]					
1280–2560 ft^3 or 6400–12,800 mR/h	$105,673	$99,399	$88,743	$83,515	$78,200
Disposal rates					
Volume: $/$ft^3$	$152.20	$114.00	$115.50	$127.10	$98.70
Shipment: $/manifested shipment	$14,650	$13,510	$13,750	$13,370	$14,740
Container: Container/manifest	$10,320	$7790	$7560	$8960	$7080
Dose rate at container surface					
Less than or equal to 200 mR/h	$40	$24	$92	$17	$177
>200 mR/h < 1000 mR/h	$2844	$1706	$6540	$1209	$12,580
>1000 mR/h < 10,000 mR/h	$11,310	$6750	$26,200	$4850	$50,400

Notes:

[a] Disposal at Texas Facility
(1) Curie Inventory Charge $0.55 per MCi
(2) Max Curie Charge per shipment - $220,000 per shipment
(3) Carbon-14 Inventory – $1.00 per mCi
(4) Special Nuclear Material Charge - $100 per gram

[b] Disposal at South Carolina Facility
(1) Access to Atlantic Compact Regional Waste
(2) Dose multiplier based on weight
(3) Millicurie surcharge

[c] Disposal at Richland Washington Facility
(1) Access to facility limited, rates are site availability charge
(2) mR/hr per hour at container surface is the sum of all containers received during the year

R refers to roentgen, a legacy unit to measure radiation exposure. 1 R is equivalent to ionization of air producing $2.58 \times 10^{(-4)}$ coulombs of charge per unit mass (1 kg) of air.

13.8 Mixed Waste

Mixed waste refers to both radioactive and chemically toxic waste. More specifically, mixed waste refers to LILW that is also contaminated with chemically toxic material. The liquid HLW waste from spent fuel reprocessing as a mixture of nitric acid and fission products and actinides is by nature a mixed waste. However, as the main hazard of HLW is from radionuclides, it is handled under the rules of HLW and excluded in the discussions of mixed waste. Mixed wastes are generated from NPP operations and decommissioning. In terms of volume, mixed waste represents a very small portion, about 1% of total LILW generated from NPPs.

Common examples of mixed wastes generated from NPP operation (IAEA 2002) include radioactively contaminated spent solvents, discarded lead shielding, cadmium (control rod), beryllium (neutron source), and scintillation cocktails. From NPP decommissioning, radioactively contaminated asbestos and PCB are important mixed waste along with decontamination chemicals/sludge, radioactive scrap metals, and contaminated concrete debris. The main source of asbestos is piping insulation representing the largest volume of mixed waste from NPPs. Mixed waste PCBs are found in paints and rubbers used in NPPs.

Due to the dual nature of contamination, mixed wastes are to meet government regulations for both radioactive waste and hazardous waste. For example in the U.S. mixed waste are under NRC's 10CFR Part 61 as well as EPA's 40 CFR 261 (Subpart C or D). 40 CFR Part 261 contains the requirements under the RCRA (Resource Conservation and Recovery Act). But if mixed waste is generated and managed under a single NRC or Agreement State License, then the waste is conditionally exempt from RCRA requirements during storage and treatment. Mixed waste can also be exempt from RCRA requirements for transport and disposal if the waste container meet specified criteria as defined by the NRC. Disposal of mixed waste must meet the acceptance criteria of low level radioactive waste disposal facility under 10CFR Part 61 as well the RCRA requirements of 40 CFR 264 and 40 CFR 270. The mixed waste container must thoroughly describe the hazardous component of the waste, not just the radioactive component.

A desirable way of treating mixed waste is to separate radioactive and hazardous components. For example, after decay-in-storage, when the waste is not considered radioactive, the mixed waste becomes hazardous waste. Then the waste is handled under the rules for hazardous waste. If such separation is not possible, then the waste is either incinerated for the removal of toxic components or solidified/immobilized for disposal.

For example, beryllium, after intermediate storage to allow the decay of most of ^{60}Co, is encapsulated with cement. Asbestos waste is first supercompacted and encapsulated in a cementitious matrix or through vitrification. Cadmium waste is disposed of mainly through encapsulation in cement. Mercury is commonly encapsulated through amalgamation if distillation is not applicable. Contaminated oil or solvents are treated with incineration. PCBs are incinerated or treated with other high temperature processing methods or solidified in cement. In the case of lead, if lead cannot be decontaminated for recovery and reuse, it is encapsulated by using cement. After these encapsulation operations, the mixed waste can be disposed of under the rules for LILW.

13.9 Conclusion

This chapter provided an overview of the approaches used in the management of LILW. Although low in radioactivity contents in comparison to HLW, LILW with widespread presence of its generators in various sectors of society, demands foresight and careful planning and coordination for its safe management and disposal. An interesting observation made in this chapter is that generation, classification, treatment/stabilization, and disposal of LILW are closely related to each other, depending on the availability of technologies or the regulatory approaches taken. For example, while there are similarities in waste streams and to some degree the processes used to manage the waste, the volumes of waste disposed can vary significantly based on the waste disposal stabilization approaches required by the regulations. This is because the use of waste form, waste loading per package, and package type are dependent upon the disposal facility's waste acceptance criteria. Comparison of the practices between the U.S. and European countries clearly shows this point. US regulations are less prescribed than those of most European countries. In the US, if the privately or state owned and operated disposal facility can meet the intent of the law they have the flexibility to use the approach that best matches their site performance requirements. As such, in the US, wastes are rarely solidified, but instead are placed in concrete overpacks or modular concrete containers. In this situation, the processing of waste depends on economic viability and availability of disposal space. However, countries such as Spain and France, the majority of their wastes are stabilized, and in some cases stabilized and placed in concrete containers to meet government or quasi government operated disposal facility requirements. Regardless of the how waste site stability requirements are achieved, these waste acceptance criteria have evolved over time taking into account the lessons learned of earlier disposal facility experiences, nationally and internationally. Finally, in most countries, as public acceptance of these facilities is still a contentious issue, utility power companies and regulators both see the benefit of reducing waste volumes from generation through processing (which does not minimized activity levels) and utilizing available disposal capacity responsibly and efficiently.

Homework

Problem 13.1: Discuss the pros and cons of current U.S. nuclear waste classification scheme. If we ought to make any changes for improvement, what would be your suggested changes?

Problem 13.2: Based on the US LLW classification scheme, determine the class of the LILW with the following mix of nuclide concentrations.

(a) ^{63}Ni at 15 Ci/m^3 & ^{90}Sr at 50 Ci/m^3
(b) ^{63}Ni at 30 Ci/m^3 & ^{90}Sr at 100 Ci/m^3

Problem 13.3: In a nuclear power plant, 5 kg of wastes with a total activity of 2 μCi was accumulated of which 1 μCi was from ^{241}Am. The rest of the activity is due to isotopes with $Z < 92$. What is the class of this waste according to the US LLW classification scheme?

Problem 13.4: The steady state (saturation) concentration of the fission production ^{131}I ($t_{1/2} = 8.07$ day) in PWR fuel during irradiation is 0.023 Ci/watt of thermal power. During normal operation, 0.1% of the fuel rods in a PWR core fail each year. Assume that the entire inventory of iodine in each failed fuel rod is discharged into the primary reactor coolant. It has been proposed to install an anion exchange column to remove the iodine from the primary coolant. The column would process a side-stream of 5% of the total primary coolant flow and would achieve a decontamination factor of 20 (i.e., 95% of the iodine entering the column would be sorbed on the ion exchange resin).

Calculate the amount by which the steady-state iodine concentration in the primary coolant would be reduced if the column were installed. Base your calculation on a 1000 MWe PWR. Use the following reactor parameters: Thermal efficiency = 0.33; Primary coolant volume = 375 m^3; Primary coolant circulation time = 12 s.

Problem 13.5: A 55 gallon (200 liter) drum containing ion-exchange resins is labeled LLW and is found to have a total of 100 Bq/g activity concentration emitting 1 MeV gamma-rays. Determine the dose rate at the measurement location at 50 cm from the surface of the container. Assume a resin density of 0.8 g/cm^3, mass absorption coefficient of air for 1 MeV gamma-ray is 0.0280 cm^2/g, mass attenuation coefficient of air for 1 MeV gamma-ray is 0.0636, and density of air is 0.001225 g/cm^3.

Problem 13.6: By using the information given in Table 13.13, determine the total fee to pay at each of the U.S. LLW disposal facilities for the disposal of the following LILW. (Use \$6/ft^3 for Atlantic Coast Compact administration charge at the South Carolina facility)

(a) Ten of 20 ft. Sea land container (B-25 Steel Boxes, 92.86 ft^3 each)

Dose rate on container: 210 mR/hr. on each box
Total activity: 1000 mCi
^{14}C activity: 1 mCi
Density of waste: 50 lbs./ft^3
Total weight of S/L: 48,000 lbs.

(b) One High Integrity Container (HIC) in Type A Cask (120 ft^3)

Dose rate: 5 R/hr. on HIC contact, 100 mR/hr. on Type A cask contact
Total activity: 10 Ci
14C activity: 100 mCi
Waste weight: 10,000 lbs.
Density of waste: 84 lb./ft^3

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Chapter 14
Decommissioning a Nuclear Power Plant

Abstract Decommissioning is the process of taking a nuclear power plant out of operation and shutting it down permanently. Therefore, it is a key component of environmental stewardship in the use of nuclear energy. Major options of decommissioning include DECON (decommissioning through immediate dismantlement), SAFSTOR (delayed dismantlement and decommissioning) as well as the choice between unrestricted and restricted site release. This chapter describes the overall steps and activities of decommissioning toward site release and the factors affecting decommissioning decisions.

Keywords Decommissioning options · Key radionuclides · Survey and sampling · Site release determination · Decommissioning decision factors

Decommissioning is the final step in the life cycle of a nuclear power plant. It is the process of taking a plant out of operation and shutting it down permanently. Therefore, it is a key component of environmental stewardship in the use of nuclear energy. As the word decommission means “to remove from service” (Meriam Webster Dictionary), normally decommissioning takes place at the end of the service life of a nuclear power plant. Decommissioning also occurs before the expected service life of the plant under special circumstances. Such circumstances may arise due to accidents or an undue financial burden. If an accident at a plant caused unrecoverable damage to the reactor system, the plant goes through decommissioning. Even if the damages can be repaired but the necessary repairs are judged infeasible due to economic, safety or technical reasons, the plant is decommissioned. A major cost escalation due to back fitting requirements under new regulations or from poor plant operational performance may force the shutdown of the plant. Technological obsolescence due to new technical developments may also cause the plant to go through decommissioning.

The purpose of this chapter is to explain the process of decommissioning and to provide an overview of key factors and activities in the decommissioning process. Key elements controlling these processes include regulations, safety (facility personnel, contractors, and the public), radiological dose (collective and individual),

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time to complete tasks, risk levels (radiological and industrial hazards), and cost and resource control. Establishing an integrated management system for safe and efficient decommissioning, including training and human performance improvement, is also an important part of decommissioning. To the extent possible, insights into each of these areas are addressed, directly or indirectly, as appropriate.

14.1 Options for Decommissioning of a Nuclear Power Plant

Decommissioning of a nuclear power plant requires the safe removal of a facility, and reducing radioactive contamination to a level that allows termination of the operating license. The degree of contamination reduction at the site is determined based on the end use of the site. The end use determines the degree to which the site is accessible by the public.

The first step in every decommissioning project, regardless of the decommissioning process selected, is the removal of spent fuel from the reactor core which contains the vast majority of radioactivity in the power plant. The fuel is transferred to the spent fuel pool where it will remain for several years before removal.

Once the fuel is removed, more than 95% of the remaining activity is in the reactor vessel and its internals. This includes the reactor coolant water system, and the related components, which contain the largest concentration and mobile source of radioactivity in a nuclear plant. Therefore, the next step in decommissioning is to remove this activity. The selected timeline (e.g., immediately or in 50 years) for removing these components and other contaminated structures will determine which decommissioning option will be used. The available options include immediate decontamination and dismantling (sometimes called DECON), deferred dismantlement called safe storage (abbreviated as SAFSTOR), and encapsulating the radioactivity in the structure – entombment.

Under the DECON scenario, all fuel assemblies, radioactive fluids and waste, and other materials having activities above the accepted unrestricted activity levels are removed from the site, followed by dismantling the buildings and equipment to permit regulatory control removal. Resulting radioactive wastes are transported and disposed of at appropriate disposal facilities. Thus dismantlement could take place immediately after cessation of operation. The end state of dismantlement is most likely an unrestricted release of the site (although some restrictions may apply), depending upon the nature and conditions of the site.

The procedures followed in dismantlement include the following

- Reactor shutdown
- Fuel removal
- Cooling water system, water purification, removal of spent fuel and reactor coolant, resins, and sludges

- Dismantling of reactor pressure vessel (RPV)
- Dismantling of primary circuit components
- Dismantling of biological shield
- Dismantling of other radioactive equipment
- Remove steam generator(s) (whole or dismantled)
- Decontamination
- Conditioning, packing, and transportation of wastes
- Dismantling of buildings
- Removal of contamination on nuclear plant walls
- Demolition of a reactor containment building
- Soil remediation

The safe storage option (SAFSTOR) refers to “putting the facility in a state of protective storage” as a deferred dismantlement option. Under this option, after removing all spent fuel and reactor coolant and existing radioactive waste from the site, the plant is placed in a safe stable condition. Thus, the plant is in storage without the removal of the reactor vessel and plant structures, but with plant security in place. During the storage period (perhaps ranging between 30 and 60 years), the inventory of radioactive materials is reduced through radioactive decay. Accordingly, the burden of decontamination and worker exposure control is reduced. During the storage period, radiation monitoring, environmental surveillance and appropriate security procedures are established under a possession-only license. Decontamination and dismantlement of the reactor vessel and the rest of the plant systems removal then follow.

The third option, entombment, requires the plant to be securely encased in a concrete structure to prevent access and be maintained with continued surveillance until the radionuclides decay to a level that permits elimination of regulatory controls. Entombment is exercised after having all spent fuel, reactor coolants and existing radioactive wastes, and certain selected components shipped offsite. All the remaining highly radioactive or contaminated components (e.g., reactor vessel and reactor internals) are sealed within a biological shield, typically with concrete and steel reinforcements. The structure should provide integrity over a period of radioactive decay, where the radioactive materials reach unrestricted release levels. An appropriate surveillance program should be established and continued but at a reduced level than what’s required for SAFSTOR. However, due to the uncertainties about the regulatory viability of the option and the concern of potential release of radioactive materials, entombment has been unattractive or unused. To date, there have been five entombment cases: St Lucens in Switzerland; Chernobyl 4 in Ukraine; BONUE, Hallam and Piqua in the USA. Except the Chernobyl unit which occurred in 1986, these entombment cases occurred in the 1960s. In modern nuclear power plant decommissioning, the option has not been exercised and not encouraged by the IAEA. In the U.S., there is a requirement of completing decommissioning within 60 years of permanent cessation of plant operation (10 CFR 50.82(a)(3)) (NRC 2007). Entombment can be approved only if shown necessary to protect public health and safety.

As the goal of nuclear decommissioning is to terminate the operating license of a nuclear reactor while ensuring public safety after license termination, decommissioning must include a plan for the end-state of the site. If the end state is unrestricted release for public use, it is called “greenfield”. If there is any restriction, it is called “brownfield”. The brownfield option could include some sort of site reuse with restrictions. For example, the site can be used as a parking lot, museum, or an industrial facility. The site could even be used for the operation of a conventional power plant or reused for a new nuclear power plant. Even in the absence of any reuse, the brownfield option implies the presence of institutional control for the foreseeable future after decommissioning. Under such a scenario, building foundations and other underground structures or piping may be left at the site if the residual contamination levels deem very low.

In the case of DECON or SAFSTOR, the end state of the site is either “greenfield” or “brownfield”. With entombment, the likely end state is a de facto waste disposal facility.

14.2 Radionuclides of Concern in Decommissioning

Characterizing the presence of radionuclides provides the basis for decommissioning through identification of contamination, assessment of potential risks, and developing plans for the removal of radioactivity and worker radiation protection. Typical radionuclides present in the structure of nuclear reactor components or plant building are listed in Table 14.1 capturing the products of activation of base materials such as carbon steel, stainless steel, and concrete. As decommissioning of nuclear power plants takes place within relatively short time period of about 100 years after the shutdown of a reactor, mostly the shorter-lived radionuclides are of concern as a source of radiation exposure to workers.

The most important nuclide in decommissioning is ^{60}Co, a strong gamma emitter. On average, ^{60}Co is known to be responsible for over 80% of the dose to workers during the operation of the plant. Cobalt-60 remains dominant in controlling the radiation levels for up to 30 years. Along with ^{60}Co, ^{58}Co is also an important nuclide at reactor shutdown (and during the first few years) as key dose contributor. With the half-life of 100 years, ^{63}Ni, a beta (66 keV) emitter, takes an important place in the plant’s radioactivity content for up to 700 years. Its contribution to dose is through surface activity as ^{63}Ni is self-shielded in metals and concrete. It is relatively easy to shield against ^{63}Ni. Beyond hundreds of years, ^{59}Ni and ^{94}Nb each with a half-life of 8×10^4 years and 2×10^4 years respectively, become an important source of radiation dose in the plant components. Furthermore, depending on plant operation (e.g., fuel failure history), there could be significant fission product contamination, especially in areas such as the reactor cavity, spent fuel pool and any soils that have had leakage from fuel pool. If the plant had been well operated and maintained without incidents of major spill, contamination by fission products will be a minor issue.

If decontamination and decommissioning take place soon after the plant shutdown, ^{60}Co will dominate the worker dose during the decommissioning operations.

Table 14.1 Typical radionuclides of concern in neutron-activated structural materials

Nuclide	Half-life (year)	Means of production	Emission	Energy (MeV)	Base material
^{14}C	5730.0	^{14}N(n,p)	β^-	0.156	Carbon steel, stainless steel, concrete
^{49}V	0.906	^{52}Cr(p,α)	γ, β^-	0.6[a]	Carbon steel, stainless steel
^{54}Mn	0.856	^{56}Fe(d,α)	γ	0.835	Carbon steel, stainless steel, concrete, aluminum
^{55}Fe	2.6	^{54}Fe(n,γ)	γ	0.23[a]	Carbon steel, stainless steel, concrete, aluminum
^{59}Ni	8×10^4	^{58}Ni(n,γ)	ε	1.06[a]	Carbon steel, stainless steel, concrete
^{63}Ni	100.0	^{62}Ni(n,γ)	β^-	0.066	Carbon steel, stainless steel, concrete
^{65}Zn	0.667	^{64}Zn(n,γ)	γ, ε, β^+	1.115, 1.352, 0.325	Carbon steel, stainless steel, concrete, aluminum
^{58}Co	0.194	^{55}Mn(α,n)	β^+, γ	0.474, 0.810	Carbon steel, stainless steel, concrete
^{60}Co	5.263	^{59}Co(n,γ)	β^-, γ, γ	0.314, 1.17, 1.33	Carbon steel, stainless steel, concrete, aluminum
^{93}Mo	3.5×10^3	^{92}Mo(n,γ)	γ (EC)	Nb *x*-rays	Carbon steel, stainless steel, concrete
^{94}Nb	2×10^4	^{93}Nb(n,γ)	β^-, γ, γ	0.49, 0.702, 0.871	Carbon steel, stainless steel, concrete
^{95}Nb	0.096	^{95}Zr decay	β^-, γ	0.16, 0.765	Carbon steel, stainless steel, concrete
^{95}Zr	0.175	^{94}Zr(n,γ)	β^-, γ, γ	0.396, 0.724, 0.756	Carbon steel, stainless steel
^{35}S	0.238	^{34}S(n,γ)	β^-	0.167	Concrete
^{36}Cl	3.01×10^5	^{35}Cl(n,γ)	β^-, ε,	0.714, 1.018[a]	Concrete
^{37}Ar	0.0953	^{36}Ar(n,γ)	ε	0.81[a]	Concrete
^{39}Ar	269.0	^{38}Ar(n,γ)	β^-	0.565	Concrete
^{40}K	1.28×10^9		β^-, γ	1.314, 1.46	Concrete
^{41}Ca	8×10^4	^{40}Ca(n,γ)	γ (EC)	K *x*-rays	Concrete
^{45}Ca	0.446	^{44}Ca(n,γ)	β^-	0.257	Concrete
^{46}Sc	0.229	^{45}Sc(n,γ)	β^-, β^-, γ, γ	1.48, 0.357, 0.889, 1.12	Concrete, aluminum
^{59}Fe	0.122	^{58}Fe(n,γ)	β^-, γ, γ	1.57, 1.1, 1.29	Concrete
^{54}Fe	0.122	^{52}Fe(n,γ)	β^-, γ, γ	1.57, 1.1, 1.29	Aluminum
^{110m}Ag	0.69	^{109}Ag(n,γ)	β^-, γ[b]	0.087, 0.6577	Aluminum
^{152}Eu	13.48	^{151}Eu(n,γ)	β^-, γ	1.477, 1.408	Bioshield concrete, core graphite
^{154}Eu	8.59	^{153}Eu(n,γ)	β^-, γ	1.968, 1.274	Bioshield concrete, core graphite

Source: Moghissi et al. (1986, p. 499)

[a] Continuous spectrum of x-ray energies below this number, due to Bremsstrahlung

[b] Energy of most probable energy β- and most probable energy γ given

However, since ^{60}Co has a half-life of 5.2 years, waiting for about 40 years will have the benefit of most of the ^{60}Co having decayed away. This is the main rationale for the SAFSTOR option. Thus, the choice between immediate or delayed decommissioning involves tradeoffs between costs of maintaining the facility in a storage status versus the increased costs of handling the higher radiation levels associated with earlier decommissioning. In the case of the entombment option, the presence of long-lived radionuclides is a major consideration. If significant inventory of ^{59}Ni and ^{94}Nb inventory exist in a reactor facility resulting from a long operating history, the required time to achieve unrestricted release would be too long, making this option unrealistic.

Table 14.2 show the time-dependent changes in radioactivity inventory of key contributing radionuclides in PWR fuel assembly. Although the materials of the assembly, Zircaloy and Inconel, do not represent all materials of reactor equipment, this data may still be useful to show the trend of radioactivity inventory changes of in-reactor metallic components during the period of decommissioning.

In the case of reactor pressure vessel, contributions from principal radionuclides to the activity are represented in Fig. 14.1. The figure is based on the results from Trino BWR in Italy (after 26 years of operation at 260 MWe) (IAEA 1998). The figure indicates that the activity of ^{60}Co is the largest (^{55}Fe as the second) at the time of reactor shutdown but, after about 20 years, the activity of ^{63}Ni becomes the dominant one. The results show similar trends of key radionuclide activity with the data in Table 14.2.

If we project the dose from these radionuclides to workers at the immediate vicinity of nuclear reactor component for a typical PWR, the expected result as a function of time after reactor shutdown can be shown in Table 14.3 for the key contributing radionuclides. The table indicates that initially after reactor shutdown, the dose is dominated by ^{60}C. Therefore, if immediate decontamination and dismantling is exercised, ^{60}Co is the primary source of worker exposure to radiation. This trend changes after 50 years with ^{63}Ni becoming the dominant nuclide. If the plant is decommissioned through SAFSTOR, ^{63}Ni will be the primary dose contributor to workers. If the plant structure remains intact through the entombment option, ^{59}Ni may emerge as an important source of radiation exposure at later years during long term storage or disposal.

Along with these activation products, contamination of soils at the plant site by ^{137}Cs and ^{90}Sr, the fission products with the half-life of about 30 years, is often observed.

14.3 Steps in Nuclear Power Plant Decommissioning

Steps in actual plant decommissioning include (1) a transition phase, (2) characterization and survey, (3) segmentation and dismantling, (4) decontamination and remediation, (5) materials and waste management, (6) final site characterization, and (7) environmental monitoring (NEA 2014).

Table 14.2 Inventory (Ci) of radionuclides in PWR assembly as a function of time since reactor discharge (Alexander et al. 1977)

	At discharge	1 year	2 year	10 year	30 year	100 year
^{55}Fe	2.572×10^{3}	1.970×10^{3}	1.509×10^{3}	1.788×10^{2}	8.644×10^{-1}	6.793×10^{-9}
^{58}Co	3.157×10^{3}	9.062×10^{1}	2.601×10^{0}	1.198×10^{-12}	0.0	0.0
^{60}Co	3.629×10^{3}	3.181×10^{3}	2.788×10^{3}	9.716×10^{2}	6.964×10^{1}	6.867×10^{-3}
^{59}Ni	2.118	2.118	2.118	2.117	2.117	2.116
^{63}Ni	3.061×10^{2}	3.038×10^{2}	3.015×10^{2}	2.839×10^{2}	2.442×10^{2}	1.441×10^{2}
^{95}Zr	2.142×10^{4}	4.489×10^{2}	9.406×10^{0}	3.498×10^{-13}	0.0	0.0
^{95}Nb	2.190×10^{4}	9.494×10^{2}	2.085×10^{1}	7.543×10^{-13}	0.0	0.0
^{119m}Sn	3.082×10^{3}	1.097×10^{3}	3.902×10^{2}	1.005×10^{-1}	1.066×10^{-10}	0.0

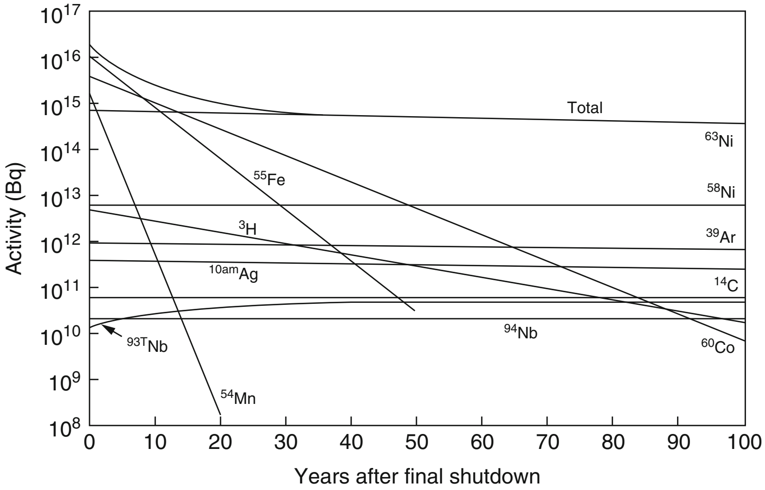

Fig. 14.1 Activity of principal radionuclides of the reactor pressure vessel. (From Trino BWR) (IAEA 1998)

14.3.1 Transition Phase

Before taking on decommissioning of a nuclear power plant, the plant goes through a transition phase at the end of reactor operation. During the transition phase a strategy and plan for decommissioning are developed including the identification of options for spent fuel and waste management. The transition phase continues until the completion of planning and the planned implementation begins.

During the transition period, spent fuel is removed from the reactor and spent fuel pool, the reactor coolant water is purified and removed along with demineralizer resins and filter sludges to reduce the holdup of radioactive material. The cost of decommissioning is also estimated and the necessary funding is secured.

14.3.2 Characterization and Survey

Understanding the problem to be solved is one of the key steps in nuclear decommissioning. Understanding the problem means the understanding of the physical, radiological and non-radiological characteristics of the plant and the site to be decommissioned. Such understanding comes from site characterization. Site characterization is done at several different stages including pre-dismantlement characterization as well as during and post dismantlement characterization and final status survey (MARSSIM 2000).

Table 14.3 Estimated contact dose as a function of years after shutdown in a PWR (unit: R/h)

		Co-60		Ni-63	Ni-59	Fe-55	Total	
Time since shutdown	Location	γ	β	β	γ/x-ray	γ/x-ray	γ/x-ray	β
2 years	Core shroud	3.6E+05	9.1E+04	2.3E+03	3.2E-02	2.7E+01	3.6E+05	9.3E+04
	Core barrel	1.1E+05	1.5E+04	3.8E+02	5.2E-03	4.4E+00	1.1E+05	1.5E+04
	Upper core plate	3.2E+04	3.4E+03	8.6E+01	1.2E-03	1.0E+00	3.2E+04	3.5E+03
	Lower core plate	6.5E+04	6.8E+03	1.7E+02	2.4E-03	2.0E+00	6.5E+04	7.0E+03
	Vessel cladding	3.8E+01	7.7E+02	2.0E+01	2.8E-04	2.3E-04	3.8E+01	7.9E+02
50 years	Core shroud	6.5E+02	1.6E+02	1.6E+03	3.2E-02	2.5E-05	6.5E+02	1.8E+03
	Core barrel	2.0E+02	2.7E+01	2.3E+02	5.2E-03	4.2E-06	2.0E+02	2.6E+02
	Upper core plate	5.9E+01	6.1E+00	5.9E+01	1.2E-03	9.5E-07	5.9E+01	6.5E+01
	Lower core plate	1.2E+04	1.2E+01	1.2E+02	2.4E-03	7.9E-06	1.2E+04	1.3E+02
	Vessel cladding	7.0E-02	1.4E+00	1.4E+01	2.8E-04	2.2E-07	7.0E-02	1.5E+01
100 years	Core shroud	9.0E-01	2.3E-01	1.1E+03	3.2E-02	–	9.3E-01	1.1E+03
	Core barrel	2.8E-01	3.8E-02	1.6E+02	5.2E-03	–	2.9E-01	1.6E+02
	Upper core plate	8.1E-02	8.5E-03	4.1E+01	1.2E-03	–	8.2E-02	4.1E+01
	Lower core plate	1.6E-01	1.7E-02	8.1E+01	2.4E-03	–	1.6E-01	8.1E+01
	Vessel cladding	9.7E-05	2.0E-03	9.6E+00	2.8E-04	–	3.8E-04	9.6E+00
150 years	Core shroud	1.3E-03	6.4E-05	7.5E+02	3.2E-02	–	3.3E-02	7.5E+02
	Core barrel	3.9E-04	5.2E-05	1.1E+02	5.2E-03	–	5.6E-03	1.1E+02
	Upper core plate	1.1E-04	1.2E-05	2.8E+01	1.2E-03	–	1.3E-03	2.8E+01
	Lower core plate	2.3E-04	2.4E-05	5.6E+01	2.4E-03	–	2.6E-03	5.6E+01
	Vessel cladding	1.4E-07	2.8E-06	6.6E+00	2.8E-04	–	2.8E-04	6.6E+00

14.3.2.1 Types of Surveys/Characterization

Pre-dismantlement characterization identifies the types, quantities and physical properties of systems, structures and components (SSC) of the plant to be removed or demolished. The activities performed at the stage of pre-dismantlement characterization start with a historical site assessment to form a conceptual site model through interviews with staff, review of records, and site visits. A conceptual model describes the expected presence of radioactive materials in terms of type (radionuclides), size, locations, levels and the media with the site diagram and identifies the relevant migration and exposure pathways.

The historical site assessment differentiates impacted areas from non-impacted areas, and provides input to scoping and characterization survey designs. The historical site assessment should identify locations of previous spills, leaks, and it should identify all buildings and areas that are currently used or had been used in the past for radioactive material handling or storage. Locations of potential background concentrations (i.e., uncontaminated) are thus identified.

This is followed by efforts to quantify the levels of hazards and to prioritize the follow-on activities. These efforts include scoping surveys and characterization surveys. A scoping survey is performed to make a preliminary hazard assessment by classifying the site according to the level of expected contamination and evaluating the survey plans to support characterization survey design. A characterization survey is to determine the nature and extent of the contamination and to evaluate the necessary remedial actions and approaches. Depending on the results from the surveys, the projected cost of decontamination may need to be adjusted if there are surprises in the results.

Surveys, assays and sampling are conducted to define and verify the extent and levels of contamination. More detailed and targeted characterization efforts can also be made on high priority or high risk SSCs including computer code execution and analysis of materials properties to develop a detailed distribution of radioactive contamination.

During and after demolition, contamination is characterized through surveys, assays and sampling of the materials removed to verify contamination levels and to ensure the protection of workers, the public and the environment during the decommissioning process. This characterization also supports classification of waste to facilitate onsite management of the waste and to plan for future waste shipments. Surveys are conducted to confirm the removal of impacted materials and to assess the level of residual contamination or need for further remediation. This takes the form of a remedial action support survey which is to support any remediation activities needed and to get the site ready for the final status survey.

All of the surveys performed eventually form the basis for the final site status survey. All of the data collected and the resulting estimates of site-specific parameters are utilized to plan the final status survey to determine the compliance for license termination. The final status survey is to demonstrate that the potential dose or risk from the residual contamination at the site is below the release criterion specified by the regulatory authority. Detailed activities of final status survey are described in Sect. 14.3.6.

To successfully design a survey, the radionuclides of concern must first be identified. Identifying these radionuclides is based on surveys conducted during plant operation, analysis of radiological effluents and radioactive waste from the site, and the literature on similar nuclear installations. The effort to develop derived concentration guideline levels (DCGL) as the goal of site cleanup will follow based on this list of radionuclides. This list will also support the analysis of the samples with the understanding of the minimum detectable activity.

14.3.2.2 Design of Surveys and Sampling

The surveys conducted for a nuclear decommissioning project determine the need for decontamination or whether the license can be terminated or the site can be released for its intended future use. Thus, the levels of radioactive contamination at the site and the plant need to be well characterized.

In nuclear decommissioning site surveys, the site or the plant areas are classified according to the expected level of contamination. This preliminary classification becomes the basis for determining the necessary level of effort in sampling and later remediation. According to the MARSSIM guidelines of the U.S., a site or plant to be decommissioned is divided into four classes of areas: Class 1, Class 2, Class 3, and non-impacted areas (MARSSIM 2000).

A Class 1 designation is applied to an area with the highest potential for radioactive contamination with levels greater than the allowable levels of residual activity. Examples of Class 1 areas include locations where radioactive spills or leaks are known to have occurred, former burial or disposal sites of radioactive materials, or any areas of radioactive waste storage at the site. If no information is available at an area, the area by default is designated as Class 1 requiring extensive investigations through surveys. Class 2 is an area where radioactive contamination is expected but with levels lower than the allowable levels of residual activity. The allowable level of residual activity is called DCGL (derived concentration guideline level). Thus, DCGL becomes the criterion for the release of the site to control the post-release human health risk to acceptable levels. Class 3 is the area potentially impacted by the plant's operation but with a low probability of radioactive contamination. Examples of Class 3 areas include buffer zones around Class 1 or Class 2 areas or areas with very low potential for residual contamination but with no verification. Non-impacted areas are the areas with no potential for residual contamination thus not included in the survey coverage.

Within each class, an area is divided into survey units. A survey unit is a physical area specified at a site or inside a plant where the extent of contamination is related to the exposure pathways under consideration for an individual or groups of individuals. Survey units share a common history or other characteristics during the plant's operation, or are naturally distinguishable from other areas of the site. For example, in the case of indoor areas classified as Class 1, each room could be designated as a survey unit. Thus its entire physical area within the unit becomes the object of a single licensing decision with regards to whether the regulatory release criterion can be met.

In the case of land, the suggested size of survey units is up to 2000 m^2 for Class 1, 2000 to 10,000 m^2 for Class 2, and no size limit for Class 3. In the case of building structures, the suggested size of survey units is up to 100 m^2 for Class 1, 100 to 1000 m^2 for Class 2, and no size limit for Class 3.

In each of the survey units, sampling is performed to determine the level of radioactive contamination along with an estimation of the uncertainty. As sampling the entire area is not feasible, the key goal in designing the sampling program is to ensure representativeness of the samples. Here representativeness is the measure of the degree to which data accurately and precisely represent a characteristic of a population, i.e., the levels of contamination and their variability within the survey unit (EPA 2002). The number of samples needed for representativeness may vary depending on the characteristics of the contamination within the survey unit. Also depending upon the goal of sampling, different sampling designs can be employed.

The methods of sampling design include judgmental sampling, simple random sampling, stratified sampling, systematic and grid sampling, ranked set sampling, adaptive cluster sampling, and composite sampling. The sampling designs differ in terms of selected sampling units (the number and locations), and/or timing of sample collection. Brief description of each sampling method and their use are summarized in Table 14.4.

14.3.3 Segmentation and Dismantling

As part of the effort for removing radioactively contaminated SSC (systems, structures, and components), segmentation and dismantling is performed. This step involves removal and segmentation of the reactor vessel and internals, steam generators, pressurizers, reactor coolant pumps, and their associated piping. The procedures tend to be a very labor intensive step requiring manual labor to perform mechanical cutting and handling of the materials. Therefore improving efficiency and safety while reducing cost is important in the procedures.

The technologies used for the removal of large reactor components include milling cutters, plasma arc torch, high pressure abrasive grit, diamond wire saws, electric discharge machining, or metal discharge machining (NEA 2014). Often off-the-shelf technology is utilized and adapted for the intended segmentation operation.

Milling cutters are a self-propelled circular milling machine cutter. It is mounted on a track attached to a specially designed support fixture and powered pneumatically, hydraulically, or electrically. They can be used for cutting large components such as the reactor vessel and steam generators, and piping. They were used at the Rancho Seco NPP in the U.S. However, their slow cutting speed and frequency for changing the bit and blade may require newer technology.

Table 14.4 Comparisons of different sampling designs (EPA 2002)

Sampling method	How sampling is done	Main uses	Issues
Judgmental sampling	Based on previous knowledge and professional judgment	As a screening effort for a relatively small-scale problem	Interpretation is limited.
Simple random sampling	Sampling locations and times randomly selected over the unit area using random numbers	Good to estimate the presence of rare characteristics	Implementation can be problematic if precisely identifying random geographic locations is difficult. Could be costly.
Stratified sampling	Sampling from each non-overlapping subpopulations after dividing the unit area into subunits that are expected to be homogeneous	Good to estimate a population mean achieving same precision with fewer samples and lower cost	Need to identify homogeneous subunits relative to the environmental medium or the contaminant
Systematic and grid sampling	Samples are taken at regularly spaced intervals over space or time	Used to search for hot spots and to infer means, percentiles, and to estimate spatial patterns or trends over time	Could miss rare characteristics
Ranked set sampling	Uses a two-phase sampling design that identifies sets of field locations, utilizes inexpensive measurements to rank locations within each set, and then selects one location from each set for sampling.	This design results in more representative samples and is highly useful and cost efficient in obtaining mean estimates of concentration levels	The cost of locating and ranking locations in the field should be lower compared to laboratory measurement cost
Adaptive cluster sampling	After taking samples using simple random sampling take additional samples at locations where measurements exceed some threshold value	Good for searching for rare characteristics or boundaries of a contaminated area, appropriate for inexpensive, rapid measurements	Depends largely on the results of initial random sampling
Composite sampling	Volumes of material from several of the selected sampling units are physically combined and mixed in an effort to form a single homogeneous sample, which is then analyzed	Needed to get the mean when samples are non-homogeneous (e.g., concrete), most cost effective when analysis costs are large relative to sampling costs	Should not be used if the integrity of the individual sample changes with physical mixing of samples; there should be no safety hazards in mixing samples

Other common technologies used for segmentation of reactor vessel and internals include plasma arc torch (used for the Yankee Rowe plant in the US) and high pressure abrasive grit (used at Connecticut Yankee, Main Yankee, and San Onofre NPPs). A plasma arc torch ejects high velocity plasma from the torch nozzle to blow the molten metal away for cutting. With the plasma arc torch, controlling the generated fine particulates is difficult. The heat from the plasma torch results in the rise of high activity particulates to the surface of the water causing workers to be exposed to radiation.

High pressure abrasive grit is based on forcing a jet stream of highly pressurized water mixed with an abrasive (called grit) through a wear-resistant nozzle on a metal for cutting. Use of high pressure abrasive grit was found to have the problem of generating large quantities of secondary waste due to the contaminated grit.

Diamond wire saws use a diamond-embedded wire to cut metals. The length of the wire can be very long enabling any size cut. Moreover, the system can be applied when the work place is limited as the power supply unit can be placed several meters away from the work area. Diamond wire saws were successfully used at the Rancho Seco plant to cut the steam generators in half. It has also been utilized for cutting reactor vessels.

Electric discharge machining or metal discharge machining is also available for cutting large components. Their cutting is based on localized heating of metals. Electric discharge machining (EDM) uses an electrode (typically graphite) for the localized heating of metals positioned at a fixed distance above the surface of the item to be cut. With metal discharge machining (MDM), localized heating is achieved by using a vibrating electrode contacting the metal surface. Very high energy deposition is achieved by the generation of cutting pulses and passing the current between the electrode and the metal. EDM and MDM were used at Yankee Rowe for the cutting of piping. While slow cutting speed may limit their application, they are used predominantly for high precision cuts and have the advantage of minimal generation of secondary wastes. Though MDM is faster, it is less precise than EDM in metal cutting.

Other technologies for segmentation and dismantling of metallic components include robot-supported cutting, laser cutters, and an arc saw. Robot-supported cutting uses robotic and intelligent machines (RIM) with the mounting of different cutting technologies on the robots depending upon the needs of the cutting equipment. For example, the weight of the cutting head, cutting reaction force, articulation and degree of freedom, positioning accuracy and repeatability, access limitations, etc., can be key to robotic selection. Use of robots can significantly reduce worker dose or be applied in hard-to-reach locations.

Using lasers to cut materials is widely exercised. CO_2 lasers, solid-state lasers, or fiber lasers are used for this purpose with known precision and accuracy. Application of laser cutting in nuclear decommissioning has been demonstrated with active investigations underway. An arc saw is a thermal treatment technique applied to cut any conducting metal. The cutting is achieved by maintaining a high current electric arc between a circular, toothless saw blade and the materials to be cut. In this case, there is no physical contacts or reaction forces between the two. The technology can be operated under water and in air. Underwater application is preferred as the in-air use produces significant amounts of smoke, producing greater noise levels and a rougher cut. However, with improvements in speed and efficiency, it could find its application extended to segmentation of reactor vessels and internals.

In general, thermal techniques are easy to apply but require substantial effort for contamination control of the aerosols produced. Thus, their use becomes problematic or unsuitable in contaminated areas. Use of mechanical tools generally produce larger sized particulates whose contamination control is easier but not suitable in confined spaces. For underwater tasks, many of the thermal and mechanical cutting techniques are applicable.

For the demolition of concrete structures, various off-the-shelf techniques are available. These techniques include controlled blasting with explosives, wrecking ball with a crane, cyclic water pumps, flame cutting techniques using oxygen and fuel gas, rock splitter using hydraulically powered cylinders, drilling, and non-explosive demolition agents. A non-explosive demolition agent is a non-explosive technique using a chemically expanding compound poured into predrilled holes, thus causing concrete fractures upon hardening.

While the segmentation technologies have advanced to such a state that only minor development to suit an individual situation is needed, a number of tools may still need case-by-case adaptations.

14.3.4 Decontamination and Remediation

As part of decommissioning, decontamination is performed on the contaminated metals and concrete of SSCs. Decontamination reduces radiation exposure to workers and the public and may allow recycling and reuse of materials, equipment, or the site itself. Decontamination also reduces the volume of waste to be disposed, unless the secondary waste generation becomes significant.

Decontamination involves the use of a wide range of technologies and methods. These can be divided into three groups: (1) in-situ technologies for the decontamination of intact systems and structures in place; (2) methods used for material and equipment as part of handling and processing of them, (3) approaches used for decontamination and remediation of contaminated soils and groundwater.

Standard practice of decontamination is based on either chemical or mechanical treatment or a combination of both. In general, for the first group, chemical methods are widely used, in particular for metals. For the second group, mostly mechanical treatments are used for decontamination of concrete. For the third group, various physical and chemical methods are used, i.e., electrochemical, biological, sonic technologies or hybrid approaches. Available approaches can combine very different technologies.

Depending upon the levels of radioactive contamination, the site soils will also go through segregation or decontamination. Cleanup of groundwater or limiting the spread of contamination in groundwater may be necessary (AECOM 2017).

Groundwater cleanup efforts are typically extensive, including identification of groundwater flow relative to possible leakage sources (e.g., spent fuel pool). The related sampling programs extend over many months and cleanup can be complex. The effort also include evaluation that the groundwater has been effectively cleaned up.

14.3.4.1 Chemical Decontamination

Chemical decontamination is less labor intensive, is applicable to inaccessible surfaces, produces few airborne hazards, and allows remote application. However, it is not usually effective on porous surfaces. Chemical decontamination generates potentially large volumes of secondary waste (regeneration of the decontamination chemicals can reduce secondary waste generation), including possible generation of mixed waste.

Chemical treatment can be either “hard” or “soft” and is based on dissolving either the base metal or the contaminated film covering the base metal. Both alkaline and acidic solutions are used for the treatment. Soft decontamination treatment is based on using relatively dilute solutions that ease waste management concerns, lower corrosion concern but result in lower decontamination performance. Hard treatment uses concentrated chemical solution for decontamination. Although hard treatments can achieve a high level decontamination, they can generate a larger volume of secondary waste involving a high management cost. Typically soft approach is preferred in chemical decontamination.

Application of chemical decontamination can be categorized into one of the following six approaches: (1) alkaline oxidation and dissolution, (2) alkaline oxidation followed by acidic dissolution, (3) acidic oxidation and dissolution, (4) acidic oxidation followed by acidic dissolution, (5) acidic dissolution, and (6) acidic reduction and dissolution (DOE 1994).

For the application of alkaline treatment, alkaline salts are widely used. They include potassium hydroxide (KOH), sodium hydroxide (NaOH), sodium carbonate (Na_2CO_3), trisodium phosphate (Na_3PO_4), ammonium carbonate ($(NH_4)_2CO_3$), and alkaline permanganate (NaOH and $KMnO_4$). Use of alkaline salts has several purposes including removal of grease, oil films, rust, paint and other coatings, to neutralize acids and provide the right chemical environment for other agents, and as a solvent for species soluble at high pH. In particular, alkaline permanganate (AP) is widely used for decontamination of metal surfaces. Use of alkaline salts has the benefit of being low cost, easy to store, easier to handle than acids, and applicable for ceilings and walls in the form of gels. Nevertheless, their slow reaction time, destructive effects on aluminum, and potential safety hazard from burning upon contact with humans must also be noted.

Commonly used strong acid solutions include hydrochloric acid (HCl), nitric acid (HNO_3), and phosphoric acid (H_3PO_4). These can also be used as dilute solutions in mixtures with other compounds. Sometimes, acid salts are used in place of acids as they provide the advantage of being less corrosive. Weak acids commonly used are oxalic acid ($C_2H_2O_4$), citric acid ($C_6H_8O_6$), and sulfamic acid (HSO_3NH_2). Weak acids are used on metal surfaces for the dissolution of the metal oxide film and solubilizing the metal ion.

In addition to acids and alkaline solutions, other agents used in chemical decontamination include complexing agents, oxidizing/reducing agents, detergents and surfactants, and organic solvents. Complexing agents (e.g., EDTA) are chemical species that form a stable complex with a metal ion. By sequestering the solubilized metal ions in a stable complex with complexing agents, their redeposition on metallic surfaces can be prevented. Use of oxidizing/reducing agents are used to condition metal oxide films. Such conditioning facilitates the removal of radioactivity on the surfaces (e.g., on equipment and piping of primary coolant circuits in a nuclear power plant).

Alkaline oxidation and dissolution is usually done with the use of an alkaline permanganate (AP). AP dissolves chromium oxides and attacks various hard-surface alloys, organics, and copper. While AP is noncorrosive to stainless steel, it is corrosive to carbon steel.

Alkaline oxidation followed by acidic dissolution is commonly performed for decontamination of metals with the use of AP. Use of AP is followed by citric acid or any other acid or acidic salts. In this case, using AP is mainly a pretreatment to condition the film of corrosion products and application of the acid does most of the decontamination. Use of ammonium citrate as the acid is effective with stainless steel but highly corrosive to carbon steel. The addition of EDTA is shown to have the benefit of improving process effectiveness and prevents the redeposition of contamination. Using a mixture of citric acids and oxalic acids (called Citrox) after AP pretreatment is effective for decontaminating stainless steel, carbon steel, as well as Inconel. Citrox treatment was shown to be noncorrosive to carbon steel decontamination. Using AP, then rinsing it and applying sulfamic acid was also effective for the treatment of stainless steel, carbon steel, and Inconel.

The application of acidic oxidation and dissolution can be achieved using nitric acid to remove uranium oxide fuel debris. In this case, nitric acid is both oxidant and acid. Nitric acid is also used for the application of acidic oxidation followed by acidic dissolution. An example of this application is the removal of fuel and fission product debris. For this application, nitric acid is used as oxidant and Citrox or another acid is used for dissolution. Additionally this can be used for corrosion product removal from metal if little or no chromium is present.

Depending upon the choice of acids, acidic dissolution can be used for the decontamination of stainless steel, carbon steel, copper alloys, or aluminum alloys. For copper alloys, hydrochloric acid, sulfamic acid, and phosphoric acid can be used. With aluminum alloys, nitric acid or sulfamic acid can be used. Stainless steel can be treated with oxalic acid or nitric acid, while carbon steel can be treated using sulfamic acid.

Use of acidic reduction and dissolution is not common. However, for the treatment of high-temperature stainless steel it is reported as used with the reducing decontamination solution, hydrazine.

14.3.4.2 Mechanical Decontamination

Mechanical decontamination includes a wide range of non-chemical methods, i.e., using mechanical forces. Mechanical forces involving abrasive scraping/grinding, high pressure flushing, vibration, etc. are utilized to decontaminate specific components, equipment, and surfaces of piping and equipment. For decontamination of concrete, a mechanical process of removing a thin layer of concrete from a structure, called scabbling, is commonly used. The mechanical process is based on using compressed air, water jets, microwave, and lasers. Use of ultrasonics, steam cleaning, foams, gels, strippable coatings, and CO_2 blasting is also exercised. Table 14.5 shows the list of highly effective techniques for different decontamination applications. Advantages and disadvantages of the methods are summarized in Table 14.6.

Decontamination of graphite is an important issue in the case of gas cooled reactors. In this case, thermal treatment technologies and steam reforming methods along with chemical methods are employed for the separation of volatile radionuclides such as ^{3}H, ^{14}C, and ^{36}Cl (NEA 2014).

Usually, the most commonly used methods for decontamination of materials and equipment are labor-intensive and high-waste-generating technologies. As an alternative, use of robots for decontamination applications is actively investigated as the technology has become mature and intelligent with much wider application potentials. Characterization of physical-chemical process of decontamination is still under development and implementation of technologies is often based on trial and error. As reviewed in this section, selection of specific methods of decontamination should be based on the examination of not only contaminant characteristics and effectiveness

Table 14.5 List of highly effective non-chemical methods for various decontamination applications

Applications	Highly effective methods
Loose particulate contamination	Dusting/wiping/scrubbing, vacuuming, turbulator, strippable coatings, steam cleaning, sponge blasting, CO_2 blasting, wet abrasive cleaning, electropolishing, ultrasonic cleaning, vibratory finishing, light ablation
Bare and painted concrete	Sponge blasting, ultra high pressure water, shot blasting, grit blasting, grinding, scarifier, milling, drill and spall, paving breaker/chipping hammer, expansive grout, light ablation, microwave scabbling, flaming, flame scarifying, plasma torch, electrical resistance
Metal surfaces	Metal-based paint removal, sponge blasting, CO_2 blasting, wet ice blasting, hydroblasting, ultra high pressure water, wet abrasive cleaning, grit blasting, scarifier, milling, electropolishing, ultrasonic cleaner, vibratory finishing
Components of all sizes	Steam cleaning, sponge blasting, CO_2 blasting, wet ice blasting, hydroblasting, ultra high pressure water, wet abrasive cleaning, grit blasting

(continued)

Table 14.5 (continued)

Applications	Highly effective methods
Pipe and tank internals	Flushing with water, hydroblasting, ultra high pressure water, grit blasting
Embedded materials and some oxide surfaces	Hydroblasting, ultrahigh pressure water, grit blasting, drill and spall, paving breaker/chipping hammer, expansive grout, electropolishing
Small hand tools	Turbulator, wet abrasive cleaning, electropoloshing, ultrasonic cleaning, vibratory finishing
Special equipment (e.g., motors)	Vacuuming, sponge blasting, CO_2 blasting

Table 14.6 Advantages and disadvantages of mechanical decontamination methods

Method	Advantages	Disadvantages
Flushing with water	Easy to apply, readily available, little of no surface damage, easy training, remote operation	Difficult to remove contamination from crevices, cross contamination possible, high worker exposure, labor intensive, solvents/detergents could complicate waste management, large secondary waste
Dusting/wiping/scrubbing	Easy to apply, readily available, little of no surface damage, easy training, can be applied wet or dry	Difficult to remove contamination from crevices, high worker exposure, labor intensive, solvents/detergents could complicate waste management
Vacuuming	Easy to apply, readily available, easy waste handling/disposal, little of no surface damage, easy training, can be applied wet or dry	High worker exposure, labor intensive
Turbulator (using fluid turbulence for surface cleaning)	Little of no surface damage	Solvents/detergents could complicate waste management, large secondary waste
Metal-based paint removal	Easy waste handling/disposal, easy training	Difficult to remove contamination from crevices, cross contamination possible, high worker exposure, labor intensive, solvents/detergents could complicate waste management
Strippable coatings	Easy to apply, readily available, little of no surface damage, easy training	Difficult to remove contamination from crevices, high worker exposure, labor intensive, solvents/detergents could complicate waste management
Steam cleaning	Little of no surface damage, remote operation	Cross contamination possible, labor intensive
Sponge blasting	Little of no surface damage, remote operation, can be applied wet or dry	Cross contamination possible, labor intensive, large secondary waste

(continued)

Table 14.6 (continued)

Method	Advantages	Disadvantages
CO_2 blasting	Remote operation	Cross contamination possible, labor intensive
Wet ice blasting	Easy waste handling/disposal, remote operation	Cross contamination possible, labor intensive
Hydroblasting	Remote operation, can penetrate crevices	Cross contamination possible, solvents/detergents could complicate waste management, large secondary waste, possibility of excessive erosion or surface roughening
Ultra high pressure water	Remote operation, can penetrate crevices	Cross contamination possible, solvents/detergents could complicate waste management, large secondary waste, possibility of excessive erosion or surface roughening
Shot blasting	Remote operation	Cross contamination possible, possibility of excessive erosion or surface roughening
Wet abrasive cleaning	Can be a good compliment to chemical decontamination	Solvents/detergents could complicate waste management
Grit blasting	Readily available, remote operation	Cross contamination possible, large secondary waste, possibility of excessive erosion or surface roughening
Scarifier	Readily available, little of no surface damage, easy training, remote operation	Cross contamination possible, possibility of excessive erosion or surface roughening
Milling	Use on all metals, air and underwater environments, remote operation feasible	Cross contamination possible, possibility of excessive erosion or surface roughing
Drill and spall	Concrete demolition	Cross contamination possible, labor intensive, possibility of excessive erosion or surface roughening
Paving breaker/ chipping hammer	Readily available, easy training, can penetrate crevices	Cross contamination possible, high worker exposure, labor intensive, possibility of excessive erosion or surface roughing
Expansive grout	Nonexplosive cracking agent	Labor intensive, possibility of excessive erosion or surface roughing
Electropolishing	High decontamination factor, can decontaminate planar areas, corners, recessed geometries, etc.	Typically multistep process, solvents/detergents could complicate waste management, large secondary waste
Ultrasonic cleaning	Readily available, little of no surface damage	Solvents/detergents could complicate waste management, large secondary waste
Vibratory finishing	Little of no surface damage	Solvents/detergents could complicate waste management, large secondary waste

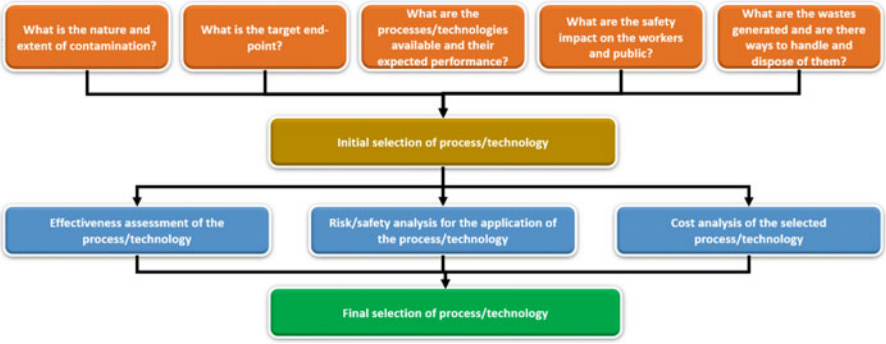

Fig. 14.2 Logic diagram for selecting a decontamination technique

the method but also cost, worker dose reduction, secondary waste minimization, and consideration of the target end-state. The overall process to follow in method selection is depicted in Fig. 14.2.

14.3.4.3 Remediation of Contaminated Soils and Groundwater

Remediation of radioactively contaminated soils typically involves removal of contaminated soil through excavation and disposal in an off-site disposal facility. How the contaminated soil should be handled depends upon the level of contamination. When contamination levels are low but require removal from the site, the soil may qualify for disposal at a low activity waste (LAW) facility. For example, in the US, an operating hazardous waste disposal facility in Idaho can accept LAW. On the other hand when the soil has higher levels of contamination and the volume of the contaminated soils is very large, the excavated soils may go through treatment either ex-situ or in-situ before disposal to minimize the volume. The treatment could be done by simple segregation or through soil washing or electroremediation. The following technologies are used or being developed for such use at a variety of nuclear facility decommissioning projects.

Segregation is done by using real-time gamma detectors to examine the presence of gamma-emitting nuclides in the soil. Typically, a threshold level is defined for a reference gamma emitter. Using the detectors, contaminated soils are segregated from the uncontaminated bulk. In soil washing, the contaminated soil particles are physically segregated (while being suspended in water) from the contamination-free bulk materials. Chemical treatment can be used for soil washing where complexants, acids and alkalis are used for leaching of radionuclides from the soils. The contamination-free bulk materials are recycled as backfill or disposed of as less hazardous material than the original soil.

Electroremediation uses a low level direct electric current to drive the movement of water and ions in the soil to remove contaminants. In this case, applying an electric field in the soil causes convective movement of water through a porous medium and migration of the dissolved ions. Using these movements, the contaminants are collected at the cathode for separation.

If removal of the contaminated soils is not feasible, stabilization, solidification, immobilization, or containment using barriers can be exercised *in situ*. In stabilizing the soil, the contaminants are stabilized by forming chemically immobile compounds. In the case of solidification, the contaminated soil is turned to a monolithic block by using a binder material such as a grout. Soil immobilization is done through *in situ* vitrification of soil by turning the soil into a glassy matrix through high temperature melting and cooling. Containment is done by using physical barriers to prevent migration of contaminants. Another related method of *in situ* soil remediation is phytoremediation. Phytoremediation uses living plants for removal or containment of contaminants through bio-uptake. Thermal processes can also be utilized in the case of soil contamination with volatile radionuclides such as tritium and ^{14}C. Such processes uses heat to separate these radionuclides through volatility. Another related method used is to prevent rainwater from passing through the soil or to slow the movement of contamination through the soil column by using an engineered cap, such as asphalt.

To improve the performance of soil remediation, other technologies such microbial treatment or using nanomaterials have also been suggested. Microbial treatment is to break down contaminants by using them as a food source for microbes. Use of nanomaterials with their large surface area to volume ratio and reactive sites for sorption of contaminants appears promising and is under active investigation.

In the case of contaminated groundwater, the contaminated water is removed through pumping and treated with demineralizers for decontamination. If pumping and treatment is not feasible, the contaminated groundwater is isolated by installing a migration barrier.

14.3.5 Waste Management in Decommissioning

Decommissioning of nuclear power plants generates a wide variety of nuclear wastes. Aside from spent nuclear fuel, the majority of the activity is in the radioactive waste from the removal of activated hardware (from the reactor vessel internals) and from piping and components contaminated with corrosion/activation products and fission products. Table 14.7 shows major types of contaminated materials to be handled (in ton) in decommissioning of PWR or GCR (gas cooled reactor).

In terms of volume, the decontamination and dismantling process and site remediation activities will produce a significant amount of nuclear waste. The wastes include contaminated concrete, pipes, ducts, miscellaneous steels, and secondary radioactive waste from decontamination and effluent treatment. If only dry decommissioning processes are used, the types of nuclear wastes are similar to LILW produced during reactor operation. A large volume of slightly contaminated soil is also expected from site remediation. The resulting waste management activities follow similar steps of LILW management activities during plant operation.

Table 14.7 Typical radioactive material generation from NPP decommissioning (unit: ton) (IAEA 2008a)

Radioactive material generation	PWR (900–1300 MWe)	GCR (250 MWe)
Irradiated carbon steel	–	3000
Activated steel	650	–
Graphite	–	2500
Activated concrete	300	600
Contaminated ferritic steel	2400	6000
Steel likely to be contaminated	1100	–
Contaminated concrete	600	150
Contaminated lagging	150	150
Contaminated technological wastes	1000	–

Mixed wastes are also produced from decommissioning which was discussed in Sect. 13.8.

Minimizing LILW generation will be a related important goal in nuclear decommissioning. The amount of waste generated from decommissioning varies depending upon the strategies used. If buildings and equipment are decontaminated to clearance levels, the waste generation will be the lowest. If little decontamination of buildings and equipment is performed, the volume will be high. If buildings are decontaminated with little decontamination of equipment, the generated waste volume will be in between. This is given in Table 14.8 to show the volumes of LILW generated from the U.S. decommissioning projects. A comparison is also made to the case of European plant where extensive decontamination is expected.

14.3.6 Final Site Characterization and Environmental Monitoring

Through the application of appropriate cleanup efforts, a decommissioned nuclear power plant is made ready for license termination examination or for any intended future use of the site. The decision to release the site requires a release criterion based on the consideration of the resulting human dose impact. The residual contamination level at the release criterion should result in a dose less than the prescribed regulatory limit. The dose-based release criterion is then translated to acceptable levels of residual concentration. This level is called the *derived concentration guideline level* (DCGL) as explained in the next section. This is followed by measurements of the levels and distributions of residual contamination at the site. Based on the measured levels of residual contamination, a decision is made regarding whether the site meets the release criterion. These steps are depicted in the following Fig. 14.3.

Table 14.8 Volumes (m^3) of radioactive waste generated from selected U.S. decommissioning projects (Reid 2015)

Waste type (US classification)	Plant name					
	Connecticut Yankee	Maine Yankee[a]	Rancho Seco[b]	San Onofre Unit 1	Trojan	Estimate for European Plant[c]
Class A	106,318	91,290	17,237	47,588	Not available	2911
Class B & C	293	600	93	30	Not available	2459
Greater than class C (GTCC)	Not available	Not available	11	3	Not available	109
Total	106,611	91,860	17,341	47,621	7790	5479

[a] Little decontamination of buildings and equipment

[b] Decontamination of buildings, little decontamination of equipment

[c] Decontamination of buildings and equipment to clearance levels

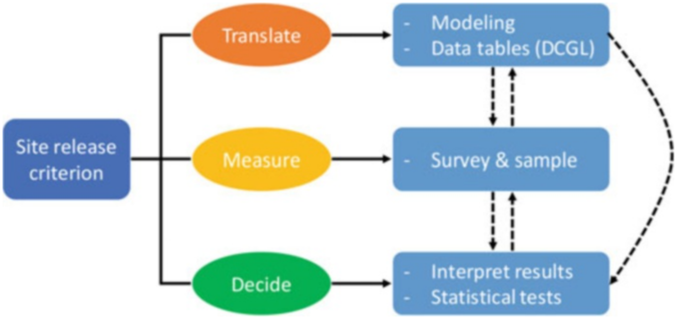

Fig. 14.3 Steps involved in site release decision. (Redrawn from MARSSIM 2000)

14.3.6.1 Determination of the Acceptable Level of Residual Contamination

The main criterion for site release is to show that any radiation exposure due to the presence of any residual radioactivity poses a very low level risk. Such low risk levels are translated into the corresponding dose levels becoming a release criterion. Therefore, a release criterion is expressed in terms of annual dose limit in mSv/year or mrem/year

In the case of the US, the dose limit as the release criterion is 250 μSv/year (25 mrem/year). This dose limit needs to be converted into the corresponding residual contamination level at a specific site. For such conversion, exposure pathway modeling of the site is necessary. Exposure pathway modeling uses the level of residual contamination at a site as the source and analyzes the fate, transport and exposure to humans to determine the annual dose. The residual contamination level as the concentration of radionuclides that results in the dose at the regulatory limit

(250 μSv/year) is the DCGL. The units for the DCGL are Bq/kg or pCi/g for soil and Bq/m^2 or dpm/100 cm^2 for building surface contamination.

Exposure pathway modeling requires the understanding of the behaviors of the local population in terms of their food consumption and time spent in different microenvironments (i.e., indoor, outdoor). Therefore, determining DCGLs requires site specific analysis. The conditions of the site as well as the characteristics of local population need to be taken into account in developing the site-specific DCGL values.

At the same time, given the complexity involved in a site specific analyses, the regulatory agencies (U.S. NRC, IAEA) offer an alternative approach of using screening analysis to site release determination. The screening analysis is based on the use of very conservative models and data in exposure pathway modeling without relying on site specific information. The goal of the screening analysis is to make credible effort not to underestimate the derived dose from the given contamination levels.

Based on the use of screening analysis, generic values of DCGL have been suggested by the U.S. government. These values are listed in Tables 14.9 and 14.10 for the cases of building-surface contamination and soil contamination, respectively (NRC 2001). Use of these generic DCGLs allows decommissioning projects to expend minimal efforts for site specific analysis. These generic values were derived based on selection of the 90th percentile of the resulting dose distribution for each radionuclide under consideration. The input values for human behavior modeling were selected as the mean of the distribution of the conservatively assumed critical group. Adopting the generic DCGL values for site release determination then puts more emphasis on site cleanup efforts.

Another set of screening levels as generic clearance levels are suggested by the IAEA (IAEA 2004). These levels are also the derived levels of activity concentrations in soil for site clearance based on the dose limit of 10 μSv/year (1 mrem/year). The values are shown in Table 14.10 along with the US NRC's screening DCGL values for comparison. It can be seen that the screening levels of the DCGLs from the U.S. NRC are in general more conservative than the clearance levels of the IAEA. This may indicate that the level of conservatism embedded in the exposure pathway modeling approaches is higher with the US NRC approach.

14.3.6.2 Residual Contamination Measurements

Using the predefined target levels, i.e., the DCGL for site release, cleanup efforts are made in nuclear decommissioning through decontamination and site remediation. Such cleanup efforts are followed by a final status survey to examine the status of residual contamination at the site with respect to the site release decision. Levels and distribution of residual contamination are measured by employing suitable field and/or laboratory measurement techniques. The data obtained from the previously conducted surveys are also utilized.

Table 14.9 Allowable levels of residual building surface contamination for license termination for major radionuclides (in Bq/m^2 or dpm/100 cm^2) – the screening levels (NRC 2001)

Radionuclide	Bq/m^{2a}	In dpm/100 cm^{2a}
^{3}H	2.0E+08	1.2E+08
^{14}C	6.2E+06	3.7E+06
^{22}Na	1.6E+04	9.5E+03
^{35}S	2.2E+07	1.3E+07
^{36}Cl	8.4E+05	5.0E+05
^{54}Mn	5.3E+04	3.2E+04
^{55}Fe	7.5E+06	4.5E+06
^{60}Co	1.2E+04	7.1E+03
^{63}Ni	3.0E+06	1.8E+06
^{90}Sr	1.5E+04	8.7E+03
^{99}Tc	2.2E+06	1.3E+06
^{129}I	5.8E+04	3.5E+04
^{137}Cs	4.7E+04	2.8E+04
^{192}Ir	1.2E+05	7.4E+04

[a] These values are based on the assumption that the fraction of removable surface contamination is equal to 0.1. If higher fraction of surface removal is possible, the values can be increased in proportion to the removable surface contamination. For example, if 100% of the surface contamination is removable, then the values can, and therefore the screening levels should be decreased by a factor of 10

The final status survey begins with a plan for data collection based on data quality objectives (DQOs). DQOs are predefined requirements for the quality and quantity of the data collected. For example, an acceptable probability of obtaining a false positive or negative decision is set as part of DQOs.

A false positive or false negative decision refers to decisions made against the true state of the site. If the site is clean enough based on the regulatory requirements, but the decision becomes to reject the request for site release, it is a false positive decision. If the site is not ready for release requiring further cleanup work, but the decision is to declare compliance and to grant site release, it is a false negative decision. The acceptable probability to make a false positive or false negative decision is a choice made by the responsible entity. Values like 1%, 2.5%, 5% or 10% is used as acceptable probability of making such wrong decision to occur.

The survey plan is implemented through data collection including direct measurements and sampling based on the survey unit classification (as discussed in Sect. 14.3.2). This is followed by verification and validation of the survey results combined with an assessment of the data with respect to the data quality objectives. Determination of the average levels of contamination along with their distributions are examined both qualitatively (through a visual representation of the radionuclide distribution to identify patterns or potential anomalies) and quantitatively (through statistical analysis) with respect to regulatory compliance demonstration.

To quantitatively assess the contributions of the plant's operation to the radiation inventory at the site, determining the background levels of radionuclides present,

Table 14.10 Allowable levels of residual soil surface contamination for license termination for major radionuclides (in Bq/kg or pCi/g) – Comparison of the screening levels between US NRC and IAEA (NRC 2001; IAEA 2004)

Radionuclide	US screening level (pCi/g)	US screening level (Bq/kg)	IAEA generic clearance level (Bq/kg)
^{3}H	1.1E+02	4.1E+03	1.0E+05
^{14}C	1.2E+01	4.4E+02	1.0E+03
^{36}Cl	3.6E-01	1.3E+01	1.0E+03
^{54}Mn	1.5E+01	5.5E+02	1.0E+02
^{55}Fe	1.0E+04	3.7E+05	1.0E+06
^{60}Co	3.8E+00	1.4E+02	1.0E+02
^{59}Ni	5.5E+03	2.0E+05	1.0E+05
^{63}Ni	2.1E+03	7.8E+04	1.0E+05
^{90}Sr	1.7E+00	6.3E+01	1.0E+03
^{94}Nb	5.8E+00	2.1E+02	1.0E+02
^{99}Tc	1.9E+01	7.0E+02	1.0E+03
^{129}I	5.0E-01	1.9E+01	1.0E+01
^{137}Cs	1.1E+01	4.1E+02	1.0E+02
^{152}Eu	8.7E+00	3.2E+02	1.0E+02
^{154}Eu	8.0E+00	3.0E+02	1.0E+02
^{210}Pb	9.0E-01	3.3E+01	1.0E+03
^{226}Ra	7.0E-01	2.6E+01	1.0E+03
^{232}Th	1.1E+00	6.3E+01	1.0E+03
^{235}U	8.0E+00	3.0E+02	1.0E+03
^{238}U	1.4E+01	5.2E+02	1.0E+03
^{238}Pu	2.5E+00	9.3E+01	1.0E+02
^{239}Pu	2.3E+00	8.5E+01	1.0E+02
^{241}Pu	7.2E+01	2.7E+03	1.0E+04
^{241}Am	2.1E+00	7.8E+01	1.0E+02
^{242}Cm	1.6E+02	5.9E+03	1.0E+04
^{243}Cm	3.2E+00	1.2E+02	1.0E+03

apart from the operation of nuclear power plant is necessary for the survey. In this case, the surveys for background radiation is made mostly for the naturally existing nuclides such as ^{40}K, ^{14}C, ^{3}H, uranium, thorium, and their decay products including radium. Cesium-137 and other fission products may also be included as part of the background measurements if the impacts from nuclear weapons fallout are considered non-negligible. For such purpose, one or more background reference areas are selected from areas that are not affected by NPP operations while having similar physical, chemical, geological, radiological, and biological characteristics with the affected areas at the NPP site (NRC 1998).

The techniques for sampling contaminated and activated surfaces and materials are well established (IAEA 2008b). At the same time, new techniques are emerging with enhanced capabilities for specific applications. These techniques include in-situ

3D gamma spectrometers with visual images, automated data collection techniques for large readings, combination of radiation measurements with GIS (geographic information system) maps, use of robots with radiation probes into pipes and small spaces, etc. Mature development of these techniques could be integrated into the decision support system for systematic demonstration of regulatory compliance.

14.3.6.3 Compliance Determination

Measured radionuclide concentrations at a site from the final status survey along with background measurement data are used for comparison against the target cleanup levels, i.e., DCGL. A compliance determination for site release is made for each of the survey units. Such a determination is made under the consideration of the following issues:

- Presence of contaminants in background radiation
- Acceptable probability to make a false positive decision (this is called α).
- Acceptable probability to make a false negative decision (this is called β).

If the radionuclides of concern are also present as part of the background radiation at the site, the background levels of these radionuclides need to be subtracted from the measured values of the radionuclide for comparison against the DCGL. In this case, the difference between the maximum value of the measured data in the survey unit and the background level in the survey unit is compared with the DCGL. If the difference is less than the DCGL, the survey unit meets the release criterion. If the difference is greater than the DCGL, then the survey unit does not meet the release criterion. If the difference of the survey unit average value and the background level is less than DCGL, but there are measured data point(s) with the value of the difference between the measurement and the background level being greater than DCGL, the situation requires a statistical analysis (the Wilcoxson Rank Sum test) for a compliance determination.

Similar steps are also followed in the case where the radionuclides of concern are not present in the background data. If all measured data from a survey unit are below the DCGL, the unit meets the release criterion. If the average value of measured residual contamination of a survey unit is greater than the DCGL, then the unit does not meet release criterion. In a situation where the average value of the measurements are lower than the DCGL but the average includes the values greater than DCGL, performing a statistical test (the sign test) is necessary.

When a mixture of radionuclides is present at a site, the compliance determination is based on the "sum of fractions". In this case, the combined fractional concentration limit of the radionuclides is determined. If this sum is less or equal to one, as shown in the following relationship, then the compliance is demonstrated.

$$\sum\nolimits_{i=1}^{n} \frac{C_i}{DCGL_i} = \frac{C_1}{DCGL_1} + \frac{C_2}{DCGL_2} + \cdots + \frac{C_n}{DCGL_n} \leq 1 \tag{14.1}$$

where C_i is concentration of radionuclide i ($i = 1, 2, \ldots$, n) as residual level of contamination, $DCGL_i$ is derived concentration guideline level as the predefined target cleanup level for each individual radionuclide.

The statistical test used to examine whether there is a difference between the residual contamination levels and the levels prescribed by the DCGL looks for a difference that is statistically significant. Only when the difference is statistically significant, then the difference is accepted. Such test also requires the specification on the acceptable probability to make false positive (α) or false negative (β) decision.

For this testing, the sign test is suggested for the case of the contaminants are not present in the background (MARSSIM 2000). Also, if the contaminants are present in the background, use of the Wilcoxon Rank Sum (WRS) test is suggested. Both of these tests are to examine differences between groups of data. They are a nonparametric method and applied to various types of data without requiring special features in the data such as symmetry, normality, etc.

The data under examination by the sign test are pairs of data, i.e., the measured concentration of a radionuclide and the DCGL value for the radionuclide under consideration. The sign test is based on the counts of signs, i.e., + or − using the difference between the DCGL value of a radionuclide and the corresponding measured concentration. If the DCGL value is greater than the measured value, the sign is + (i.e., it is a positive difference; the measured concentration is lower than the target cleanup level). If vice versa, the sign is − (i.e., a negative difference; the residual contamination level is higher than the target cleanup level). If the difference is zero, the measured value (or the pair) is dropped from the test. The total counts of + signs becomes the test statistic. If the number of total counts of + signs is greater than a specified number (called the critical value) for the total sample size in the survey unit and the acceptable level of false positive (see Table 14.11), then the survey unit pass the test and is declared compliant for release. If the number of total counts of + signs is smaller than the critical value, the survey unit fails to pass and is declared noncompliant.

Example 14.1: Survey Unit Examination Using the Sign Test
The following 22 data points were collected in a final status survey from a survey unit at a plant site as the residual concentration of ^{60}Co.

Measured data of the residual activity (Bq/kg): 121, 140, 144, 110, 125, 133, 122, 115, 124, 149, 115, 113, 127, 133, 148, 130, 119, 136, 129, 125, 141, 129

By using 2.5% as the acceptable probability of making a false positive and negative decision, determine if the survey unit is ready for release. Use the

(continued)

Table 14.11 Examples of critical values when using the sign test for a compliance determination (MARSSIM 2000)

Critical values for the sign test corresponding to each sample size

Sample size	**α = 0.01**	**α = 0.025**	**α = 0.05**	**α = 0.1**	**Sample size**	**α = 0.01**	**α = 0.025**	**α = 0.05**	**α = 0.1**
4	4	4	4	3	**28**	20	19	18	17
5	5	5	4	4	**29**	21	20	19	18
6	6	5	5	5	**30**	21	20	19	19
7	6	6	6	5	**31**	22	21	20	19
8	7	7	6	6	**32**	23	22	21	20
9	8	7	7	6	**33**	23	22	21	20
10	9	8	8	7	**34**	24	23	22	21
11	9	9	8	8	**35**	24	23	22	21
12	10	9	9	8	**36**	25	24	23	22
13	11	10	9	9	**37**	26	24	23	22
14	11	11	10	9	**38**	26	25	24	23
15	12	11	11	10	**39**	27	26	25	23
16	13	12	11	11	**40**	27	26	25	24
17	13	12	12	11	**41**	28	27	26	25
18	14	13	12	12	**42**	28	27	26	25
19	14	14	13	12	**43**	29	28	27	26
20	15	14	14	13	**44**	30	28	27	26
21	16	15	14	13	**45**	30	29	28	27
22	16	16	15	14	**46**	31	30	29	27
23	17	16	15	15	**47**	31	30	29	28
24	18	17	16	15	**48**	32	31	30	28
25	18	17	17	16	**49**	33	31	30	29
26	19	18	17	16	**50**	33	32	31	30
27	19	19	18	17					

Example 14.1 (continued)
U.S. screening level (DCGL) for the compliance determination (MARSSIM 2000).

Solution:

The DCGL for ^{60}Co, the radionuclide under consideration, is 140 Bq/kg. Using the sign test:

Number	Measured data (Bq/kg)	Difference between DCGL and the measured data (Bq/kg)	Sign
1	121	19	+
2	140	0	(dropped from the sample data)
3	144	−4	–
4	110	30	+
5	125	15	+
6	133	7	+
7	122	18	+
8	115	25	+
9	124	16	+
10	149	−9	–
11	115	25	+
12	113	27	+
13	127	13	+
14	133	7	+
15	148	−8	–
16	130	10	+
17	119	21	+
18	136	4	+
19	129	11	+
20	125	15	+
21	141	−1	–
22	129	11	+
Total number of (+) = 17			

The number of positive signs from the comparison as shown above is 17 (out of 21 samples).

The critical value is 15 for 21 samples, with $\alpha = 0.025$ (from Table 14.11)

Since the test statistic (17) > the critical value (15), the survey unit passes the test (declared compliant).

The WRS test is suggested when background contamination is present because the test can handle a data set that is not necessarily paired. In other words, the number of measurements from the survey unit can be different from the number of background measurement data in the reference area.

Let's assume that we are investigating a case where only one radionuclide is involved in a survey unit. There are m measured data points for the radionuclide from the survey unit. The corresponding background measurements for the radionuclide has n data points. For regulatory compliance, it should be demonstrated that the measured residual contamination levels minus the background levels is lower than the DCGL (i.e., the residual contamination level – the background level < DCGL).

For this demonstration using the WRS test, we first add the value of the DCGL to each of the n measured background concentration data. Thus we are to examine if the measured contamination levels are less than the DCGL plus background. The n data points of background concentrations plus the DCGL are now called the n adjusted reference measurements. We are testing whether the m measured concentration data of the radionuclide from the survey unit is different from the n adjusted reference measurements. If the measured concentration of the radionuclides is lower than the adjusted measurements, then the survey unit can be considered compliant for release.

The WRS test is based on ranking the $m + n$ (the survey unit measurements + the adjusted reference measurements) data points. Each data point has a rank starting from 1 and going up to $m + n$. If the rank values of any of the data are equal, then assign a rank as the average value (e.g., if two are tied at rankings 3 and 4, then assign a rank of 3.5 to both). Then, we add all the values of the survey unit measurements. Call this sum W_s.

Subtract this W_s from the total sum of the ranks of the $m + n$ data (i.e., $(m + n)\times(m + n + 1)/2$) to obtain the test statistic, W_t. Therefore, W_t becomes $(m + n)\times(m + n + 1)/2 - W_s$ (this is equal to the sum of the ranks of the adjusted reference measurements). If the value of W_t is greater than the critical value for the given sample size and the acceptable false positive probability (see Table 14.12), then the survey unit is judged to be in compliance for release. This is shown in Example 14.2.

Example 14.2: Wilcoxon Rank Sum Analysis

From a final status survey at a site, the following data of residual contamination (Bq/kg) are obtained in a survey unit (MARSSIM 2000):

540, 488, 508, 514, 467, 539, 493, 545, 482, 524, 477 ($m = 11$)

It was also found that the radionuclide is present in the background from a survey in a reference area. The measured background levels data (Bq/kg) are as follows:

182, 233, 233, 213, 228, 249, 192, 239, 218, 244 ($n = 10$)

If the DCGL value for the radionuclide is 400 Bq/kg, determine if the survey unit is ready for release (use 0.25 as the acceptable probability of the false positive decision (α)).

(continued)

Table 14.12 The critical values for the Wilcoxon Rank Sum Test per a given sample size (m = sample size of the residual concentration measurements, n = sample size of background measurements) if the acceptable probability of a false positive decision is 2.5% or 5% (α = 0.025, and 0.5 respectively) (MARSSIM 2000)

If the acceptable probability of false positive decision is 2.5% (α = 0.025)

m	**n = 2**	**n = 3**	**n = 4**	**n = 5**	**n = 6**	**n = 7**	**n = 8**	**n = 9**	**n = 10**	**n = 11**	**n = 12**	**n = 13**	**n = 14**	**n = 15**	**n = 16**	**n = 17**	**n = 18**	**n = 19**	**n = 20**
2	7	9	11	13	15	17	18	20	22	23	25	27	29	31	33	34	36	38	40
3	12	15	18	20	22	25	27	30	32	35	37	40	42	45	47	50	52	55	57
4	18	22	25	28	31	34	37	41	44	47	50	53	56	59	62	66	69	72	75
5	25	29	33	37	41	44	48	52	56	60	63	67	71	75	79	82	86	90	94
6	33	37	42	47	51	56	60	64	69	73	78	82	87	91	95	100	104	109	113
7	42	47	52	57	63	68	73	78	83	88	93	98	103	108	113	118	123	128	133
8	51	57	63	69	75	81	86	92	98	104	109	115	121	126	132	137	143	149	154
9	62	69	76	82	88	95	101	108	114	120	126	133	139	145	151	158	164	170	176
10	74	81	89	96	103	110	117	124	131	138	145	151	158	165	172	179	186	192	199
11	87	95	103	111	118	126	134	141	149	156	164	171	179	186	194	201	208	216	223
12	100	109	118	126	135	143	151	159	168	176	184	192	200	208	216	224	232	240	248
13	115	125	134	143	152	161	170	179	187	196	205	214	222	231	239	248	257	265	274
14	131	141	151	161	171	180	190	199	208	218	227	236	245	255	264	273	282	292	301
15	148	159	169	180	190	200	210	220	230	240	250	260	270	280	289	299	309	319	329
16	166	177	188	200	210	221	232	242	253	264	274	284	295	305	316	326	337	347	357
17	184	197	209	220	232	243	254	266	277	288	299	310	321	332	343	354	365	376	387
18	204	217	230	242	254	266	278	290	302	313	325	337	348	360	372	383	395	406	418
19	225	239	252	265	278	290	303	315	327	340	352	364	377	389	401	413	425	437	450
20	247	261	275	289	302	315	329	341	354	367	380	393	406	419	431	444	457	470	482

(continued)

Table 14.12 (continued)

If the acceptable probability of false positive decision is 5% (α = 0.5)

m	**n = 2**	**n = 3**	**n = 4**	**n = 5**	**n = 6**	**n = 7**	**n = 8**	**n = 9**	**n = 10**	**n = 11**	**n = 12**	**n = 13**	**n = 14**	**n = 15**	**n = 16**	**n = 17**	**n = 18**	**n = 19**	**n = 20**
2	7	9	11	12	14	16	17	19	21	23	24	26	27	29	31	33	34	36	38
3	12	14	17	19	21	24	26	28	31	33	36	38	40	43	45	47	50	52	54
4	18	21	24	27	30	33	36	39	42	45	48	51	54	57	59	62	65	68	71
5	24	28	32	35	39	43	46	50	53	57	61	64	68	71	75	79	82	86	89
6	32	36	41	45	49	54	58	62	66	70	75	79	83	87	91	96	100	104	108
7	41	46	51	56	61	65	70	75	80	85	90	94	99	104	109	113	118	123	128
8	50	56	62	67	73	78	84	89	95	100	105	111	116	122	127	132	138	143	148
9	61	67	74	80	86	92	98	104	110	116	122	128	134	140	146	152	158	164	170
10	73	80	87	93	100	107	114	120	127	133	140	147	153	160	166	173	179	186	192
11	86	93	101	108	115	123	130	137	144	152	159	166	173	180	187	195	202	209	216
12	99	108	116	124	132	140	147	155	165	171	179	186	194	202	209	217	225	233	240
13	114	123	132	140	149	157	166	174	183	191	199	208	216	224	233	241	249	257	266
14	129	139	149	158	167	176	185	194	203	212	221	230	239	248	257	265	274	283	292
15	146	157	167	176	186	196	206	215	225	234	244	253	263	272	282	291	301	310	319
16	164	175	185	196	206	217	227	237	247	257	267	278	288	298	308	318	328	338	348
17	183	194	205	217	228	238	249	260	271	282	292	303	313	324	335	345	356	366	377
18	202	215	226	238	250	261	273	284	295	307	318	329	340	352	363	374	385	396	407
19	223	236	248	261	273	285	297	309	321	333	345	356	368	380	392	403	415	427	439
20	245	254	267	279	291	303	315	327	339	351	363	375	387	399	410	422	434	446	458

Example 14.2 (continued)

Solution:

To analyze the data using the Wilcoxon Rank Sum test, we add the value of DCGL to the background measurement data. This becomes the adjusted reference data. Then we compare the measured residual contamination data with the adjusted reference data. If the measured residual contamination levels are less than the adjusted reference levels, the site meets the compliance requirement, i.e., (contamination level) – (background) < DCGL.

Number	Measured data (Bq/kg)	Data type	Adjusted data	Rank	Residual level data rank	Background data rank
1	182	Background	582	13		13
2	233	Background	633	17.5		17.5
3	233	Background	633	17.5		17.5
4	213	Background	613	14		14
5	228	Background	628	16		16
6	249	Background	649	21		21
7	192	Background	529	9		9
8	239	Background	639	19		19
9	218	Background	618	15		15
10	244	Background	644	20		20
11	540	Residual level	540	10.5	10.5	
12	488	Residual level	488	4	4	
13	508	Residual level	508	6	6	
14	514	Residual level	514	7	7	
15	467	Residual level	467	1	1	
16	539	Residual level	539	10.5	10.5	
17	493	Residual level	493	5	5	
18	545	Residual level	545	12	12	
19	482	Residual level	482	3	3	
20	524	Residual level	524	8	8	
21	477	Residual level	477	2	2	
	Sum =			231	69	162

In this example, the test statistic is 231–66 = 162 (this is equal to the sum of the ranks of the background data). This value is compared to the critical value for the case of m = 11, n = 10, and α = 0.025. Table 14.12 shows that the corresponding critical value is 149.

Since the test statistic (W_t) 162 is greater than the critical value, 149, the survey unit passes the test and is declared compliant.

14.4 Policy Issues in Decommissioning

14.4.1 *Historical Trends in Nuclear Shutdowns*

Since the global nuclear power industry started, 173 nuclear power plant units have been permanently shut-down. These plants were shutdown not necessarily because of aging as some of them were shutdown prematurely. The timing of the shutdown of these reactors varies, but there are a few patterns. As shown in Fig. 14.4, nuclear reactor shutdowns started in the 1960s and continued through the 1970s, meaning these shutdowned reactors were still relatively new.

Historically, there were mainly two peaks of reactor shutdown events. One was around 1990 and the other 2011. The sharp rise in the 1990s was mainly in the U.S., Germany, Russia, and various countries in Europe (Italy, Ukraine, Spain, Sweden, etc.). These shutdowns occurred after the 1986 Chernobyl accidents largely influenced by the anti-nuclear political developments after the accidents. Similar observation can be made for the 2011 peak in association with the Fukushima accidents. This time, the shutdowns occurred mainly in the U.S., Japan, and Germany. In particular, Germany, following the 2011 Fukushima accidents, reinstated their nuclear phase-out policy. The adoption of very strict safety regulations in Japan discouraged many nuclear reactors from restarting after being forced to shutdown in 2011. Poor safety and economic performance of the reactors may have contributed to the decision to shutdown the reactors in the U.S.

There was also a continued rise in shutdowns between 1995 and 2010. This rise may be the result of a mixture of various factors including post-Chernobyl developments including the adoption of stricter safety regulations, decline in public support for nuclear power, governments' nuclear phase-out decisions. Retirement of U.K.'s Magnox reactors also contributed to this trend.

Another important trend hidden in the figure was the delayed shutdown of U.S. nuclear reactors. People were expecting a sharp rise in nuclear shutdowns in the U.S. in the 2000s as many of the reactors were built in the 1960s and 1970s with a 40-year operating license. However, deregulation of the electricity market in the mid to late 1990s in the U.S. pushed the merger of utility companies resulting in large utility companies dominating the market. After the acquisitions, the large utility companies were able to invest in enhancing the safety and operating performance of older nuclear units and became successful in extending the licensed operating periods of the acquired nuclear units. This effectively pushed the curve of the reactor shutdowns toward the right.

In recent years, reactor shutdowns in the U.S. are on the rise. This trend reflects the aging of many of the operating reactors. The trend is also influenced by the very low price of natural gas which makes the operation of old nuclear units economically non-competitive in the electricity market. The U.S. utilities have announced the permanent shutdown of 18 nuclear reactors since 2013. Since then, the decision on five of these units were reversed (FitzPatrick in New York, Clinton and Quad Cities-1 & -2 in Illinois and Palisades in Michigan). The reversal was due to legislative

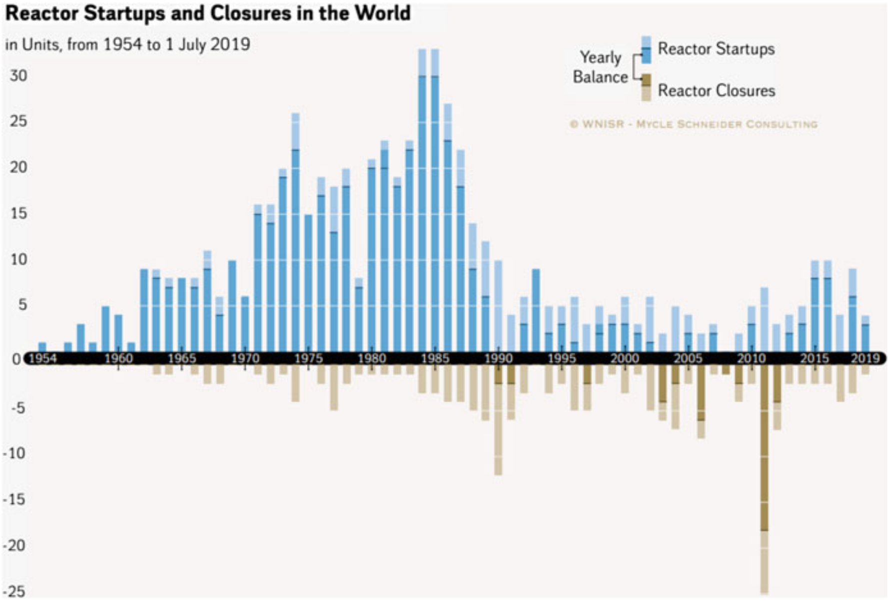

Fig. 14.4 Worldwide reactor startups and closures (1954 – July 2019), where closure means no more energy production (not permanent shutdown). (Source: Schneider et al. 2019)

efforts by the states, to provide zero carbon emission credits to nuclear power plants as part of the plan for greenhouse gas reduction. Under such legislation, nuclear utilities received financial support for the continued operation of uneconomic nuclear units.

These observations indicate that safety and economic performance of nuclear reactors are very important part of nuclear reactor shutdown decisions. Also government policy driven by social and political factors have a large impact on the shutdown decision.

Table 14.13 shows the history of U.S. nuclear reactor shutdowns starting in 1963 through 2018 (based on the data from NEI).

14.4.2 Selection of Nuclear Decommissioning Strategies

14.4.2.1 Selection of Decommissioning Options

Among the 173 shutdown reactors worldwide, 115 units have gone through decommissioning while the rest are awaiting a decision on which specific option to pursue. As discussed, there are mainly two options for decommissioning today, i.e., DECON (immediate dismantlement) and SAFSTOR (deferred dismantlement).

Historical data indicates that SAFSTOR was preferred in many cases while DECON was used only in a few countries (e.g., Germany, the U.S.).

Selection of a decommissioning option is affected by a large number of factors (Suh et al. 2018). These factors include available funds, the expected hazards to workers, the public attitude and opinion, availability of radioactive waste disposal facilities, availability of qualified workers and corporate memory retention, expected cost and time requirements for the project, expected changes in the regulatory requirements, and the concern over environmental protection. In particular, availability of radioactive waste disposal facilities is likely to support DECON which has been the case in the U.S. Other factors related to reactor characteristics, such as operating periods, reactor types, and safety performance, are also important in the selection of an option.

14.4.2.2 Selection of the End-State of Decommissioning

Regarding the choice of the end-state (i.e., greenfield vs. brownfield), the decision is also influenced by various national, local, and plant specific factors. Along with the aforementioned factors, national policy on the future use of nuclear power, safety performance record of the reactor, presence of multiple units at a site, the societal issues associated with the reason for reactor shutdown, and availability of a highly trained nuclear workforce are also expected to affect the decision.

The role of these factors in the selection of decommissioning options can be understood by examining historical cases in various countries (Suh et al. 2018). For example, when nuclear energy was positively accepted by the public, SAFSTOR and brownfield have been preferred. Favorable public support for nuclear energy may translate into less demand for immediate or complete removal of the nuclear legacy from the site. Also if the public perceives significant economic benefit of site reuse, the likely decision is to select brownfield as such reuse can improve the standard of living for future generations. If the level of public acceptance of nuclear energy is low, such consideration would play a minor role. Moreover, the perceived benefit would be offset by the public's concern over radiation risk. In this case, any levels of residual radiation may not be acceptable to the public resulting in the demand for complete removal of radioactive materials from the site. Also, a strong anti-nuclear movement may push for the DECON strategy and the greenfield.

The presence of multiple units at a site is likely a motivator to select SAFSTOR and brownfield. In this situation, minimizing the impact of the decommissioned unit on the other operating units becomes an important consideration. The utility would prefer to optimize the usefulness of the decommissioning project by turning it into an asset, for example by building an independent spent fuel storage installation (ISFSI) to support the operation of the existing units. Since the decommissioned unit may not be ready for an unrestricted release, it will beneficial to put the decommissioned unit under active security surveillance in connection with performing support services for the other unit(s). In this case, reusing the site even with restrictions on the permit would be preferred.

Table 14.13 History of U.S. nuclear reactor shutdowns

Reactor name	Type	MWt	Location	Operating license	Shutdown	Status
GE VBWR	BWR	50	Pleasanton, CA	8/31/1957	12/9/1963	SAFSTOR
Hallam	SCGM	256	Hallam, NE	1/2/1962	9/1/1964	ENTOMB
Piqua	OCM	46	Piqua, OH	8/23/1962	1/1/1966	ENTOMB
CVTR	PTHW	65	Parr, SC	11/27/1962	1/1/1967	DECON completed
Pathfinder	BWR	190	Sioux Falls, SD	3/12/1964	9/16/1967	DECON completed
Elk River	BWR	58	Elk River, MN	11/6/1962	2/1/1968	DECON completed
Bonus	BWR	50	Punta Higuera, PR	4/2/1964	6/1/1968	ENTOMB
NS Savannah	PWR	74	Baltimore, MD	8/1/1965	11/1/1970	SAFSTOR
Saxton	PWR	24	Saxton, PA	11/15/1961	5/1/1972	DECON completed
Fermi 1	SCF	200	Newport, MI	5/10/1963	9/22/1972	SAFSTOR
Indian Point 1	PWR	615	Buchanan, NY	3/26/1962	10/31/1974	SAFSTOR
Peach Bottom 1	HTG	115	Peach Bottom, PA	1/24/1966	10/31/1974	SAFSTOR
Humboldt Bay 3	BWR	200	Eureka, CA	8/28/1962	7/2/1976	DECON in Progress
Dresden 1	BWR	700	Morris, IL	9/28/1959	10/31/1978	SAFSTOR
Three Mile Island 2	PWR	2770	Middletown, PA	2/8/1978	3/28/1979	SAFSTOR
Shippingport	PWR	236	Shippingport, PA	NA	1/1/1982	DECON completed
LaCrosse	BWR	165	Genoa, WI	7/3/1967	4/30/1987	DECON
Rancho Seco	PWR	2772	Herald, CA	8/16/1974	6/7/1989	DECON completed
Shoreham	BWR	2436	Wading River, NY	4/21/1989	6/28/1989	DECON completed
Fort St. Vrain	HTG	842	Platteville, CO	12/21/1973	8/18/1989	DECON completed
Yankee Rowe	PWR	600	Franklin Co., MA	12/24/1963	10/1/1991	DECON completed
Trojan	PWR	3411	Ranier, OR	11/21/1975	11/9/1992	DECON completed
San Onofre 1	PWR	1347	San Clemente, CA	3/27/1967	11/30/1992	SAFSTOR
Zion 2	PWR	3250	Zion, IL	11/14/1973	9/19/1996	DECON in Progress
Haddam Neck	PWR	1825	Meriden, CT	12/27/1974	12/5/1996	DECON completed

(continued)

Table 14.13 (continued)

Reactor name	Type	MWt	Location	Operating license	Shutdown	Status
Maine Yankee	PWR	2700	Wiscasset, ME	6/29/1973	12/6/1996	DECON completed
Zion 1	PWR	3250	Zion, IL	10/19/1973	2/21/1997	DECON in Progress
Big Rock Point	BWR	240	Charlevoix, MI	5/1/1964	8/29/1997	DECON completed
Millstone 1	BWR	2011	Waterford, CT	10/31/1986	7/21/1998	SAFSTOR
Crystal River 3	PWR	2609	Crystal River, FL	12/3/1976	2/20/2013	SAFSTOR
Kewaunee	PWR	1772	Kewaunee, WI	12/21/1973	5/7/2013	SAFSTOR
San Onofre 2	PWR	3438	San Clemente, CA	2/16/1982	6/7/2013	DECON in Progress
San Onofre 3	PWR	3438	San Clemente, CA	11/15/1982	6/7/2013	DECON in Progress
Vermont Yankee	BWR	1912	Vernon, VT	3/21/1972	12/29/2014	SAFSTOR
Fort Calhoun	PWR	1500	Ft. Calhoun, NE	8/9/1973	10/24/2016	SAFSTOR
Oyster Creek	BWR	1930	Forked River, NJ	7/2/1991	9/17/2018	SAFSTOR

Source: NEI; https://www.nei.org/resources/statistics/decommissioning-status-for-shutdown-us-plants

In the case of operating periods, SAFSTOR and "greenfield" may be preferred with longer operating periods. If the operating periods are long, the contamination levels in the plant will be higher thereby raising the cost of DECON due to high radiation exposure to workers. If the operating periods of a reactor are longer than 30 years, plant workers familiar with site operations and contamination episodes may not be available for the tasks of decommissioning due to retirement. This reduces the benefits of pursuing immediate dismantling and can favor the SAFSTOR option. Also longer operating periods may mean greater monetary gain from the plant operation and potentially a more soluble decommissioning fund to support the project.

With respect to the role of operating history, experiencing a major nuclear accident during the plant's operating history is likely to lead to brownfield. This may be due to practical difficulties in completely removing contamination from the site. It could also be related to the public's elevated concern over the level of residual radioactivity at the site from their experiences of the accident. So far the reactors with major accidents, i.e., the TMI unit 2 in the U.S. and the nuclear units in Fukushima Daiichi are under SAFSTOR while the Chernobyl unit 4 is undergoing entombment.

The *type of reactor* to be decommissioned is expected to have a high correlation with the decommissioning decisions. History tells that decommissioning of non-LWR type reactors, such as gas cooled reactors, graphite moderated reactors (RBMK), and fast breeder reactors were placed under SAFSTOR. These reactors have elevated levels of contamination and require long periods of radioactive decay or careful demolition/decommissioning preparations. Decommissioning of these reactors mostly occurred in Germany, the U.K., France, Japan, and the former Soviet Union. In Germany and the U.S., greenfield was mostly advocated while in the former Soviet Union, France, and Japan, no decisions or brownfield have been selected.

Having a high decommissioning experience level so far has resulted in a preference toward DECON and greenfield in the U.S. and Germany. Nonetheless, country-specific issues may not be necessarily represented in these examples. So far, the availability of nuclear waste facilities was not found to be a significant factor in the choice between greenfield and brownfield.

14.4.3 Examples of Nuclear Decommissioning in the U.S.

So far, the largest number of nuclear power plant decommissioning occurred in the U.S. As of 2017, ten commercial nuclear power plants have completed decommissioning. These reactors include Shippingport (1957–1989), Yankee Rowe (1957–1992), Big Rock (1962–1999), Pathfinder (1966–1967), Connecticut Yankee (1968–1996), Maine Yankee (1972–1997), Fort St. Vrain (1979–1989), Rancho Seco (1975–2009), Trojan (1976–1992), and Shoreham (1986–1989) in the order of plant operating dates (year commissioned – year decommissioned)

(Schneider et al. 2019). Today, these ten commercial nuclear power plants either have achieved license termination with full site release or have their only remaining responsibility as managing their Independent Spent Fuel Storage Installation (ISFSI).

The Shippingport nuclear power plant in Beaver County (near Pittsburgh), Pennsylvania was the world's first full-scale nuclear power plant dedicated only to commercial operations. It was a 60 MWe PWR (light water moderated with a Breeder reactor), starting its operation in December 1957 and was shutdown in October 1982. The plant went through DECON between 1985 and 1988. The site was released for unrestricted use in December 1989. It was a test case demonstration of decommissioning to prove that a nuclear reactor can be safely decommissioned and achieve a greenfield designation.

While Shippingport can be considered a demonstration reactor of PWR, Yankee Rowe was the "first fully commercial PWR". It was a 167 MWe PWR, located in Rowe, Massachusetts, starting its commercial operation on November 1960 and was voluntarily shutdown February 1992. With a lifetime capacity factor of 74%, it was one of the best performing commercial nuclear plants in the world. The reason for the shutdown was a concern over reactor pressure vessel embrittlement from neutron exposure. The plant went through DECON and its license was terminated in 2007 with the site designated as greenfield.

Big Rock Point was a 67 MWe BWR located near Charlevoix, Michigan. It was commissioned in March 1963, and decommissioned in August 1997 after 35 years of operation. The plant was shutdown 3 years before the end of operating license period due to low financial benefit of continued operation. After DECON operations, eight spent fuel casks remain onsite. The remainder of the decommissioned site is now a state park.

The Pathfinder plant was a 59 MWe BWR located near Sioux Falls, South Dakota. It was commissioned in July 1966 as a 'proof of concept' nuclear plant for steam superheating. However, technical difficulties led to the plant's retirement in October 1967. The plant was then converted to a gas/oil fired plant in 1968 which operated until 2000.

The Connecticut Yankee plant located in Haddam Neck, Connecticut (also called the Haddam Neck plant), was a 560 MWe PWR. It was commissioned in January 1968 and decommissioned in December 1996. The plant's lifetime capacity factor was 73.5%. The plant was shutdown as its operation was no longer cost effective and the site was returned to greenfield through DECON.

Maine Yankee was a 860 MWe PWR plant located in Wiscasset, Maine. The plant operated from December 1972 till its retirement in August 1997 with a lifetime capacity factor of 68.2%. The plant was shutdown over safety concerns. When the cost of addressing those concerns became too expensive, the plant went through DECON and returned to greenfield.

The Fort St. Vrain Generating Station, located in Platteville, Colorado was a 330 MWe HTGR. It operated from 1979 untill 1989. As the first commercial gas cooled reactor in the U.S., the plant experienced numerous problems throughout its operating periods (e.g., water infiltration, corrosion, electrical system issues, etc.).

Operation and maintenance of the plant became very expensive which led to its decommissioning and conversion to a gas turbine plant in 1996.

The Rancho Seco plant was a 913 MWe PWR located in Clay Station, near Sacramento, California. The plant started commercial operation in April 1975. Three years into its operation (on March 20, 1978), the plant experienced steam generator dryout due to a failure in the power supply of the plant's non-nuclear instrumentation system. This raised significant safety concerns over the plant's operation. The plant continued its operation while experiencing multiple annual shut-downs, cost overruns, mismanagement, and incidents involving radioactive steam releases. The plant was shutdown by public vote on June 7, 1989 over concerns of nuclear safety, with a lifetime capacity average of only 39%. The plant was decommissioned using DECON and the majority of the site was released for unrestricted public use in 2009. A small part of the site containing spent fuel and LILW remains under an NRC license.

Trojan was a 1095 MWe PWR, located in Rainier, Oregon. The plant was commissioned in May 1976 and continued its operation until November 1992 with a lifetime capacity factor of 53.6%. The plant experienced steam generator tube leaks. The plant owner estimated that the cost of repairs and related delays would result in the plant's not being economically competitive against the plentiful hydroelectric power available in the region. As a result, a decision to early close the plant was made. Adverse public sentiment towards the plant's continued operation also affected the decision. The plant went through DECON which was completed with license termination in 2008.

The Shoreham plant was a 809 MWe BWR, located in Wading River, Long Island, New York. It was commissioned in August 1984 and in 1986 the Long Island Lighting Company received federal permission to conduct a low-power (5%) test. From the beginning, local public opposition to the plant was severe causing extensive delays in licensing and authorization for commercial operation. In 1989, the concern over severe accident made the local community refuse to sign the plant's evacuation plan. In addition, New York's new governor ordered state officials not to approve the evacuation plan. Then, the owner of the plant agreed not to operate the newly built plant under an agreement with the state. Most of the cost-recovery to the power company was by passing the cost to Long Island residents. The plant was decommissioned in 1994 and was converted to a gas turbine plant in 2002.

14.5 Conclusion

The current status of NPP decommissioning indicates that no insurmountable problems exist in decommissioning of commercial power reactors using present day technologies. The radiological and industrial hazards associated with decommissioning can be significant but can be managed. Such hazards level depends on the number of years of operation, the plant type, the experiences (e.g., spills, leaks, accidents) occurred during the operation of the plant. This chapter presented the options in NPP decommissioning along with the discussions of the necessary steps to take, available technologies, and considerations to support the decisions. In terms of how decommissioning a facility is done, the options are between DECON (decommissioning through immediate dismantlement) and SAFSTOR (delayed dismantlement and decommissioning) as well as the choice between unrestricted and restricted site release. The DECON and SAFSTOR options have different implications in terms of worker dose and resulting costs. The DECON option involves higher worker dose and perhaps higher project costs but may be preferred if the necessary funds and waste disposal spaces are available. In SAFSTOR, the activity of the plant is allowed to decay over years, resulting in lower radiation fields, and thereby reducing workers' radiation exposure but requiring the cost of long-term monitoring and surveillance. The decision between restricted and unrestricted site release is affected by national, local, and plant specific factors. Regardless of the option selected, each facility will have regulatory, legal, financial, industrial and environmental responsibilities which must be demonstrated at every step of the decommissioning processes. Sufficient transparency must exist to ensure the public to have confidence in the processes. The lessons learned from the previous decommissioning experiences and the status of technology developments should be actively shared to support worldwide success in NPP decommissioning.

Homework

Problem 14.1: Describe the key features of DECON, SAFSTOR, and Entombment and the differences among them. List key nuclides as the source of radiation exposure to workers with each approach.

Problem 14.2: Through a final status survey of a site of a nuclear power plant under decommissioning, the following concentrations of radionuclides are found. Assuming that there are no other radionuclides present in the site soils above the background, determine if the site is ready for unrestricted release.

Radionuclide	Residual concentration (Bq/kg)	Background (Bq/kg)
H-3	350	3
C-14	320	5
Fe-55	200	
Co-60	50	
Ni-63	200	
Sr-90	10	
Cs-137	10	

Problem 14.3: The following 20 measured data were obtained as the residual concentration of ^{14}C of a survey unit at a nuclear power plant site.

Measured data of the residual activity of ^{14}C in the site soil (Bq/kg):

352, 451, 477, 437, 470, 497, 436, 375, 383, 454, 410, 404, 428, 422, 363, 459, 392, 370, 408, 446.

The measured background levels of ^{14}C in the soil of the reference area are as follows:

4.1, 4.9, 4.8, 3.5, 5.0, 4.8, 5.1, 4.0

By controlling the probability of false positive to be less than 2.5%, determine if the survey unit is ready for release. Use the U.S. screening level (DCGL) for the compliance determination.

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Chapter 15
Cross-Cutting Systems Issues: Economics, Nuclear Nonproliferation and Security

Abstract Cost consideration is an important part of nuclear waste management. Also, due to the presence of special nuclear materials in spent nuclear fuel, the issues of nuclear nonproliferation and security must be addressed in nuclear waste management. These issues are particularly salient if the scheme of spent fuel recycling is adopted. This chapter describes how cost of spent fuel recycling is estimated in relation to various options of nuclear fuel cycle. International approaches to nuclear nonproliferation and security are also discussed along with the examination of the role of national policy in addressing these cross-cutting issues.

Keywords Fuel cycle economics · Proliferation resistance · Nuclear safeguards · Nuclear security · Determinants of reprocessing policy

In addition to the technical issues discussed in the previous chapters, management of nuclear waste requires consideration of cross-cutting system-wide issues. One of the key system-wide issues to be examined is economics as cost is an important factor to consider in the management of nuclear waste. Also, in connection with the presence of special nuclear materials in spent nuclear fuel, the issue of nuclear nonproliferation and security must be carefully addressed in nuclear waste management. These issues are closely connected to the specifics of a country's nuclear fuel cycle capabilities with respect to spent fuel recycling. For example, establishment of infrastructure for fissile separation from spent fuel and fuel refabrication would be under heavy scrutiny of international nuclear governance regime.

This chapter examines how economics of nuclear waste management is aligned with various options of nuclear fuel cycle. The concept of nuclear nonproliferation and security is discussed within the existing international governance framework. National policy consideration and their impacts on the choice of nuclear fuel cycle are also discussed.

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15.1 Economics of Nuclear Fuel Cycles

15.1.1 The Concepts and Implications of Spent Fuel Recycling

15.1.1.1 Fuel Cycles Concepts Involving Spent Fuel Reprocessing

The spent fuel from an LWR contains 0.7 ~ 0.8% ^{235}U and about 1% plutonium (see Sect. 7.1). While these fissile materials are thrown away in the once-through cycle (also called the open cycle), a fuel cycle based on recycling of spent fuel has also been implemented. Such spent fuel recycling-based fuel cycle is further divided depending on the degree of recycling, the modified open cycle or the closed cycle. In the modified open cycle, a limited recycling is done, with at least one re-burn of the fissile materials typically in a LWR. In the closed cycle, the recycling is done to its fullest limit, with full recycling of fissile materials through repeated re-burns of fissile. This scheme is based on the use of a fast reactor. Achieving highest degree of uranium resource utilization is pursued with the closed cycle.

The current commercial implementations of spent fuel recycling in the world are based on recycling spent fuels only once, i.e., the modified open cycle, by using the PUREX technology (see Chap. 8). This is also called the twice-through cycle. The routine recycling of spent fuel in PWRs in France is an example of this. In France, extracting more energy at least once from spent fuel is found to providing enough economic gain to sustain the program.

The closed fuel cycle is to perform multiple recycling of spent fuels using fast reactors until all the fissile and fissionable materials with a fuel value are fully utilized. According to the scheme, only fission products and some minor actinides are disposed of as waste in the end. Using a fast reactor is required in this case for repeated recycling as the fast neutron spectrum enables the fission of various fissionable elements. Using fast reactors also enables transmutation of most actinides through neutron irradiation. No commercial implementation of the closed cycle has been demonstrated yet.

As the modified open cycle or the closed cycle requires the infrastructure for spent fuel recycling, key differences in nuclear fuel cycle options lie in the back-end. As discussed in Sect. 6.1, the back-end nuclear fuel cycle in the open cycle (OC) covers storage, transportation, and disposal of spent fuel. In the modified open or the closed cycle, the back-end includes treatment (reprocessing) of spent nuclear fuel for fissile recycling, re-conversion and enrichment of uranium, and fabrication of uranium and plutonium into mixed (i.e., plutonium and uranium mixed) oxide (MOX) fuel. Refabricated MOX fuel can be utilized either in a thermal or fast reactor.

The schematics of different fuel cycle concepts are depicted in Fig. 15.1. In the twice-through fuel cycle, LWR spent fuel is reprocessed and plutonium and uranium are separated from minor actinides and fission products. The separated plutonium is used for MOX fuel fabrication. The produced MOX fuel is fed to an LWR for

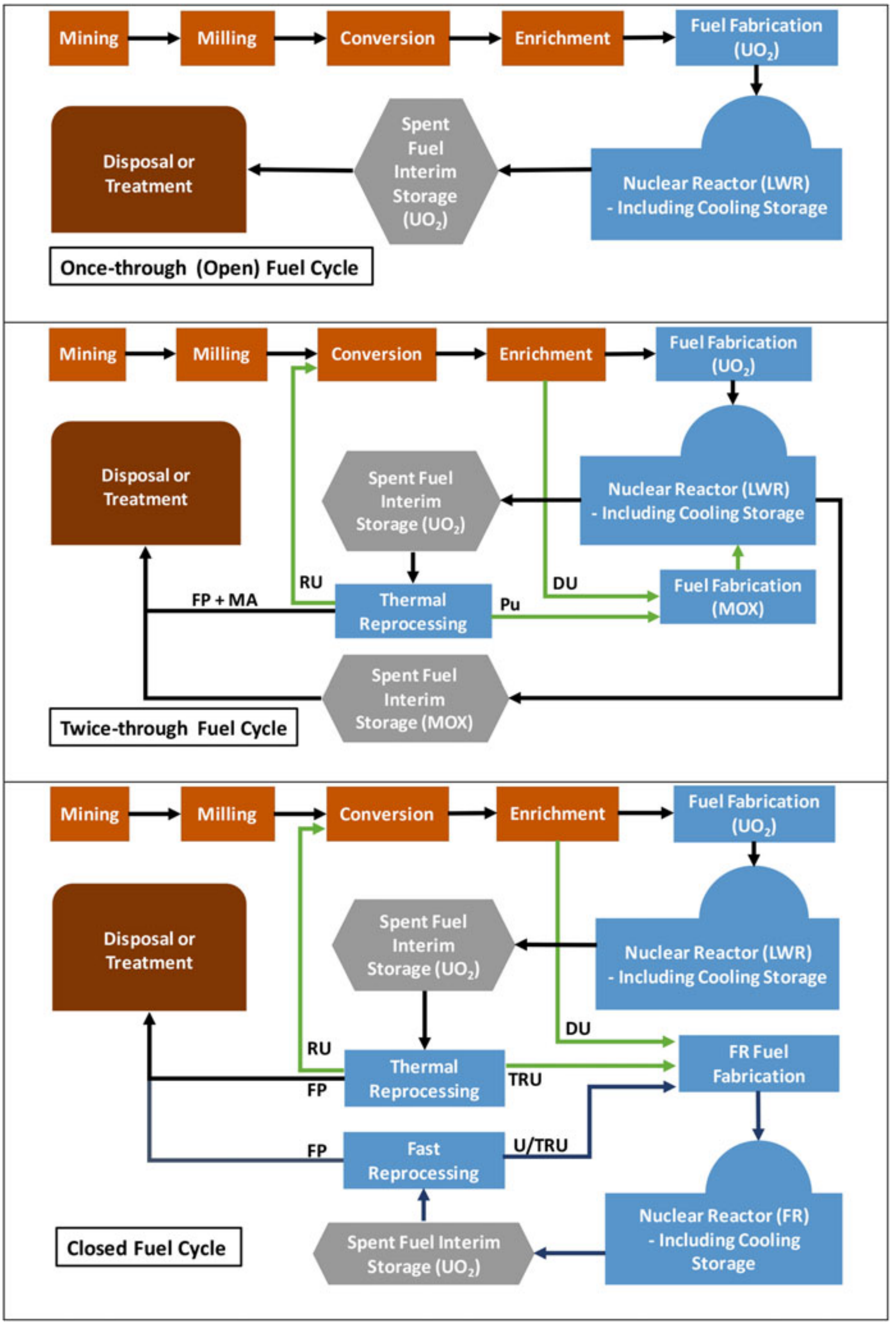


Fig. 15.1 Schematics of nuclear fuel cycles

electricity generation. The separated uranium goes through conversion and enrichment and is used as UO_2 fuel for an LWR. The LWR allows an option of using only the MOX fuel or the mixture of MOX and UO_2 fuel to support reactor operation. The separated minor actinides and fission products are stored and ultimately disposed of in a geological repository. With recycling of spent fuel, volume reduction of nuclear waste to be sent to a geological repository is achieved.

In the case of the closed fuel cycle, the separated plutonium and the minor actinides from the reprocessing of LWR spent fuel are mixed and fabricated into a fuel for a fast reactor. The separated uranium goes through conversion and enrichment for new UO_2 fuel. The separated fission products are disposed of as waste. The spent fuel from the fast reactor is also reprocessed and the separated uranium and plutonium and the minor actinides are refabricated into a new fuel. Thus the fuel for a fast reactor includes the plutonium and the minor actinides from LWR spent fuel as well as uranium, plutonium, and the minor actinides from fast reactor spent fuel. This scheme is repeated until the depletion of fuel materials in the spent fuel. Compared to the twice-through cycle, further reduction in the volume of nuclear waste to be sent to a geologic repository is achieved with the closed cycle.

15.1.1.2 Spent Fuel Recycling without Reprocessing

It is conceivable to construct the modified open cycle without relying on spent fuel reprocessing. In this case, nonconventional ways of fuel recycling can be employed. These nonconventional ways include the DUPIC cycle or melt-refining. In both cases, separation of fissile is not necessary.

The DUPIC (Direct Use of Pressurized water reactor spent fuel In CANDU) cycle refers to recycling of light water reactor (LWR) spent fuel in a CANDU reactor. In DUPIC, LWR spent fuels are dry-processed with oxidation of UO_2 to U_3O_8 at $400 \sim 500\ ^\circ C$ which then is reduced to UO_2 powder at $600 \sim 850\ ^\circ C$. In the process, volatile fission product gases are removed and uranium, plutonium, fission products and the minor actinides are all kept together in the fuel powder and repackaged into a new fuel bundle to be loaded to a CANDU reactor.

The melt-refining (Burris 1959) technology refers to melting of spent fuel mixture materials followed by re-solidification. In the process, the spent fuel is disassembled, chopped and decladded into zirconia crucible and heated up to 1400 °C to achieve melting of the mixture. The gaseous and volatile fission products are released and some solid fission products are partially removed through melting. The remaining elements in the crucible are solidified through injection casting into fuel. The resolidified fuel is then used in a fast reactor.

The DUPIC fuel cycle concept was proposed, developed, and demonstrated by the Republic of Korea (ROK). Implementation of the DUPIC cycle, however, has not been pursued as building new CANDU reactors discontinued in the ROK after the installation of four units. To operate the DUPIC cycle, one CANDU reactor is needed to support two large LWRs under an equilibrium situation. As of 2020, the

ROK operates 21 PWRs and 3 CANDU reactors. No other countries have shown serious interest in DUPIC.

Melt refining was used in the U.S. from 1964 through 1969 at Argonne National Laboratory for the purpose of recycling the spent fuels from EBR-II, one of the prototype fast breeders. Recently, TerraPower and Argonne National Laboratory are reexamining the option as part of fuel fabrication strategy for the Breed and Burn Reactor. The method is also considered for high burnup fuel application in a fast reactor to improve fuel integrity.

15.1.1.3 Fast Reactors for Spent Fuel Recycling

Use of fast reactors as part of the nuclear fuel cycle has been envisioned from the beginning of nuclear technology development. Initially, the purpose of fast reactor was to breed new fuel. Fast reactors produce higher number of neutrons per fission compared to thermal reactors. The additional neutrons available in a fast reactor beyond what is needed to maintain self-sustaining fission chain reactions are used to convert fertile into fissile, i.e., to breed plutonium.

Also, with no moderator, the neutrons in fast reactors stay fast, which enables fissioning of most minor actinides. Therefore, the minor actinides are effectively utilized in a fast reactor as fuel allowing transmutation of them while producing energy.

As discussed in Chap. 8, a fast reactor can achieve either fuel breeding or waste transmutation. This difference can be quantified by using term called conversion ratio. The conversion ratio (CR) refers to the ratio of the amount of fissile produced to the amount of fissile consumed in a reactor. If the CR is less than one, the reactor is called a burner with the rate of consumption of fissile faster than its production. In this case, the reactor is a device for transmutation. As mentioned in Sect. 8.4.3, Generation IV reactors are developed as burners for the purpose of nuclear waste transmutation. If the CR is higher than one, the reactor is a breeder and produces more fissile than what is consumed. If the CR is equal to one, the rate of production of fissile is the same as the rate of its consumption and, the reactor runs in a self-sustaining manner.

Fast reactors were developed by a number of countries, mostly for a breeder application. For example, countries like the U.S., Russia, France, the U.K., Japan, Germany, India, and China have designed, constructed, and operated fast reactors at different stages of development including research reactors, experimental reactors, demonstration or prototype reactors, and the commercial scale reactors. Out of these efforts, three commercial scale fast reactors emerged in the world, such as Superphenix reactor of France (1240 MWe, 1985–1996), and BN-800 and BN-1200 of Russia. Currently, only one commercial scale fast reactor, BN-800, is under operation (864 MWe, 2014-present). BN-1200 (1220 MWe) is also under construction in Russia with expected operation in 2025. About 400 reactor-years of fast reactor operation has accumulated in the world from these developments. All of the commercial scale fast reactors are sodium-cooled.

Sodium, the common coolant used in fast reactors, is an excellent heat transfer material and is used under the atmospheric pressure with no pressurization requirement. Sodium is also relatively cheap and inert and compatible with the fuel and structural materials even at high temperatures. However, in the event of sodium leakage, a violent reaction between sodium and water (or air) takes place which can lead to explosion or a fire. To eliminate such concern, a sealed coolant system has been implemented. The sodium-cooled fast reactor operates using either oxide fuel or metallic fuel achieving reactor coolant outlet temperature of 510–550 °C.

Under the current Generation IV nuclear reactor development (GIF 2020), the global collaborative effort of developing advanced nuclear reactors, including sodium cooled fast reactor (SFR), is underway. Other types of advanced reactors developed under the GIF effort include gas-cooled fast reactor (GFR), lead-cooled fast reactor (LFR), and molten-salt fast reactor (MSFR).

The GFR uses helium as coolant and uses composite ceramic fuel and operates at about 5 MPa (~47 times atmospheric pressure). Helium is chemically inert, good for high temperature application, compatible with other reactor materials, and has low neutron cross section. With the availability of high temperature coolant with a reactor outlet temperature of 850 °C, cogeneration of electricity and hydrogen is possible. The main challenge with GFR is the reactor materials compatibility issue at high temperature.

The LFR uses molten lead or lead-bismuth eutectic (LBE) as coolant with the loading of metallic or nitride fuel, operating at atmospheric pressure. It has reactor outlet coolant temperature of 500–550 °C (with possibility of going up to 800 °C). Lead features excellent cooling properties with low neutron cross section and no concern of coolant boiling. However, it has to be maintained at high temperature (above 400 C) to prevent its solidification and lead is opaque not allowing visual inspection and monitoring. LBE is an alternative coolant to lead with lower (about 200 °C) operating temperature requirement. The primary challenge with LFR is corrosiveness of coolant. Also, the LFR system gets contaminated by polonium during operation which presents hazards to workers through inhalation. Polonium is an activation product of bismuth which is present as component of coolant or impurities in lead.

In MSFR, molten salt is a coolant and at the same time a fuel containing dissolved fissile materials. With homogeneous isotopic composition of fluid fuel, MSFR provides unique capability for actinide burning for fuel recycling. A MSFR operates under atmospheric pressure with large negative reactivity coefficient (i.e., decreasing reactivity with temperature increase) and core outlet temperature of 700–800 °C. Possible diversion of fluid fissile is a major concern with MSFR along with corrosiveness of salt.

Table 15.1 Comparison of nuclear fuel cycles with respect to reduction in natural uranium demand and repository capacity requirement to support a nuclear fuel cycle

	Once-through cycle	Twice-through cycle	Closed cycle		
			CR = 0.75	CR = 1.0	CR = 1.23
Reduction in natural uranium demand to support a nuclear fuel cycle	0% (the base case)	16.1%	32.9%	43.5%	46.6%

Source: Kazimi (2011)

15.1.1.4 Implications of Spent Fuel Reprocessing on Uranium Resource Utilization

The twice-through cycle or multiple recycling of spent fuel with the closed cycle, improves uranium resource utilization, thus reduces the demand for uranium for fuel production. Table 15.1 shows how much reduction in natural uranium demand can be achieved with different fuel cycle options (Kazimi et al. 2011). The comparison is based on operating nuclear power program for a period of 100 years using specific type of fuel cycle to provide the same amount of electricity. The twice-through cycle provided 16.1% reduction in natural uranium demand while the closed cycle provided 32.9–46.6% reduction. The variation in the result for the closed cycle are due to the different CR valued assumed with the fast reactors. Apparently, the higher the value of CR, the higher the reduction in uranium demand is.

If the price of uranium remains low with no concern over long-term supply of uranium, there is no economic incentive for spent fuel recycling. This is the case of today’s nuclear industry. Global inventory of uranium to support nuclear reactor operations is not expected to be constrained soon. With the current size of global fleet of nuclear reactors, the annual demand for natural uranium is about 60,000 ton. With the current worldwide uranium resource estimated at about 15 million ton, the annual demand can be met for about 250 years into the future (NEA 2001). This assumes no growth in nuclear power generation. The concern of long-term uranium supply is also alleviated as uranium is available from the seawater providing additional 4 billion tons. With the current trend of low cost stable uranium supply continuing, most countries are expected to operate their nuclear power program under the once-through cycle.

15.1.1.5 Implications of Spent Fuel Reprocessing on Repository Space Utilization

In relation to the economics of nuclear fuel cycle, an issue to be considered is geological repository space utilization. This is because spent fuel recycling has a significant impact on the utilization of the available repository space. With very high cost of building a geological repository, effectively utilizing the available repository disposal space for spent fuel or HLW disposal can have an impact on the overall cost of the nuclear fuel cycle.

Capacity of a geological repository is a measure of how much mass (as metric ton) of nuclear waste can be accommodated in the facility. While a large size facility would generally mean large capacity, what actually determines the capacity of a repository is the total heat inventory to be contained in the repository within the given footprint size. If nuclear waste contains low heat content, more waste can be accommodated in the given repository space. Accordingly, reducing the heat content of nuclear waste can effectively increase the capacity of a repository.

As discussed in Sect. 10.4, important consideration in the design of a geological repository is to limit the temperature of the rocks surrounding nuclear waste. The purpose of using rock temperature limits is to maintain the physical integrity of the waste packages and the rocks of the repository systems, therefore, not to compromise the performance of waste packages as waste isolation barrier. These limits can be met by constraining the total amount of heat to be kept in a repository.

The thermal energy carried by delayed beta particles and photons released from the disintegration of fission products is the source of the heat in spent nuclear fuel or HLW. As discussed in Sect. 7.1.4, the decay heat from a typical LWR spent fuel is dominated by the radionuclides such as ^{90}Sr/^{90}Y, ^{137}Cs/^{137m}Ba, during the first 100 years after reactor discharge. After ~100 years, the contributions from ^{90}Sr/^{90}Y and ^{137}Cs/^{137m}Ba gradually decrease and ^{241}Am ($t_{1/2} = 432$ years) emerges as the dominant heat producer until ~1000 years. Beyond 1000s of years, plutonium isotopes (e.g., ^{240}Pu, ^{239}Pu) and curium become the important source of decay heat production.

The opportunity to selectively remove these high heat generating radionuclides is provided by spent fuel reprocessing. Once these high heat generating radionuclides are removed, the repository can take more HLW (in terms of mass) into the available disposal space.

Table 15.2 shows the effect of removing high heat generating fission products and actinides through spent fuel recycling on the capacity of a geological repository. While the specifics of the results vary depending on the details of the recycling strategies (e.g., types of fuel, reactor, separation schemes, etc.) or the rock types of the geological repository, the closed cycle is shown to have the largest impact on reducing repository space requirements (about 52–71% reduction). With the twice-through cycle, 4–8% reduction in repository space requirements is expected.

Table 15.2 Comparison of nuclear fuel cycles with respect to repository capacity requirement

	Once-through cycle	Twice-through cycle	Closed cycle
Expected reduction in the repository space requirements	0% (the base case)	4 ~ 8%	52 ~ 71%

Source: NEA (2006), Wigeland and Bauer (2004)

15.1.2 Comparison of Economics of Nuclear Fuel Cycles

Better utilization of uranium resource reduces the demand for uranium mining and eventually reduces the mass of materials to be disposed of as waste. Reduction of decay heat in spent fuel through selective removal of high heat generating nuclides allows a geological repository to support higher amount of electricity generation per the given footprint size of the repository. These observations indicate that different degrees of uranium resource and repository space utilization will have different economic implications of nuclear fuel cycle. At the same time, spent fuel recycling using reprocessing demands additional processing and handling of the highly radioactive materials with addition costs involved.

For instance, to produce one ton of MOX fuel from recycling of spent fuel from LWRs, about 7 tons of spent fuel need to be reprocessed (Kazimi et al. 2011). If the cost required for spent fuel reprocessing is higher than the reduction in cost achieved by better uranium and repository space utilization, the scheme of spent fuel recycling does not present economic incentive. To examine economic advantage or disadvantage of spent fuel recycling, systematic evaluation of the economics of different nuclear fuel cycles is needed.

Evaluating the economics of nuclear fuel cycles is based on determining the needed amount of nuclear fuel materials, the associated cost for their acquisition through using various industrial services, and the cost to manage the resulting waste. This results in the estimation of the fuel cycle cost. Therefore, the fuel cycle cost includes not only the cost to provide fuel for nuclear reactor operations (this is the front-end cost) but also the cost to manage the resulting waste (this is the back-end cost). The front-end cost includes the cost for the purchase of uranium and the services for its conversion, enrichment, fuel fabrication, and transport of fuel to a nuclear reactor. The back-end cost includes the cost of storage and transport of spent fuel, the spent fuel reprocessing and reuse cost (if employed), and the cost for final disposal of nuclear waste.

15.1.2.1 Calculation of the Fuel Cycle Cost

Fuel cycle cost calculation begins with the determination of the needed amount of nuclear fuel (in mass of uranium) to support the energy production at the rated power level for a given period (typically a year). Then for the needed amount of fuel, the amount of uranium materials to be processed at each stage of the front-end fuel cycle (i.e., mining, milling, conversion, enrichment, fuel fabrication) is determined. The cost of processing of uranium materials at each front-end fuel cycle stage is also determined. This cost (called the unit cost) is multiplied by the mass of the uranium processed to give the cost of required mining, milling, conversion, enrichment, and fuel fabrication for reactor operation. Summing all of them gives the annual materials service cost of the front-end fuel cycle (in the case of the once-through cycle). Similar calculations are made for the discharged spent fuels by multiplying the cost of storage, transportation, and disposal with the respective amount of the spent fuel materials handled. Summing them gives the annual materials service cost of the back-end fuel cycle (again for the once-through fuel cycle).

If reprocessing of spent fuel is involved in the fuel cycle, the front-end fuel cycle includes the steps of conversion and enrichment of reprocessed uranium, and fabrication of fuel from reprocessed uranium and plutonium (see Fig. 15.1). The back-end fuel cycle in this case includes reprocessing of spent fuel from thermal reactor(s) (and also fast reactor in the closed cycle).

The fuel cycle cost [$/MWh] is calculated by summing the total annual cost of the front-end and the back-end fuel cycle services [$/year] and then dividing it by the annual electricity production [MWH/yr]. This is shown in the following equation.

$$FCC\left[\frac{\$}{MWh}\right]=\frac{\sum_{all\ components} unit\ cost\left[\frac{\$}{KgHM} or \frac{\$}{SWU}\right]\cdot amount\ of\ material\ serviced\ \left[\frac{KgHM}{yr} or \frac{SWU}{Yr}\right]}{annual\ electricity\ production\ \left[\frac{MWh}{yr}\right]} \tag{15.1}$$

Dynamic Model Vs. Equilibrium Model

Two different approaches can be employed in calculating the flow of nuclear materials in the fuel cycle. The first one is the dynamic model. The dynamic model simulates the flow of materials in a time dependent manner. The model starts from uranium mining and ends with final disposal taking into account the time periods involved at each service step including the time gap between development and deployment of fuel cycle stages. The second approach is the equilibrium model. The equilibrium model assumes that the whole fuel cycle is in steady state and that all of the fuel cycle steps are available instantly for service. Therefore, the time needed to reach equilibrium is ignored. In this case, calculation of uranium materials flow is based on the set amount of electricity production. The needed uranium materials to produce the required amount of electricity at each fuel cycle service step form the mass balance of the fuel cycle. Typically, the equilibrium model is used for the purpose of cost comparisons of different fuel cycles.

Unit Costs for Fuel Cycle Services

The unit cost is the cost of handling nuclear materials in each service stage of the fuel cycle (unit cost is defined per unit mass). The unit cost, as the cost of fuel cycle service, varies depending upon the demand for the service, technological progress, financial arrangement of the service provider, competition in the market, and the political and social environment surrounding the fuel cycle service. Given the fluctuations in these conditions, the value of the unit cost can be assumed to be a random variable. Table 15.3 gives the range of the values of the unit cost (as random variables) for each step in the nuclear fuel cycle. The specified values include the minimum, the maximum, the mean, and the mode (as the nominal values) based on the report from US DOE's Advanced Nuclear Fuel Cycle studies (Shropshire et al. 2009). The report lists the unit cost of advanced reprocessing technologies such as

Table 15.3 Values of the unit cost for fuel cycle services

	Value				
Phase	Minimum	Maximum	Mean	Mode (Nominal cost)	Reference cost contingency (+/− %)
Uranium ($/kgU)	30	260	122	75	NA
Conversion ($/kgU)	5	15	10	10	+/− 2
Enrichment ($/SWU)	85	135	110	110	+/− 25
UO_2 fuel fabrication ($/kgU)	200	300	250	250	N/A
MOX fuel fabrication ($/kgHM)	3000	5000	3733	3200	10–40%
Interim spent fuel storage (wet) ($/kgHM)	100	500	300	300	NA
Interim spent fuel storage (dry) ($/kgHM)	100	300	173	120	NA
Spent fuel reprocessing with PUREX (with vitrification & storage) ($/kgHM)[a]	782 ~ 1043 (assuming average annual inflation rate of 2.68% and the exchange rate of 1:1 between US dollar and EU euro)				
Spent fuel reprocessing with COEX (with vitrification & storage) ($/kgHM)	755 (1108)	1096 (1619)	925 (1370)	925 (1370)	NA
Spent fuel reprocessing with UREX+1a (with vitrification & storage)	903 (1494)	1339 (2214)	1120 (1850)	1120 (,850)	NA
Spent fuel recycling with pyroprocessing (with fast reactor fuel fabrication) ($/kgHM)	1800 (3000)	2700 (9000)	2300 (6000)	2300 (6000)	NA
Spent fuel packaging for shipping to reprocessing, storage, or disposal ($/kgHM)	50	130	93	100	NA
Spent fuel geological disposal ($/kgHM)	400	1000	683	650	NA

Source: Shropshire (2009)
[a]NEA (1994, in 2007 $)

COEX, UREX+1, and pyroprocessing as part of the consideration for advanced fuel cycles (but PUREX is not considered in the study). The values are, however, based on projections with no actual operating experiences and expected to have large uncertainty. For PUREX reprocessing, the values from the OCED/NEA study (NEA 1994) are listed.

As seen from the table, the most expensive step in the once-through cycle is spent fuel disposal. This is followed by fresh fuel fabrication, spent fuel dry storage, enrichment, etc. The uncertainty in the cost of spent fuel disposal is very high due to the difficulty in estimating the project completion time along with the site-specific nature of the task. The uncertainty in the cost of uranium purchase is also relatively high. This variable nature of uranium price reflects the fluctuating market situation.

If recycling of spent fuel were employed, the stages of reprocessing and fuel fabrication of recycled materials are expected to be the most expensive step. The unit cost of this step is about 15 times higher than that of fresh fuel fabrication. Due to this high cost of recycling, the modified open cycle or the closed cycle may not be found economical unless the price of uranium purchase or geologic disposal of waste becomes very high.

The Levelized Cost Calculation

Due to the time value of money involved, estimating the fuel cycle cost requires the calculation of the levelized cost. The levelized cost considers the fact that each step of the fuel cycle services and the necessary payment occur at different times with different lead times or lag times with respect to nuclear reactor operation. This calculation is noting that any payment made before the revenue generation involves financial services with interest payment and that the revenue in the nuclear fuel cycle is only generated from selling electricity from nuclear reactor operation. Therefore, depending on the timing of payment, the value of money paid for fuel cycle service varies with different amount of interest involved. Accordingly, the levelized cost of fuel cycle is to represent the present value (or at a selected reference time point) of the total fuel cycle cost. Using the levelized cost covering the net present values of all services over the life-time of a nuclear power plant, therefore, allows the comparison of different fuel cycles with different life spans of facilities and capacities on an equivalent basis.

Most countries do not own or operate all of the facilities needed in a fuel cycle, thus rely on the services provided by other countries' facilities. Therefore, the levelized fuel cycle cost can be considered a proxy of the average price for a nuclear power plant owner to pay for all of the fuel cycle services to break even to operate the nuclear power plant over its lifetime. Here it is assumed that the cost of providing a service and its market price are very closely related, assuming a fully competitive market. Cost is typically the expense incurred for a service being sold by a service provider. Price is the amount a customer is willing to pay for the service. The difference between the price paid and the cost incurred is the profit to the service provider.

Calculation of the levelized fuel cycle cost requires the use of discount rate. Discount rate is the interest rate used to estimate the value of an investment, as the current value, based on the expected future return (see also Sect. 2.4.1). For the services of the front-end of the nuclear fuel cycle, a nuclear power plant owner is making investments by paying for each of the services in expectation for the return by selling electricity.

This calculation requires the specification of the lead time for the front-end fuel cycle services. Lead time refers to the time gap between the payment for the service and the loading of fuel into the reactor. In contrast, lag time is applied in the back-end of the fuel cycle and refers to the time gap between the payment for the back-end service and the discharge of spent fuel.

While the front-end fuel cycle services incur interest payments during the lead time, the back-end fuel cycle services do not require interest payment. This is assuming that the related service cost is a sunken cost, set aside during the revenue generation, i.e., reactor operation by the plant owner. The set aside fund instead can accrue interest before making the payments for the corresponding services.

Calculation of the levelized cost using discount rate can be made by using either the discrete discount method or the continuous discount method. The discrete discount method assumes that the interest is compounded every year. The continuous discount method considers the interest as continuously applied while electricity is being generated. Due to the simplicity in calculation, the discrete discount method is commonly used. The discount rate used apparently is subject to the terms of contracts and might be different between countries or between government and private utilities. The rate generally takes into account the effect of tax and rate of return of funds.

The equation for the discrete discount method, using annual cost of fuel cycle and annual production of electricity, is given as

$$LFCC = \frac{\sum_{t=1}^{n} \frac{F_t}{(1+r)^t}}{\sum_{t=1}^{n} \frac{E_t}{(1+r)^t}} \tag{15.2}$$

where, *LFCC* is the levelized fuel cycle cost covering the entire periods covered by the fuel cycle, F_t is the total fuel cycle cost in year t, E_t is the total electricity generated in year t, r is the discount rate, and n is the life of the plant in years.

As electricity is generated more or less continuously during the reactor life, using a continuous discount method could better represent the cost. If we redefine the discount rate r as r' with $r' = \ln(1 + r)$, the discount factor is replaced by the exponential form for the continuous discount method:

$$\frac{1}{(1+r)^t} = e^{-r't} \tag{15.3}$$

Then the summation terms in the Eq. 15.2 can be replaced by the integration of the annual fuel cycle cost multiplied by the exponential form over the period of electricity generation as,

$$LFCC = \frac{\int_A^B F(t) \mathrm{e}^{-r't} dt}{\int_A^B E(t) \mathrm{e}^{-r't} dt} \tag{15.4}$$

where, r' is the continuously compounded discount rate (as $r' = \ln(1 + r)$), A is the beginning time of fuel cycle activities and B is the end time of fuel cycle activities, *F (t)* is the total cost of fuel cycle as a time dependent quantity, and *E(t)* is the profile of electricity production between A and B.

The Levelized Fuel Cycle Costs with and Without Spent Fuel Cycling

Table 15.4 shows examples of the levelized fuel cycle cost for two different fuel cycles, the once-though cycle and the twice-through cycle. The numbers are based on the 1994 OECD study (NEA 1994) using the case of a 1390 MWe PWR with the fuel burnup of 42,500 MWD/t, the capacity factor of 75%, and the plant lifetime of

Table 15.4 The levelized fuel cycle cost (mills/kWh) of the once- and twice-through cycle with a PWR

Component	Lead/lag time	Once-through	Twice-through
Uranium	24 months lead	1.64	1.64
Conversion[a]	18 months lead	0.21	0.21
Enrichment[a]	12 months lead	1.85	1.85
Fuel fabrication[a]	6 months lead	1.00	1.00
Subtotal for front-end		**4.70**	**4.70**
Storage/transport of spent fuel	5 years lag	0.51	0.11
Reprocessing	6 years lag	Na	1.66
Waste disposal	40 year (OT), 56 year (TT) lag	0.25	0.02
Subtotal for back-end		**0.76**	**1.79**
Uranium credit[b]		Na	−0.18
Plutonium credit[b]		Na	−0.08
Subtotal for credit		**0.00**	**−0.26**
Total cost		**5.46**	**6.23**

Data source: NEA (1994)

[a] Loss factor of 0.5%, 1.0%, and 2.0% assumed for conversion, fabrication, and reprocessing

[b] Uranium credit is at 70% of the cost

30 years. Here capacity factor is the ratio of the energy produced by the reactor over a given period divided by the energy it would have produced at its full power capacity over the same period. The values are in 1991 US dollars assuming the exchange rate of 1:1 between US dollar and EU euro. The discount rate of 5% per year was used in the analysis.

In this calculation, the unit cost of the front-end fuel cycle services was assumed at \$50/kg U for uranium purchase, \$8/kg U for conversion, \$110/SWU for enrichment, and \$275/kg U for fuel fabrication. For the back-end, the unit cost was assumed at \$230/kg U for transport and storage of spent fuel, and \$610/kg U for encapsulation and disposal in the once-through cycle. Under the twice-through cycle, the assumed unit cost values were \$50/kg U for spent fuel transport, \$720/kg U for PUREX reprocessing, and \$90/kg U for vitrified HLW disposal. The unit cost for reprocessing included the prices for the associated spent fuel receipt, the waste conditioning/storage services, and the disposal of low and intermediate level waste. Also, the cost for MOX fabrication was assumed to be four times higher than that of the fresh uranium fuel fabrication.

The results in Table 15.4 indicate that the twice-through cycle is 14% more expensive than the once-through cycle using the levelized fuel cycle cost.

Changes in the relative fuel cost of various fuel cycle services as a function of time are captured in Fig. 15.2 with the lead and lag times specified for both the once-through and the twice-through cycle. The calculation assumed that in the once-through cycle, spent fuels were in storage for 40 years including 5 years of cooling at the reactor site and then were directly disposed. In the twice-through cycle, spent fuels were reprocessed after 6 years of cooling. The value of the credit of recovered uranium and plutonium (as the value of new fuel) contained in the spent fuel was

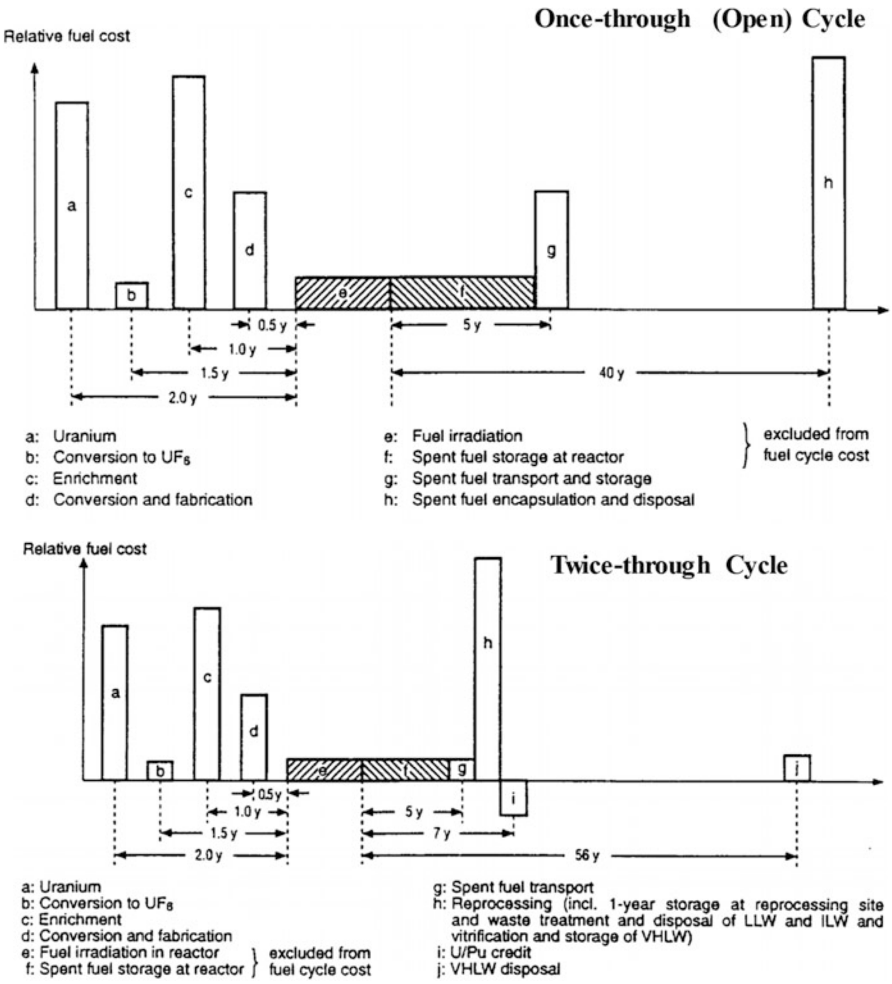
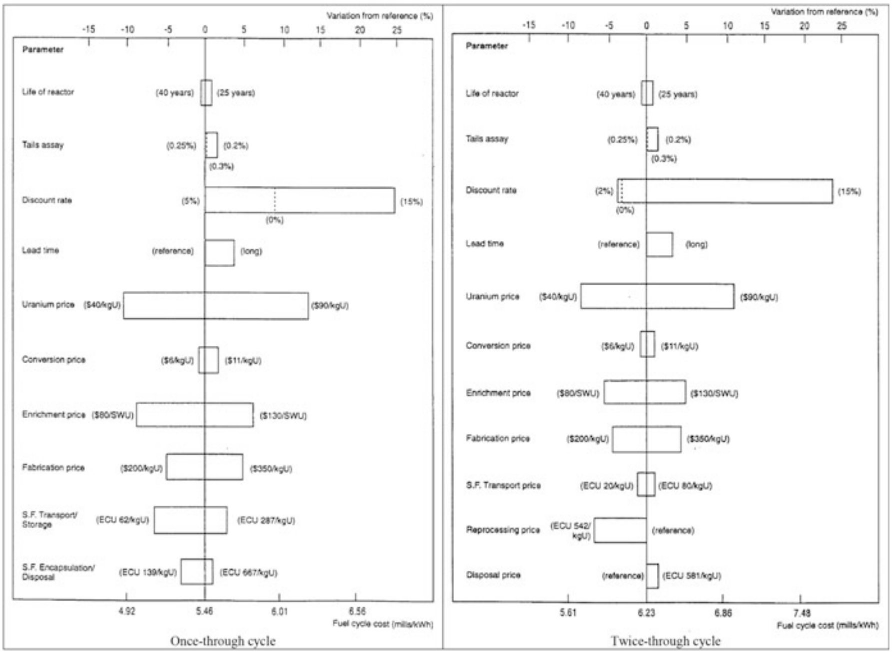


Fig. 15.2 Depiction of time flow of the fuel cycle cost for the once-through cycle and the twice-through cycle. (Source: NEA 1994)

assumed at 0.18 and 0.08 mills/kWh, respectively (1 mill = 0.1 cent). The recovered uranium and plutonium were assumed to be recycled right away in a reactor similar to the reference PWR with the same burn-up of the reference fuel. In the calculation, a single recycle of the recovered material would correspond to approximately 15–20% loading of MOX in the core, reducing natural uranium demand by approximately 20–25% (NEA 1994).

Sensitivity Analysis of the Fuel Cycle Costs with and without Spent Fuel Recycling

The 1994 OECD study also included the sensitivity analysis of the levelized fuel cycle cost. The parameters considered in the sensitivity analysis include the life of reactor, the discount rate, the concentration of ^{235}U in the depleted uranium stream



Once-through cycle

Twice-through cycle

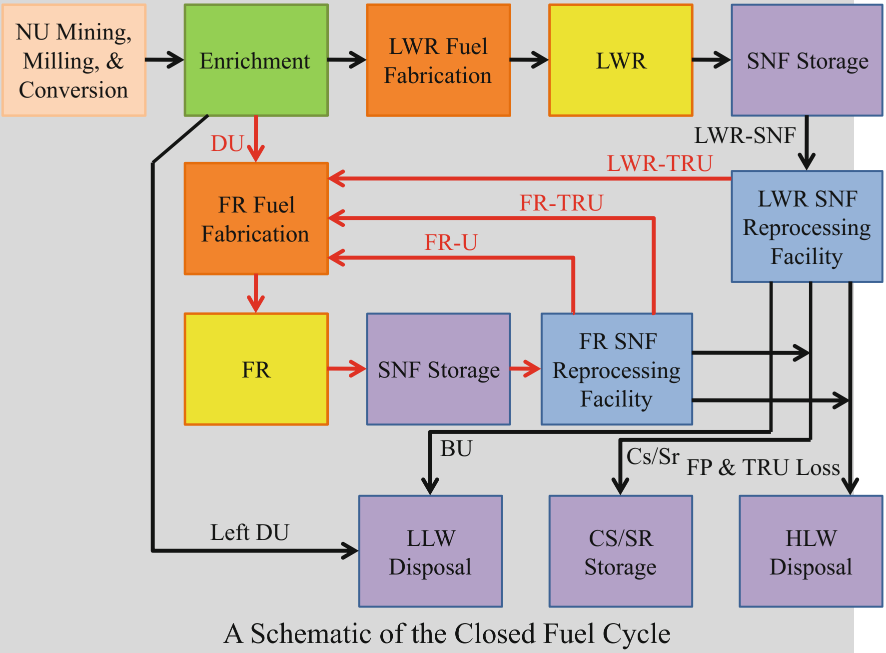
Fig. 15.3 Results of sensitivity analysis of the levelized fuel cycle cost for major parameters. (Source: NEA 1994)

(called tails assay), the price of fuel cycle services including uranium purchase, conversion to uranium hexafluoride, enrichment, fabrication, spent fuel transport/ storage, and disposal, and the lead times of each fuel cycle service (uranium purchase, conversion to uranium hexafluoride, enrichment, and fabrication) relative to the date of fuel loading into the reactor. The results are shown in Fig. 15.3. The numbers represented by the bars in the figure show the variations in the fuel cycle cost when the values of the corresponding parameters varied within the given range. The variation ranges are specified next to the bars in the parenthesis in the figure except the lead time. The lead times were increased in the sensitivity analysis from the reference value to the maximum assumed value as follows: uranium purchase (24–42 m), conversion (18–34 m), enrichment (12–22 m), and fabrication (6–12 m).

From the analysis, the most important parameters controlling the value of the fuel cycle cost were the discount rate and the price of natural uranium, in the case of the once-through cycle. The same results were also observed for the twice-through cycle. The other parameters of importance included the price of enrichment and fuel fabrication. The price of reprocessing service followed them in the case of the twice-through cycle. These results indicate that if the cost of front-end fuel cycle services were much higher, spent fuel recycling would become economical. Also if the price of uranium were very high, the modified open cycle or the closed cycle would become economically feasible, although this is an unlikely scenario at the present time.

If the available footprint of a geologic repository is very limited or if the cost of developing a geological repository is prohibitively high, use of the modified open cycle or the closed cycle may also become economically competitive. In this case, the reduction in the needed space of the repository must provide enough economic incentive to overcome the price burden associated with spent fuel recycling.

Example 15.1: Examination of Fuel Cycle Cost
The purpose of this exercise is to calculate the cost of nuclear fuel cycle. The fuel cycles considered are the once-through cycle (OTC) and the closed cycle (CC). The figure below shows the schematic of nuclear materials movements as the fuel cycle mass flow under the CC. To support the calculation, a spreadsheet model is provided. In the model, the time value of money is ignored and the unit cost values are assumed to be constant. The fuel cycle model, given in the following links, tracks the material flow in the fuel cycle from uranium mining to final nuclear waste disposal. The model is an excel-based mass tracking model. In the CC, the spent fuel from PWR reactor (APR-1400) is reprocessed and the plutonium and minor actinides are recycled into the fuel for a fast reactor. The spent fuel from the fast reactor is also reprocessed and recycled. The fuel for the fast reactor incudes the recycled TRU from a fast reactor, the recycled TRU from a PWR, and the depleted uranium as tailing from an enrichment facility.



A schematic of the closed fuel cycle

(continued)

Example 15.1 (continued)

The annual fuel cycle cost is given as,

$$FCC\left[\frac{\$}{year}\right] = \sum_{all\ components} unit\ cost\left[\frac{\$}{KgHM}or\frac{\$}{SWU}\right]\cdot$$
$$amount\ of\ material\ serviced\left[\frac{KgHM}{yr}or\frac{SWU}{Yr.}\right]$$

The unit cost values used in this example are the nominal values listed in Table 15.3.

(a) According to the historical data, the cost of uranium mining and milling varies between \$30 and \$260 per kgU, determine how this variation affects the fuel cycle cost of the OTC and the CC.

(b) If reprocessing is performed by using PUREX, the nominal cost of reprocessing is about \$800/kgHM. As a comparison, based on the experiences at the Rokkasho-mura facility, the cost of reprocessing could be over \$2000/kgHM. Determine how the reprocessing cost change between \$800 and \$2200/kgHM affects the FCC of the CC.

(c) The cost of a geological repository varies widely between countries. The following table shows this variation (as the estimated values). Determine the FCC of the OTC and the CC if the cost of SNF/HLW disposal is \$105/kgHM (the low case of Canada) or \$1250/kgHM (a slightly higher value than that of Switzerland).

Cost estimates for geologic disposal of HLW/SNF (Shropshire et al. 2009)

Country	Geologic medium	Size (MTHM)	Total Cost (2007 \$)	Unit Cost
Belgium	Clay	4900	\$1.72B	\$361/kgHM (SNF) \$140/kgHM (HLW)
Canada	Unknown	96,000–192,000[a]	\$13B–\$20B	\$105–\$140/kgHM (SNF)
Czech Republic	6 possible locations	3724	\$1.6B	\$437/kgHM (SNF)
Finland	Granite	5600	\$4.5B	\$800/kgHM (SNF)
France	Granite/clay	45,000	\$19.8B	\$440/kgHM (HLW)

(continued)

Hungary	Unknown	1320	$1.3B	$984/kgHM (SNF)
Japan	Granite	29,647	$25B–$26B	$851–885/kgHM (SNF)
Sweden	Granite	9741	$3.4B	$350/kgHM (SNF)
Switzerland	Granite/clay	2000	$3.6B	$1203/kgHM (SNF)

[a]Note: CANDU SNF has low burnup (10,000 MWD/MTU) compared to LWR SNF

The fuel cycle model is provided as supplementary material with the online version of this chapter. See note "Supplementary Information" on page 733 for access details.

Solutions:

(a) The estimated fuel cycle costs of the OTC and the CC are shown in the following table when U mining and milling unit cost varies between $30 and $260 per kgU:

Cost ($)	30	50	75	100	125	150	175	200	250	260
OC	4.33	4.68	5.11	5.55	5.99	6.42	6.86	7.29	8.17	8.34
CC	5.77	6.01	6.31	6.61	6.97	7.21	7.51	7.81	8.41	8.53

Note that the fuel cycle cost of the OTC increases rather rapidly compared to that of the CC with the variation in the unit cost of uranium mining and milling.

(b) The estimated fuel cycle costs of the CC are given in the following table when the reprocessing cost changes between $800 and $2000 per kgHM

Cost ($)	800	1000	1250	1500	1850	2000	2200
CC	4.71	5.01	5.39	5.78	6.31	6.54	6.85

The fuel cycle cost of the CC is strongly dependent on the reprocessing cost. When the cost of reprocessing is low, the fuel cycle cost of the CC becomes comparable to that of the OTC.

(c) The estimated fuel cycle costs of the OTC and the CC are given below when the cost of SNF/HLW disposal varies between $105 and $1250 per kgHM

Cost ($)	105	200	350	500	650	800	950	1100	1250
OC	4.12	4.29	4.57	4.84	5.11	5.39	5.66	5.93	6.21
CC	6.18	6.20	6.24	6.27	6.31	6.35	6.38	6.42	6.46

The fuel cycle cost of the OTC is rather sensitive to the variations in the SNF/HLW disposal cost. In contrast, the fuel cycle cost of the CC is insensitive to the variations of the SNF/HLW disposal cost. If the cost of SNF/HLW

(continued)

Example 15.1 (continued)
disposal is very high, the gap in the fuel cycle cost between the OTC and the CC becomes small.

15.1.2.2 Comparison of Fuel Cycles Using Total Electricity Generation Costs

There is another dimension to consider in the comparison of nuclear fuel cycles. As different fuel cycle concepts involve using different types of nuclear reactors, i.e., thermal reactors (the once- or twice-through cycle) or thermal and fast reactors (the closed cycle), ignoring the cost of using these reactors would not give a fully meaningful comparison of the economics of fuel cycles. The total cost of electricity generation includes not only the cost of the fuel cycle but also the cost of building and operating different types of nuclear reactors depending on the types of fuel cycle arrangements utilized.

According to an estimate of cost breakdown of different activities in generating electricity using a typical nuclear reactor given by NEA (2001), the fuel cycle cost is about 20% of the total cost of electricity generation. This 20% fuel cycle cost can be further divided into 5% in uranium mining and milling, 1% in conversion, 6% in enrichment, 3% in fuel fabrication, and 5% in the back-end fuel cycle. The estimate indicated that the cost associated with building a nuclear power plant is about 57% with the rest of the electricity generation cost (23%) belonging to operation and maintenance of nuclear reactors.

A more recent estimate (NEA 2020), however, indicated that the cost of building a plant (the capital cost and the associated financing cost) could be much higher (78% of total) mainly due to the increase in the construction time of a nuclear power plant and the interest payment. According to this estimate, the breakdown becomes 13% in O&M (operation and maintenance), and 9% in fuel cycle.

If these costs of building and operating nuclear power plants are included for cost comparison between the fuel cycles, we use the total cost of electricity generation as follows.

$$LCOE = \frac{\sum_{t=1}^{n} \frac{R_t+O_t+F_t}{(1+r)^t}}{\sum_{t=1}^{n} \frac{E_t}{(1+r)^t}} \tag{15.5}$$

where, $LCOE$ is the levelized cost of electricity, R_t is the cost of building nuclear reactors in year t, O_t is the cost of operation and maintenance of nuclear reactors in year t, F_t is the total fuel cycle cost in year t, E_t is the total electricity generated in year t, r is the discount rate, and n is the life of the plants in years.

In this case, the cost is based on the levelized cost of electricity generation using the discrete discount method. The equation includes the cost of building a nuclear power plant and its operation and maintenance, along with the fuel cycle. The cost of building a nuclear power plant is called the capital cost. The capital cost includes overnight construction cost covered by an investment through a bank loan and the associated financing cost. The overnight construction cost refers to the capital cost of plant construction if no interest was incurred during construction, as if the project was completed “overnight”. The O&M cost includes the required spending to operate, maintain, and decommission a nuclear power plant. This includes the cost of services, supplies, and salaries for the plant personnel.

Comparison of the levelized cost of electricity generation for different nuclear fuel cycles is shown in Table 15.5. The fuel cycles compared include the once-through cycle, the twice-through cycle, and the closed cycle. For the closed cycle, a fast reactor with the conversion ratio of 1.0 was assumed. The values of the unit cost of fuel cycle services and other input parameters used in the calculation are shown in Table 15.6. As the numbers reflect more recent trends, the values in this example are higher than what is given in Table 15.3.

Table 15.5 The levelized cost of electricity (in mills/KWh) (1 mill equals to 10^{-3} $) for different fuel cycles

		Twice-through cycle		Fast reactor cycle (CR = 1)	
	Once-through cycle	1st pass – UOx fuel in a LWR	2nd pass – MOX fuel in a LWR	LWR	Fast reactor
Front end fuel cycle cost	Raw uranium: 2.76	Raw uranium: 2.76	Depleted U: 0.03	Raw uranium: 2.76	Depleted U: 0.02
	Fuel production: 4.35	Fuel production: 4.35	Plutonium: −4.39	Fuel production: 4.35	Transuranics: −19.72
			Fuel production: 7.38		Fuel production: 4.05
	Subtotal: 7.11	Subtotal: 7.11	Subtotal: 3.02	Subtotal: 7.11	Subtotal: −15.66
Capital cost	67.68	67.68	67.68	67.68	81.22
O&M cost	7.72	7.72	7.72	7.72	9.26
Back-end fuel cycle cost	1.3	Reprocessing: 2.36	6.96	Reprocessing: 2.36	Reprocessing: 2.66
		HLW disposal: 0.40		HLW disposal: 0.40	HLW disposal: 0.34
		Reprocessed U:-0.14		Reprocessed U: −0.14	Reprocessed U: −0.01
		Plutonium: 0.25		Transuranics: 1.43	Transuranics: 8.75
		Subtotal: 2.87		Subtotal: 4.06	Subtotal: 11.74
LCOE total	**83.81**	**85.38**	**85.38**	**86.57**	**86.57**
LFCC	**8.41**	**9.98**	**9.98**	**11.17**	**−3.92**

Source: Kazimi (2011)

Table 15.6 Values of the unit cost and input parameters used in the MIT fuel cycle study

Step/input parameter	Value
Front-end fuel costs	
Natural uranium	80 ($/kgHM)
Depleted uranium	10 ($/kgHM)
Conversion of natural U	10 ($/kgHM)
Enrichment of natural U	160 ($/SWU)
Fabrication of UOx from natural U	250 ($/kgHM)
Conversion of reprocessed U	200%
Enrichment of reprocessed U	10%
Fabrication of UOX from reprocessed U	7%
Fabrication of MOX	2400 ($/kgHM)
Fabrication of fast reactor fuel	2400 ($/kgHM)
Reactor costs	
LWR capital (overnight)	4000 ($/kWe)
LWR capacity factor	85%
Fast reactor capital premium	20%
Fast Reactor O&M premium	20%
Fast reactor capacity factor	85%
Reprocessing costs	
UOX, PUREX, UREX+, or TRUEX	1600 ($/kgHM)
Fast reactor fuel, pyroprocessing	3200 ($/kgHM)
Waste costs	
Interim storage of LWR spent fuel (UOx, MOX)	200 ($/kgHM)
Disposal of spent fuel (UO2)	470 ($/kgHM)
Disposal of spent MOX fuel	3130 ($/kgHM)
Disposal of HLW from UOX (PUREX)	190 ($/kgHM); 3650 ($/kg fission products)
Disposal of HLW from UOX (TRUEX)	190 ($/kgHM)
Disposal of HLW from fast reactor	280 ($/kgHM)
Discount rate	7.6%

The estimated levelized cost of electricity (LCOE) of the once-through cycle, the twice-through cycle, and the closed cycle was 8.38, 8.54, and 8.66 cents/KWh, respectively. Results showed the costs of the twice-through cycle and the closed cycle are higher than the once-through cycle by 0.157 and 0.276 cents/KWh, respectively.

Within the twice-through cycle, the levelized cost of reprocessing was estimated at 0.236 cents/kWh. Although this cost was partially offset by the credit (−0.079 cents/kWh) provided by the recycled plutonium (as fuel value), the overall cost of electricity generation was still higher than the once-through cycle.

In the case of the closed cycle, the cost of fast reactors and spent fuel reprocessing was much higher than the partial credit provided by the recycling of plutonium, resulting in the cost of electricity to be higher by about 3% than the once-through cycle. This increase in LCOE appears not significant. This is due to the fact that the fuel cycle costs are small part of the total cost of electricity generation.

Uncertainty in the estimation of the fuel cycle cost or the cost of electricity generation also needs to be recognized. These uncertainties come from the variable nature of the actual cost of fuel cycle services. In particular, the costs of reprocessing and spent fuel disposal are expected to be quite uncertain as they represent the cost of the future with unknown prospects of development (depending on nation specific situations). The values given in the table may not reflect the reality of the future.

It should also be noted that whether reprocessing of spent fuel is an acceptable choice or not is more than an issue of economics. The related decision includes the consideration of how the increased cost of reprocessing compares against other non-economic factors such as energy security or fast reactor development for waste transmutation. In particular, if a country views the issue of energy security a high priority, they may want to maximize the utilization of domestic nuclear fuel resources through recycling. If the country has a concern over long-term stable supply of nuclear fuel, they may see reprocessing desirable even though the related cost is relatively high.

However, pursuing the option of spent fuel reprocessing as part of national nuclear fuel cycle development is a very complicated matter. Building a new reprocessing facility is highly scrutinized under the current international nuclear governance regime. Such scrutiny is driven by the concern of nuclear proliferation, i.e., concern over potential misuse of reprocessing technology. This issue is further discussed in the next section.

15.2 Nuclear Nonproliferation

15.2.1 Risk of Nuclear Proliferation

Commercial utilization of nuclear technology began with the vision of providing unlimited energy to humans. To realize such vision, preserving uranium resources was viewed important at the start of global nuclear industry (in the 1950s and 60s). With uncertainty in securing natural uranium resources for the long-term future, recycling of spent fuel was envisioned as necessary for sustainable nuclear power development. Accordingly, spent fuel reprocessing became a part of the design of nuclear fuel cycle. Using fast reactors was also considered necessary for the breeding of fissile for unlimited fuel supply.

This direction suddenly changed in 1970s. The change was due to an unexpected testing of nuclear weapons by India. India exploded their first nuclear weapon in 1974 out of an ostensibly peaceful nuclear power program. International community suddenly realized that civilian nuclear fuel cycles can be utilized (misused) for the production of weapons usable nuclear materials.

Uranium enrichment is a necessary step for nuclear fuel preparation in a fuel cycle. Reprocessing of spent fuel is also essential if spent fuel recycling is adopted.

Both steps produce fissile materials. When enough fissile materials become available from the use of either uranium enrichment technology or reprocessing technology, weaponization of the materials can be started. If ^{235}U from uranium enrichment or the plutonium (mainly ^{239}Pu) from spent fuel reprocessing and the related know-how are diverted for non-civilian purpose, i.e., military purposes, so-called nuclear proliferation takes place. Preventing nuclear proliferation has been the main goal of the international nuclear nonproliferation regime since the beginning of global nuclear power development.

Along with ^{235}U and ^{239}Pu, ^{233}U or ^{241}Pu are also weapon-usable fissile materials. These fissile materials, after being assembled into a fast critical mass, undergo explosive prompt fission reactions (see Table 7.7 for the amount of critical mass, or the discussion in Sect. 15.2.4). The design used for such device includes a gun-type (when ^{235}U or ^{233}U is used) or an implosion-type (when plutonium is used) as nuclear explosive. In the case of uranium, the low levels of spontaneous neutron and heat generation allows the use of fissile uranium in a gun-type weapon design. In the case of plutonium, use of an implosion-type design is necessary due to the presence of large number of spontaneous neutrons and considerable amount of decay heat with plutonium.

The levels of spontaneous neutron generation and heat are particularly high with the even number isotopes of plutonium such as ^{238}Pu, ^{240}Pu, and ^{242}Pu (as noted in Table 7.7). Presence of large number of spontaneous neutrons makes it difficult to initiate critical chain reactions with precision timing. Presence of large amount of heat complicates the use of chemical explosives to detonate the weapon. Therefore, exploding the implosion-type bomb requires sophisticated expertise of bomb designs. In addition, testing of bomb is necessary with the implosion-type bomb for functional verification.

The fissionable materials (^{237}Np, ^{231}Pa, ^{241}Am, ^{243}Am, ^{244}Cm, ^{245}Cm, ^{246}Cm, ^{247}Bk, and ^{251}Cf) are also weapons usable through incorporation into the implosion type bomb. The so-called reactor-grade plutonium in commercial spent fuel can be also utilized, although not as desirable as the weapons grade plutonium, as part of an implosion-type nuclear bomb. Here weapons grade or reactor grade refers to the content of ^{240}Pu in spent fuel. The weapons grade plutonium is the one with the ^{240}Pu contents lower than 7% of total plutonium. The reactor grad plutonium is with ^{240}Pu at higher than 19%. As the reactor grade plutonium is still usable for nuclear weapons, a civilian nuclear fuel cycle, in theory, can support the ambition of nuclear weapons development.

It should also be noted that obtaining the access to the fissile materials is only a necessary step and does not dictate the course of nuclear proliferation. Sustained government funding over a number of years is a requirement for a nuclear weapon program to actually achieve the intended outcome. Among all the factors involved, the most important one in nuclear proliferation is the political decision made by a government. Such government decision is driven mainly by three motivational factors, i.e., national security concerns, international political power aspiration, and domestic political incentives. The potential proliferant country may perceive some likelihood of future security conflicts or disputes with a country possessing nuclear weapons or an adversary country with overwhelming superiority in conventional forces. In that case, nuclear weapons can be considered to counterbalance such superiority of the adversary. The international political power aspiration comes from the belief that nuclear weapons somehow magnify a nation's image and a country may want to obtain regional or global power status through acquisition of nuclear weapons. Domestic political incentives exist if a country perceives nuclear weapon development as a way to gain political support or to divert domestic energies away from domestic problems.

Proliferation decisions are also weighed against possible cons. Acquiring nuclear weapons can be very costly in terms of international economic trade, diplomatic relations, military alliance, or simply due to the maintenance requirements. Paying high prices of nuclear weapons development was demonstrated in the past through the examples of Iraq, Iran, North Korea, etc. who tried to develop nuclear weapons as non-nuclear weapons states (see also Sect. 15.2.3 for the definition). Attempts to develop nuclear weapons are typically followed by sanctions and economic isolation which may be unbearable under today's highly connected international trade environment. Earning the reputation of a pariah state in the international system is also very detrimental. The country may have to forgo the security assurance provided by a powerful ally by pursuing their own nuclear weapons.

The balancing by a state between the pros and cons of nuclear weapons development would be strongly related to the existing international security situations, diplomatic relations with neighbors and the nuclear weapons states, and domestic political or economic situations.

The decision of nuclear proliferation is also affected by the perceived difficulties associated with overcoming the challenges in nuclear weapons development. If the time and degree of illicit activity needed to obtain special nuclear materials/related technical capabilities are considered too significant, the state may be hesitant. This will also be the case if breaking the international norms and safeguards system is perceived to be too damaging to the state. The process of making the related decisions will also be a function of regime type (authoritarian vs. democratic), leaders' psychology (in particular in the case of authoritarian regime), and the number of veto players present in the country's political decision making system.

Given these considerations, elevating the level of difficulty in any diversion attempt of misusing civilian nuclear power capability for nuclear weapons development can have major impact on nuclear nonproliferation. In this regard, establishing large technical and institutional barriers against the misuse of civilian nuclear power program will be effective to prevent nuclear proliferation. This point is further discussed in the next section.

15.2.2 Proliferation Resistance of Nuclear Fuel Cycle

The interest of nuclear industry and government in establishing large technical and institutional barriers against the misuse of civilian nuclear power program led to examination of proliferation resistance of nuclear power technology. "Proliferation resistance refers to that characteristic of a nuclear energy system that impedes diversion of undeclared production of nuclear material, or misuse of technology, by States intent on acquiring nuclear weapons or other nuclear explosive devices" (IAEA 2003). Such proliferation resistance results from intrinsic or extrinsic features of a technology in terms of their effect on the difficulty in acquiring nuclear material by an entity (e.g., national or sub-national groups). For example, the isotopic, chemical, and radiological characteristics of nuclear materials and their material forms can influence the degree of difficulty in an effort to separate fissile nuclear materials from spent fuel.

According to a report from US DOE (TOPS 2000), these barriers are classified into 11 different types: the isotopic barrier, the chemical barrier, the radiological barrier, the mass & bulk barrier, the material detectability barrier, the facility unattractiveness barrier, the facility accessibility barrier, the available mass barrier, the facility diversion detectability barrier, the skills/expertise/knowledge barrier, and the time barrier. This list of barriers indicate that not only the characteristics of materials but also the design and operation of facilities can have major implications in proliferation resistance.

A study by the U.S. DOE (DOE 1980) also noted substantial differences in proliferation resistance among different fuel cycles. Such differences can be particularly salient if the technologies are deployed in non-nuclear weapon states. Findings from the report can be summarized as follows.

Materials used in the once-through cycle are not directly weapons-usable until separation of plutonium takes place. Such effort requires constructing out-of-system facilities. Once such facility becomes operational, it could produce weapons-usable material from spent fuel within weeks of removal. Spent fuel accumulation in large quantity may create demand for reprocessing. Such demand may be for waste management or in anticipation of recycling or fast-breeder systems. Spent fuel under such development scenarios would represent a greater proliferation risk unless adequate safeguards are in place. To reduce the covert proliferation potential of the nuclear material, stringent safeguards on spent fuel in storage and in transit should be applied. Establishing an international or multinational storage system would be desirable as an alternative to alleviate the pressures of increasing accumulations of spent fuel.

A conventional PUREX-based system for reprocessing of spent fuel from LWR's (as discussed in Chap. 8) is found to have three important technical proliferation vulnerabilities. The first is that weapons-usable material is available in transit and in national facilities in forms that are relatively easy to exploit for weapons purposes. The second is the difficulty in nuclear safeguards of plutonium-bearing materials in bulk form. The third is the spread of sensitive expertise and knowledge through deployment of reprocessing and MOX fuel-fabrication facilities. These facilities, although started as civilian facility, can become starting points for a nuclear-weapons program or provide enhancement in an independent military program capability. These vulnerabilities indicate the strong needs for strengthened technical and institutional controls of reprocessing technologies. Use of advanced *technologies such as pyroprocessing or the UREX+ method is expected to enhance the proliferation resistance characteristics in reprocessing but these technologies have not been commercialized yet.*

If an enrichment plant is established in a non-nuclear weapons state with potential proliferation ambition, the situation presents very significant proliferation risk. While emphasizing cooperative arrangements with restrictions on technology transfer, limiting the number of enrichment facilities is necessary to maintain the current level of proliferation resistance associated with the once-through fuel cycle.

Since the 1977 Executive Order by President Carter in April 7th, 1977, the U.S. nuclear fuel cycle has remained once-through-cycle. This was mainly an outcome of considering proliferation resistance of nuclear fuel cycles. It should also be noted that depending on who owns and operates reprocessing or enrichment facility, the prospects of nuclear proliferation would be widely different.

15.2.3 International Regime for Nuclear Nonproliferation

International regimes are in place to control the risk of nuclear proliferation with respect to civilian application of nuclear technology. The most important one is the Nuclear Non-Proliferation Treaty (NPT). NPT was finalized in 1968 by the United Nations and went into effect on March 5, 1970. NPT has three-pillars, i.e., non-proliferation, disarmament, and the right to peacefully use nuclear technology. Within the NPT framework, countries are divided into nuclear weapons states (NWS) and non-nuclear weapons states (NNWS). The five states that had already tested nuclear weapons before January 1 1967 became NWS (the U.S., the Soviet Union, Britain, France, and China). The rest became NNWS. NNWS were asked to commit to nuclear nonproliferation by agreeing to refrain from any attempt to develop or acquire nuclear weapons. NWS were also committed to eventual nuclear disarmament. The right to peacefully use nuclear technology was given to NNWS as the compromise between NWS and NNWS. However, NNWS has to accept internationally administered safeguards on nuclear facilities. In 1995, the NPT was extended indefinitely.

The second important one is the International Atomic Energy Agency (IAEA) safeguards. IAEA safeguards was proposed as part of the Atoms for Peace Initiative announced by U.S. President Eisenhower on December 8, 1953. In October 1954, the IAEA Statute was approved and in July 1957 entered into force. IAEA safeguards on nuclear facilities began in 1959 on a limited scale. Then, in connection with the NPT, the so-called Comprehensive Safeguards Agreement (CSA) (INFCIRC/153) was developed in 1972 (IAEA 1972). Under the Agreement, all nuclear materials in peaceful uses in the state (as IAEA member) are under IAEA safeguards. States are required to meet domestic and international obligations by establishing and maintaining a State system of accounting for and control of nuclear material (SSAC).

IAEA safeguards were further strengthened in 1993 through the development of Strengthened Safeguards. This was after learning the presence of massive nuclear weapons infrastructure in Iraq in 1990 and to prevent covert nuclear proliferation attempts among IAEA compliant nuclear operations. This also led to the adoption of Additional Protocol (AP) in 1997 (IAEA 1997). The Additional Protocol marks the transition of the IAEA's safeguards approaches. The transition is from a quantitative system focused on accounting for known quantities of materials and monitoring declared activities to a qualitative system to develop a comprehensive picture of a state's nuclear and nuclear-related activities. Fuel cycle-related research and development and all nuclear-related imports and exports are also included as part of its coverage. IAEA is given the authority to visit any facility, declared or not, with multi-entry visas and access to modern means of communications to investigate questions about or inconsistencies in a state's nuclear declarations. As a way to reduce cost while improving efficacy, Integrated Safeguards was then introduced in the late 1990s and adopted in 2002. Under the system, a state's compliance with safeguards agreements is evaluated comprehensively, instead of on a facility-by-facility basis (as long as the state is a member of both CSA and AP).

Export control is another important component of the global nuclear nonproliferation regime. Between 1971 and 1974, international exports of nuclear technology were primarily controlled by the NPT Exporters Committee (known as the Zangger Committee). The control was through the use of a so-called trigger list of sensitive exports. After the Indian nuclear explosion in 1974, members of the NPT recognized the need to further strengthen export control and international safeguards. This led to the establishment of the Nuclear Suppliers Group (NSG, or called London Club) in 1975 which expanded the trigger list by including dual-use items. Export of the listed items to non-nuclear weapons states was only allowed if certain IAEA safeguards were agreed to. In contrast to the NPT Exporters Committee members, NSG members are not required to be parties to the NPT, but must adhere to instruments that contain equally binding commitments. Through this development, export of sensitive technology such as uranium enrichment and plutonium reprocessing plants was restrained (GAO 2002).

Various other international treaties and agreements for nuclear nonproliferation also exist. These include treaty for nuclear weapons free zones, Complete Test Ban Treaty (CTBT), UN Security Council resolutions (such as UNSCR 1540), and bilateral agreements between states regarding nuclear cooperation. Nuclear weapons free zones have symbolic significance in nuclear nonproliferation but don't have direct bearing with the issues of sensitive nuclear fuel cycles. CTBT was first negotiated in 1994 to prevent nuclear weapons test of any type by all states. Although 184 states have signed and 168 states have ratified it as of 2019, the treaty has not entered into force as eight of the key states with nuclear power reactors or research reactors have not approved it (China, Egypt, Iran, Israel and the United States have signed but not yet ratified; India, North Korea and Pakistan have not yet signed it). Bilateral agreements between states usually deal with the issues of nuclear fuel cycle and also play significant role in controlling the use of sensitive nuclear fuel cycle technologies. One of the key examples of such bilateral agreements is the

U.S. 123 Agreement. The 123 Agreement refers to Section 123 of U.S. Atomic Energy Act (US Congress 1954) which restricts the activities related nuclear fuel cycle by any cooperating states receiving nuclear materials or assistance from the U.S. There is also Treaty on the Prohibition of Nuclear Weapons (2017). The treaty is the first legally binding international agreement to comprehensively prohibit nuclear weapons, with the goal of leading towards their total elimination. As of July 2019, 24 nations have ratified the treaty and once 50 nations have ratified or acceded to it, it will enter into force. However, no nuclear-armed nation has expressed support for the treaty.

15.2.4 Principles of Nuclear Safeguards

As one of the key missions of the IAEA, the objective of nuclear safeguards is timely detection of diversion of significant quantities of nuclear material and deterrence of such diversion by early detection (IAEA 1997). This goal of detecting significant quantity of missing special nuclear material in a given time frame was pursued by defining “significant quantity” and “timeliness factor” based on ease of conversion to weapons. The ease of conversion to weapons as nuclear materials depends on the type of the material (e.g., metal, compounds, irradiated fuel, etc.) and generation of spontaneous neutrons and heat from the material.

The “significant quantity” (SQ) is defined as “the approximate amount of nuclear material for which the possibility of manufacturing a nuclear explosive device cannot be excluded”. The “timeliness factor” is defined as “the time required to convert different forms of nuclear material to the components of a nuclear explosive device”. Sometimes the term, special nuclear material, is used to refer to fissile material. The definition of significant quantity mainly deals with special nuclear material but also include the materials from which fissile can be derived such as natural uranium, depleted uranium (both for ^{235}U), and thorium (for ^{233}U).

The significant quantities for different nuclear materials defined under the CSA (INFCIRC/153) are given in Table 15.7 (IAEA 1972). As shown, 1 SQ is defined as 8 kg for plutonium. For HEU with ^{235}U enrichment level greater than 20%, 1 SQ is defined as 25 kg. The timeliness factors of different material forms indicate the time needed for conversion of the material into a nuclear explosive device and are given in Table 15.8. For example, the timeliness factor is defined as 7–10 days for metals and 1–3 weeks for oxides. Intervention within the timeliness factor is expected to be effective in preventing nuclear proliferation using these materials.

These nuclear safeguards goals are achieved by applying various measures including nuclear materials control and accounting, containment and surveillance, inspection by visit, and environmental sampling.

Nuclear materials control and accounting (MC&A), sometimes called nuclear materials accountancy (NMA), refers to the activity for material balance accounting of nuclear materials in a user facility to confirm the their presence and detect potential theft, loss, or diversion of them that trigger an appropriate response.

Table 15.7 Values of SQ for different nuclear materials (IAEA 1972)

Material	SQ
Direct use nuclear material	
Pu (with less than 80% ^{238}Pu)	8 kg
^{233}U	8 kg
HEU (^{235}U $\geq$ 20%)	25 kg
Indirect use nuclear material	
LEU (^{235}U < 20%)	75 kg
Natural U	10 ton
Depleted U	20 ton
Th	20 ton

SQ: "the approximate amount of nuclear material for which the possibility of manufacturing a nuclear explosive device cannot be excluded"

Table 15.8 Timeliness factors for different beginning nuclear materials forms

Beginning material form	Timeless factor (conversion time)
Pu, HEU, or ^{233}U metal	Order of days (7–10)
PuO_2, $Pu(NO_3)_4$, or other pure Pu compounds	Order of weeks (1–3)
HEU or ^{233}U oxide or other pure U compounds	
MOX or other non-irradiated pure mixtures containing PU, U (^{233}U + ^{235}U > 20%)	
Pu, HEU and/or ^{233}U in scrap or other miscellaneous impure compounds	
Pu, HEU or ^{233}U in irradiated fuel	Order of months (1–3)
U containing less than 20% ^{235}U and ^{233}U; Th	Order of months (3–12)

Timeliness factor: "the time required to convert different forms of nuclear material to the components of a nuclear explosive device"

Here, nuclear materials refer to the materials included in the definition of "SQ" with potential use in a nuclear weapon. NMA is based on examining quantities and transfers of all nuclear materials in and out of a facility by preparing and maintaining accounting records, performing measurements, and analyzing the information

A key concept in MC&A is MUF (material unaccounted for). MUF is the missing inventory of nuclear materials defined as the difference between the measured inventory and what is expected to be in the inventory based on the previous inventory and measured flows into and out of the process. The MUF is calculated as,

$$\begin{aligned} MUF = {} & Beginning\ Inventory + Receipts - Shipments \\ & - Ending\ Inventory \end{aligned} \tag{15.6}$$

Calculating MUF at a facility is conducted by defining so-called material balance area (MBA). MBA can be a single room or an entire facility, depending upon how nuclear materials are handled in what form in the facility. Within a defined MBA, the physical inventory of nuclear material must be determined to establish material balance. The overall mass balance within a facility is based on combining the results from all MBAs and must be checked against MUF. A MUF at a facility is not zero because of the presence of measurement errors. However, the MUF at a facility must be maintained below 1 SQ to be compliant. If significant discrepancies and anomalies exist, the situation must be promptly identified and reported to IAEA.

Containment and surveillance (C/S) is a supplemental system to complement NMA. Here containment refers to structural features of a facility, containers, or equipment used to determine the physical integrity of an area or items. Surveillance refers to collecting information mainly through instrumental observation to monitor operations or detect movements of nuclear material or other items including any interference with containment, surveillance, or tamper indicator tools. The key objective of C/S is to provide and maintain critical knowledge about nuclear materials throughout a nuclear facility. Providing and maintaining critical knowledge about nuclear materials throughout a nuclear facility is also called continuity of knowledge (CoK). Preventing undetected access to, or movement of the nuclear material through surveillance within the containment becomes an important part of maintaining CoK. Most common surveillance tools are cameras and video cameras. Various tamper indicators are also used to identify unauthorized attempts to access or transfer nuclear materials. These are typically called seals. These include the metal cap seal, the variable-coded fiber optic seal, and electro-optical sealing system. CoK regarding the movements of nuclear materials and changes in the physical integrity of the materials plays a key role in timely detection of diversion of significant quantities of nuclear material.

Inspection is to have physical access to the locations of nuclear materials by IAEA inspectors through on-site visits. Records and reports on nuclear material accountancy are compared and existence of nuclear materials as recorded is verified through inspection.

Further verification related to the activities of handling and processing of nuclear materials is made by environmental sampling. Environmental (swipe) samples are collected and analyzed to verify the absence of undeclared nuclear material. This is in contrast to other safeguards verification methods which aim to verify the presence of declared nuclear material.

15.2.5 Nuclear Safeguards and Nuclear Waste Management

The most important type of nuclear waste as the target of nuclear safeguards is spent fuel. This is because spent fuel presents opportunities to extract special nuclear material. Any facilities where spent fuels or special nuclear materials are handled, stored, or treated become significant in terms of nuclear safeguards. These facilities include nuclear power plants and the facilities for spent fuel storage, reprocessing, and MOX fuel fabrication.

Within the context of current civilian nuclear power program, the key goal of nuclear safeguards in the context of nuclear waste management is to detect the diversion of 1 SQ of special nuclear materials in the handling or processing of spent fuel within the timeliness limits. This goal can be translated into detecting diversion of 8 kg of plutonium within 3 months of possible diversion and 75 kg of LEU (uranium containing less than 20% of ^{235}U) within 1 year of possible diversion.

As mentioned in the previous section, application of nuclear safeguards for NMA is through designation of a nuclear material balance area (MBA). How a MBA is designated in a facility depends on how the spent fuel is handled or treated. For example, when spent fuel is in storage, each spent fuel exist as assemblies in containers. In this case, the spent fuel storage facility is considered an item facility with the whole facility designated as one MBA. In the case of reprocessing, spent fuel is segmented and the contents exist as bulk material in solutions. Then, the facility is treated as bulk-handling type facility which is composed of several distinct MBAs.

Nuclear materials going in and out of a MBA must go through non-destructive measurements at a so-called key measurements points (KMPs). The measurement results at each KMP provides the basis for verification of nuclear materials inventory at the facility. In a spent fuel storage facility, there are only two KMPs as the whole facility is a single MBA. In a reprocessing facility, there are multiple KMPs to verify the transfer of nuclear materials inventory between MBAs.

Verification of facility design information declared by the facility operator is another important part of nuclear safeguards for spent fuel. The verification is to assure that the operation of a facility does not create opportunities for nuclear proliferation. The facility operator must prepare and submit so-called Design Information Questionnaire (DIQ) through the national authorities when the facility is constructed. DIQ is a form or process to support the verification of design information and can be updated as required. Through inspection, IAEA inspectors go through a process called Design Information Verification (DIV) to verify the correctness of the DIQ to ensure that the operation of the facility followed the declarations made by the operator regarding handling, movement, or treatment of spent fuel. Through this process, opportunities for nuclear proliferation through facility upgrades or modifications are prevented. The current DIV activities are labor intensive with reliance on personal experiences and capabilities. To improve the practice, new tools such as 3-dimensional laser-based surveyor, Compton gamma radiation imaging, and geophysical methods using ground penetrating radar have been developed.

Nuclear Safeguards for Spent Fuel Storage

Spent fuels are typically stored at on-site storage pools of operating nuclear reactors. If the storage pools reaches the capacity limit, spent fuels are moved to AFR (away-from-reactor) storage facility or Independent Spent Fuel Storage Installations (ISFSIs) for interim storage. As explained, the spent fuel storage facility is taken as single MBA for nuclear materials accountancy. This is shown in Fig. 15.4 designating one MBA and two KMPs for the facility.

All nuclear material during spent fuel storage is contained in identifiable items such as fuel assemblies, sealed canisters, or storage casks. For NMA, the quantity of nuclear material contained in each item must be determined through measurement or

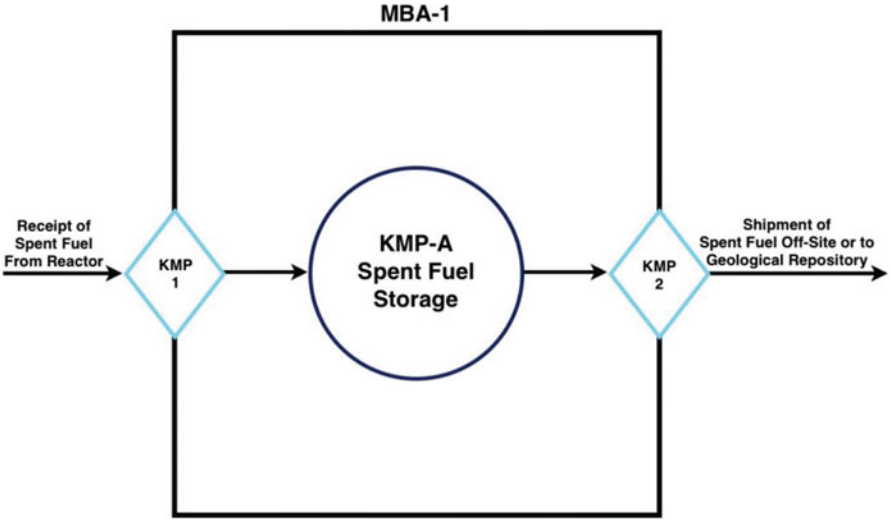


Fig. 15.4 Schematic of material balance area (MBA) and key measurement points (KMPs) at an AFR spent fuel storage facility. (Source: Durst 2012)

estimation. It is assumed that the content of nuclear materials during storage does not change, except for the changes due to radioactive decay. Under such considerations, nuclear safeguards of spent fuels are based on item counting and identification, item integrity examination, and non-destructive measurements of the contents. This accounting procedures are supplemented by C/S measures through the use of seals and cameras to provide CoK.

Nuclear Safeguards for Reprocessing

To assure non-diversion of 1 SQ (i.e., 8 kg) of plutonium, the reprocessing plant, as a bulk-handling facility, is typically divided into three MBAs (See Fig. 15.5): the first one includes the receipt and storage area, the head-end processes, and fuel dissolver; the second one is the main process areas for the separation processes; the third one includes the areas for waste treatment and product storage. In between the MBAs, KMPs are set up to perform measurements of nuclear materials inventory. In particular, input accountability tank is set up as part of KMPs between MBA1 and MBA2 to measure the fissile content in the dissolved liquids (through sampling from homogeneously mixed solution) that moves to separation processes. The result determines how much fissile goes into the separation processes.

As plutonium in solution flows in large quantity through the system, accurate material-accountancy measurements to detect diversion in high-throughput aqueous operations is a significant challenge. To cope with such challenge, process monitoring and near-real time accountancy (NRTA) are employed to improve the accuracy associated with the measurements. Process monitoring is to obtain real-time data on inter-vessel transfers as a way to ensure proper operation of the facility according to the operator’s declarations. Process monitoring also provides input data for NRTA.

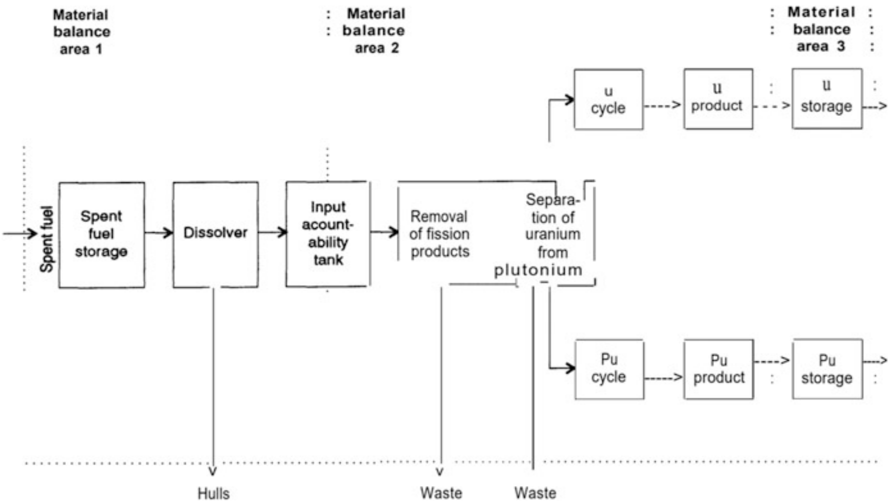


Fig. 15.5 Material balance areas for spent fuel reprocessing plant. (Source: OTA 1995)

NRTA is a method to address potentially large uncertainty in inventory characterization through improvements in timeliness and diversion detection sensitivity. In a very simplistic sense, NRTA could be viewed as performing nuclear materials accountancy in a very frequent manner. In NRTA, process data are collected in near-real time by the computerized nuclear materials control and accounting system, and the data are analyzed using statistical techniques. Computerized evaluation algorithms are then employed to calculate variations in the material inventory to determine MUF uncertainty and diversion detection probability. Availability of measurement techniques that provide the required sensitivity and timeliness is necessary to implement NRTA. Such approach has been shown to be capable of detecting diversion and unexpected losses from the process area (Cobb 1981; Shipley 1982).

These approaches of nuclear materials accountancy is further supplemented by C/S techniques using seals and camera or video surveillance. The C/S techniques are important to keep track of items in the receiving (input storage) area, product storage area, and in transit between the input storage area to head end processing. In addition, safeguards-relevant design information for the facility is verified through inspections. Presence of any physical means for material removal should be detected through verification of plant design information.

Nuclear Safeguards for Geologic Disposal

Nuclear waste eventually goes into a geological repository for final disposal. Due to the presence of fissile materials in nuclear waste, nuclear safeguards remains important in geological repositories. As the wastes are contained in closed disposal canisters (i.e., as discrete items), the geological repository is an item accountancy facility with one MBA. Nevertheless, application of nuclear safeguards to geological repositories to detect any diversion attempts presents unique challenges.

The repository facility is located entirely underground. Therefore visibility is not given. Due to the difficulty in applying conventional C/S techniques for long time periods, maintaining CoK about nuclear wastes emplaced in the tunnels is not easy. Unlike other nuclear facilities, geological repositories are not built in their entirety before commencing operations. Even after waste emplacement operations begin, construction of tunnels continues. If unanticipated underground conditions such as faults are encountered during construction, changes to the design may be necessary. Once emplacements are completed, human access is prohibited and inspection of a completed facility prior to operation is not allowed. Also, spent fuel packages cannot be re-verified by humans once they are emplaced in the facility. It is also hard to verify the emplacement of the waste/spent fuel as declared with no presence of inspectors. Even if a hidden, closed, sealed tunnel for spent fuel diversion is present, identifying the presence of such tunnel is difficult. Even if detection of any diversion attempts is made, the timeliness requirement may not be met.

While these challenges are recognized, nuclear safeguards in a geological repository is pursued by various means (IAEA 2018): (1) Verifying design information during design, construction, and operation of the facility to confirm that the physical structure and operations remain the same as declared; (2) Verifying receipts and flows of nuclear materials; (3) Verifying nuclear materials contents of incoming spent fuel containers before emplacement, and; (4) Maintaining CoK of the spent fuel inventory and on the contents of nuclear materials through C/S.

Design information verification for an underground facility is performed potentially three to six times per year by using underground mapping techniques and remote sensing methods. These methods include seismic and acoustic monitoring and ground-penetrating radar technique. Satellite imagery would also be used to detect suspicious construction activities or any changes over time at the site on the surface. The C/S system for a geological repository is an integrated system consisted of motion and radiation detectors, optical surveillance, safeguards seals, and wireless tags to monitor any movements.

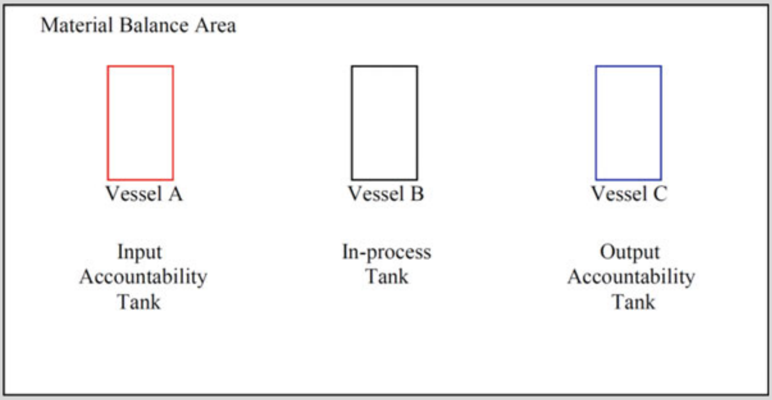
Example 15.2: Nuclear Materials Accountancy Using MBA Analysis (Goergen 2010)

A Material Balance Area (MBA) in a hypothetical facility for processing special nuclear material (SNM) is depicted in a figure below. The inventory is tracked by the mass of SNM while transfers are made into and out of the MBA during the inventory period. The MBA has a number of vessels for processing material including Vessel A (the input accountability tank), Vessel B, the in-process tank, and Vessel C (the output accountability tank). The inventory difference is calculated at the end of the material tracking period

(continued)

Example 15.2 (continued)
using the following relationship. The inventory difference should be less than 1 SQ.

$$\begin{aligned} Inventory\,Difference &= Beginning\,Inventory + Transfers\,In - Transfers\,Out \\ &\quad - Ending\,Inventory \end{aligned}$$



Material balance area

Ideally the actual inventory difference is zero at the end of the material tracking period. However, this is hard or unrealistic to achieve since there are always small quantities of material that may not be accounted for along with the presence of measurement errors. One example is holdup. Holdup is material that is located in the process that is undetected by usual measurements. Examples may include plating of material on the surfaces of equipment and solids that might get lodged in inaccessible areas. Theft or diversion of material would be another example of unaccounted for material. Also, measurement and sampling errors will make the calculations inexact. The limit of error (LOE) is a measurement of the uncertainty to be expected in this system for calculating an inventory difference. A description of inventory and transfer measurements for the tracking period is given in the Table 15.9 below.

Characterizing errors in these measurement are important part of nuclear materials accountancy. The following Table 15.10 lists both systematic errors and random errors in the related measurements in all three vessels. Here systematic errors refer to errors in a measurement due to systematic issues such as mistakes in observation or imperfect instrument calibration. Random errors refer to errors in a measurement due to unknown and unpredictable

(continued)

Table 15.9 Measurements during the material tracking period

Vessel	Transaction Type	Concentration (g/L)	Volume (L)	Mass (g)
A	Beginning inventory	10.1	100	1010
A	Transfer in	11.8	10	118
A	Transfer in	9.1	50	455
A	Ending inventory	9.4	60	564
B	Beginning inventory	6.9	80	552
B	Ending inventory	8	90	720
C	Beginning inventory	8.1	10	81
C	Transfer out	9.9	35	346.5
C	Ending inventory	9.9	60	594

Table 15.10 Description of errors in measurements

	Measurement type	Systematic error (σ) in %	Random error (σ) in %
Vessel A	Sampling	0.0	5.0
	Concentration	0.5	1.0
	Volume	1.0	2.0
Vessel B	Sampling	0.0	5.0
	Concentration	1.0	2.0
	Volume	2.0	2.0
Vessel C	Sampling	0.0	5.0
	Concentration	1.0	2.0
	Volume	1.0	2.0

Example 15.2 (continued)
causes such as noise in the instrument signal, effect of random changes in temperature or moisture.

Based on the given information, answer the following.

(a) What is the beginning and the ending inventory in kilograms?
(b) What is the inventory difference for the material tracking period?
(c) Determine the limit of error (LOE). (LOE = 2*Sigma of inventory difference)
(d) Did the absolute value of the inventory difference exceed the limit of error?
(e) Which measurement types listed in Table 15.10 were the largest contributors to the limit of error?
(f) What conclusions can be drawn from the results?

Solutions:

(continued)

Example 15.2 (continued)

(a) **What is the beginning and the ending inventory in kilograms (kgs)?**

An MBA inventory is the mass of SNM in the MBA at a given time. In a large facility, the inventory is usually expressed in kilograms (*kgs*) of SNM. The MBA inventory is the sum of the inventories at all the separate locations that constitute the MBA. In this example problem, there are three locations: Vessels A, B, and C.

$$\text{Inventory}_{\text{MBA}} = \sum_{\text{All MBA Locations}} \text{Inventory}_{\text{Location i}}$$

The most common way of determining the mass of SNM in a vessel, having all of its SNM dissolved in a well-mixed solution, is by drawing a representative sample of the solution on which to perform a concentration measurement and then measuring the volume of the solution left in the tank. If the concentration and volume measurement units are conformable (e.g., the concentration is in grams per liters and the volume is in liters, so no volume unit conversion is necessary), and the mass of SNM in the vessel equals their product:

$$\text{Inventory}_{\text{Location i}} = \text{Concentration}_{\text{Location i}} \times \text{Volume}_{\text{Location i}}$$

The inventory calculations appear in the table below (*g* and *L* refer to grams and liters, respectively; mass must be converted to *kg* in the end). Using the above formulas, we obtain the following.

Beginning Inventory = (10.1×100) + (6.9×80) + (8.1×10) = 1010 + 552 + 81 = 1643 g = 1.643 kg

Ending Inventory = (9.4×60) + (8.0×90) + (9.9×60) = 564 + 720 + 594 = 1878 g = 1.878 kg

Vessel	Transaction Type	Concentration (g/L)	Volume (L)	Mass (kg)
A	Beginning inventory	10.1	100	1.010
B	Beginning inventory	6.9	80	0.552
C	Beginning inventory	8.1	10	0.081
All vessels	Beginning inventory	**1.643**		
A	Ending inventory	9.4	60	0.564
B	Ending inventory	8	90	0.720
C	Ending inventory	9.9	60	0.594
All vessels	Ending inventory	**1.878**		

(continued)

Example 15.2 (continued)

The ending inventory (1.878 kg) exceeded the beginning inventory (1.643 kg). A difference between inventories is not unexpected since there have been transfers into and out of the facility during the material tracking period.

(b) **What is the inventory difference for the material tracking period?**

The total mass of SNM transferred into and out of this MBA during the material tracking period is as follows.

$$\mathit{Transfer\ In} = (11.8\times10) + (9.1\times50) = 118 + 455 = 573 \text{ g} = 0.573 \text{ kg}$$
$$\mathit{Transfer\ Out} = (9.9\times35) = 346.5 \text{ g} = 0.3465 \text{ kg}$$

The inventory difference (*ID*) is a mass balance calculated as follows.

$$\mathit{ID} = \mathit{Beginning\ Inventory} + \mathit{Transfers\ In} - \mathit{Transfers\ Out} - \mathit{Ending\ Inventory}$$
$$\mathit{ID} = \mathit{1.643} + \mathit{0.573}\text{–}\mathit{0.3465} - \mathit{1.878} = -\mathit{0.0085kgs}$$

A negative ID reflects a loss. Since these calculations were based on sampling and measurements which are subject to error, it remains to substantiate whether the apparent loss of material can be explained by the magnitude of errors (noise in the system) or if the size of the ID is larger than can be explained by the magnitude of the sampling and measurement errors.

(c) **Determine the limit of error (LOE). (LOE = 2*Sigma of inventory difference)**

The limit of error (LOE) = 2*Sigma of inventory difference = $2^*\sqrt{\mathit{Variance\ of\ inventory\ difference}}$

Variance of inventory difference = Variance (from systematic sampling (ss) error) (1) + Variance (from random sampling (rs) error (2) + Variance (from systematic concentration (sc) error) (3) + Variance (from random concentration (rc) error) (4) + Variance (from systematic volume (sv) error) (5) + Variance (from random volume (rv) error) (6)

Variance from systematic sampling error (1) = $\sum\limits_{i=A,B,C} (ID)_i^2 \cdot \left(\frac{\sigma_{ss,i}}{100}\right)^2 =$
$(1,010 + 118 + 455 - 564)2\left(\frac{0}{100}\right)^2 + (552 - 720)^2\left(\frac{0}{100}\right)^2 +$
$(81 - 594 - 346.5)^2\left(\frac{0}{100}\right)^2 = 0$

Variance from random sampling error (2) = $\sum\limits_{i=A,B,C}\left(\sum\limits_{j=1}^{\mathit{number\ of\ transactions}}\left(mass_i^j\right)^2\right)\cdot\left(\frac{\sigma_{rs,i}}{100}\right)^2 =$

$\left((1,010)^2 + (118)^2 + (455)^2 + (-564)^2\right)\left(\frac{5}{100}\right)^2 +$

(continued)

Example 15.2 (continued)

$$\left((552)^2+(-720)^2\right)\left(\tfrac{5}{100}\right)^2+\left((81)^2+(-594)^2+(-364.5)^2\right)^2\left(\tfrac{5}{100}\right)^2=$$
$7,154,271\ g^2$

Notice, in the case of systematic errors, it is assumed that the systematic error affects the mass involved with each transaction equally. Therefore, the mass in each transaction is simply added without considering the mass dependent relative variance effect of each transaction. However, in the case of random errors, it is assumed that the mass in each transaction is affected by random error. Therefore, mass dependent relative variance is calculated for the mass involved with each transaction. Similar approaches are taken below.

Variance from systematic concentration measurement error (3) =
$$\sum_{i=A,B,C}(ID)_i^2\cdot\left(\tfrac{\sigma_{sc,i}}{100}\right)^2=(1,010+118+455-564)^2\left(\tfrac{0.5}{100}\right)^2+$$
$$(552-720)2\left(\tfrac{1}{100}\right)^2+(81-594-346.5)^2\left(\tfrac{1}{100}\right)^2=131.5347\ g^2$$

Variance from random concentration measurement error (4) =
$$\sum_{i=A,B,C}\left(\sum_{j=1}^{number\ of\ transactions}\left(mass_i^j\right)^2\right)\cdot\left(\tfrac{\sigma_{rs,i}}{100}\right)^2=$$
$$\left((1,010)^2+(118)^2+(455)^2+(-564)^2\right)\left(\tfrac{1}{100}\right)^2+$$
$$\left((552)^2+(-720)^2\right)\left(\tfrac{2}{100}\right)^2+\left((81)^2+(-594)^2+(-364.5)^2\right)^2\left(\tfrac{2}{100}\right)^2=$$
$676.9398\ g^2$

Variance from systematic volume measurement error (5) =
$$\sum_{i=A,B,C}(ID)_i^2\cdot\left(\tfrac{\sigma_{sv,i}}{100}\right)^2=(1010+118+455–564)^2\left(\tfrac{1}{100}\right)^2$$
$$+(552–720)^2\left(\tfrac{2}{100}\right)^2+(81-594-346.5)^2\left(\tfrac{1}{100}\right)^2=188.9997\ g^2$$

Variance from random volume measurement error (6) =
$$\sum_{i=A,B,C}\left(\sum_{j=1}^{number\ of\ transactions}\left(mass_i^j\right)^2\right)\cdot\left(\tfrac{\sigma_{rs,i}}{100}\right)^2=$$
$$\left((1,010)^2+(118)^2+(455)^2+(-564)^2\right)\left(\tfrac{2}{100}\right)^2+$$
$$\left((81)^2+(-594)^2+(-364.5)^2\right)^2\left(\tfrac{2}{100}\right)^2=1,144.683\ g^2$$

By using these estimated variances:

Variance in inventory difference = (1) + (2) + (3) + (4) + (5) + (6) = 0 + 7,154.271 + 131.5347 + 676.9398 + 188.9997 + 1,144.683 = 9296.428 g^2

(continued)

Example 15.2 (continued)

$\sqrt{Variance\ of\ inventory\ difference} = \quad = \quad \sqrt{9296.428} = 96.418\ g = 0.0964\ kg$

$\text{LOE} = 2^{*}\sqrt{Variance\ of\ inventory\ difference} = 2^{*}0.0964\ \text{kg} = 0.193\ \text{kg}$

(d) **Did the absolute value of the inventory difference exceed the limit of error?**

The absolute value of the inventory difference is 0.0085 kg. This is less than the LOE of 0.193 kg.

(e) **Which measurement types listed in** Table 15.10 **were the largest contributors to the limit of error?**

From the results in part c), the biggest contributor was the random sampling variance (7154.271 g2) within the total 9296.428 g2 (contributing 77% of the total variance).

(f) **What conclusions can be drawn from the results?**

The inventory difference is much less than the limit of error and appears to be not different from zero. The confidence interval as $ID \pm 2\sigma$ is (−0.0085–-0.193 = −0.201 kg, −0.0085 + 0.193 = 0.185 kg). There seems to be no gain or loss of special nuclear material during the mass tracking period.

Therefore, there is no concern in terms of NMA at the facility.

15.3 Nuclear Security

15.3.1 Basic Concepts of Nuclear Security

A closely related concept to nuclear nonproliferation is nuclear security. Nuclear security refers to the activities related to protection of nuclear materials or other radioactive substances against sabotage or theft from the identified threats (IAEA 2013). While nuclear safeguards focuses on the intentional misuse of special nuclear materials by state actor to build nuclear weapons, nuclear security focuses on the intentional misuse of nuclear or other radioactive materials by terrorists (non-state actors) to cause harm.

During the early days of nuclear technology development, the need for physical protection of nuclear materials in various facilities was recognized. Guns, guards, gates, and fences were used for such purpose. Since then, design of a physical protection system has evolved over the years with technological advances and the objectives of preventing, detecting, delaying, responding to, interrupting, and neutralizing a malevolent human adversary (including international actors). Today nuclear security is much more broadly defined referring to all activities related to minimizing/reducing nuclear security risk.

Nuclear security risk is the likelihood of an attack or sabotage by an adversary involving nuclear materials resulting in harmful consequences. Such risk can be represented by the following relationship.

$$Risk = Threat \times Vulnerability \times Consequence \tag{15.7}$$

This relationship indicates that nuclear security risk depends on three major components: threat, vulnerability, and consequence.

Threat represents the likelihood that an attack could occur by an adversary. The attack could be a sabotage on a nuclear facility (nuclear power plant or nuclear fuel cycle facilities) to result in release of radioactive materials. The attack could also directly involve using nuclear devices such as nuclear weapons or radiological dispersal device (RDD, so-called dirty bomb). Likelihood of such attack occurring will depend on the adversary's intention and capability to carry out an actual attack. Vulnerability reflects the likelihood of the attack, if launched, being successful. Therefore vulnerability depends on the effectiveness of protection system or the adversary's attack plan, resources and capabilities. Consequences are the impacts caused by an attack, including both tangible and non-tangible ones. Tangible impacts include deaths, disease occurrences, damage to properties, or economic losses. Non-tangible impacts include the effects on consumer confidence or national pride or anything that would negatively affect the country under the attack. Consequences will depend on specific targets being attacked, readiness of the national infrastructure to handle the attack (e.g., medical systems for emergency response), government's capability to continue effective governance, and natural weather or atmospheric conditions at the time of attack. Threat is also affected by the vulnerability of the target and consequences as the decision to attack is linked to the effectiveness of the protection system and the magnitude of the consequences.

Once threat in nation specific situations is defined, it constitutes so-called design-basis threat (DBT). DBT provides a common basis for determining security requirements and system performance evaluation.

The best course of action in dealing with nuclear security risk is to prevent attack. However, if prevention is not attainable, strategies are needed to decrease the threat, vulnerabilities and consequences. The strategies applied to decrease the threat are called countermeasures. The related process to copy with nuclear security risk include: 1) Assessing risk, 2) Identifying countermeasures, 3) Weighing cost/benefit of each countermeasure, 4) Implementing countermeasure plan, 5) Evaluating impact, and; 6) Adaptation as necessary.

Implementing nuclear security principles in nuclear waste management is through the application of appropriate countermeasures to prevent the acquisition of nuclear materials by adversary or to avoid radioactive material releases from nuclear waste by sabotage. Examples of such countermeasures are to provide adequate security at facilities or during transit and better accounting of nuclear materials at facilities, or to design the nuclear waste package system to withstand large external impacts.

15.3.2 International Regime for Nuclear Security

As a matter of national sovereignty, each state takes the responsibility for nuclear security by defining and managing potential threat to any facilities handling nuclear materials within the territory. At the same time, as the ambitions of nuclear terrorism by terrorist groups like Al Qaeda, Chechen rebels, or Aum Shinrikyo were disclosed, nuclear security emerged as global issue. In particular, the September 11, 2001 terrorist attack in New York clearly demonstrated the potential impact of a major terrorism incident on a global scale. The incident not only resulted in a large number of casualties but also caused enormous damage to international trade, global tourism, labor productivity, etc. Close international collaboration is required to deal with nuclear terrorism.

The Fukushima nuclear power plant accidents in 2011 was also an important milestone in nuclear security developments. About a half of Japan Ground Self Defense Force was mobilized to handle emergency response measures after the accident. For an extended period, the nation was under crisis situation while radioactive materials were being released to the atmosphere and a large group of people were relocated. The accident reminded of the possibility of large, cross-border releases of radioactive materials through a terrorist attack on a nuclear facility. The accident also indicated that nuclear safety and nuclear security are closely related.

One of the earliest international legal instruments to address nuclear security is IAEA's INFCIRC/225 in 1972 (revised in 1977, 1989, 1993 and 1998). It is entitled, "The Physical Protection of Nuclear Material and Nuclear Facilities". The document serves as recommendation to provide a guideline on establishing a framework for managing nuclear security.

The international legal framework against nuclear terrorism is constituted by the international conventions and UN Security Council resolutions such as the Convention on Physical Protection of Nuclear Materials (CPPNM), International Convention for the Suppression of Terrorist Bombings (ICSTB), International Convention for the Suppression of Acts of Nuclear Terrorism (ICSANT), and UN Security Council Resolution 1540.

With the focus on physical protection of nuclear material, the CPPNM establishes State obligations to protect nuclear facilities and materials in peaceful domestic use, storage, and in transport. The CPPNM was first adopted in 1979 through IAEA, entered into force in 1987 and has been amended in 2005. With the amendment, it was renamed the Convention on the Physical Protection of Nuclear Material and Nuclear Facilities. As of 2018, 157 State Parties plus the European Atomic Energy Community joined the Convention as signatory.

ICSTB was designed to criminalize terrorist bombings and requires State Parties to make effort to prevent, investigate and punish those acts. ICSTB was adopted in 1998 at the UN General Assembly and became effective as of 2001 with 170 state parties.

ICSANT also criminalizes acts of international nuclear terrorism and requires States Parties to make effort to adopt appropriate measures to ensure the protection of nuclear material and to investigate and punish those acts. ICSANT was adopted in 2005 at the UN General Assembly and brought into effect in 2007. It has 114 State Parties as of 2018.

UN Security Council Resolution 1540 requires the members of UN to implement domestic legislation and control to prevent the proliferation of Weapons of Mass Destruction (WMD) and their means of delivery and related materials. It also created a process to monitor compliance by the states. A notable provision is about State's responsibility to prevent non-state actors (i.e., subnational groups) from developing, acquiring, possessing, transporting, transferring or using WMDs and their means of delivery. For seizure of illegal material transfers, international authorization was provided and use of sanctions or military force in response to international security threats was granted to the Security Council. The resolution was adopted by the United Nations Security Council in 2004.

UN Security Resolution 1373 (2001) also has an important relevance to nuclear terrorism through establishing the process for counter-terrorism including prevention and suppression of the financing of terrorism and promotion of international cooperation.

While a number of international instruments exist as listed above, no single one addresses nuclear security in a comprehensive manner yet. Also a large number of states remain outside of these international agreements. Also some of the member states have not followed through with implementation of them in their national legal and regulatory frameworks. These gaps in the global system remain a concern.

15.3.3 Physical Protection System for Nuclear Facilities

To perform the necessary task of nuclear security for nuclear waste management, the physical protection system is designed and implemented at the facilities where nuclear materials are present. These facilities include nuclear power plants, uranium enrichment plants, spent fuel storage facilities, reprocessing plants, MOX fabrication plants, and geological repositories.

Design of a physical protection system starts with the determination of system objectives based on the understanding of threats to the facility and potential consequences of any attacks (see Fig. 15.6). Key objective of the design is to overcome the threat by combining technology, humans, and system operations dedicated to nuclear security. Class, capabilities, and range of tactics of potential adversaries, and the outcome of threat identification must be considered as part of defining what is required in the design.

Given the system objectives, initial design of the system is developed. The design should meet the objectives within the operational, safety, economic, and legal constraints of the facility. In this regards, an effort is made to identify the critical assets or areas to be protected to prevent undesirable events. Such effort is called vital area identification (VAI). Here, the undesirable events refer to the adversary

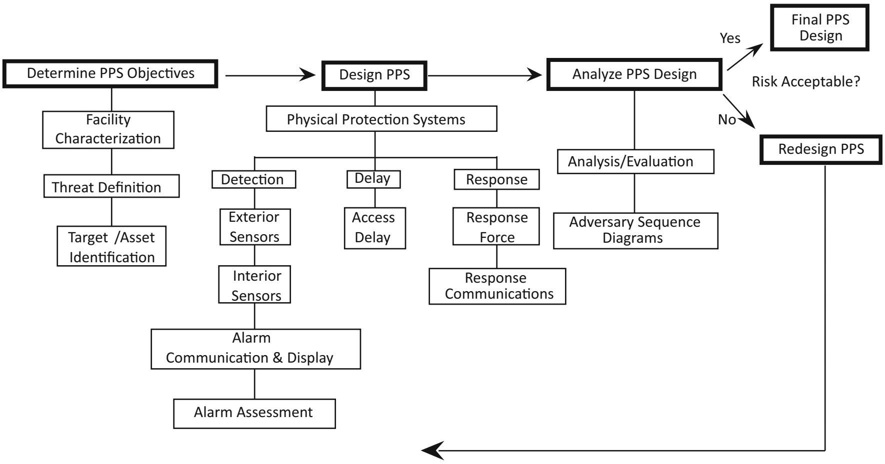


Fig. 15.6 Design of physical protection system. (Source: Duran and Cipiti 2009)

obtaining the fissile material or the adversary succeeding in releasing radioactive materials to the environment from a nuclear waste handling facility.

VAI is a structured approach of target identification typically based on using so-called fault trees. In a traditional sense, fault tree analysis refers to an analysis by using graphic representation of the combinations of components and subsystem events leading to a specified undesired state. Therefore, specific combinations of events that lead to undesired state(s) are identified. For example, in VAI, sabotage fault trees are constructed and used to depict the combinations of security failures of rooms leading into the access to the special nuclear material by terrorists in the facility. Through the analysis, minimum sets of locations are identified to prevent an adversary from accomplishing successful sabotage. This minimum sets are the most economical set of locations to protect for the prevention of sabotage. Therefore, the rooms in this selected set become vital areas to be protected. For example, the vital areas in nuclear waste management would be the rooms where fissile materials are present in readily accessible packages.

Once the vital areas are identified, the next step is to design the system to secure these vital area(s). The design is based on performing key security functions such as detection, delay, and response to protect the vital areas.

Detection, the first function of a physical protection system, is to discover an adversary action. The action by an adversary could be both overt and covert. The means for detection include visual observations, entry/access control, video surveillance, electronic sensors, accountancy records, tamper indicating devices (e.g., seals), and process monitoring systems. Also, performing the task of detection is more than sensing the action but includes activities of generating an alarm, communicating the alarm, and correctly assessing the alarm for response decisions.

Delay, the function following detection of adversary action, is to impede an adversary's attempt to gain unauthorized access so that the response force has adequate time to interrupt the adversary from completing their desired task. The means of delay include delay barriers such as, fences, gates, entry/access control, or vehicle barriers, the barriers as part of building structures (e.g., walls, locked doors, cages, windows, roofs, or floors), and the barriers used in dispensable forms (e.g., rapidly dispensable rigid foams, sticky foams, aqueous foams, sticky sprays, slippery sprays, sand columns, noise, lights, smoke, and rubble piles).

Response is to prevent the adversary from completing their task through neutralization. Examples of neutralization include forcing the adversary to flee, capturing the adversary, or incapacitating the adversary. The task is performed by the response force which includes on-site guards or off-site agencies such as law enforcement or military personnel with the use of arms. The capability and deployment of response force should be commensurate to the capabilities of potential adversaries to achieve successful neutralization of them. The nature of successful neutralization depends on the target of attack and must be defined in the objectives of the design process.

Another potential function to be considered in design is deterrence, i.e., to dissuade the adversary from launching an attack. However, it is difficult to measure success in deterrence. Its use in design is also subjective. Therefore, deterrence is not explicitly considered as part of the physical protection system design. A suitably robust nuclear security system (based on detection, delay, and response) should help to deter a malicious act (IAEA 2013).

The selected design is evaluated with respect to the system performance measures. The performance measures include the probability of detection or probability of system failure, time to respond, response force effectiveness after deployment, and cost. If the system is found inadequate in meeting the performance measures, a redesign or refinement of the system is needed and its performance is re = evaluated. For example, if the probability of system failure is greater than what is specified in the design objectives, the design is not acceptable. If the time to respond is less than the time needed by the adversary to gain access to the target or to sabotage the facility, the design has to be revised.

Figure 15.7 shows the details of how the performance measure of "time to respond" is assessed in comparison to adversary task time. The figure indicates that "response force time" begins only after the detection of adversary action. Therefore, there exists a time gap between first sensing and detection of adversary action. In contrast, the adversary task time starts when the adversary start the action of attack ("begins the task"). The design has to ensure that the response force time is less than adversary task time for the success of the physical protection system.

The process of physical protection system design is iterative and continues until the requirements on performance measures are met. Figure 15.6 captures this iterative nature of the physical protection system design process.

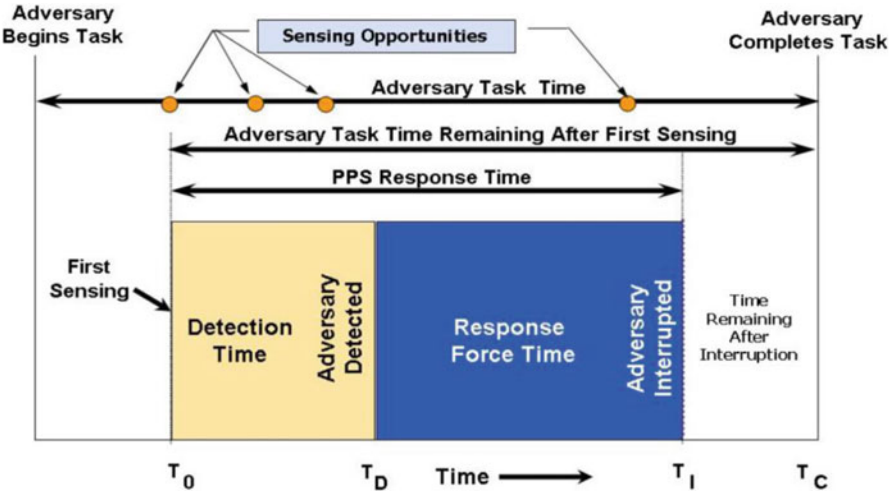


Fig. 15.7 Consideration of physical protection system design with respect to adversary task time (where T_0 is First alarm occurs, T_D is Time at which alarm is assessed to be valid, T_I is Time at which Response Force interrupts adversary actions, and T_C is Adversary task completion time). (Source: SNL 2018)

15.4 Role of Policy in National Choices on Reprocessing

15.4.1 Comparison of National Policies on Reprocessing

Reprocessing has been a subject of policy debate in many countries and in the international community. Reprocessing requires breaking of fuel cladding, an important physical barrier for nuclear safety and increases the possibility of accidents due to increased handling of spent fuel. It also opens the possibility of misusing special nuclear materials by producing and allowing access to them. Unless the reactor-based recycling system or the necessary integrated waste management system is ready, reprocessing adds to the burden of managing the stockpile of plutonium from which nuclear explosives could be fabricated. Any country interested in reprocessing should consider nuclear nonproliferation and security implications of the technology.

The current status of different nuclear power countries' choice on reprocessing is summarized in Table 15.11. Most countries' nuclear power program is based on direct disposal of spent fuel (i.e., under the once-through fuel cycle). Out of the 31 countries listed in Table 15.11, seven countries uses reprocessing as part of national nuclear fuel cycle. These countries include France, Japan, Russia, Ukraine, U.K., India, Bulgaria, and Netherlands. Two countries, Germany and Belgium, transitioned from reprocessing-based fuel cycle to the once-through cycle. These two countries also have the plan for nuclear phase out. Then there are a total of seven countries with a plan or interest in pursuing reprocessing. These include the U.S., the

Table 15.11 Current nuclear power countries and their nuclear fuel cycle policy

Nation	NPP units	# under construction	Capacity (MWe) in 2014	Policy for fuel cycle and spent fuel
U.S.	100	5	99,081	Direct disposal but reprocessing prospect revived
France	58	1	63,130	Reprocessing
Japan	48	2	42,388	Reprocessing
Russia	33	10	23,643	Reprocessing
ROK	23	4	20,721	Direct disposal, interested in reprocessing
China	21	28	17,978	Developing reprocessing
Canada	19	0	13,538	Direct disposal
Ukraine	15	2	13,107	Reprocessing
Germany	9	0	12,068	Changed from reprocessing to direct disposal
Sweden	10	0	9474	Direct disposal
U.K.	16	0	9243	Reprocessing
Spain	7	0	7121	Direct disposal
Belgium	7	0	5927	Reprocessing (stopped)
India	21	6	5308	Reprocessing
Taiwan	6	2	5032	Direct disposal, interest in reprocessing
Czech Republic	6	0	3884	Open policy, NPP operators to decide
Switzerland	5	0	3308	No clear policy
Finland	4	1	2752	Direct disposal
Bulgaria	2	1	1906	Reprocessing and direct disposal
Hungary	4	0	1889	Direct disposal
Brazil	2	1	1884	Interim storage, interest in reprocessing
South Africa	2	0	1860	Interim storage, interest in reprocessing
Slovakia	4	2	1815	Direct disposal
Argentina	3	0	1627	Interim storage, interest in reprocessing
Mexico	2	0	1570	Interim storage, further decision pending
Romania	2	2	1300	Direct disposal
Iran	1	2	915	Spent fuel return to Russia
Pakistan	3	4	690	Interim storage, interest in reprocessing
Slovenia	1	0	688	Direct disposal (possibly abroad)
Netherlands	1	0	482	(overseas) reprocessing, policy change decision pending
Armenia	1	0	375	Interim storage, further decision pending
World	**437**	**72**	**374,704**	

ROK, Taiwan, China, Brazil, Argentina, Pakistan, and South Africa. China is making rapid progress to use reprocessing for its spent fuel management with the construction of a commercial scale facility. In several other countries, such as Czech Republic, Switzerland, Mexico, and Armenia, the decision for the fuel cycle is pending while relying on interim storage of spent fuel. Due to declining economic benefits of and political support for reprocessing, the number of countries planning spent fuel reprocessing is expected to decrease.

While the countries with large nuclear power program tend to opt for or remain interested in reprocessing in general, Canada is an exception. In Canada, use of natural uranium as fuel for CANDU reactors presents little incentive for spent fuel recycling. This is due to the fact that very small amount of fissile uranium remain in spent fuel.

15.4.2 Determinants for Spent Fuel Reprocessing Policy

Based on a comparison of national policies of 32 countries (including Italy where nuclear phase out is complete), a study identified factors affecting a country's choice on spent fuel recycling (Högselius 2009). These factors were military ambitions, technological culture, political culture, geological conditions, and energy policy. How each of these factors affects a nation's spent fuel policy is explained below.

Military ambitions: The countries with current reprocessing capability or the record of having pursued spent fuel reprocessing largely coincide with the nuclear weapons states or the states with nuclear weapons ambition. In several cases, the plan for reprocessing was cancelled when the country's nuclear weapons ambitions were given up (e.g., Sweden, South Korea, and Taiwan). Gaining access to reprocessing technology may still remain attractive for a country with interest in nuclear weapons capability.

Technological culture: Technological culture may be defined as the culture dominated by technological optimism. The post WWII industrial developments in the world can be characterized as being dominated by technological culture. Many countries in the 1960s with an interest in nuclear power had an ambition of mastering all steps of the nuclear fuel cycle. Strong technological culture surrounding nuclear power development may push the country toward reprocessing development as a way to achieve domestic self-reliance in nuclear technology.

Political culture: The type of the political regime of a country (i.e., democratic vs. autocratic) has a strong effect on their choices of spent fuel strategies. It is easier for countries with authoritarian and semi-authoritarian regime, such as Russia and North Korea, to sustain the national program for reprocessing. Also, if strong anti-nuclear movements are absent in a country, the lobbying efforts by the nuclear industry have in general been successful in favor of spent fuel recycling. In contrast, if anti-nuclear movements are strong in a country, the country is less likely to support reprocessing due to the perceived political difficulty or high social cost.

Usually, reprocessing program is often under heavy public criticism in a democratic country.

Geological conditions: Finding the site for geological repository and making the necessary developments is a daunting task. Part of difficulty with geological disposal of spent fuel comes from the difficulty in finding suitable geological formations within the country. Developing geological repository also demands for necessary scientific and technological experiences adding to the difficulty. Countries without suitable geological conditions thus lacking the necessary experiences with them may have interests in seeking alternatives to direct disposal, i.e., reprocessing. For instance, unstable geology combined with a very high population density discouraged Japan to pursue direct disposal of spent fuel. A country like South Korea with limited choice of suitable geological formations or strong public opposition against siting a geologic repository also has interest in reprocessing to alleviate the burden of geological disposal. Reprocessing still demands the option of geological disposal but with less space needs.

Energy policy: Energy policy of a country is driven mostly by the concern over long-term energy security. Nuclear power appears attractive, in that regard, especially for those countries lacking natural energy resources. Reprocessing can also be part of the country's strategy to better conserve nuclear fuel for energy security. The countries with active fast breeder reactor programs such as India, Russia, and China keep reprocessing as an essential part of nuclear fuel cycle.

The Fukushima accident in 2001 made a huge impact on a number of countries' energy policy. Some countries such as Germany, Switzerland, and Italy, decided to phase out nuclear power after the accident (Germany and Belgium had a nuclear phase-out policy before the accident and solidified the plan after the accident). A number of nuclear aspiring countries cancel the plan for nuclear power development. These include Peru, Venezuela, Uruguay, Dominican Republic, Haiti in South/Central America, Thailand, Singapore, Myanmar in East Asia, Israel in the Middle East, and Senegal, Tanzania, Tunisia, and Libya in Africa. At the same time, a large number of countries are also continuing their nuclear power program. The current users with continuing development include three in Northeast Asia (China, Japan, South Korea), five in North/South/Central America (the US, Canada, Brazil, Argentina, Mexico), five in Western Europe (France, the U.K., Spain, Finland, Netherlands), five in Eastern Europe (Armenia, Bulgaria, Czech Republic, Hungary, Slovakia, Romania, Russia, Slovenia, Ukraine), two in Central and South Asia (India, Pakistan), one in the Middle East (Iran), and one in Africa (South Africa). In addition, several countries are becoming or have plan to become a new nuclear power country such as United Arab Emirates, Kingdom of Saudi Arabia, and Poland. There are also about 30 countries who aspire to have nuclear power with plans for its development.

It is not clear whether the continuing demand for nuclear power from both the existing nuclear power countries and aspiring states will increase the demand for spent fuel recycling. Given the high cost of reprocessing, most newcomer countries are expected to pursue the option of direct disposal. A small number of newcomer countries may be interested in reprocessing. However, the associated difficulty under the current international nuclear governance regime may dissuade these countries from pursuing reprocessing. Therefore, the demand for reprocessing is not expected to grow in the future.

15.4.3 Multilateral Approaches to Spent Fuel Reprocessing

From the international perspectives, one of the key issues in spent fuel reprocessing is the concern over misuse of technology. The international community has been discussing the establishment of multilateral infrastructure for reprocessing, given the needs for spent fuel reprocessing under the specific national situations. Such approach is based on a scenario of several countries working together to build and operate a reprocessing facility possibly along with a MOX fuel fabrication plant. This approach has the benefit of enhanced nuclear security with built-in transparency, thus reducing the risk of nuclear proliferation. New technologies such as UREX or pyroprocessing could be utilized as part of the approach to enhance proliferation resistance in the system. In the long run, such arrangements could help to support sustainable use of nuclear power when the number of countries interested in nuclear power continue to grow. Nevertheless, fundamental challenges in multilateral spent fuel facility development remain. With highly politicized risk of spent fuel, any country to become the host of an international spent fuel facility will face major challenges. Although the arrangement provides business opportunities and boost in national capability building, oppositions are expected in the host country from local government, local communities, environmental groups, as well as from neighboring countries (e.g., due to the international transit issue with the necessary transportation of spent fuel). In the end, such arrangement requires agreements on assurance of continued service, securing transportation of nuclear materials, specifying the roles of the participating governments, coordination with national legislations of the participating countries, and the benefits to the host community and host country.

15.5 Conclusions

Countries with large nuclear power programs tend to favorably consider spent fuel recycling as part of nuclear fuel cycle. While such consideration can be well supported by the technologies available today, the decision on national fuel cycle requires complex balancing of pros and cons of the choices. This balancing will depend on economics, domestic technological capabilities, nuclear waste management needs, national energy policy, domestic politics, and international nonproliferation and security considerations. The issue of economics is typically the key predicament in the pursuit of reprocessing. At the same time, country's strategic calculus in favor of long-term energy security could override the cost argument. Ultimately, international nuclear governance may dominate the decision process given the dual-use nature of spent fuel reprocessing technology. In this regard, development of a multinational facility for spent fuel recycling deserves serious considerations by the international community.

Homework

Problem 15.1: Using the uranium mass balance given for the once-through fuel cycle of Example 6.6, determine the annual fuel cycle cost of operating one 1000 MWe nuclear reactor: 1) by ignoring time value of money, and 2) by considering time value of money. Use the following data and the discrete discount method for the calculation.

Discount rate: 3%, 5%, 7%

Lead time: 24 months for uranium purchase, 18 months for conversion, 12 months for enrichment, 6 months for fabrication.

Problem 15.2: Based on the problem given in Example 15.1 and by using the fuel cycle model provided, determine how the following variations affect uranium utilization efficiency and fuel cycle cost of 1) the once-through cycle and 2) the close cycle.

(a) Product fuel enrichment (at 3%, 4%, and 5%)
(b) Capacity factor of the reactor (60%, 70%, 80%, 90%, and 95%)
(c) Separation efficiency of elements (at 90%, 99%, 99.9%, and 99.99%)

Problem 15.3: Define proliferation resistance and describe the relationship between proliferation resistance and a country's decision to pursue nuclear weapons.

Problem 15.4: Describe the relationships between civilian nuclear power program and nuclear proliferation in a country.

Problem 15.5: Describe how material balance areas (MBAs) can be set up for a small research nuclear reactor.

Problem 15.6 (Goergen 2010)**:** Four containers containing solid plutonium with combined mass of 1 kg are received by a facility. The uncertainty on the material is ±5% (@ 2 sigma). So the quantity of material could fall between 950 gm and 1050gm (with a 95% Confidence Limit).

The material in the containers was dissolved and put into a tank. The processed solution in the tank was measured for the volume and concentration. That is, after dissolution a level reading of the tank is obtained and a volume determined from a calibration chart and a small sample is obtained and analyzed for the concentration. The results were: 92.5 liter of volume and 10.0 g/L of plutonium concentration.

Questions:

(a) How much material is found after dissolution?
(b) Is the difference between what was expected and what was found within measurement uncertainty, or has there been a potential theft/diversion of material?

Assume that the volume determination has an uncertainty of ±4% (@2 sigma) and the uncertainty in the analytical technique for concentration measurement is ±1.3% (@ 2 sigma).

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Chapter 16
Social Aspects of Nuclear Waste Management

Abstract The biggest challenges to successful nuclear waste management are societal. Although technologies for nuclear waste management have significantly advanced, there is a large gap between the scientific community's achievements and the public's perception toward nuclear waste. This chapter describes how public perception or attitude is formed through human cognitive processes under the influence of society and culture. Challenges to risk communication efforts are also discussed highlighting the differences among the stakeholders of nuclear waste management.

Keywords Basic human needs · Risk perception · Human information processing · Heuristics and biases · Risk communication challenges

If people say that the course of nuclear-power development has been contentious, people would also agree that the course of nuclear waste management has been even more contentious. The biggest source of contention in nuclear waste management is in the social arena. Ranging from gaining legitimacy of the problem to obtaining acceptance on decisions, social issues dominate the dialogues surrounding nuclear waste. One of the key issues among them is public's risk perception and attitude. These issues along with the examination of social, cultural and political influences are examined in this chapter.

16.1 Social Aspect of Risk of Nuclear Waste

With ever-increasing use of technologies in modern society, risk from technologies and the resulting environmental impacts are part of today's living. These technological and environmental risks affect all citizens due to their pervasive nature in human surroundings.

Technological and environmental risks can be grouped in four broad categories (Lewis 1992): (1) The common, familiar high risks, (2) the low probability, high

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consequence risks, (3) the remote, unexperienced risks with very high consequences, and (4) Known risks that are increased slightly by technology.

The first category of risk, the common familiar high risks, is something people accept as part of their life. People are familiar with this type of risks and the information about their consequences is widely available. An example is the risk from driving motor vehicles. Large-scale societal efforts are made to deal with the risk of automobile accidents. These efforts include developing laws, issuing government regulations and license procedures, etc.

The second category, the low probability high consequence risks, is something people thinks real and consider seriously, but there is no sense of urgency. The probabilities of events occurring and their consequences are uncertain. People tend to do little about the risk. An example is the risk from a large earthquake or dam failure.

The remote, unexperienced risks with very high consequences, the third category, are about an event or phenomenon that never happened but with lingering possibility of occurrence. An example is the risk from a nuclear war or global warming. If such events were to occur, the consequences would be devastating. Depending on the policy choices taken, making major efforts to cope with this type of risk is possible. However, the policy to prevent or minimize this type of risk could be unpopular. The policy decision would depend on the stance on whether precautionary approaches are to be taken to handle the risk.

In the case of the fourth category, the known risks that are increased slightly by technology, the probabilities and consequences of risk events are not well characterized. While people may be familiar with the effects, the damages show up slowly later in time. Therefore, the damages may be masked by naturally occurring hazards. Thus quantifying the risk is difficult. Examples in this category include the health risk from natural or commercial chemicals or low-level ionizing radiation. This risk category is often associated with fear among the public. The government and the public often have very different views about the risk or how to handle the risk. Fear among the public may drive policy regardless of the reality or experts' opinions.

Risk of nuclear waste is an example of the fourth category of risk. While government must deal with it, very different approaches can be taken depending on who or what drives the policy. In this regard, it is essential to understand how social issues and public thinking affect the decisions regarding nuclear waste.

16.2 Psychological Aspects of Risk

Humans have basic innate needs. According to Abraham Maslow, basic human needs are classified in five hierarchical categories such as physiological needs, safety needs, need for belongingness and love, need for esteem, and need for self-actualization. These needs are in hierarchy: The lower level needs are most urgent and must be satisfied in order for an individual to be motivated toward higher level needs.

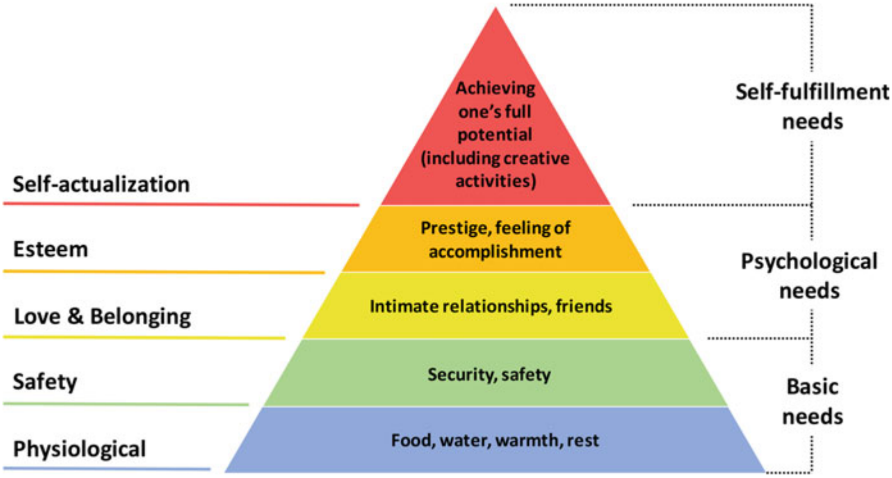

Fig. 16.1 Maslow's hierarchy of human needs

As the most primitive one, physiological needs are the lowest level need. Safety needs, the second most primitive one, cover not only physical safety but also a feeling of being safe and secure from physical and emotional harm. These two most basic needs are followed by the need for feeling loved and accepted and the esteem needs in sequence. Eventually humans strive for feeling that we are living up to our potential as the highest-level need (Fig. 16.1).

Maslow wrote (Maslow 1971):

> All the basic needs which have been fully gratified tend to be forgotten by the individual and to disappear from consciousness. Gratified basic needs just simply cease to exist in a certain sense, at least in consciousness. Therefore, what the person is craving, wanting, and wishing for tends to be that which is just ahead of him in the motivational hierarchy.

Once a need is satisfied, it ceases to act as a motivator. However, if satisfaction of the needs is threatened, they can again be activated. Maslow's concept is particularly pertinent to the study of risk that is perceived to threaten a person's safety. For example, if the person were destitute in daily financial needs through unemployment or homelessness or worry about crimes in his neighborhood, technological risks with potential threat to the person's safety would take low priority. However, once the person becomes well off with no concern over personal safety, then perception of technological or environmental risk can activate his/her need for safety. The resulting motivation for safety can dominate that person's behavior.

16.3 The Concept of Risk Perception

A survey study sponsored by U.S. DOE (Flynn et al. 1990) examined the U.S. public's attitude toward a nuclear waste repository. The survey participants were residents of Southern California, Nevada, Phoenix, or the general U.S. nationals. They were asked to indicate the first thoughts or images coming to mind when they think about an underground nuclear waste repository. The dominant answers were "dangerous/toxic", "death/sickness", "environmental damage", "bad/negative", "scary", "unnecessary/opposed", "leakage", and "not near me". This indicates the presence of extremely negative images of nuclear waste repository among the public. As the public perception of nuclear waste repository is closely tied with negative emotions, we can expect that they would perceive the risk of nuclear waste very high.

Risk perception is the subjective judgement made by people about a risk based on its perceived characteristics and severity. As people are very sensitive to anything that threatens their safety, any event or an object that triggers personal safety needs will have high-perceived risk. This indicates that risk perception is strongly affected by psychological factors.

Studies have shown that there are many factors or qualitative characteristics associated with the risk source that are known to affect risk perception. These factors and characteristics can cause the perceived risk to be higher or lower regardless of the actual level of hazard involved. These factors or characteristics in relation to a person's perceived risk can be explained as follows.

- Familiarity: Depending on how familiar the person is with the object, the risk perceived about the object vary. Less familiarity leads to higher perceived risk.
- Understanding: Depending on how well the mechanisms or processes of harm from the risk source are understood, the perceived risk would be different. Lack of understanding leads to higher perceived risk.
- Uncertainty: Depending on the degree of uncertainty involved with the harm from the risk source, the perceived risk would be different. Higher uncertainty usually makes people believe that the risk in question is greater than what the risk source actually represent.
- Controllability (by self or trusted expert): Depending on the perceived controllability of the harm associated with the risk source, the perceived risk would be different. If the person think the mechanisms or processes related to the harm from the risk source is not controllable, the perceived risk will be higher.
- Voluntariness of exposure: If the exposure to the risk source occurs involuntarily, the risk is perceived to be higher.
- Dread: If there is dread/fear associated with the harm associated with the risk source, the perceived risk is higher.
- Trust in institutions: Depending on the level of trust a person has in the institution responsible for managing the risk source, the perceived risk would be different. If the person trusts the institution, the perceived risk could be lower.

- Effects manifestation: If the adverse health effects associated with the risk source are not immediate but delayed or lingering for long-time, the perceived risk may be higher.
- Reversibility: If the harmful effects associated with the risk source are irreversible, the perceived risk would be higher.
- Equity: Depending on the level of equity in the distribution of risk, the perceived risk would vary. Less equitable risk carries higher perceived risk.
- Origin: Depending on whether the risk event is caused by human actions or natural causes, the perceived risk could be different. Human induced risk carries higher perceived risk.
- Media attention: Depending on the level of attention given by the media to the risk source, the perceived risk varies. High level media attention causes the perceived risk to be higher.
- Accident history: If there were major or even minor accidents publicized in the management of the risk source, the perceive risk would be higher.
- Benefits: If there are clear benefits with the use of risk source, the risk is perceived to be lower.
- Personal stake: Depending on the level of personal stake in the risk source, the perceived risk could be different. If the person has the experience of being negatively affected by the risk source, the perceived risk would be higher.
- Catastrophic potential: If the harmful effects from the risk source are perceived catastrophic, the perceived risk would be higher.
- Effects on children or future generations: If children or future generations are specifically at risk by the risk source, the risk is perceived to be higher.
- Victim identity: Depending on how identifiable are the victim, the perceived risk could be different. If there are no clearly identifiable victims, the perceived risk could be slightly higher.
- Physical extent of damage: If the physical area affected by the harm caused by the risk source is large, the perceived risk would be higher.
- Social extent of damage: Depending on the number of people involved with the harm from the risk source, the perceived risk would be different. If larger number of people are involved, the perceived risk would be higher.

According to these observations, the main features that increase perceived risk can be summarized as:

- The technology is unfamiliar;
- The mechanisms and processes involved with the risk are not understood;
- The person thinks the process associated with the risk source is uncontrollable;
- The exposure is involuntary;
- Manifestation of the effects are delayed;
- Future generations are adversely affected;
- Effects are dreaded;
- There is lack of trust in the responsible institutions, and;
- The harmful effects are irreversible.

These observations indicate that risk perception is multidimensional. This means many factors are involved in forming a person's risk perception. This "multi-dimensionality" of risk complicates the dealings with the risk source or the related risk management activities. This "multi-dimensionality" of risk explains why single-dimensional technical approach to risk communication is not effective in the dialogues with the public.

The risk of nuclear waste is clearly multidimensional. It is unfamiliar, hard to understand, uncertain, beyond their capability to control, appears to be dangerous to their children, and is usually imposed even when unwanted. Arousal of fear is expected as the public first thinks of dreadful hazard of radiation when they consider nuclear waste. Most individuals view radiation as qualitatively different from other health risk sources. People even associate nuclear waste with radiation sickness, physical deformities, and genetic mutations due to the widely circulated misinformation in the social media networks. In addition, the public often have low level trust toward their government or the industry who are responsible for managing nuclear waste. A very negative perception of risk is expected among the public about nuclear waste.

Further research on the factors or dimensions involved with risk perception found ways to better characterize the multidimensionality of risk by using two key characteristics. These two characteristics are "dread" and "unknown". "Dread" covers fear, catastrophic potential, fatal consequences, lack of control, and inequitable distribution of risk and benefits. "Unknown" covers the feature of risk being unknown or uncertain, new, and delayed in effect manifestation. The unknownness of risk may cause the person to take overly protective approaches against the risk source. This "unknown" nature of the risk from nuclear waste may drive its perceived risk to be higher than that of nuclear power or even nuclear weapons.

16.4 Human Cognition Toward Risk Attitude

While risk perception is affected by basic human needs, empirical studies also reveal additional features of risk perception regarding its cognitive aspects. For example, a study by Fischhoff showed the following features of human risk perception (Fischhoff 1985):

> People simplify. Once people's minds are made up, it is difficult to change them. People remember what they see. People can't readily detect omissions in the evidence they receive. People disagree more about what risk is than about its magnitude. People have difficulty in detecting inconsistencies in disputes about risk. People find it hard to evaluate expertise.

This statement indicates the possibility of flaws in our cognition regarding risk perception. As human cognition not only affects perception or risk but also attitude, our risk perception or attitude can be formed on a flawed ground.

Attitude is a mental position of a person with regard to a fact, object, or state. Perception of an object affects the person's own view about the object, which becomes an attitude. A person's beliefs and feelings about an object also become the basis of attitude formation. These also indicate that depending upon how the process of cognition takes place, risk perception or attitude could be manipulated.

16.4.1 Human Information Processing

Human's cognitive information processing directly controls risk perception and attitude formation. As today's individuals are exposed to a large amount of information, information received is often ignored. Only when a person pays attention, the information pass the so-called attention filter. Whether a particular piece of information passes the attention filter or not depends on the receiver's interests or the nature of the information. Novelty of the information, specific symbolic keywords contained (e.g., mentioning of prestigious persons or institutions), or signals that are related to the receiver's personal motivation tend to support the initial selection of information. Through this attention filtering, the receiver becomes "aware" of the information. Then the receiver selects the relevant parts of the information and will interpret or analyze the meaning of the information. He or she will also compare the interpretation with other messages from other sources or previous experiences or existing beliefs. He or she then evaluates the potential effect on personal life or the potential for personal involvement depending on the perceived accuracy of the information. From these, the person forms specific beliefs about the subject of the message. The result could be simply reassuring previously held beliefs or having question about it. The person may form a new perception or belief about the subject of the message if the person didn't have any preexisting one or had very weak ground for the previously held one. The receiver may generate intentions for future actions in accordance with the beliefs.

For mental processing of the information, individuals are likely to evaluate whether it is necessary to study the content of the information in detail or to make a fast judgment according to some salient cues contained in the message received. Selection between the two choices is referred to as taking two different information processing routes, i.e., the central route of information processing or the peripheral route of information processing (Petty and Cacioppo 1986).

This selection depends upon the *ability* and *motivation* of the receiver. If the receiver has the relevant ability or motivation to examine the information, the receiver is expected to pay attention to the details of the information following all of the steps listed above. This is called information processing through the central route. The credibility of each argument in the message is examined and tested by referring to previous experiences, plausibility, and perceived motives of the communicator.

If the receiver does not have the relevant ability or personal motivation, the individual may not bother with the details of the message. In this case, the receiver looks for easily accessible cues in the information to make fast judgment on the whole package. This type of evaluation is called information processing through the peripheral route. In this case, there is no serious deliberation on the information. The cues used for fast judgment can be about the source, message, transmitter, or the context of the message. Regarding the source of the information, reputation, credibility, or social attractiveness of the message source are examples of the related cues. In terms of the message itself, proximity and affinity of the information to one's own interests are an example of such cues. Presence of symbolic signals that trigger immediate emotional responses is also an example. The transmitter related cues are the perceived neutrality or social credibility and the personal experiences with the transmitter (e.g., newspaper or TV) in the past. Personal alignment with the political or ideological position of the transmitter is another related cue. The context-related cues include presence of competing messages, controversy among experts, or general social consensus about of the issue.

In the cases of risk perception of nuclear waste, people normally use the peripheral route for information processing. This is because the public in general have low level motivation to examine the issue or lack the ability to examine the details of the information. Presence of environmental controversies, disagreement among experts, negative stereotypes, and widely available negative information from the social media may provide enough cues for the public to form perception or opinion about the risk of nuclear waste. It should also be noted that the perceptions or attitudes formed through peripheral processing of information are subject to change.

16.4.2 Influence of Heuristics and Biases

The term "heuristics" refers to simple and efficient rules used by people to form judgments. When information is presented to a receiver, the individual uses heuristics as mental shortcuts to focus on one aspect of an object for judgment, ignoring other aspects of the object. This occurs because using heuristics helps making reasonable judgment without requiring much mental work. Use of heuristics can occur in both central and peripheral routes of information processing and can lead people to be in error.

Heuristics are particularly important in the discussion of risk perception as they can become a source of bias or misconception in comprehending the presented information. Major types of heuristics include availability, representativeness, and anchoring. These heuristics present biases in the inferencing process of risk perception.

The availability heuristic is used when people estimate how likely or how frequent an event is. Availability refers to the ease with which a particular concept can be brought to mind. An infrequent event but with dramatic and vivid image can be brought easily to a person thus is mentally available. Frequent, mundane events with no dramatic memory are hard to bring to mind and less mentally available. According to this heuristic, events or activities coming to mind immediately are more probable than the events and activities that are less mentally available. For example, most people can easily recall one or two serious nuclear reactor accidents

(Three Mile Islands accident, Chernobyl accident, or Fukushima accidents), but they may not recall any dam failures. Therefore, they would judge that nuclear accidents are more probable and riskier than dam failures although, in reality, many more fatalities have been caused by dam failures than by nuclear accidents. With the availability heuristic, people may perceive accident as typical with nuclear energy.

Representativeness as heuristic is to use categories. People makes quick judgments on the objects according to the category to which the objects seem to belong. For example, people or technologies can be categorized as good or bad based on personal experiences or social reputation. In the case of nuclear technology, some people perceive it as something bad or even evil in association with nuclear bomb and/or radiation sickness. The waste from nuclear technology as a source of radiation would easily be perceived as something very bad or dangerous.

Anchoring effects refer to a tendency to estimate probability of an event on the basis of the information presented for other events: In estimating the probability of an event, people tend to use the readily available number as the “anchor” and adjust it to reach an answer. This heuristic occurs when people lack background knowledge or relevant prior experience. For example, if a person were told that 530 people on average die annually from dam failures and then is asked to estimate how many people die from nuclear reactor accident per year, the answer would be a comparably small number. However, if the person were first told that more than 780,000 people die prematurely in the world per year as a result of coal power plant related air pollution, their answer about the nuclear related death would become comparably large. In reality, the total recorded number of death due to civilian nuclear reactor accidents in the world is 42 so far. This includes 28 short-term death, 11 latent death, and 3 non-radiation death over the last 62 years (UNSCEAR 2013). When people are faced with a question with inadequate knowledge, their answer tends to anchor on a specifically mentioned fact even though the information has no relevance to the subject matter under consideration.

Anchoring effect also affects risk judgement through political orientation. A study on the U.K. public’s energy choice (Costa-Font et al. 2008) showed that the people with left wing political views tend to oppose nuclear energy while the people with right wing political views tend to support nuclear energy. This result implies that people with inadequate knowledge about the risk of nuclear energy employ the heuristics of political anchors. They rely on their political affiliation to decide on their attitudes towards nuclear energy. Therefore, opposition to nuclear power is likely to depend upon individuals’ political stance. People’s worldviews or ideology also works as anchors in attitude formation. This is further explained in the next subsection.

Avoidance of cognitive dissonance is another heuristic employed for simple judgement. It refers to people’s tendency of downplaying new information when their preexisting beliefs are challenged. This occurs as a way to avoid a psychologically uncomfortable state of arousal after being faced with new information. Accordingly, a person with negative attitudes towards an event or activity will tend to prefer to seek information that reinforces the initial position and reject the information challenging the already formed opinion. Therefore, people with negative attitude toward nuclear power would be reluctant to accept new information with positive views on nuclear power. This indicates the difficulty in changing a person's attitude once an attitude is formed.

There are also other psychological biases that cause error in cognitive information processing. These include the overconfidence bias, the negativity bias, the compression bias, and false consensus.

Overconfidence means that a person's subjective confidence in his or her judgements is greater than the objective accuracy of those judgements. Such overconfidence keeps people from realizing flaw in their thinking.

The negativity bias means that humans are more sensitive to negative/bad news. Thus, things of a more negative nature (e.g. fear, unpleasant thoughts/emotions, and harmful/traumatic events) have a greater effect on one's psychological state than the neutral or positive ones. According to this bias, a negatively framed information will carry added weight and have larger impact on the processing of the information. Risks are perceived to be higher if the related activities of an object are framed negatively. The related effects of framing are found important in risk perception and attitude of the public. Framing refers to the context into which information is placed or the way how information is presented. For example, if a treatment of disease with a new drug provides 40 percent chance of survival, there is 60 percent chance that the patient would not survive. Depending on whether the information is framed negatively or positively, acceptance of the treatment by the patient may be different. The same information but framed differently is found to have different reaction to the information from people.

The compression bias refers to tendency to overestimate small frequency risks and underestimate large frequency risks. According to this bias, risks characterized by low probabilities of occurrence would be perceived as more threatening than the actual level. A related observation is people's prejudice that a very low risk estimate carries less credibility and appears highly uncertain.

False consensus is an egocentric bias. People tend to believe their opinions are much more common than they are in reality. Therefore, they would overestimate the extent to which their opinion is shared by the rest of the population. If a person thinks the public is against nuclear power, he/she may overestimate the proportion of others who actually oppose nuclear power.

How a person perceives the risk is also affected by the perceived vulnerability of the person to the risk source. The judgment on vulnerability will depend on the level of trust he or she has in the person or party responsible for the management of the risk source. Therefore, trust emerges as an important factor in relation to the role of heuristics and biases. Trust is a person's confidence in another person or party's future actions to be beneficial, favorable, or at least not detrimental to himself/herself. At the same time, when people recognize that there are limits to how much the trusted party knows or can handle about the risk, the role of trust in risk perception is limited.

16.4.3 Influence of Worldviews, Interpersonal Relations and Ethics as Cultural Biases

16.4.3.1 Influence of Worldview

As social beings, humans are influenced by society and its culture. At the same time, the way a person interacts and relates to society and culture depends on his/her worldview.

A worldview is the fundamental cognitive orientation of an individual. People see things through worldview. Worldview controls a person's point of view and his/her political, ideological positions. Worldview is also closely related to individual's beliefs and values. Worldview affects the perceived credibility of the information received, independent of the information itself, depending on who the source or the transmitter is. Therefore, worldview affects people's perception of different types of risks.

Consider a worldview about social culture. It is the belief system a person holds about social culture and experiences. Such cultural worldview can be broken down into three groups, guilt-innocence, honor-shame, and power-fear (Muller 2000). Every individual is expected to hold a part or a mix of these worldviews and how a person perceive risk will be affected by them. If a person holds a worldview derived from a guilt–innocence focused culture, the person is likely to see issues often as black and white. The person is likely to rely on the representativeness heuristic in perceiving risk. Under the influence of an honor-shame worldview, a person's risk attitude would be more relationship-driven. In this case, political anchoring or trust would be important. If a person has a power–fear worldview, role of trust is expected to play a prominent role.

16.4.3.2 Influence of Worldview Through Interpersonal Relations

Another way of classifying different worldviews is based on patterns in interpersonal relations. As part of personal way of life, people with different interpersonal relations patterns may choose what to fear (and how much to fear it) in different ways. These groups include fatalists, hierarchists, egalitarians, individualists, and hermits. Differences are expected in the way a person in each category perceive risk. This categorization may help to predict people's behaviors and reactions in dealings with technological risk.

The categorization of people into five groups is in association with two characteristic dimensions: *group* and *grid*. The *group* dimension is the extent to which an individual is involved in social groups and represents how much a person is "individualized" or "collectivized". The *grid* dimension is the extent to which an individual is involved in hierarchical arrangements and covers the spectrum between "egalitarian" and "hierarchical". The combination of these two dimensions can illustrate the nature of the people in different categories.

Fatalists are the strongly negative *group* (actively resist social involvement) and the strongly positive *grid* (willingly accept involvement in hierarchical arrangements) category people. They tend to avoid and ignore information about risks because this enables them not to worry over things they believe they can do nothing about. For them there are no such options as “accept” or “reject” risk. They tend to consider risk as a “fact of life”.

Hierarchists represent the strongly positive people in both *group* and *grid.* They are active in social involvement and willing to accept involvement in hierarchical arrangements. They emphasize obedience to authority and believe in the role of technology to improve the quality of life. They trust in experts and expect them to make right decisions about risk assessment and management. For them to accept a risk, the uncertainty must be lower. If risks cannot be avoided completely, they must be spread.

Egalitarians are the strongly positive *group* and strongly negative *grid* people. They are active in social involvement and actively resist involvement in hierarchical arrangements. They value equality and perceive the dangers associated with technology very large. They believe that an inegalitarian society tends to insult the environment the same way as it exploits poor people. They blame the system imposing hidden, involuntary dangers on people and show strong aversion to all technological risks.

Individualists are strongly negative in both *group* and *grid*. They actively resist involvement in society and hierarchical arrangements. They are usually pragmatic materialists and prefer self-regulation over any authority. They see technology as a vehicle for unlimited individual enterprise and perceive technological risk small. Therefore, risk is seen as opportunity to them. For individualists, risk is the price to pay for personal progress and improvement.

Hermits are also strongly negative in both *group* and *grid* as an extreme case of individualists. They are different from individualists by being autonomous and more resigned. They distinguish themselves by being disengaged. They deliberately choose to keep their involvement in socially binding relationships to a minimum. Their risk attitude will depend on the expected level of social involvement. If their rejection or avoidance of risk would require social involvement, they would rather accept risk as lesser of the two evils.

Egalitarians are more sensitive to risk issues and ask for a substantive, equal distribution of risk, as they care more about others. They demand all interested and affected persons to have an equal voice. According to their belief, democratic procedures should not violate the interests of minority in the decision processes of risk management. A hierarchist, in contrast, would support ignoring the safety of some minority of people for the sake of dominant majority. A hierarchist employs technical rationality (see 16.5.2) and tends to trust government or industry in the management of risk. An individualist would care more about the personal benefit in their risk attitude. A fatalist tends to be pessimistic about social systems and perceives more risk in them but may not resist technological risk.

In general, people who resists social hierarchy, e.g., egalitarians and individualists, would perceived more risk in technologies than people accepting hierarchy, i.e., fatalists and hierachists.

16.4.3.3 Influence of Ethics

Worldviews are also closely related to ethics. Ethics deals with the issue of how one should live and act and make decisions. Therefore it is an important dimension in every human's experience in life.

As indicated, egalitarians are driven by strong ethical conviction regarding their decisions on risk. Egalitarianism emphasizes treating individuals equally. According to them, risk associated with nuclear waste must be distributed equally not only among individuals but also with respect to social groups, regions, and generations. Those who live near a nuclear waste repository should not bear more risk than those who live farther away from it. Also, members of future generations should not bear more risk than the present generation. In making risk decisions, all persons with a stake should have an equal voice. The procedure must not violate the interests of the minority.

A contrasting ethical belief is utilitarianism. Utilitarianism emphasizes providing the greatest safety/welfare or the least risk for the greatest number of people. By the reasons of utility, efficiency, or expediency, some minority or certain groups of people may be treated inequitably.

From the social justice point of view, the egalitarian view is certainly appealing as it strives for equal justice, equal protection, equal opportunity, and equal access to the decision process. However, implementation of such view often faces unsurmountable challenges as time and resources available to manage risk are not unlimited. Gaining social consensus through political processes to support the egalitarian approach is a hard task.

16.5 Challenges of Risk Communication

When strong negative risk perception exists among the public about technology, a common explanation by the technical community is that the public is uneducated or poorly informed. Against such lack of understanding, the technical community often emphasizes the role of risk communication. In this context, risk communication is referring to educating the public.

In the 1970s and 1980s, governments of several nuclear power countries made major efforts for public education on nuclear technology. Such effort was based on a belief that providing information would increase public's confidence in nuclear power and improve its public acceptance. However, these efforts were found unsuccessful. The so-called risk communication efforts did not work.

In conducting the efforts, risk communication was considered a one-way process: An expert is feeding information to a receiver. The receiver, i.e., the members of the public, should listen, understand and change.

However, it is now recognized that for risk communication to be relevant, it has to be a two-way process. The word "communication" in Latin is communicare, which means "to share" or "to make common". It is like laying a bridge to connect two sides being separated. Communication is a process of understanding each other.

16.5.1 Understanding the Differences Between Experts and the Public

16.5.1.1 Risk Information Gap Between Experts and the Public

The discussions given earlier note a gap in the perceived risk of nuclear waste between the experts and the public. When this gap is not filled with proper scientific information, it does not remain as vacuum but is filled by social learning with information from readily available sources such as news and stories from various social media. The news and the stories from these sources are often alarming and the public frame or solidify their negative views based on them. The information and perspectives given by various social or political groups or individuals promoting their own interests also fill this gap resulting in suspicions, concerns, mistrust, and fear among the pubic.

Such developments may also lead to so-called social amplification of risk. Social amplification of risk refers to social processes that exaggerate (or understate) risk. Information systems such as scientists communicating the risk assessment, the media, and interest groups can intensify (or weaken) risk information. Intensification depends on the volume of information flow, the degree to which individuals or groups dispute factual information, and dramatization involved (Kasperson et al. 1988). Regardless of the accuracy of the information, if large information flow takes place repeatedly in the media with disputed arguments and dramatization of impact, the risk associated with the information will be amplified. The amplified risk leads to behavioral responses such as refusing to accept new information about the risk, taking negative attitude, and developing social network to promote political agenda against the risk source. Therefore, social amplification of risk may be manifested in the form of social phenomena or syndrome (e.g., NIMBY).

16.5.1.2 Differences in the Way of Thinking Between the Experts and the Public

Differences in risk perception between experts and the public not only come from psychological aspects but are also attributed to the differences in the way of thinking.

This is because the notion of risk may be framed differently between scientific experts and the general public.

Those entities who are eager to convince the public on the safety of nuclear waste often operate on the premise that they and their audience share a common framework of thinking. However, there are differences between the experts and the public in the way of thinking, evaluating, and interpreting technological risk information. These differences are linked to one's belief with one's reason to believe, termed rationality: Rationality in thinking within the perspectives of the experts is different from that of the general public. The experts work under so-called technical rationality while the public are under so-called cultural rationality (Plough and Kimsky 1987).

Technical rationality is based on trust in scientific norms. In this approach, use of scientific methods with explanations and evidences are important. A theory or an assumption must be tested through data gathering and analysis. Objective (non-personal) data rather than subjective (experiential) information must be used to provide the basis of analysis and judgment. Boundaries of analysis are narrowly defined and statistical average is emphasized in the analysis. The risk is depersonalized and comparison of risk events are relevant through the use of quantitative measures. Consistency and universality are important in the interpretation and application of the numbers. Impacts that cannot be articulated or anticipated are irrelevant. If any controversy or disagreement arises, resolution is made according to the hierarchy of scientific expertise. Technical rationality is also a way of thinking by the government authority and the scientific community.

Cultural rationality significantly downplays the use of scientific methods. Use of evidences, expertise, and statistical tools is not emphasized. Boundaries of analysis are broad in this approach and unanticipated or unarticulated risks are relevant and deserve investigation. Use of analogy and historical precedent are important and the impacts of risk on the family and community are emphasized. Most importantly, risks are personalized. Consistency or universality is not necessary and particularity is focused. If any controversy or disagreement arises, resolution should be pursued through democratic processes. Scientific expertise should not dictate the process.

Cultural rationality defies the authority of experts in a traditional sense. This cultural way of thinking is based on the notion that an expert and popular approaches to a risk event can each be logical and coherent on their own terms. Any disagreements are believed to be due to differences in how the problem is articulated. It could be argued that cultural rationality uses technical knowledge in a sense relevant within a broader social framework.

Differences in the way of thinking also affect communication through the use of different languages. The language used by experts is full of quantitative terminology and technical jargons. It is based on the concept of probability and focuses on acceptable risk, comparative risk and pays little attention to the lives of individuals. The language used by the public is simple, plain, and intuitively grounded but is not quantitative. It focuses on safety and wants the yes or no answer. Discrete events and personal consequences are important. It matters how a person dies.

Both languages are used in daily communications of risk. One cannot replace the other. Consideration should be made on how to translate scientific findings and probabilistic information into plain terms in qualitative dimensions. Acknowledging the limits of scientific knowledge is also necessary while importance of uncertainty needs to be shared. Building trust and/or credibility of experts by the public would also be an essential part of the efforts to reconcile the differences.

16.5.2 Difficulties with Science

Scientific expertise play an important role in risk communication as their analysis forms the basis of the communication effort. For scientific expertise to be effective in the effort of communication, the followings should be noted.

Scientific analysis for risk decisions is interdisciplinary. It needs contributions from diverse disciplines (i.e., biology, toxicology, environmental science, engineering, etc.). The results are often laden with uncertainties. The uncertainties in risk analysis are not only scientific but also beyond science. In fact, the problem of characterizing the risk of nuclear waste often involves "trans-scientific" issues. Trans-scientific means that question can be asked but is unanswerable by science. Biological effect to humans from very low levels of radiation is an example of trans-scientific issue as the problem demands extrapolation of scientific data into the policy realm. Predicting future state of things for the safety in geologic disposal of waste is another example of trans-scientific issue. Providing safety to the generation in the long-term future is also trans-scientific.

In this case, disagreements among experts are often observed and achieving consensus is difficult. Such disagreements arise from differences in professional training, disciplinary orientation, intellectual style, and experiences. Difficulties with articulation, presence of cognitive biases, and use of different paradigms also contribute to the disagreements. The public may be confused at observing such disagreements among experts. Scientific analysis is also not necessarily value neutral and objective as it can be dictated by the worldview of the person conducting the work. When science loses its aura of objectivity and rationality, the public lose confidence in science.

The limitations in science often leave an important scientific or technical issue to become a public policy choice. In this case, the scientific/technical expertise plays little role as the decision process is driven by the interests of the stakeholders.

16.5.3 Issues with the Role of Media

Technological risks today's society is facing are rarely experienced by the public in person but often learned through the social media or social networks. The social media or networks can construct a universe of their own which does not necessarily reflect the reality. At the same time, as the social media is powerful and well-accepted, information presented by the social media is rarely questioned by the public. When the public is regularly exposed to certain type of social media reporting of technological risks, they will keep the same kind of opinion or bias carried by the media. Therefore, the mass media can shape people's perception of technological risks and often impede or sometimes promote technological innovation processes.

The objectives of the social media, in particular, mass media such as TV and newspapers are different from those of scientific community. The objectives of mass media are to inform or entertain and to make profit. They make profit by reflecting society. This particular nature drives the media to focus on short-term events of major public interest or concern. The content must draw attention by the public. Therefore, the negativity bias and dramatization of stories are often necessary.

Science in dealings with risk has different objectives. Scientific experts are to find the truth by creating understanding through complex scientific analysis. With the created knowledge, they want to educate the public and make a positive difference in the society. While addressing societal concerns, they are mostly interested in long-term perspectives, continuation of research, and improving public wellbeing.

To improve risk communication with the public, the scientific community must be willing to work with the media. Most reporters do not have the scientific background or expertise, or even time to evaluate the complex scientific information. They often lack the ability to discern the disagreements surrounding the risk debates. They work under extremely tight deadlines and tend to rely on sources that are readily accessible and available. Sources that are difficult to contact or reluctant to provide the necessary information are often left out. Truth in journalism can be different from that in science. From the perspectives of journalism, there are different or conflicting views and claims and their goal is to present them with balance.

The mass media must recognize their responsibility in society. They can create artificial crisis and drive the public towards unhealthy policy decisions. The journalists must cover risk stories as fairly as possible by maintaining objectivity. They must work diligently not to distort the facts. Use of sensationalism and appeal for negativity bias must be minimized with a sense of social stewardship. Also the public must be aware that not all media are alike. There are differences in objectives and goals. These differences exist in political culture and orientation between print and electronic, national and international, local and regional organizations, and even among the same categories of media. The public must be aware that the media exists to make profit.

16.6 Conclusions

Sustainable use of nuclear power can only be possible with success in nuclear waste management. Such success can only be realized when the public is adequately informed and their meaningful participation in decision making is allowed. As a pre-requisite for such developments, understanding of how the public perceives risk and forms attitudes is necessary. This chapter described how public perception or attitude is formed through human cognitive processes based on basic human needs and under the influence of the society and culture. Any effort to affect public risk perception or attitude toward nuclear waste must address psychological and social and cultural aspects of the information and its delivery. The supporting risk communication efforts should be more than information delivery but must be based on mutual understanding. A genuine and candid communication effort based on the goal of achieving societal good based on trust will make a difference.

Homework

Problem 16.1: Explain Maslow's basic human needs and describe how they are related to the discussions of nuclear waste management.

Problem 16.2: Explain why the public perceives the risk of nuclear waste very high and negative. Explain this based on your understanding of key factors and characteristics of risk information in affecting human risk perception. List also the key heuristics and biases used by humans in making judgment about information and explain how these heuristics and biases affect public perception of nuclear waste risk.

Problem 16.3: Based on the understanding of human information processing, describe how people may develop their opinion or attitude towards nuclear waste.

Problem 16.4: Explain key differences between technical rationality and cultural rationality.

Problem 16.5: Based on your understanding of the design and characteristics of a high level waste geological repository, *create a public information brochure* to explain the safety of spent fuel disposal. Your brochure should be less than 2 pages in length and be effective in demonstrating the concept of safety of spent fuel disposal. An average adult person with high school education should be able to understand the contents of the brochure. Please be "public friendly" in the organization and delivery of the information.

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Chapter 17
Addressing Key Challenges in Nuclear Waste Management

Abstract Difficulties in nuclear waste management arise from the very long-term nature of the problem, inadequacies of human institutions to handle the problem, and difficulties in deriving social consensus. This chapter explores possible ways to overcome these difficulties and include discussions on building legitimacy, establishing stability in human institutional approaches, creating risk acceptability among the public, and considering alternative ways to addressing long-term safety in nuclear waste disposal.

Keywords Legitimacy of the problem · Stability in institutional measures · Risk acceptability · Long-term safety · Social stewardship

While much efforts and progresses have been made, there are challenges remaining in the management of nuclear waste. Some of these challenges belong to the technical domain but the more formidable ones are in the social arena.

A good majority of the public do not recognize the problem of nuclear waste as a legitimate social problem to solve. They may think it belongs to someone else. When they see problems occur with nuclear waste, they blame the nuclear industry or the government who started it. It is not surprising to witness lack of participation or cooperation of the public in dealings with nuclear waste.

The existing risk perception of nuclear waste among the public is a key barrier in making social decisions regarding nuclear waste. Meaningful interactions of the stakeholders are restricted when the members of the public are driven by fear-associated risk perception. Such perception led to the phenomena such as NIMBY (“Not In My Back Yard”), LULU (“Locally Unwanted Land Uses”), NIMTOO (“Not In My Term Of Office”), NIABY (“Not In Anyone’s Back Yard”) and even BANANA (“Build Absolutely Nothing Anywhere Near Anyone”). Presence of these phenomena hampers efforts in risk communication and social decision making.

While the importance of good public policy is well recognized, developing good policy for nuclear waste management is not easy nor simple. With many stakeholders involved with the policy process driven by different and divergent motivations, achieving consensus or attaining robust outcome is expected to be very

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difficult. Inadequacies of human institutions to maintain long-term commitment (e.g., funding, government support) towards the policy goals are also noted.

One of the inherent challenges in nuclear waste disposal is the need for the demonstration of very long-term safety. Technologies must not only perform successfully for very long time but such long-term performance must be demonstrated through quantitative analysis. The time frame to be covered in the demonstration goes well beyond the time horizon of engineering analysis.

In this chapter, each of these challenges is discussed and potential ways to address them are elaborated.

17.1 Recognizing Nuclear Waste as a Legitimate Problem to Solve

Cooperation of stakeholders is an essential requirement in nuclear waste management. The cooperation among federal government, local governments, the nuclear industry, various interest groups, and the citizens of communities are necessary, starting from initial development of policy until the final disposal stage. The most essential aspect of this cooperation is the general public's participation. The general public drives political process and the related social activities, and ultimately bears their consequences. Nonetheless, unless their own community is affected or involved, the general public is in general disinterested in the issue of nuclear waste. They think that they have done their share by paying the utility bills. The rest belongs to government or the industry. Perhaps they might not even have endorsed nuclear power. They may think that there are alternative ways to generate electricity or to handle nuclear waste. That is something government must figure out.

The current concern over global climate change is changing the perceived need for nuclear energy in a number of countries. Even some environmental activists are supporting nuclear energy as a way to secure low carbon energy. If the world is headed in this direction, the problem of nuclear waste deserves more attention. Without solving the problem of nuclear waste, nuclear power is not sustainable nor legitimate.

Consider the issue of legitimacy. To view nuclear waste as a legitimate social problem, consent is required. In *Two Treatises of Government* (1689), John Locke indicated that legitimacy comes from consent of the governed. The public have to feel that they have a voice in the matter. The authority has to be fair in handling the matter.

For the public to feel that they have a voice, they must be given opportunities to be part of the decisions. For the public's voices and concerns to be heard as part of the decision-making process, they need to be informed of the issues involved and potential consequences of the decisions. The process cannot be justified if they are left out until the last minute. If the public is informed under the atmosphere of mutual understanding, their choice is usually not different from the recommendation by the scientific experts. Such procedural justice-based approach establishes legitimacy of the decision.

Government has to address fairness by being mindful about the distributions of risks and benefits. The people who are at risk due to the presence or handling of nuclear waste versus the people who have benefited from the activities leading to nuclear waste production must be treated fairly.

Consider siting of a geological repository. The procedure for selecting a site must be based on social equity. The potential host communities must see enough merits in taking the burden of being a host. The merits should be directly beneficial to the members of the community. Other citizens who are not affected by the presence of geological repository must share the burden by contributing to the compensation package to the host community. The industry who generated the waste should take the bulk of the burden, not only financially but also morally and culturally. The members of the potential host communities must be afforded a substantive opportunity to influence the decisions based on their own risk and benefit balancing. The residents of the participating communities must feel the sense of pride, instead of feeling like being dumped on the problem because they are losers. They must feel that they are making important contributions to solving an authentic national problem. The agency who is responsible for siting the facility must be credible and regard the citizens of potential host communities with dignity.

As nuclear waste is a long term problem, the issue of intergenerational equity must also be addressed. The current generation is the beneficiary of the energy generated by running nuclear power plants. If the problem of nuclear waste is not properly taken care of now, the current generation is leaving the issue to future generations without their informed consent. Therefore, the current generation's needs are satisfied at the expense of future civilizations. The fact that the current generation enjoyed the benefit of nuclear power makes nuclear waste a legitimate problem to be solved by them.

There is also an issue of alternatives. If nuclear power were not used, the alternatives would be to use coal, natural gas, hydro power, solar power, or wind power. For nuclear waste problem to be legitimate, use of nuclear energy has to be viewed a responsible choice among them.

Human society cannot be sustained without energy. In particular, electricity, the most convenient form of energy, drives virtually every sector of today's society. In particular, with heavy reliance on information technology in virtually all sectors of society, electricity has become an essential commodity of our life and key component of national security infrastructure.

Electric utilities use various energy technologies to meet the time varying energy demand. In particular, electric utilities use a certain portion of electric generating capacity as base load to manage electric grid. Base load is the minimum amount of electric power that an electric utility must maintain all the time, i.e., 24 h a day, 7 days a week. For a typical grid, the base load power covers about 35–40% of the maximum demand in a year. Usually the most cost-effective or dependable plants are used as the baseload units. This is because the base load units must maintain continuous energy generation regardless of weather or any other adverse climate

Table 17.1 CO_2 equivalent emissions from the selected electricity supply resources (gCO_2eq/kWh)

Energy sources	Average	Minimum	Maximum
Coal – PC	820	740	910
Gas – combined cycle	490	410	650
Biomass – cofiring	740	620	890
Biomass – dedicated	230	130	420
Geothermal	38	6.0	79
Hydropower	24	1.0	2200
Nuclear	12	3.7	110
Concentrated solar power	27	8.8	63
Solar PV – rooftop	41	26	60
Solar PV – utility	48	18	180
Wind – onshore	11	7.0	56
Wind – offshore	12	8.0	35

Data source: Schlömer et al. (2014)

situations. Utilities also use the intermediate load capacity and the peaking capacity to effectively manage the variations in electric demand. More expensive and/or less reliable options are used as the intermediate or peaking units. The peaking units should respond fairly quickly to the changes in electrical demand (i.e., for frequency control) to maintain the quality of electricity to be high.

Currently, coal or nuclear power plants are the predominant baseload units. Depending on the local conditions, hydro or geothermal power can also be used as baseload. Both wind and solar power are considered as intermediate due to their intermittent power generations. Natural gas or sometimes oil is often used for peaking. With the continuing trend of low gas price, natural gas is now considered an intermediate or even a base load option.

Extensive use of fossil fuel energy today is potentially causing irreversible damages to the global ecosystems through greenhouse gas-induced climate change. As part of the effort to reduce CO_2 (one of the key greenhouse gases) emissions, use of coal or oil as electric power source is seriously challenged. Even the use of natural gas (which emits about half of CO_2 from coal) is being questioned. With the movements towards low carbon energy systems, energy planners are pushing renewable energy hard. As no greenhouse gases are directly emitted from energy generation, nuclear energy is also seen as a necessary part of future energy mix.

Table 17.1 gives the comparison of CO_2 emissions from various energy technologies. The emission figures in the table are for the lifecycle and include the emissions from the production of materials for plant construction. The average lifecycle emission of CO_2 from coal and natural gas is 820 and 490 g/kWh, respectively, compared to 12 g/kWh from nuclear, 11 ~ 12 g/kWh from wind, and 41 ~ 48 g/kWh from solar power (Schlömer et al. 2014).

According to the U.S. Energy Information Administration, nuclear power accounted for 36% of the voluntary greenhouse gas reductions reported in the

U.S. by the electric power sector in 2005. According to Nuclear Energy Institute, nuclear power plants generate 73% of all carbon-free electricity in the U.S.

Renewable energy is very important for energy system development for the future. In fact, fully utilizing the potentials of renewable energy technologies is essential to sustain human civilization. At the same time, renewable energies have the inherent limitations of low energy density and intermittency. Large scale deployment of solar or wind energy requires coverage of large areas. To make up for intermittency, solar or wind power requires backup power or energy storage. The backup technologies used are often fossil fuel based which defeats the purpose of sustainable development. Energy storage system today is very expensive and the technology is not ready to handle large storage capacity needs to support national electric grid.

All energy generating technologies produce waste as a byproduct. The wastes generated from conventional fossil fuel-based technologies come in large volumes. Due to high energy content of the fuel, nuclear power produces far less amount of waste. For example, the Kingston Fossil Plant, a coal power plant in eastern Tennessee, in 2007, emitted 10.9 million tons of carbon dioxide, 51,000 tons of sulfur dioxide, 12,500 tons of nitrogen oxides, 1700 tons of hydrochloric acid aerosol, 230 tons of hydrogen fluoride, 330 tons of sulfuric acid aerosol, 11 tons of ammonia, and 780 tons of toxic heavy metals in airborne particulates. With the use of low-sulfur blend coals and low-NOx burners and installation of air pollution control technologies (scrubbers and selective catalysts to reduce NOx), the emissions in 2015 were reduced to 4.78 million tons of carbon dioxide, 1470 tons of sulfur dioxide, 1490 tons of nitrogen oxides, 44 tons of hydrochloric acid aerosol, 4 tons of hydrogen fluoride, 227 tons of sulfuric acid aerosol, 22 tons of ammonia, and 380 tons of toxic heavy metals in airborne particulates (TVA 2020). In comparison, a nuclear power plant at Watts Bar a few miles away from the Kingston Plant, produced 26 tons of spent fuel waste along with less than 100 tons of low and intermediate level waste in the same year.

Use of solar power, such as solar photovoltaic, also carries the burden of waste management. Solar panels produce 300 times more waste than nuclear power plants while producing the same amount of electricity. Solar panels often contain heavy metals as part of impurities in the glass of the panel and release toxic materials into the soils upon breakage of the panels during disposal. While solar panels are typically disposed of in regular landfills, no proper regulation is presently in place to manage the waste in many countries. Wind power comes with much less burden on waste management as most of the wastes from wind turbines can be recycled. However, turbine blade materials (made of resin and fiberglass) are rarely recycled today. These turbine blades are very long and large and the associated disposal cost can be high.

Although the wastes generated from nuclear energy are very hazardous, they come with small volume, (~10 m^3 of spent fuel and ~30–100 m^3 of low level waste per 1000 MWe/y). Being small in volume, it is conceivable to develop a dedicated system to *isolate and contain* nuclear waste as discussed in previous chapters. Natural decay characteristic of radionuclides can be utilized in the design of the systems to provide the necessary level of safety. In contrast, there is no decay in the hazards of heavy metals or toxic chemicals.

Continued use of fossil fuels may make the planet earth uninhabitable. Renewable energy alone may not sustain human developments in the future Although nuclear energy has its own problems, use of nuclear power would leave much less burden to future generations. The problem of nuclear waste is a legitimate problem to solve to sustain human lives in this planet.

17.2 Difficulties with Human Institutions in Dealings with the Problem of Nuclear Waste

Developing a comprehensive national policy for nuclear waste management is a tall task. In particular, in democratic societies with frequent changes of public officials, the short term nature of the leadership of responsible institutions or government personnel makes developing a comprehensive policy challenging. The task at hand requires managing complex, tightly coupled systems of relations, technologies, and issues that requires long-term coordination and commitment for the attainment of the desired outcome. For the national policy to address all of the necessary coordination and balancing, concerted efforts among the stakeholders are necessary. The reality is that the elected officials of the government often have only specified terms which may not provide the longevity needed to carry out the tasks. The syndromes like “not in my term of office” indicate the pervasive nature of elected officials being reluctant to touch upon unpopular issues like nuclear waste during their terms.

There is also potential for abuse of political process in policy making. While the importance of participatory decision making well noted, the system of electing officials and representatives through democratic processes puts special premium on what the public demands. Accordingly, the expressed preferences of the citizens could dominantly drive the political process leaving important but less popular concerns of societal sector ignored. This implies the likelihood of politically biased positions taking over the policy process at the cost of resource mismanagement. This indicates the importance of healthy dialogues among the stakeholders as part of the policy process.

Even after the policy for nuclear waste management is set up, execution of the policy takes time. The execution can occur over the period of several decades. With government administration changing frequently, the leadership of different administrations may have different views or goals on the issue of nuclear waste. The situation may open the possibilities of the policy being reversed during the policy execution phase. The necessary resource to support policy execution can also be scarce, as the funding is often subject to annual appropriation process. This may destabilize or delay the project. Even worse, after achieving national consensus through a long arduous negotiation processes, the project may get cancelled. This is what happened to the Yucca Mountain project of the U.S. as reviewed in Sect. 2.2.

Ensuring long-term social and political commitment to the management of nuclear waste is an important component in policy execution. This is simply because the problem requires long-term custody of institutions. The national policy should survive different administrations. For instance, stability is embedded in the national policy for national security as law. This is to prevent manipulations per administration changes. Policy for nuclear waste needs similar provisions if a country is serious about making progress. Providing stability in project funding is also desired. Instead of being subject to annual appropriations, stable funding should be sustained with strong accountability requirements through periodic audits at each major milestone point. Guidelines for coordination and management of the project regarding intergovernmental interactions should be given with specifications on the limits or conditions of administrative power in changing the course of policy execution.

Needless to say, for any country with nuclear power, a well-thought-out policy for nuclear waste management is a prerequisite toward success in nuclear energy use. However, policy for nuclear waste usually does not take priority at the outset. This was the case in nuclear power developments in many countries. Most countries were focused on plant operations or technology developments but paying much less attention to the issue of nuclear waste. This is because there is no sense of urgency with the problem. The problem of nuclear waste seemed easy: It can be solved when necessary. In reality, countries who went through this path had to pay a big price when the time came to take care of the problem: Not paying enough attention early on often hampered national ability in finding solutions later on. For example, the U.S. started nuclear technology development in the 1940s. Then, it took almost 40 years for the U.S. government to promulgate the Nuclear Waste Policy Act in 1982. During these periods, episodes of spills, contaminations, and mismanagements were reported and the level of trust among the public toward government went down significantly. This made the development and execution of national policy on nuclear waste very difficult. The history of LLW management in the U.S. as summarized in Sect. 13.1 also tells that lack of government policy and leadership were the reason for haphazard practices resulted in environmental contamination from LLW in the country.

These observations indicate that national policy for nuclear waste management should be developed early, preferably as soon as nuclear wastes are generated from nuclear power plants. Starting early the process of policy making helps to avoid mistakes in later years. When a country starts a nuclear power program, the public in general welcomes it with positive attitude as such development comes with the promise for economic prosperity and a sense of national pride. Attaining national consensus to support policy development can be readily pursued under such favorable social atmosphere.

17.3 Risk Perception of Nuclear Waste

Presence of very negative risk perception of nuclear waste among the public is well noted as discussed in Sect. 16.3. Such risk perception is also closely related to radiation phobia, i.e., a fear of radiation. Radiation phobia is widespread among the public perhaps due to the memories of atomic weapons. The story of fruit flies developing massive genetic mutations (according to the 1927 experimental findings of Dr. Hermann Muller as quoted in books such as "Small is Beautiful") could have contributed to radiation phobia. As discussed in Sect. 5.2, studies over the last 60 years on the surviving population of atomic bomb explosions in Japan has found no evidence of genetic damages among the survivors' offspring.

Radiation is ubiquitously present in our lives through cosmic radiation, terrestrial radiation, and naturally radioactive materials in air, food, and buildings. Our life on the earth implicitly accepts the risk of background radiation. Therefore, life with radiation is not a reality. An argument can be made that fear of radiation is more dangerous than radiation itself due to its psychological harm or stress. Studies have indicated that the risk of radiation below certain level of dose is negligible. This is the basis of the ICRP's recommendation for 1 mSv per year as recommended dose limit for the public (as discussed in Sect. 5.3). Notice that the annual dose from background radiation ranges between 3 mSv and 5 mSv among the members of the public in the world. The radiation safety limits employed in various countries for nuclear waste disposal is at less than 0.3 mSv per year.

A related question is whether such fear of radiation or negative risk perception of nuclear waste is subject to change. People's personal experiences tells that fear can be removed or overcome through knowledge and/or experiences. Studies have also shown that risk perception is subject to change as noted in Sect. 16.4.1.

The degree of difficulty in changing risk perception depends on how the risk perception was initially formed. As discussed (Sects. 16.4.1 and 16.4.2), if the perception was formed through careful deliberations of the issue, the resulting risk perception is expected to be stable. However, if the perception is formed by using simple cues or judgment heuristics (i.e., through peripheral processing of information), such risk perception is subject to change. This may be the case with the risk perception of nuclear waste among the majority of the public. This is because the public in general lack the ability and motivation to make careful deliberations on the issue. At the same time, difficulties are still expected to change the risk perception of nuclear waste as the risk of nuclear wastes often causes fear to the minds of the public.

Even if the existing risk perception of nuclear waste still persists, consensus can be attained for the problem. The progresses made in countries like Finland and Sweden affirm that. In both countries, oppositions (worse in Sweden) initially arose from the public against any development for nuclear waste disposal. The public had very negative perception of nuclear waste risk. Both countries, however, were able to arrive at solutions. While negative risk perception may have persisted, acceptability of risk was created. In the case of these two countries, well conducted risk communication efforts and community engagement activities led to successful outcomes of facility development with social consensus.

Consider Sweden. Sweden started the civilian nuclear power program in 1964 with the operation of small (10 MWe) nuclear reactor. The program grew to operate 12 nuclear power plants in 1980s. With such growth, the need for the management of nuclear waste became strong. The country established the Swedish Nuclear Fuel and Waste Management Company (SKB) in 1977 to develop comprehensive plan for the management and disposal of nuclear waste. As part of the development, the effort to find a suitable site for a disposal facility began in 1977. This led to test drilling at potential candidate sites which soon faced intense public protests. For example, the 24 h watch was organized by the public over the roadway leading to the intended drill sites. Such confrontations led SKB to change their approach by emphasizing public engagement and communications. In 2009, SKB announced the selection of the Forsmark site as the host of the nation's geologic repository for spent fuel disposal. The decision was based on 77% of the local citizens supporting the decision. Swedish government is currently reviewing the license application for the construction of waste encapsulation plant and geological repository at the site. SKB is expected to start the construction of the geological repository in early 2020s.

It is obvious that people cannot live in a risk-free society. Even if there were no technologies around us, there always will be background risks from natural sources. Also, although risk acceptability is related with the magnitude of risk, there is no strong correlation between the two as the decision on risk acceptability is heavily affected by personal values and priorities regarding the possible outcomes within the given social context. Douglas and Wildavsky state that "we choose the risks in the same package as we choose our social institutions" (Douglas and Wildavsky 1982). As long as the project of nuclear waste is presented as an acceptable one amongst other alternatives, the risk of nuclear waste can be accepted.

As stated earlier, public responses to nuclear waste activities are commonly characterized as NIMBY or LULU. Stronger syndromes such as NIMTOO, NIABY and BANANA also exist. These responses represent a set of complex public reactions to both the risk and the social circumstances involving nuclear waste or other hazardous materials. Although these responses are usually labelled irrational by the technical community, close examinations of the phenomena reveal the opposite (Inhaber 1992). Studies of NIMBY showed that the residents' opposition is not just about risk but more about sovereignty (Stevens 1993). This means that, to address NIMBY, the sense of intellectual sovereignty should be provided.

The defining element of NIMBY is defense of people's home. For most people, home is their shelter, protector of their privacy, and expression of their personality (Lee 1993). For them, preserving their home and neighborhood determines the quality of life and the security of their investment. These local residents are defending inherent individual right and protecting their physical and ideological home. They have every incentive to oppose activities which may threaten the very existence of their home, security, personality, and privacy.

To overcome the NIMBY challenge, incentives that motivate people's behavioral change must be provided. These incentives must provide assurance that the alternatives presented can meet their personal needs and quality of their life can be preserved. For their decisions to be voluntary instead of being imposed, enough incentives should be assured based on market principles (Inhaber 1992). The people who would be most affected by the waste site should see the incentives larger than what they have to lose. The incentives should be large enough for the residents to overcome fear.

Adequate citizen empowerment is a necessary part in this process. Citizen empowerment means meaningful involvement of local citizens in understanding risk and making decisions about risk. Government agencies must seek consensus with those who must ultimately live with the consequences of the resulting decisions. Empowerment doesn't mean that residents would share the power of operating the local facility. Empowerment allows residents to exercise their territorial and risk bearers' rights. To support citizen empowerment, proper risk communication is a must. Such communication is to provide community-sensitive and technically accurate risk information.

Distinct parties involved in risk communication include the risk producer (government or industry), scientific experts (as consultant or advisors), the media and interest groups, and the general public. As the interrelations and interactions among these parties are complex, the roles, interests, needs, objectives, and constraints of the parties should be carefully examined and understood. Risk communication must be capable of assisting mediation, negotiation, or conflict resolution between the interested and affected parties coping with disagreements. Through communications, interactions, and participations, the general public becomes informed, intelligent, and even confident in making their decisions.

For any risk communication effort to be effective, the information exchange must address core issues “under contest”. A simple exchange of factual information is highly unlikely to be effective for the purpose. The exchanges must also affect the related value system of the participating individuals. Skepticism on the part of the public can be overcome if impartial, credible sources confirm the risk to be acceptably low.

In these processes of communications and interactions, trust can be a critical factor. When trust is present, individuals engage in cooperative and altruistic behaviors (PEW 1996). Lack of trust pre-determines people's interpretation of the interactions reinforcing their prior beliefs against an entity, preventing the change of perception. Perceptions of others' trustworthiness are largely history-dependent. If an organization or a person lacks trust by the public, the first thing to do is to admit past mistakes and to defy the negative stereotype by committing to different behaviors from the past. The behavior must demonstrate that the organization cares about the public and is capable of delivering on the promises. Investments must be made that are required to change the culture of the organization without making exaggerated claims and promises. Successful communication involves building trust or establishing credibility. Information exchange under trust relationship will render issue-relevant examinations non-confrontational and cooperative which may lead into changes in perception and attitude.

17.4 The Challenge of Long-Term Safety Performance Requirement

Critics of nuclear waste disposal argue that science is trying to do what the science is incapable of doing in nuclear waste management. The issue at hand is whether long-term safety can be guaranteed in nuclear waste disposal.

Limits in science and technology to demonstrate safety of nuclear waste disposal are well noted. It is not realistic to demonstrate the performance of any engineering system for periods beyond human civilization. In this regard, the regulatory requirements poorly match the technical task at hand.

One of the key concerns about the limits of science is uncertainty. There are inherent uncertainties in quantitative predictions of very long-term future. However, presence of uncertainty does not necessarily mean that the risks involved would be significant. While providing absolute guarantee of safety is not possible, serious unforeseen events leading into catastrophe happening in geologic disposal of nuclear waste is highly unlikely. It is not the detailed engineering analyses that provide or promise safety in the future. Discreet applications of scientific principles would be much more credible in providing long-term safety. Such approach is the basis of regulatory decisions in determining acceptability of nuclear waste disposal as discussed in Chap. 15.

A simple intuitive approach was suggested to examine safety of nuclear waste disposal as an example while recognizing limits of science (Cohen 1995). The suggested approach assumes that buried HLW behaves like average natural rock. Its dissolution into groundwater then contaminates food and water consumed by humans. The calculation used the national average of rainfall and elevation and ignores the protection by the multiple engineered barrier system. Then the probability of dissolution of an atom of natural rock into groundwater per year is estimated. Geochemical speciation of specific elements is ignored in the calculation. The resulting probability is 10^{-9} per year. The probability for an atom dissolved in groundwater to enter a human stomach is then subsequently calculated. This step assumes consumption of this contaminated groundwater through drinking of water from contaminated well and river, ingestion of plant food irrigated with the contaminated groundwater, and eating fish from rivers contaminated with groundwater. Therefore, presence of long travel time and decay or dilution of radionuclides during groundwater travel are ignored. The estimated probability of radioactive atom entering human stomach is 4×10^{-4} per year. Multiplying these two probabilities with corrections for the differences between HLW and natural rock, the probability for an atom of the buried HLW to enter a human stomach is estimated at 10^{-12} per year. This probability is then multiplied by the total number of atoms in buried HLW to give the number of deaths per year that would result if all of HLW were ingested by humans. The resulting estimates as fatalities per year can be added up over the time periods to provide the total number of deaths expected from HLW disposal.

This analysis produces the estimate of 0.0005 deaths over the first 500 years from the high level waste produced from the production of 1 GWe of electricity per year.

If all of the deaths occurring over millions of years are added based on the assumed approach, the estimates of death is 0.02 per GWe per year. The resulting numbers as the estimate of potential deaths in the future are low. This analysis does not consider variations in the compositions of groundwater and surface water and related geochemistry effects but attempts to characterize the long-term risk of nuclear waste disposal in a very simplistic way. The approach is in contrast to complex analysis efforts to support long-term predictions of the geological repository system behavior.

This analysis provides a way to represent the risk of nuclear waste in characteristic terms. In the end, the results may not be accurate. However, the general concepts captured by the analysis may indicate that the risk of nuclear waste disposal is not very high in contrast to what the public risk perception dictates. The approach signifies, as an example, the non-dramatic consequence of any failure in nuclear waste disposal.

Engineered systems, hydrogeological systems, as well as the surrounding natural environment of nuclear waste disposal will inevitably go through changes in the future. Therefore, the approaches to address safety in the systems may need to allow and emphasize flexibility. To provide long-term safety in the face of uncertainty, there needs to be a willingness to respond to problems as they are found along the way.

17.5 Conclusion

As a final one, this chapter reviewed key challenges in nuclear waste management and elaborated possible ways to address them. Part of the challenges comes from the long-term nature of the problem, inadequacies of human institutions to deal with the problem, and difficulties in deriving social consensus. Recent success stories in countries like Finland and Sweden highlight the importance of science/technology and human institutions closely working together to support the public. Science and technology must continue their best effort in preventing and minimizing the risk from nuclear waste. Responsible plans and approaches must be in place to render the consequences of unwanted failures in the system to be minimal. Human institutions must be vigilant in guiding the execution of science and technology through necessary policy development and establishing and maintaining the necessary infrastructure. Through cooperation among human institutions, the public, and the scientific and technical community under social stewardship, the goal of safe and responsible management of nuclear waste can be attained.

Homework

Problem 17.1: A country in Southeastern Asia wants to be a new owner of nuclear power plant. The country's government will purchase nuclear fuels from an outside supplier (without the front-end fuel cycle capability) and will be responsible for the back-end of fuel cycle based on the once-through fuel cycle concept. Therefore the types of nuclear waste the government will need to manage are spent fuel and low/intermediate level waste. Geological formations available in the country for HLW disposal include granite and clay sites.

(a) Describe key provisions needed in their national nuclear waste policy.
(b) Describe key provisions needed in their regulations and standards to ensure safety in nuclear waste management.
(c) Summarize key challenges in the country's nuclear waste management program and explain how these challenges should be addressed by the government.

Problem 17.2: Do you think nuclear waste management a legitimate social problem to solve? Explain your answer.

Problem 17.3: If the public has very negative risk perception of nuclear waste, can progresses be made toward success in nuclear waste management? Explain your answer.

Problem 17.4: Discuss the specific efforts to be made by the scientific/technical community to achieve success in nuclear waste management.

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Correction to: Introduction

Correction to:
Chapter 1 in: M.-S. Yim, *Nuclear Waste Management*,
https://doi.org/10.1007/978-94-024-2106-4_1

The original version of the content **“Especially, the case of the U.S., where the effort for nuclear waste management began in the 1950s and continued over the years with finalizing the site selection process in 2002 by announcing Yucca Mountain as the nation’s high level radioactive waste disposal site. This highlights the challenges associated with nuclear waste management”** in page 2 of chapter 1 has been revised with the below content as per the author’s request:

“Especially, the case of the U.S. highlights the challenges associated with nuclear waste management, where the relevant effort began in the 1950s and continued over the years with finalizing the site selection process in 2002 by announcing Yucca Mountain as the nation’s high level radioactive waste disposal site. This plan was subsequently cancelled in 2010.”

The correction to this chapter can be found at
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