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The Nuclear Industry

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INTRODUCTION

The objective of the nuclear industry is to produce energy in the forms of heat from either fission reactions or radioactive decay and radiation from radioactive decay or by accelerator methods. For fission heat applications, the nuclear fuel has a very high specific energy content that currently has two principal uses, for military explosives and for electricity generation. As higher temperature reactors become more widely available , the high temperature heat (>900°C) will also be useful for making chemicals such as hydrogen. For radiation applications, the emissions from radioactive decay of unstable nuclides are employed in research, medicine, and industry for diagnostic purposes and for chemical reaction initiation. Radioactive decay heat is also employed to generate electricity from thermoelectric generators for low-power applications in space or remote terrestrial locations. As well, radiation produced from accelerator-based sources is used for geologic investigation (e.g., identifying oil deposits), materials modification, and contrast imaging of dense media (e.g., security inspections in commercial shipping). Fuel from the first atomic pile is shown in Figure 21.1.

This nuclear technology is based on both the nuclear and the chemical properties of the atom. At the beginning of the twentieth century fewer than 90 chemical elements were known and there was only a dawning awareness of isotopes. Today, largely because of the nuclear industry, thousands of isotopes (or nuclides, depending on the properties of interest) have been identified. Brief definitions of several chemical and nuclear terms are given in Table 21.1.

As with other technology, nuclear technology involves a combination of science and art. However, it is unique because of the development of the atomic bomb that contributed to the ending of World War II. Many people view nuclear technology from the point of view of nuclear weapons and more recently nuclear accidents such as those at Chernobyl and Three Mile Island. This leads to the view that nuclear technology is only useful for explosive applications and that it is only with great care that it can be safely used. In reality, it is difficult to produce nuclear explosions

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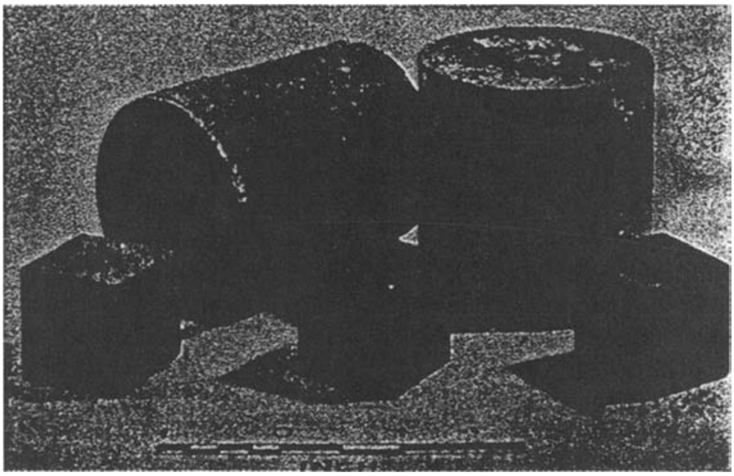


Fig. 21.1. Uranium metal fuel from the first atomic pile. This reactor contained 40 tons of uranium oxide along with 6.2 tons of uranium metal. *(ORNL News, 1-01-076.)*

TABLE 21.1 Definitions of Atoms, Chemical Elements, Isotopes, Nuclides, and Isomers

Atoms:	Elementary particles of matter composed of protons, neutrons, and electrons. In a neutral atom the total number of orbital electrons is equal to the number of protons in the nucleus.
Chemical elements:	Atoms with unique properties related to their orbital electrons.
Fissile:	Capable of undergoing nuclear fission initiated by a slow neutron.
Fissionable:	Requiring a neutron of kinetic energy above a threshold value, to initiate nuclear fission.
Isotopes:	Atoms of the same chemical element with different masses related to a different number of neutrons in the nucleus.
Nuclides:	Atoms with unique properties related to the neutrons and protons in the nucleus of the atom.
Isomers:	Nuclides with the same numbers of protons and neutron but in different energy states.
Radioactive decay:	The process by which unstable nuclei become more stable.
Radioactive half-life:	The time during which the decay rate of a radioactive nuclide decreases by a factor of two.

and safe use of nuclear energy is really only a matter of following common-sense rules of behavior no more mysterious than those involved with handling dangerous chemicals or electricity.

This chapter first considers the following subjects.

- The status and outlook of the nuclear industry
- Nuclear safety.
- The role of nuclear energy in meeting the world's energy needs.
- The nuclear processes on which the nuclear industry is based (Fig. 21.2a).
- The chemical process technologies involved in the fuel cycle for nuclear electric power and chemical process heat generation (Fig. 21.2b).

This is followed by discussions of radioactive waste management and transportation of nuclear materials, and consideration of various applications, such as nuclear power reactors used for the generation of electric power, the use of radioisotopes, and other military and civilian uses of nuclear materials (Fig. 21.2c). The processing of uranium ore, the enrichment of material for use as nuclear fuels, the production of electricity using nuclear fuel and the subsequent handling, storage, and in some cases reprocessing of the radioactively contaminated waste constitute some of the more sophisticated and challenging areas of fundamental and applied chemistry and engineering as well as requiring huge investments on the order of tens of billions of dollars for each stage in the process.

A reference section is provided for those who wish to have more detailed information.

STATUS AND OUTLOOK

Nuclear energy provides products that play vital societal roles:

- As a primary energy source for the production of electricity to meet the world's energy needs without polluting the atmosphere.[1] Currently, it is the only large-scale electrical production process that does not produce greenhouse gases (e.g., CO_2) or other pollutants such as SO_2, NO_x, mercury, or particulates.
- As stable and radioactive nuclides, which contribute significantly to research, medicine, and industry (see Table 21.2 and Fig. 21.2.).
- As radioactive decay heat sources that in conjunction with thermoelectric conversion devices provide low leves (kilowatts) of power for very long times in deep space probes and for remote terrestrial applications such as weather sensors.
- As nuclear explosives and in nuclear-powered submarines and ships, which have contributed to maintaining world peace since World War II, but have since declined in importance.
- Test reactors that are used to generate isotopes, do chemical analyses, and study materials.

TABLE 21.2 Nuclear Industry Products (2005)

	Approximate Number
Civilian:	
Worldwide electric generating plants	440
Ships (icebreakers and transports)	~10
Research reactors	280
Radioisotope applications (tracers, radiation sources, thermal sources)	Millions
Radiation modification of materials (solid-state devices, treated polymers, etc.)	*Small (mostly proprietary)*
Military:	
Weapons	20,000[a]
Submarines	160
Ships (aircraft carriers, cruisers, transports)	~80

[a] Although it is a possibility that up to 10,000 more nuclear warheads may be awaiting dismantling or are in reserve in Russia.[4]

Nuclear technology continues to be developed for military applications including nuclear explosives, submarines, and ships. Brief attention is given herein to explosives, primarily in terms of their historical role as a potential means for excavating harbors and canals and in the enhancement of gas and petroleum deposits.

It should also be noted that since the end of the Cold War, official military stockpiles of nuclear (and other) weapons have been diminished, by international agreement. At the same time, acquisition or fabrication of nuclear devices by nongovernmental terrorist groups or individual states has become a matter of increased concern.

Since the discovery of nuclear fission in the late 1930s, this technology has been developed to supply 16 percent of the world's electricity from 440 nuclear reactors located in 30 countries (2005).[2] This is an international program regulated by the United Nations' International Atomic Energy Agency, which, as of 2004, includes 137 member countries. In 2004 there were 104 nuclear units in the United States, including 69 pressurized water reactors and 35 boiling water reactors, which together generated 789 billion kWh of electricity, some 20 percent of the country's total electricity generation.[3] This represented a 214 percent increase in output since 1980. (See Fig. 21.3.)

The safety record of the nuclear industry has continued to be generally very good, despite the 1987 accident with a nuclear reactor at Chernobyl, in Ukraine, which killed a number of workers, caused the abandonment

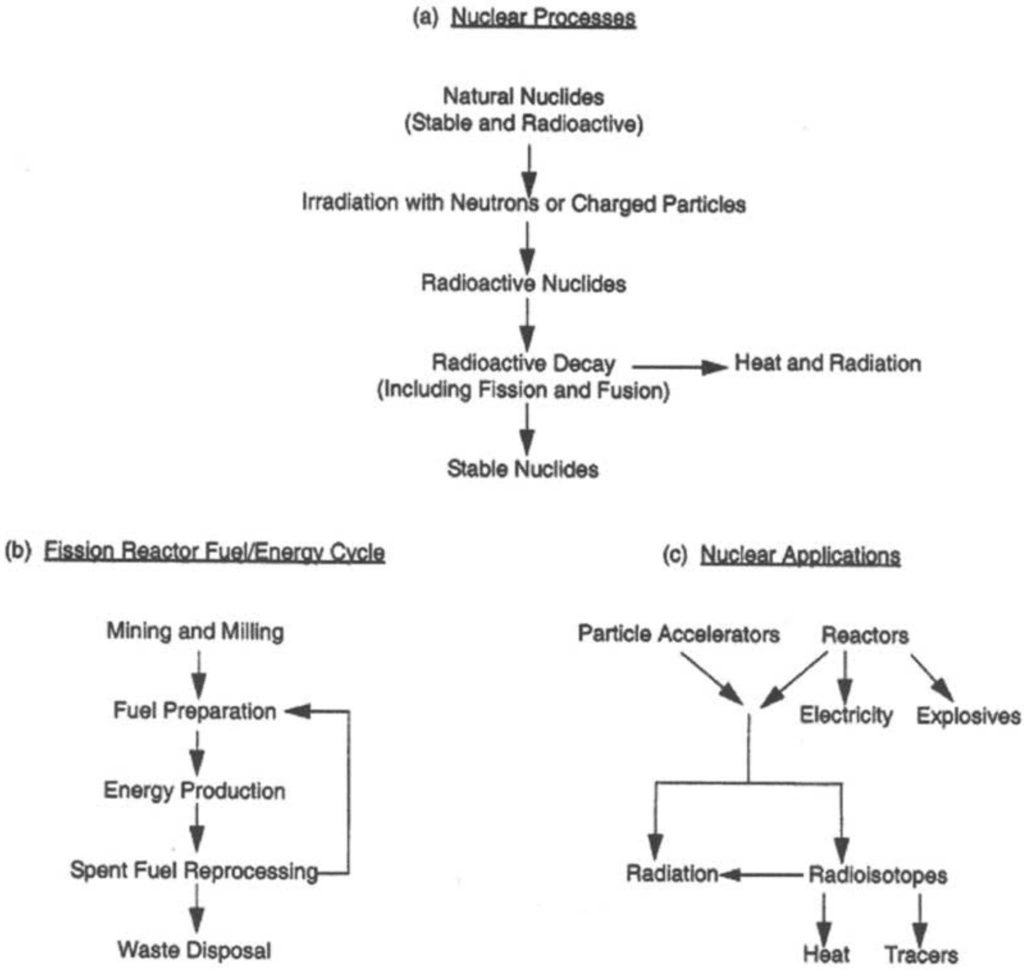


Fig. 21.2. The nuclear industry.

of adjacent communities and farmland, and resulted in radioactive fallout in neighboring countries. It should be noted that these were the first civilian nuclear fatalities since the beginning of the nuclear power industry.[5] It is generally agreed that this unfortunate occurrence was caused by serious design flaws, and that power reactors in use elsewhere are not subject to similar occurrences. Indeed, a somewhat similar yet also very different accident occurred in the United States in 1979 (Three Mile Island); but because of the reactor's inherent physics feedback mechanisms and its engineered safety features (required in the United States and recommended by the International Atomic Energy Agency), there were no injuries and no significant radiation exposure, either to workers or the public.[6]

Worldwide, aggressive public concern about nuclear safety has been delaying the development of waste disposal facilities for high-level wastes. Current intermediate-level waste facilities in the United States are nearly full, and there has been great difficulty in siting new facilities. It now appears that for many years the interim solution for the storage of spent fuel elements will be aboveground storage facilities at existing reactor sites with appropriate safeguards and security. On the energy side, increased attention to

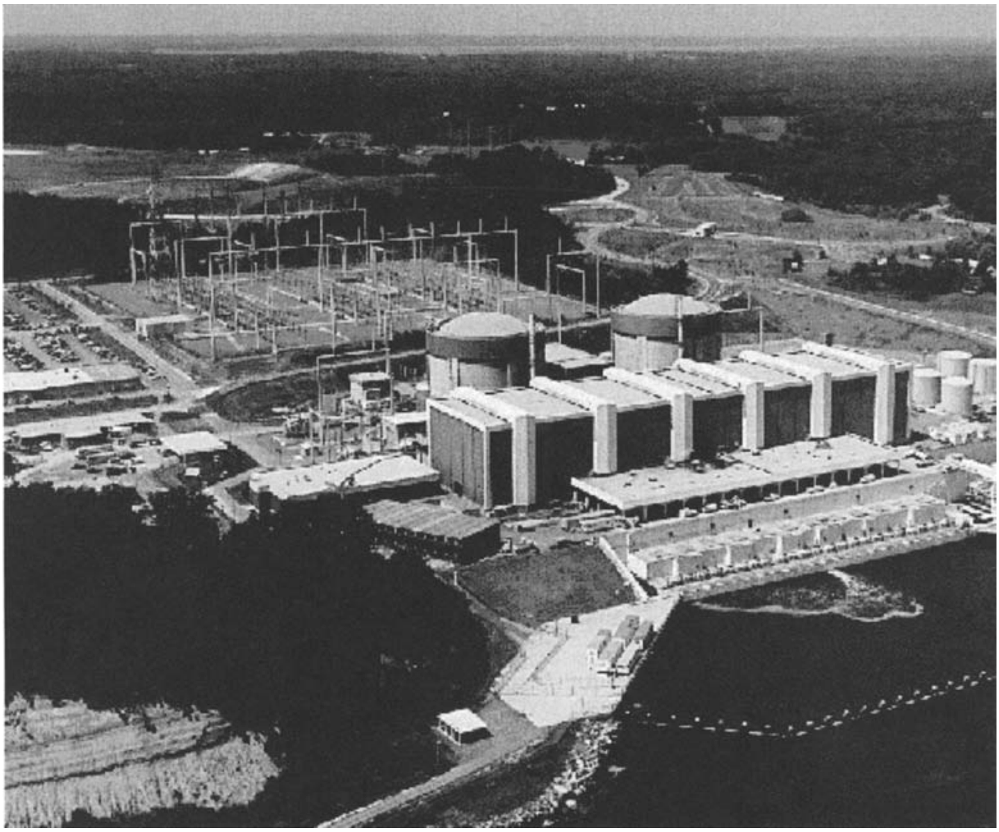


Fig. 21.3. The Calvert Cliffs Plant in Lusby, Maryland , Baltimore Gas and Electric. Capacity: 1600 MW(e) from two reactors. *(Courtesy of EEI-Electric Perspective.)*

conservation and the ready availability of natural gas in the United States that can be used in low-cost combined cycle electric generating plants have combined to end the urgent need for continued nuclear power development. However, due to declining new supplies of natural gas and increased usage for electrical power generation, natural gas prices have begun to rise with an attendant rise in the quantity of nuclear-generated electricity in 2005. Future importation of liquefied natural gas in the next few years is likely to establish a "floor" price of natural gas and provide immediate increased interest in new nuclear plants.

Recognition of the continuing instability of the oil supply was demonstrated by Iraq's invasion of Kuwait in mid-1990 and the U.S. invasion of Iraq in 2003, and the price volatility of oil has begun to lead to the realization that the lack of a United States energy policy that places less reliance on foreign oil (and soon to be natural gas) will continue to place the United States at risk of severe economic disruptions. Furthermore, the continued reliance on burning coal as a source of electricity is being questioned due to its adverse environmental impact in the short term due to NO_x, SO_x, and heavy metal emission and in the long term due to the emission of CO_2. Although natural gas use for electrical generation has increased dramatically, the higher cost of natural gas and the depletion of this limited resource that is a prime feedstock for the chemical industry for essentially short-term goals will put a damper on future economic growth.

Light water reactors (LWRs) and heavy water reactors (HWRs) have been successfully used for electric power generation throughout the world with good operating and safety records. In recent years significant advances in nuclear reactor technology have been realized. A new generation of light and heavy water reactors has been designed and licensed to further reduce the already very low probability of loss-of-coolant accidents such as occurred at Three Mile Island and Chernobyl. In addition, the enrichment level of new fuel for LWRs has been increased, and the average fuel life has thereby been extended from 33,000 to as high as 50,000 megawatt days/metric ton, and the typical reactor cycle between shutdowns has been increased from 12 to 18 months. As a result, online times (i.e., capacity factors) have increased to the low 90 percents, providing low-cost as well as reliable electrical power generation.

All the components of the nuclear-fission power system are fully operational except for ultimate waste disposal. However, spent fuel is not reprocessed in the United States because there is currently an adequate supply of natural uranium and enrichment services available domestically and from other countries at a lower cost than that of the recovered fissionable material from spent fuel. Also, the United States unilaterally declared a moratorium on reprocessing in the early 1980s in an attempt to reduce the spread of nuclear weapons. Current economics do not favor a return to reprocessing and fuel recycling in the United States at this time in as much as it does dramatically increase the amount of interim and final waste storage capacity that is required.

The nuclear industry makes available about 3000 nuclides, including both the stable and the radioactive nuclides. Approximately 50 radioactive nuclides, along with some stable nuclides that have been isotopically enriched, are essential in research, medical, and industrial applications. Many of these are now produced commercially, but several still are dependent on government facilities. Some, for economic reasons, come from other countries. Radiation processing for sterilization of disposable medical supplies is an important operation using cobalt-60 from Canada. Electron accelerators have replaced radioactive nuclides as radiation sources for polymerizing plastic coatings on wire and paper. Perhaps the greatest disappointment experienced by the industry has been the public opposition to irradiation of food to extend its shelf life. The principal current application of this type is by the Russians, who are using electron accelerators for the deinfestation of wheat, and the use in the United States for treating herbs and spices.

In the continuing research and development of advanced nuclear energy, the liquid metal reactor (LMR), molten salt reactor (MSR), high-temperature gas-cooled reactor (HTGR), and fusion are the major activities. It would now appear that these alternatives will likely increase the cost of electrical energy in comparison with the present LWR/HWR technology based on low-cost uranium ore. HTGRs have been of particular interest since 2003 when the United States declared its intent to replace the use of oil-based fuels for transportation with hydrogen. Several hydrogen production processes (e.g., the Westinghouse Process and the Sulfur Iodine Process; see Figure 21.4) are being investigated that utilize high temperature (850–900°C) heat from a reactor to provide a major portion of the energy for making hydrogen. These temperatures are only compatible with HTGRs and fusion reactors. The advantage of using these high-temperature processes is the much higher overall energy efficiency that is obtainable (see Figure 21.5). For instance, the Westinghouse Process can achieve overall efficiencies of 50 percent or greater.

Plant-scale liquid metal reactors are in operation in France and Russia to establish the engineering technology and to evaluate their role in increasing the efficiency of uranium utilization. France, without a significant primary energy supply, wants to minimize imports of uranium. The United States imports about 50 percent of its uranium. In the United States, engineering test LMRs have operated for many years and have made significant contributions to LMR technology. Whereas they were first studied to reduce natural uranium requirements to approximately 20 metric tons/gigawatt year electrical

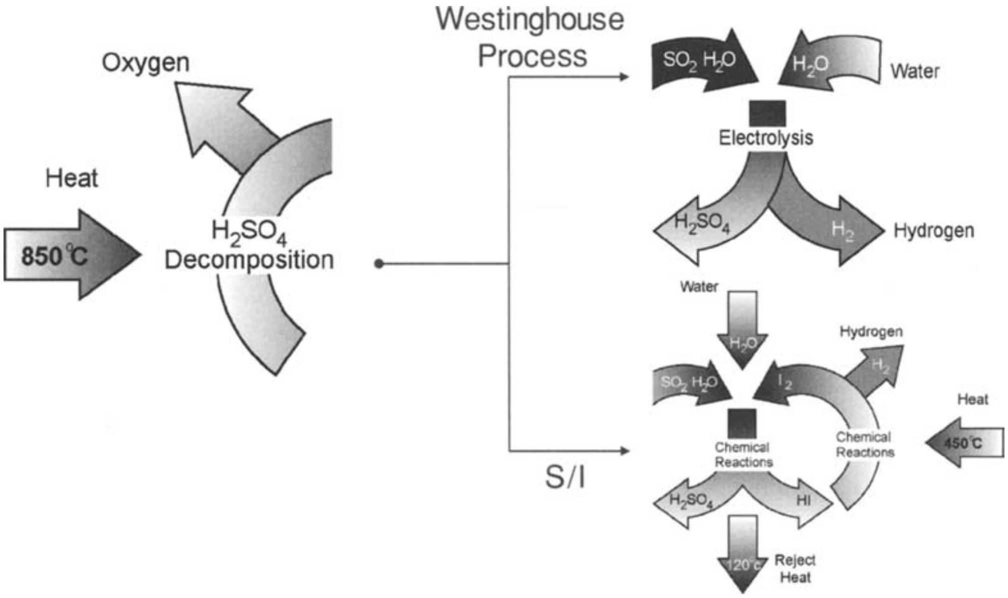


Fig. 21.4. High-temperature processes for generating hydrogen. (*Nuclear Hydrogen R&D Plan.* Final Draft, U.S. Department of Energy, March 2004.)

(GWye), development is now centered on (1) reducing plutonium in the waste, (2) the liquid metal benefit to safety, and (3) electrometallurgical reprocessing at each reactor site, thereby reducing proliferation concern. However, at this time all such LMRs have been shut down and are being decommissioned. Programs are also underway to use conventional uranium dioxide fuel containing "blended down" highly enriched uranium

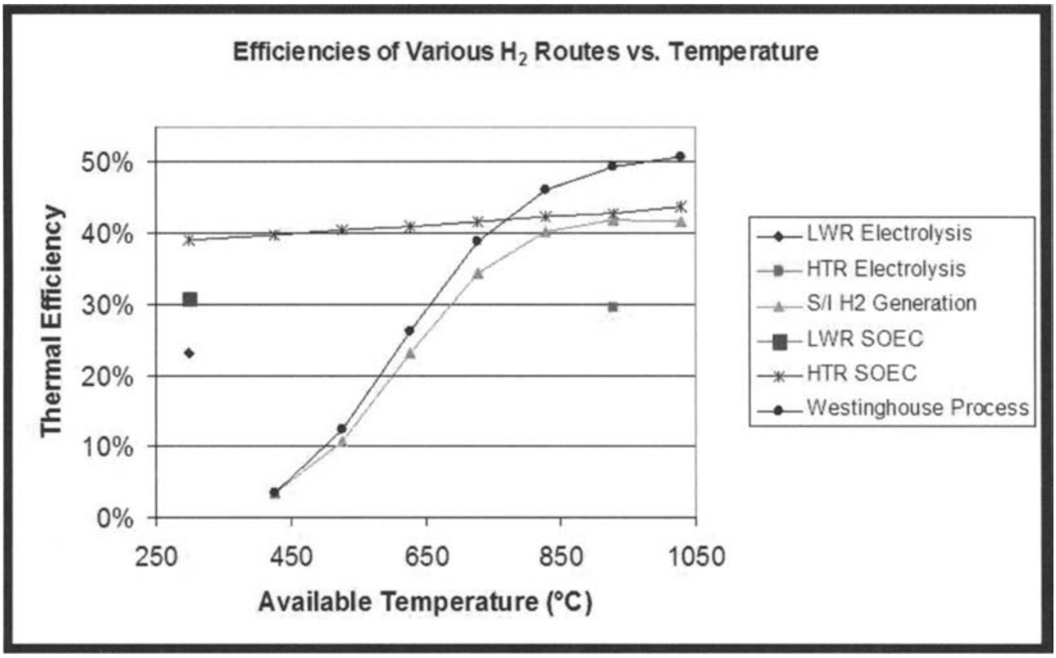


Fig. 21.5. Efficiencies of various H2 production processes.

(HEU) or mixed oxide (MOX) fuel containing oxides of uranium and plutonium in commercial nuclear power reactors, in order to dispose of nuclear weapons materials. Other theoretical reactor concepts are also being investigated for disposal of actinides.

It is difficult to assess the progress of fusion research, a significant international research program. Russia, the European Community, Japan, and the United States are the principal participants. It is now clear that fusion will be a high-cost source of energy and not economically justified within the foreseeable future. However, it is essential for the major world nations to cooperate responsibly in its development because of the severe technical challenges and high cost of research and development of this technology. The basic research has yet to achieve controlled ignition of the fusion reaction along with energy parity (energy input less than output).

At this time (2005), the outlook for the nuclear industry is difficult to project. France, Korea, and Japan, countries without domestic energy resources, are continuing to move ahead to establish efficient nuclear systems for electric generation; and although they have a concerned public, their fears regarding waste disposal and proliferation are not overriding their need for electricity. China has embarked on an aggressive nuclear build program, but its projected electricity demand is growing so rapidly that this aggressive program will only result in about 5 percent of electricity from nuclear in the next 15 years. In the United States, even with abundant coal, it will probably be essential to follow their lead because of energy security and environmental concerns.

NUCLEAR SAFETY

Safety first has always been and continues to be the basic policy of the nuclear industry. This includes reactor safety by design as well as activities to discourage the proliferation of nuclear weapons and to prevent sabotage of nuclear facilities. This policy has been successful; the chance of death from a nuclear accident is over a million times less than death from ordinary human activities, and over a thousand times less than death from natural events (see Table 21.3). Safety remains the most challenging responsibility of the nuclear industry.

TABLE 21.3 Average Risk of Fatality by Various Causes in the United States (1975)

Accident Type	*Annual Total Number*	*Individual Annual Risk*
Automobile accidents	56,000	1 in 4,000
Falls	18,000	1 in 10,000
Fires	7,500	1 in 25,000
Drowning	6,200	1 in 30,000
Gun accidents	2,300	1 in 100,000
Air travel	1,800	1 in 100,000
Electricity	1,200	1 in 160,000
Lightning	160	1 in 2,000,000
Hurricanes	94	1 in 2,500,000
Tornadoes	91	1 in 2,500,000
Nuclear reactors	—	1 in 5,000,000,000[a]
All accidents	112,000	1 in 1,600

[a] Calculation based on 100 LWRs operating under United States Nuclear Regulatory Commission supervision.

It is the public's strong perception that all nuclear activities are more dangerous than other accepted risks. Much of this public concern results from the atomic bombs and the government's secret program that produced the bombs. However, even with the early large-scale nuclear operations, there were few nuclear fatalities or life-shortening injuries. After 40 or more years of operation, these facilities are starting to be retired; however, there is a strong movement now to extend the life of most operating reactors for another 20 years. Meanwhile, considerable operating and safety experience has been gained that may very significantly reduce public concern in the future.

The nuclear safety program originally was based on both established laws regulating all industrial safety and early knowledge of radiation health effects related to radium and X-ray exposure. Since then the safety regulations have been greatly enhanced, and they are present in the Code of Federal Regulations, Title 10, Chapter 1, titled "Nuclear Regulatory Commission."[7] These regulations strictly control the management, engineering design, and operations of all nuclear activities. In addition the International Atomic Energy Agency, an agency of the United Nations located in Vienna, Austria, has established regulations as a reference for all national programs. Each country also has its own regulatory agency. There are two areas of concern in nuclear safety: radiation exposure of the public and of workers resulting from normal and accident conditions; and danger to world peace and order from nuclear weapons proliferation and sabotage.

Radiation, like air, has a ubiquitous presence in the human environment. The amount of background radiation that humans are exposed to ranges from 100 to 300 mrem/year depending on their location in the world, and an additional 60 mrem/year comes from other normal sources (medical, consumer products, etc.). The principal natural radiation sources are cosmic rays from outer space, radon from geologic sources, and potassium-40 within the human body (see Table 21.4). The radiation exposure to the public resulting from normal nuclear operations is limited by federal regulation of engineering design and management controls. There is also an overriding requirement that limits the radiation exposure to "as low as reasonably achievable (ALARA)."

TABLE 21.4 Annual Estimated Average Effective Dose Equivalent Received by a Member of the Population of the United States. (From HP Society University of Michigan site, 1/18/05.[11])

	Average Annual Effective Dose Equivalent	
Source	*(μSv)*	*(mrem)*
Inhaled (radon and decay products)	2000	200
Other internally deposited radionuclides	390	39
Terrestrial radiation	280	28
Cosmic radiation	270	27
Cosmogenic radioactivity	10	1
Rounded total from natural sources	3000	300
Rounded total from artificial sources	600	60
Total	3600	360

The annual radiation exposure of nuclear workers is limited to 5 rem, and the limit for public exposure (not radiation workers) from licensed activities is 100 mrem/year although people living near a nuclear power station are on average only exposed to <1 mrem/year.[8] The government also puts limits on fatality risk levels due to a nuclear accident:

> The risk of an immediate fatality to an average individual in the vicinity of a nuclear power plant that might result from reactor accidents should not exceed 0.1% of the sum of the immediate fatality risks that result from other accidents to which the U.S. population is generally exposed, and the risk of cancer fatalities to the population near a nuclear power plant should not exceed 0.1% of the sum of cancer fatality risks from all other causes.[9]

The levels of radiation exposure were first established on the basis of historical data, and since then they have been periodically evaluated by the International Commission on the basis of continuing experience.

In 1975 the United States Energy Research and Development Agency, the forerunner of the United States Department of Energy, sponsored the "Reactor Safety Study," the most thorough hazard analysis ever performed[10] for any technology. It concluded that on the basis of past licensing review practices, the risk from existing light water type power reactors was orders of magnitude less than other commonly accepted hazards. It further concluded that human error would be the expected cause of any accidents that might occur. This study also found that the cost of nuclear power reactor accidents would be several orders of magnitude less than the cost of other natural and human-caused events to which the public is accustomed (see Fig. 21.6).

In the 1950s the Nuclear Safeguards Program was established by the United Nations to administratively reduce the risk of nuclear proliferation. The objective of the Safeguards Program is to prevent the diversion of fissionable material, primarily plutonium,

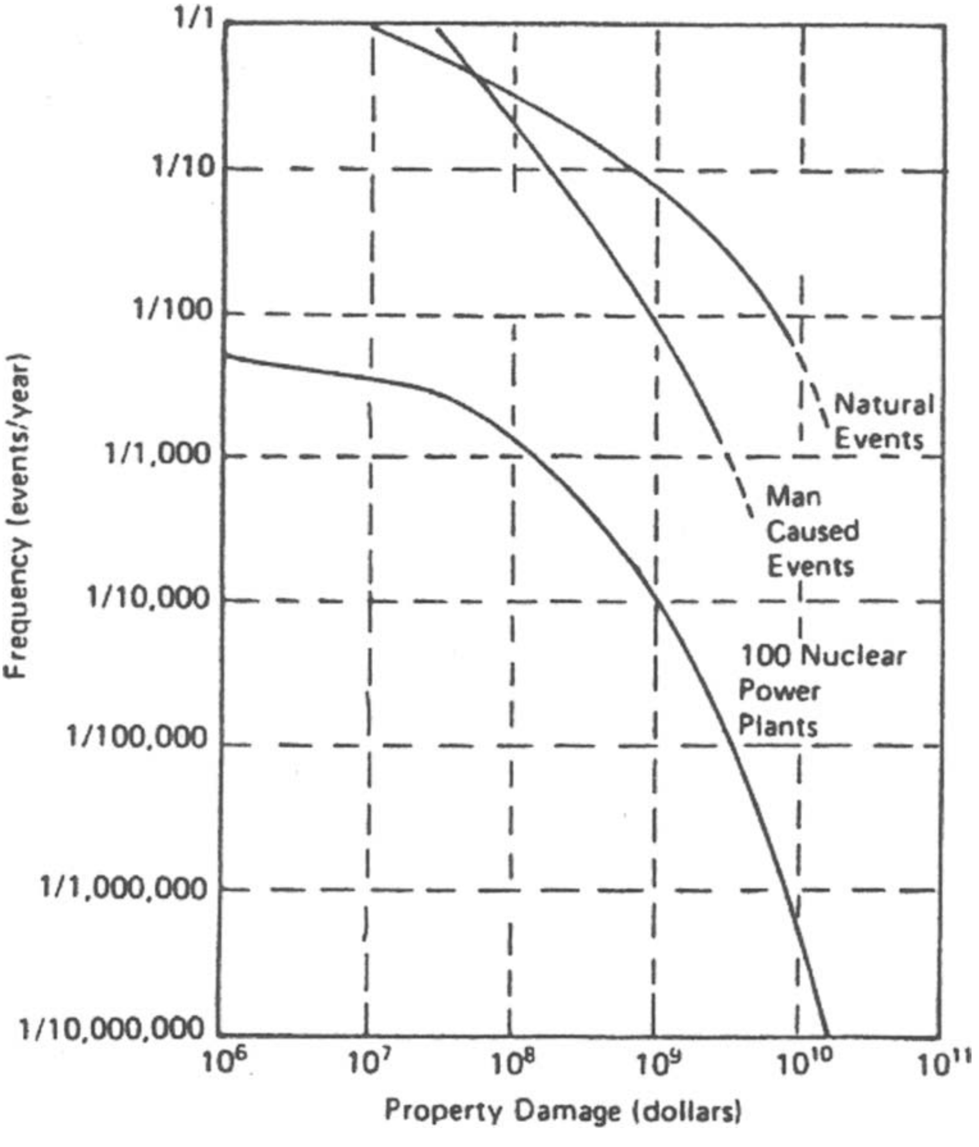


Fig. 21.6. Frequency of property damage due to natural and human-caused events. (Notes: (1) Property damage due to auto accidents not included. (2) Approximate uncertainties for nuclear events are estimated to be represented by factors of 1/5 and 2 on consequence magnitudes and by factors of 1/5 and 5 on probabilities. (3) For natural and human-caused occurrences the uncertainty in probability of the largest recorded consequence magnitude is estimated to be represented by factors of 1/20 and 5. Smaller magnitudes have less uncertainty.)

from the reprocessing of spent fuel. A force of onsite inspectors is employed by IAEA in this activity. This program is administered by the IAEA and is generally considered to be successful. Countries join this program voluntarily and do not include their weapons production facilities. India, Pakistan, and North Korea did not choose to join but developed their own nuclear weapons. It is believed that several nonmember countries have in the past or are developing nuclear weapons including South Africa and Israel.

In 1980 a study of proliferation control, titled "International Nuclear Fuel Cycle

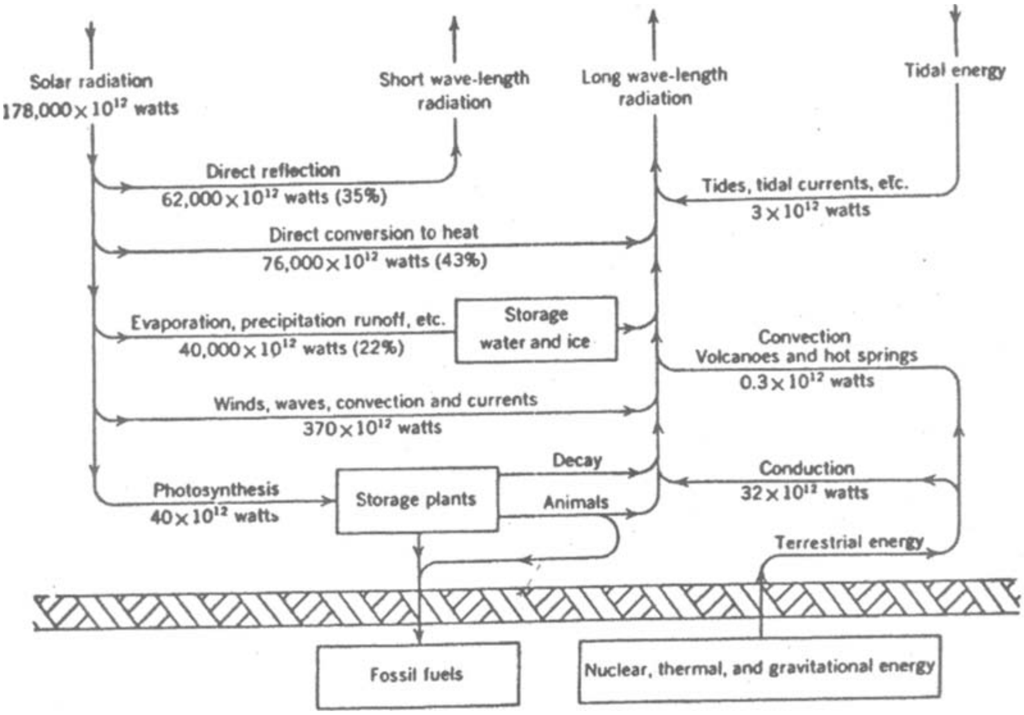


Fig. 21.7. World energy flow sheet. (*Hubbert, U.S. Geological Survey.*)

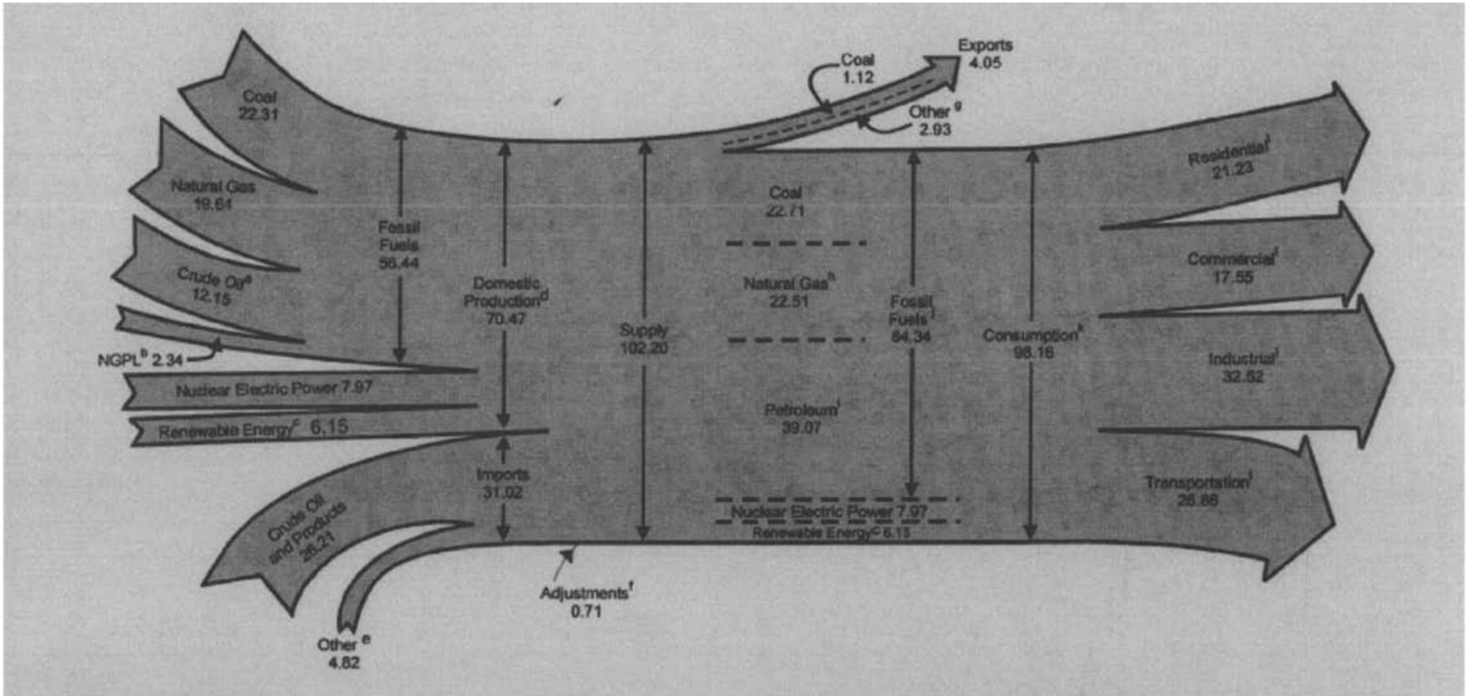
Evaluation," was carried out for President Jimmy Carter of the United States. The United States had called for this study, in which 50 countries participated and which required several years to complete. The study concluded that only administrative controls such as those already in place would be effective, and these could only delay really determined nations from diverting nuclear fuel from power reactors to military weapons. By 2003, North Korea became the first example of the subversion of test/commercial nuclear materials for use in weapons.

THE EARTH'S ENERGY SUPPLY AND USE

The earth is provided with two sources of energy, the "capital" resources deposited within the earth during its formation and the "income" resource continuously beamed onto the earth from the sun. Both are essential to human existence.

In the 1950s, global energy analyses were performed by King Hubbard for the U.S. Geologic Survey and by Palmer Putnam for the Atomic Energy Commission. Hubbard's work reported the income energy from the sun to be equivalent to 178,000 million megawatts (Fig. 21.7). At noon on a cloudless day a square meter of the earth's surface facing the sun receives approximately one kilowatt. The income resource also includes a very small contribution, less than 0.02 percent, from the gravitational forces of the sun, moon, and earth and from the thermal and nuclear sources within the earth. Part of the solar energy appears in the form of water and wind power, wood, alcohol, garbage, cow dung, solar heaters, and photoelectric generators.

Putnam's study, titled "Energy in the Future," was primarily concerned with the capital energy resources. These analyses are continued today by the Energy Information Administration in the U.S. Department of



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[a] Includes lease condensate.

[b] Natural gas plant liquids.

[c] Conventional hydroelectric power, wood, waste, ethanol blended into motor gasoline, geothermal, solar, and wind.

[d] Includes -0.09 quadrillion Btu hydroelectric pumped storage.

[e] Natural gas, coal, coal coke, and electricity.

[f] Stock changes, losses, gains, miscellaneous blending components, and unaccounted-for supply.

[g] Crude oil, petroleum products, natural gas, electricity, and coal coke.

[h] Includes supplemental gaseous fuels.

[i] Petroleum products, including natural gas plant liquids.

[j] Includes 0.05 quadrillion Btu of coal coke net imports.

[k] Includes, in quadrillion Btu, -0.09 hydroelectric pumped storage; -0.24 ethanol blended into motor gasoline, which is accounted for in both fossil fuels and renewable energy but counted only once in total consumption; and 0.02 electricity net imports.

[l] Primary consumption, electricity retail sales, and electrical system energy losses, which are allocated to the end-use sectors in proportion to each sector's share of total electricity retail sales. See note at end of Section 2.

Notes: • Data are preliminary. • Totals may not equal sum of components due to independent rounding.

Sources: Tables 1.1, 1.2, 1.3, 1.4, and 2.1a.

Fig. 21.8. U.S. energy flow, 2003 (quadrillion Btu). (*Annual Energy Review, U.S. Department of Energy 2003.*)

Energy (see Fig. 21.8). The capital resources are the fossil and nuclear fuels present in the crust of the earth. The fossil fuels were created by the interaction of the sun with the terrestrial flora and fauna. This was a very low-yield process that over the five billion years the earth has existed captured about two days of the solar energy in the form of coal, oil, gas, and other combustibles. The recoverable quantities are difficult to estimate.[12–15]

The nuclear fuels were created in the cosmic event that created the universe and were deposited in the earth as it took form. There are two families of nuclear fuels, those for fission (uranium and thorium) and those for fusion (protium $\left[{}_{1}^{1}H\right]$, deuterium, helium-3, and lithium). Only uranium fission has been developed as a commercial source of nuclear energy. Although fusion has been developed as a military weapon, the hydrogen bomb, it is premature to include the fusion fuels in the world's inventory of capital energy. The technology for controlled fusion is not available, nor is development of a controlled fusion process expected in the next several decades. When available it would increase the capital supply to a level greater than that from all other sources combined.[16]

In 1990, the National Academy of Science completed an energy study that concluded that commercial fusion power may be required by the year 2050. By that time 40 percent of the oil reserves will have been consumed, and current natural gas reserves can supply only about half of the projected needs. The discovery of additional deposits and improved production might meet the need for natural gas, but at a higher price.

Little use was made of the earth's capital energy until the nineteenth century, and now 5 to 8 percent of that resource may have been consumed. Yet even though most of the fossil fuels remain, there are increasing problems with their extraction and distribution and the pollution that is incidental to their use. The demand for energy is increasing as the world continues to industrialize. In 2002, the annual per capita consumption in the United States was 340 million Btus versus 190 million in Russia, 47.7 million in Brazil, and 13.5 million in India. The increasing demand will stem from population growth and increases in the standard of living. It will be accompanied by increased energy use for the extraction of marginal resources and the treatment of wastes before their release into the earth's environment.

Petroleum is and will remain the major source of mobile energy for the next century. However, petroleum production has passed its peak in the United States, and in several decades it may peak in the rest of the world. However, in 2001, approximately 45.8 trillion kW hours of energy were consumed worldwide using oil, and projections indicate that this number jumps to approximately 71–74 trillion kW hours in 2025, indicating a ~58.3% increase in oil consumption. Also in 2001, the U.S. energy supply from petroleum products was 11.2 trillion kW hours, and it is projected that this number will increase to 16.1 trillion kW hours, indicating a 43.8% increase.[8]

Coal can supply U.S. needs for the next several hundred years, but there is increasing concern regarding pollution from both coal and petroleum. Unfortunately for the developed nations, using pollution-free energy sources such as nuclear and solar will do comparatively little to reduce worldwide atmospheric pollution. The sources of such pollution are worldwide. About 6 billion tons of CO_2 was generated in 1985, and this pollution source is expected to increase to 30 billion tons/year by 2060. In the 1980s only 10 percent of the 6 billion tons/year came from the United States. In 2002, this increased to about 5.7 billion tons (Annual Energy Review 2003, DOE) or about 23 percent of the world's total. In 2002, about 210 million tons of avoided carbon emission was achieved by nuclear (140 million) and renewables (mainly hydro).[17]

The benefits of nuclear power obtainable with the present technology depend on the availability of uranium resources. For example, using the present light water reactor technology, each 1000-megawatt reactor requires a few thousand metric tons of uranium during its 40-year operation. Because of limited uranium ore reserves in the United States and the lower cost of uranium from other countries, in the late 1980s about 50 percent of ore consumed in the United States was imported. This number increased to 64 percent in 2003. However, U.S. reserves should increase significantly when exploration again becomes profitable. In addition, the energy extracted from the uranium can be significantly increased by reprocessing the spent fuel. If the recovered mixed uranium and plutonium were used in a heavy water reactor, there might be as much as a 68 percent increase in energy generated. A further increase by a factor of ten might be achieved going to a liquid metal cooled reactor. However, such advanced technology will result in higher electricity cost. In view of the current low cost for natural uranium, storage rather than reprocessing of spent reactor fuel creates a beneficial fission fuel reserve.

NUCLEAR PROCESSES

The science of the nuclear processes has a long history. Democritus in the fifth century B.C. started the search for the elementary particle of nature he called the atom. John Dalton, about 1803, related atomic masses to hydrogen, and Mendeleev in 1869 correlated these masses with the atoms' chemical properties and hypothesized missing elements. Many thought this ended the adventure, but in 1895 Roentgen discovered X-rays, and a year later Becquerel discovered natural radioactivity. A short time thereafter, in 1902, Rutherford and Soddy proposed that radioactivity was related to atomic changes, and in 1905 Einstein equated mass to energy. In 1932 Curie and Joliot discovered artificial radioactivity, and in 1938 Meitner and Frisch recognized that uranium fissioned when bombarded with neutrons. In 1942 Enrico Fermi and Glen Seaborg led development of the first controlled neutron fission reactor to produce plutonium-239.[18]

The nuclear processes of most interest to the nuclear industry are radioactive decay and the transmutation of nuclides. Whereas chemical processes relate to the interactions of orbital electrons of the atom, nuclear processes relate to interactions of neutrons, charged particles, and nuclides with the neutrons and protons in the nucleus of the atom. As noted above, several thousand nuclides and isomers are now known and only 287 of these are naturally occurring. More continue to be found. As Mendeleev invented the chart of the chemical elements, Emilio Segre invented the chart of the nuclides to give order to the nuclear properties and processes.[19]

Radioactive Decay

Radioactive decay occurs when an unstable atomic nucleus emits particles and/or energy to achieve a stable state. The nucleus contains neutrons and protons along with the energy that binds them together. Many reactions are involved in radioactive decay (see Table 21.5). These reactions are characterized by the type, energy, and rate of radiation emitted. A different nuclide, which may also be unstable, results from the decay process. This nuclide then will decay with its own unique radiation. Naturally occurring uranium-238 passes through 12 nuclides before reaching a stable end product, lead-210. The types of radiation include:

- Electrons, called beta particles when negatively charged and positrons when positively charged
- Helium ions, called alpha particles
- Electromagnetic energy, called gamma or X-rays, which accompanies almost all other emissions
- Neutrons
- Nuclide ions from fission and fusion

The energy of these emissions covers a wide range of values but is typically 190 million electron volts (MeV) for fission, 17 MeV for fusion, 5 MeV for alphas, 1 MeV for gammas, and 0.5 MeV for betas. The rate of radioactive decay is expressed through the half-life, the time required for the decay rate of the unstable nuclide to decrease by a factor of two. The half-lives range from less

TABLE 21.5 Radioactive Decay Process Examples[a]

	Nuclide			
Decay Process	*Parent*	*Daughter[b]*	*Half-life[a]*	*Energy (MeV)*
Alpha (α, 4/2He)	Pu-238	U-234	89.6y	5.5
Beta (β^-, °e)	Cu-64[c]	Zn-64(s)	13h	0.57
Positron ($\beta+$, °e)	Cu-64[c]	Ni-64(s)	13h	0.65
Orbital electron Capture (EC, °e)	Cu-64[c]	Ni-64(s)	13h	1.68
Internal Transition (IT)	Tc-99m	Tc-99	6h	0.14
Fission	U-235	FP[d], n	Prompt	~200
	Cf-252	FP,n	2.6y	~200
Fusion	D/T	He-4(s), n	Prompt	17

[a] Thorium-234 also is radioactive, and the decay process continues through ten more radioactive nuclides before reaching stable lead-210. Decay half-life calculated by $\tau_{1/2} = ((-\ell n\, 2\, (t - \mathrm{to}))/((\ell n\, (N\, /\, No))$ where t = time and N = disintegration/sec.

[b] Daughters always include gamma rays.

[c] Cu-64 has three decay processes (β^- 38%, β^+ 19%, and E, C 43%).

[d] Fission products.

(s)Stable nuclides; others are radioactive.

than microseconds to greater than a trillion years.

As radiation is not detected by the human senses, special detection and measurement methods are necessary. The silver halides in photographic film are sensitive to radiation as well as to the electromagnetic energy of visible light; in fact, it was photographic film detection that led to the accidental discovery of radiation. This technique continues to be used, particularly for the study of cosmic irradiation. A closely related method is the use of crystals, for example, sodium iodide and germanium silicide, that emit light when subjected to radiation. This light is measured with photoelectric cells that can be tuned to measure the strength of the radiation. Another popular method measures the discharge of electric condensers in the form of gas-filled ion chambers. The radiation passing through the chamber ionizes the gas and discharges the condenser. Ion chambers are particularly useful for soft alpha and beta radiation measurement. The Geiger counter is an ion chamber with its spontaneous discharge hooked to an audio speaker, whose resulting clicking provides a background noise. All these detection methods have special uses but at the same time a wide range of applications. For each application they are calibrated with radioactive standards that are traceable to national and international reference standards.

Fission

Fission is a relatively simple process. The process is explained in detail very well by Lamarsh.[20] Neutrons at room temperature (termed *thermal*) are captured by a fissile material producing an unstable nuclide which promptly fissions, yielding energy and fission products. (Note that certain nuclei, termed *fissionable*, are caused to fission only by the impingement of an energetic neutron.) The fission products generally include more than two neutrons, and under certain circumstances a fission chain reaction can be sustained. Under special conditions where more than one neutron per fission reaction produces a subsequent fission in the fissile/fissionable material in an uncontrolled manner, a violent explosion can result. However, considerable technical effort is required to hold the material together long enough to obtain a high-yield explosion. In a controlled nuclear fission process, such as in a nuclear power reactor, only one of the neutrons is captured by other fissile/fissionable nuclei. The remaining neutrons are used to produce more fissile/fissionable material, are absorbed in other components, or leak from the reactor.

Uranium-235. Natural uranium contains 0.711 percent uranium-235. The nuclear industry is still completely dependent on this single nuclide, which is the only naturally occurring one that can be effectively fissioned with thermal neutrons. A controlled nuclear fission chain reaction was first achieved on December 2, 1942 under the west stands of the football stadium at the University of Chicago. The Chicago event was achieved in an atomic pile, a stacked array of natural uranium bodies in channels of a large graphite block. The graphite was present to reduce the energy of the neutrons to a value where they are most readily captured in U-235 (See Table 21.6.) Approximately 85 percent of the 200 MeV released in a fission reaction is in the kinetic energy of the fission products and is instantly captured in the uranium in the form of thermal energy. The remaining energy is released in the form of radiation during decay of the radioactive fission products, e.g., strontium-90 and cesium-137. When this radiation is released, it too is largely converted into thermal energy.

The fission neutrons at birth have energies of approximately 1 to 2 MeV. In a thermal reactor the neutron energy is rapidly reduced through collisions with light nuclei to thermal (~.02 to 1 eV), to promote for more efficient capture. Besides the nuclear fuel, there are many other materials in the reactor core also competing for the neutrons, including: the moderator (the material used to slow down or thermalize the neutrons), fertile nuclides that produce additional fissile material (discussed in a later section), neutron poisons present in control rods, the coolant, fuel element cladding, and other structural materials.

TABLE 21.6 The Fission Process

Reference equation:

84% *Fission products* + ~200 MeV

$$ {}^{235}_{92}\mathrm{U} + {}^{1}_{0}n \rightarrow {}^{235}_{92}\mathrm{U} \begin{array}{l} \nearrow \; 16\% \\ \searrow \; \tau_{1/2}\text{~}207y \quad {}^{232}_{90}\mathrm{Th} + {}^{4}_{2}\mathrm{He} + 4.5\,\mathrm{MeV} \end{array} $$

Fission Cross Sections[a] vs. Neutron Energy:

Energy (keV)	*Thermal[b]*
U-235	585
Pu-239	750
U-233	531

Thermal Neutron Capture Cross Sections:

Fissionable Nuclides (includes fission)		*Moderating Materials*	
U-235	684	H_2O	0.33
Pu-239	1021	D_2O	0.0006
U-233	577	Helium	0.007
Fertile Nuclides[(c)]		Carbon	0.004
U-238	2.7	Sodium	0.01[d]
Th-232	7.4	*Neutron Poisons*	
Fuel Cladding		Boron	760
Zircaloy	0.2	Cadmium	3,300
Stainless steel	3	Gadolinium	46,000

---

[a] Cross-section in barns (10^{-24} cm^2).

[b] Thermal energy at 20°C, 2200 m/sec, 0.0253 eV.

[c] Capture neutrons to produce fissile nuclides

[d] At 100–1000 keV.

Neutron poisons, which are nuclides with very high cross-sections for the capture of neutrons but which do not produce any neutrons, are an essential part of the nuclear reactor core. By their addition and withdrawal, the nuclear chain reaction is controlled with a multiplication factor of one, that is, one fission per fission in the previous generation. To achieve high fuel burnup (utilization) in a pressurized light water reactor, boron (a poison) is present in the coolant (and in some cases on the fuel pellets) during the early part of each operating cycle. This serves to remove the excess neutrons (and therefore the excess reactivity of the fuel); subsequently, the boron is removed as the fuel is consumed and poisons from the fission products accumulate. This is the primary means of controlling the neutron multiplication throughout the operating cycle. Cadmium and/or boron is placed in control rods for fine tuning such as load-follow operation and reactor shutdown. Gadolinium and boron, which have very high cross-sections for neutron absorption, find use in emergency shutdown of heavy water reactors and as a volumetrically dispersed component within the fuel pellets of some light water reactors. Table 21.6 shows cross-sections at thermal energy for the principal fissile materials.

Plutonium-239. Plutonium-239 represents a fortuitous phenomenon. Whereas U-235 is the only significant fissile nuclide in nature, its major isotope, U-238, captures a neutron to produce another fissile nuclide, plutonium-239. A substantial amount of the energy produced during the life of uranium fuel is produced by the conversion of U-238 to Pu-239 which subsequently fissions. This process provides the basis for the nuclear breeding cycle.

Uranium-233. A second fissionable isotope uranium-233, can be produced from naturally occurring thorium. It does not present an economically attractive option at present because of its dependence on highly enriched U-235 to bring the thorium cycle into operation and the large R&D expenditures required to develop the technology (for more information see Reference 21).

Fusion

At present the only application for fusion is in thermonuclear military explosives, where the necessary temperature and pressure are achieved with a plutonium fission initiator for a very short time. The search for the science and technology needed to make fusion a viable source of energy for commercial electric power generation is the basis for one of the world's most important and challenging R&D efforts. The development of a controlled nuclear fusion reactor could provide a virtually unlimited source of energy. Like fission, fusion eliminates the atmospheric pollution associated with the use of fossil fuels. Also, it could significantly reduce the waste disposal problem of energy-producing processes.[22,23]

In the Tokamak fusion reactor depicted in Fig. 21.9, electric current to the poloidal coils on the primary magnetic transformer generates the axial current in the secondary plasma composed of deuterium and tritium ions. These ions are heated to ignition temperature and then the reaction becomes self-sustaining. The toroidal field coil suspends the plasma away from the metal conducting walls. Contact with the wall would both cool the plasma below ignition temperature and contaminate the plasma with heavy ions. The relevant reactions are given below.

Deuterium-Tritium Fusion.

1. ${}^{2}_{1}\mathrm{H} + {}^{3}_{1}\mathrm{H} \rightarrow {}^{4}_{2}\mathrm{He} + {}^{1}_{0}n + 17.6\ \mathrm{MeV}$

It should be noted that this reaction is also used to provide rugged, durable commercial sources of 14 MeV neutrons for oil-well logging and scientific research. Similarly, deuterium–deuterium fusion provides sources of 2.5 MeV neutrons.

Tritium production is accomplished in a fusion reactor blanket (or a fission reactor, as is now done to produce tritium for the weapons program).

2. ${}^{6}_{3}\mathrm{Li} + {}^{1}_{0}n \rightarrow {}^{3}_{1}\mathrm{H} + {}^{4}_{2}\mathrm{He}$

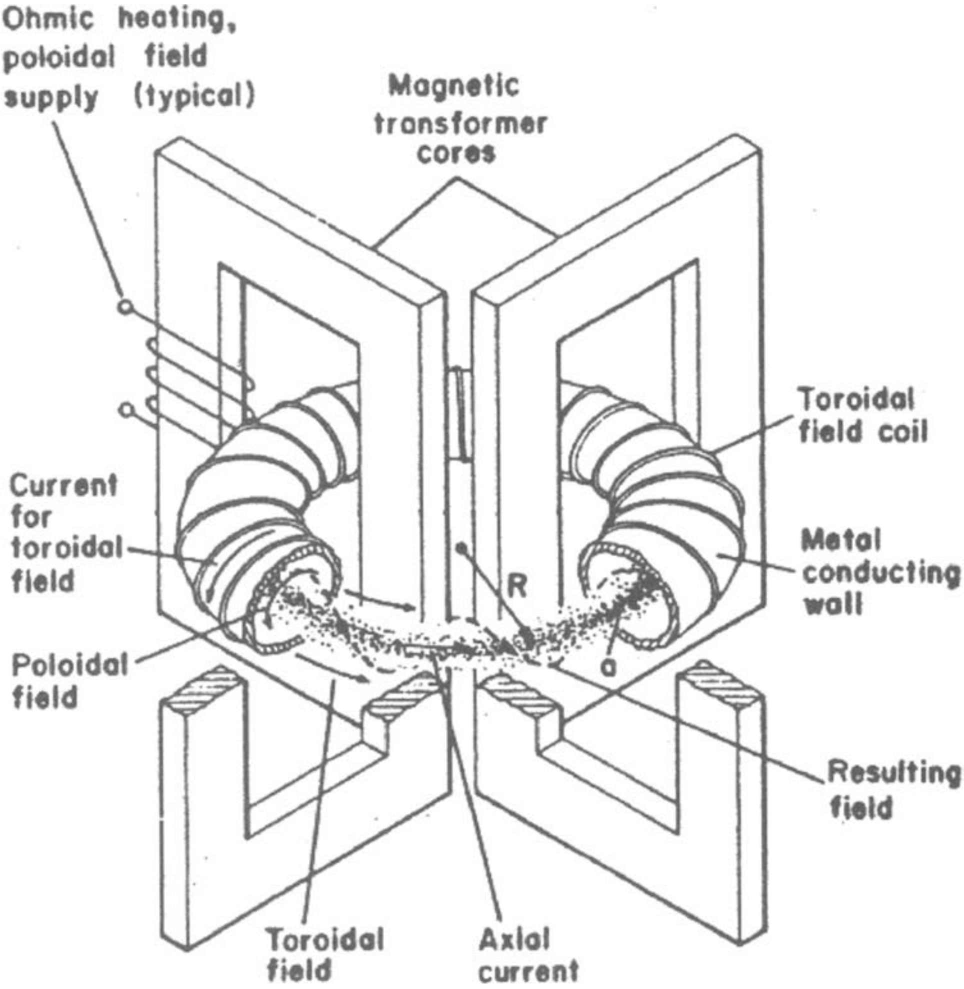

Fig. 21.9. Tokamak fusion reactor. *(Courtesy U.S. Department of Energy.)*

3. ${}^{7}_{3}\mathrm{Li} + {}^{1}_{0}n \rightarrow {}^{3}_{1}\mathrm{H} + {}^{4}_{2}\mathrm{He} + {}^{1}_{0}n$

Protium Lithium fusion would produce charged particles (90% of the energy in helium ions) for direct conversion to electricity, but higher temperatures and pressure would need to be achieved.

4. ${}^{1}_{1}\mathrm{H} + {}^{6}_{3}\mathrm{Li} \rightarrow {}^{3}_{2}\mathrm{He} + {}^{4}_{2}\mathrm{He} + 4\ \mathrm{MeV}$

5. ${}^{1}_{1}\mathrm{H} + {}^{7}_{3}\mathrm{Li} \rightarrow {}^{4}_{2}\mathrm{He} + {}^{4}_{2}\mathrm{He} + 17\ \mathrm{MeV}$

Of the several fusion reactions, deuterium tritium fusion is the most feasible, as it has the lowest ignition temperature, 40 million°K. (See Reaction 1.) Deuterium comprises 0.15 percent of naturally occurring hydrogen, whereas tritium is produced by neutron fission of lithium-6 that is irradiated in a blanket surrounding a nuclear reactor core or by the absorption of a neutron by deuterium, for example, in a heavy water reactor. Nuclide separation is required to produce the deuterium and possibly the lithium. The tritium would be produced in a surrounding lithium blanket of the fusion reactor to sustain the process (See Reactions 2 and 3). The tritium for hydrogen bombs is produced in nuclear fission reactors using these processes. The alternative fusion reactions use protium and lithium and yield charged helium ions (See

Reactions 4 and 5). These would contain 90 percent of the fusion energy and could be directly converted to electricity, but higher temperatures and pressures would be required.

There are now two approaches to developing a fusion reactor, magnetic containment and inertial confinement. The major effort, magnetic containment, uses the Tokamak concept to compress and heat the reactants in a plasma isolated in a vacuum away from the walls of the reactor. The fuel is injected as pellets and heated with electric ohmic energy to strive for the ignition temperature. Research along this line started in the early 1950s, and in 1968 the Russians discovered the Tokamak principles. (Tokamak is the Russian acronym for "toroidal chamber with magnetic coil.") Massive amounts of electrical energy are required to heat and compress the plasma. Ohmic energy now is supplemented with induction, microwave, and neutral beams techniques. The time of heating has been extended from microseconds to over a minute using these additional methods.

There are two immediate objectives in fusion research, first to achieve ignition and then to achieve parity (produce more energy than is required to maintain steady-state operation). In the 1990s the International Energy Agency (IEA) member countries spent ~\$8.9 billion on fusion R&D. Today (2005), the majority of fusion research is carried out by the European Union (EU), the United States, Russia, and Japan. The Tokamak is still the most promising reactor design to produce fusion energy. The two largest Tokamaks that are being studied are the Joint European Torus (JET) located in the United Kingdom and the Tokamak Fusion Test Reactor (TFTR) located at Princeton, New Jersey, United States. JET was first to demonstrate breakeven (output power = input power) in 1997, although ignition had not been achieved as of 2003. However, it was decided by the Soviet Union in the mid 1980s that a next generation Tokamak was needed. In 1992 the Soviet Union, Europe, Japan, and the United States agreed to collectively design an International Thermonuclear Experimental Reactor (ITER). Although the United States pulled out of the collaborative group in the late 1990s, it rejoined in 2003. The cost of ITER is thought to range from \$5–8 billion, and its construction would take 10 years. When ignition and parity are finally achieved and understood, spending will need to be substantially increased to establish commercial feasibility. It is now realized that electricity from fusion will be more costly than that from fossil and fission processes. It is only as these resources are depleted that fusion will become economically feasible.

Nuclide Production

Nuclides are produced by capturing a nuclear particle in the nucleus of the target atom, which thus is transmuted to a different atom, one that in most cases is unstable and decays, as discussed in an earlier section.[24]

The target atom may be any stable or radioactive atom. In a process termed irradiation, it is bombarded, leading to production of many of the nuclear particles discussed in the previous section. Most often the incident particle is a neutron produced in a nuclear reactor, but frequently it is a charged particle raised to the required energy in an electromagnetic accelerator. The charged particles often are protons but may be deuterons, helium nuclei (He-4 or He-3), or heavier ions.[25,26]

The rate of nuclide production is dependent on the number and the energy of particles bombarding the target, the cross section of the target and the half-life of the product (see Table 21.7). The decay of these products can be a source of significant amounts of energy, as already discussed.

Fission Products

The fission process produces radioactive as well as stable nuclides with masses ranging from 72 to 167 and with two broad peaks in the regions of 95 and 138. The masses are identified rather than the specific nuclides because in fission many short-lived nuclides are produced that quickly decay by beta

TABLE 21.7 Nuclide Production Process

$$A = N\sigma\phi(1 - e^{-\lambda t})$$

Where:

$(1 - e^{-\lambda}t)$ = Saturation factor: for small values of λ, it equals λt; for $t = \tau_{1/2}$, it equals 0.5, the optimum irradiation time.
A = Disintegrations per second; 1 Curie = 3.7 E10 d/s.
N = Number of atoms; Avogrado's number = 6.023E23 atoms/gram atomic wt.
σ = Sorption cross-section; 1 barn = 10^{-24} cm^2.
Φ = Irradiation flux, particles/sec cm^2; for electromagnetic accelerators reported in microamperes.
λ = Decay constant, natural logarithm 2/half-life.
t = Irradiation Time, Seconds
$\tau_{\frac{1}{2}}$ = Nuclide half-life.

Examples:

1. Co-60 production (Co-59 target at 1E14 sec^{-1} cm^{-2}, σ = 37 barn)

Irradiation Time (y)	*Curies Co-60/Gram Target*[a]
1.1	130
2.2	225
5.5	370

2. Pu-239 production (U-238 target at 1E14 sec^{-1} cm^{-2}, σ = 2.7 barn):

	Grams Pu-239/Metric Ton Target
1	4,800
2	9,600
3	14,400

---

[a] These quantities ignore depletion of the products through decay and conversion to other products.

emission to a long-lived or stable member of the mass chain.[19,20]

The yields of selected mass chains that result from fission of U-235 and Pu-239 are shown in Table 21.8. In addition to the fission products with masses roughly half that of U-235, neutrons, tritium, helium, and beryllium are products of ternary fission. There are significant differences in some of the yields from U-235 and Pu-239.

Although the fission products could be recovered as byproducts from the waste from spent nuclear reactor fuel, special-purpose neutron irradiation of highly enriched uranium (isotopically separated uranium-235) followed by chemical separation is the normal production method. The major products, molybdenum-99 and iodine-131 with fission yields of 6.1 and 6.7 percent, respectively, have important medical applications. Mo-99, with a half-life of 2.7 days, is particularly interesting because its short-lived decay product, technetium-99m with a half-life of 6 hours, is the product actually used. To make this short-lived product available, the Mo-99 is sorbed in a silica gel column, and the Tc-99m is eluted as needed.

During the 1980s, large-scale processing of the defense wastes at Hanford extracted and encapsulated 100 megacuries of strontium-90 and cesium-137 to reduce heat generation in the waste storage tanks. Krypton-85 has been recovered from the airborne effluents at the Idaho processing plant.

In addition, development studies have addressed the recovery of stable rhodium, xenon, technetium, and palladium because of their limited availability in nature. For palladium this would amount to about 40 kg per year from a 1000 MWe reactor. However, a

TABLE 21.8 Major Fission Products

Mass	*Long-Lived Nuclides*	*State*	*Half-Life*	*Percent Yield*[a]	
				U-235	*Pu-239*
72	Zn		46.5 hrs	0.001	0.001
79	Se	Meta Stable	3.92 min	0.044	0.044
		Ground	6.5 E4–6.5 E5 yrs		
85	Kr	Meta Stable	4.48 hrs	1.3	0.58
		Ground	10.76 yrs		
90[b]	Sr		28.78 yrs	5.8	2
99	Mo		2.7476 days	6.1	6.2
102	Pd(s)		1.02% (a/o) abundance	7.7[c]	28.1
103	Rh(s)		100% (a/o) abundance	3	7
124	Xe(s)		0.096% (a/o) abundance	23.7	27.2
131	I		8.0207 days	2.9	3.9
133	I		21 hrs	6.7	7.0
133	Xe	Meta Stable	2.19 days		
		Ground	5.243 days		
137	Cs		30.07 yrs	6.7	6.6
140	Ba, La		1.678 days	6.2	5.4
167	Dy		6.2 min	0.001	0.001

[a] Because two fission products are emitted for every binary fission, the cumulative yield for the mass chains is 200 percent.

[b] Mass-90 beta decay chain (half-life): Br(2 s), Kr(32 s), Rb(4 m), Sr(29 y), Y(2.6 d), Zn(stable).

[c] This is equivalent to 1.2 kg of palladium/metric ton of spent fuel burned to 32,000 MWd. One megawatt-day of thermal energy from fission is approximately equivalent to one gram of fission products.

(s) Stable products of the mass chains. Multiple chains contribute to the high yields for palladium and xenon.

small amount of palladium-107 (half-life 6E6y) is present that could complicate its use. This is also a problem for technetium, which is 100 percent technetium-99 with a half-life of 2E5y.

The mass-135 provides an example of a fission product chain that must specifically be considered in reactor operation. Iodine-135 with a 6.6 hr half-life decays to 9.1 hr xenon-135, which has a high cross-section for thermal neutrons. When a reactor is shut down, the xenon-135 grows in from its I-135 parent, because it is not being eliminated by neutron irradiation. For high-flux reactors this could lead to a "dead-time" of tens of hours during which the reactor cannot be restarted. These effects are controlled through the adjustment of fixed and dissolved neutron poisons during power changing maneuvers.

Neutron Transmutation Products

The production of neutron transmutation products involves the neutron irradiation of target materials followed by chemical separation. Although this promises high purity with high specific activity, the isotopic composition of the target along with other competing reactions frequently yields a spectrum of isotopes of the desired nuclide.

Plutonium-239 and tritium for use as military explosives are the two major transmutation products. The nuclear process for Pu-239 production is the same as for energy generation, but there are some differences: (a) metallic natural uranium clad with aluminum facilitates later dissolution for plutonium recovery, and the reactor operates at a relatively low temperature because of the aluminum clad and better heat transfer (due to the metallic natural uranium); (b) the irradiation cycle is limited to a few months to minimize the Pu-239 conversion to Pu-240 and Pu-241; and (c) a carbon or a heavy water moderator is used to increase the neutron efficiency.

More complex is the production of Pu-238 (used for isotopic heat sources) and californium-252 (used in research as a source of fission neutrons). For Pu-238 there are two irradiation cycles: the first starts with natural uranium but maximizes the neptunium-237 yield; then the Np-237 is separated, converted into aluminum-clad oxide targets, and irradiated to Pu-238 in a second cycle. The total process requires double neutron capture.

Californium-252 production was especially challenging, as it involved the sequential capture of 14 neutrons along with the intermediate separation and fabrication of two intermediate targets (americium and curium isotopes) when starting with Pu-239.[27] This production campaign lasted ten years, produced about 10 g of Cf-252, and then was terminated. The product was evaluated as a neutron source but had insufficient value to justify continuing production.

Neutron Activation Products

The neutron activation production cycle requires only neutron irradiation without chemical separation. The target and the product are the same chemical element but have different nuclide compositions. The specific radioactivity of the product is a function of the nuclide composition of the target, the neutron flux environment, the irradiation time, and the half-life of the product nuclide, along with the nuclear cross-sections of target and product nuclides.

The major neutron activation product is cobalt-60 with a reasonably long half-life (5.3 years). It is produced by capture of a thermal neutron by cobalt-59, the only naturally occurring isotope of that element. Cobalt-60 emits high-energy beta particles and gamma rays appropriate for radiographic, irradiation, and isotopic power applications. It is an ideal product because naturally occurring cobalt is a relatively stable metal that is uniquely monoisotopic cobalt-59. Furthermore, the target nuclear cross-section is good, and the product cross-section is lower than the target's by a factor of ten. Another factor in cobalt-60's long list of advantages is that the Canadian CANDU family of nuclear power reactors are ideal for production of Co-60.

TABLE 21.9 Major Isotopes and Their Uses

Isotope	*Use*
D	1. D2O moderator for HWR 2. Fuel in fusion reactions
Li-6	1. Source for tritium 2. Fuel in fusion reactions
Li-7	1. Water-cooled reactors use lithium-7 hydroxide as a water conditioner 2. Possible coolant in LMFBR
F-18	Used in nuclear medicine, especially in labeling D-glucose
N-13	Tests done show that when used in ammonia, helps to determine global myocardial blood flow
C-14	Used in carbon dating
N-15 C-13 O-17 O-18	1. Used as tracer in living beings 2. Using in NMR
B-10	1. Neutron absorber in nuclear reactors 2. Used in the medical field in neutron-capture therapy
B-11	Can be used in tritium production and in NMR
Na-24	Used to locate pipe leaks
Mg-27	Used to locate pipe leaks
K-42	Can measure exchanged potassium in blood
Cr-51	Used to label red blood cells
Fe-57	Used as a spin isotope in chemistry
Fe-59	1. Used in blood studies 2. Can be used to determine friction in machinery when used with steel
Co-60	Used in cancer treatment
Ga-67	Used to locate tumors
Kr-81	Used in lung ventilation studies
Tc-99	Used as a tracer in locating brain tumors
I-131	Used as a tracer when studying the thyroid gland
Yb-169	Used during brain scans
U-235	Fuel for most nuclear reactors
Pu-239	Used in nuclear weapons, fast breeder reactors, and MOX fuel reactors
Am-241	Used in smoke detectors

Other important activation products include molybdenum-99 and iridium-192. However, the Mo-99 is better obtained from fission, and already has been discussed in that section. The 74-day Ir-192 has a gamma with less energy than Co-60 and is used for radiography of less dense materials.

Charged Particle Transmutation Products

Many radionuclides can be produced in cyclotrons, thus avoiding the use of more costly nuclear reactors. Many research hospitals now have cyclotrons to provide short-lived radionuclides of carbon, nitrogen, oxygen, and fluorine. The longer-lived products are produced commercially or in government laboratories.[25,26,28]A list of major isotopes and their uses is shown in Table 21.9,

These radionuclides are produced by irradiating targets with beams of hydrogen ions (protons), but frequently deuterium ions (deuterons) are used (see Table 21.10). Some products require beams of helium-4 and helium-3 ions. The typical process involves the capture of the proton with the prompt emission of a neutron. This is called a p,n reaction. However, in other cases there may be protons, alpha particles, or up to five neutrons emitted. The resulting products decay generally by positively charged electron (positron) emission, but also decay by capture of an orbital electron.

To produce the beam of high-energy charged particles, a stream of hydrogen or helium is electrically ionized and accelerated in an oscillating magnetic field that holds the particles in a spiral path within the cyclotron. At the desired energy the particle beam is magnetically deflected to the target. The beam energy can vary from 4 to 800 MeV but is usually less than 30 MeV. The beam current ranges from 3 to 2500 microamperes. The target is designed to accommodate the ten to several hundred watts of thermal energy deposited by the beam. Gas and liquid targets are used for the short-lived products to facilitate processing for use. For the long-lived products that require longer irradiation and higher energy flux (up to a kilowatt per square centimeter), the target material may be plated on a water-cooled heat exchanger. The target materials in many cases are separated stable nuclides.

The Department of Energy's laboratories have accelerators that provide up to 800 MeV proton beams. Other machines support high-energy physics research but occasionally are used to produce special nuclides such as xenon-127.

REACTOR MATERIALS PROCESSING

Reactor materials processing is concerned with preparation of the special materials used in

TABLE 21.10 Nuclide Production by Charged Particles

Typical reaction:

$$ {}^{14}_{7}\mathrm{N} + {}^{1}_{1}\mathrm{H} \rightarrow {}^{15}_{8}\mathrm{O} \rightarrow {}^{4}_{2}\mathrm{He} + {}^{11}_{6}\mathrm{C} \xrightarrow{20\mathrm{m}} \boldsymbol{\beta}^{+} \rightarrow {}^{11}_{5}\mathrm{B} \quad \textbf{(stable)} $$

This reaction normally is reported as N-14(p,α)C-11. Natural nitrogen gas is bombarded in a cyclotron with a 30-microampre current of 4 to 13 MeV protons (p). O-15 is produced, but an alpha particle is promptly ejected, producing C-11, the desired product. The yield for a 20-minute bombardment is 0.2Ci.

$$ {}^{58}_{28}\mathrm{Ni} + {}^{1}_{1}\mathrm{H} \rightarrow {}^{59}_{29}\mathrm{Cu} - 2{}^{1}_{1}\mathrm{H} + {}^{57}_{27}\mathrm{Co} \xrightarrow[270d]{EC} {}^{57}_{26}\mathrm{Fe} \quad \textbf{(stable)} $$

Ni-58[p,2p]Co-57. Enriched Ni-58 (enriched in the Oak Ridge Calutron) is plated on a copper heat exchanger, bombarded for 5 hours with 1000 microamperes of 20 MeV protons. Cu-59 is produced but 2 protons are promptly ejected producing Co-57, the desired product. The yield for a 14 hr bombardment is approximately 28 microcuries per microampere hour.

Other (p,n) Processes: Li-7 to Be-7; Fe-56 to Co-56; Cu-65 to Zn-65; Ag-109 to Cd-109.

Other Processes and Products: N-14(d,n)O-15; O-16(p, α) N-13; Ne-20(d, α)F-18.

β^{+} represents a positron.

EC represents electron capture.

nuclear reactors. This includes the nuclear reactor fuels along with the reactor vessels, plumbing, heat exchangers, coolants, and moderators. All the materials that go into the nuclear reactors are deliberately and thoroughly controlled by standards established by the nuclear industry with the participation of the Nuclear Regulatory Commission and administered by the American National Standards Institute. These materials include the uranium oxide pellets, zirconium cladding, and zirconium or steel fittings of the fuel assemblies, the corrosion– and radiation-resistant steel used for the reactor vessels, and the various corrosion– and erosion-resistant steels for piping, valves, pumps, and heat exchangers. In addition there are the coolants (water, sodium, and helium), the moderators (water, deuterium, and graphite), and the neutron sorbers (boron, erbium, cadmium, hafnium, and gadolinium). Each of these materials requires intensive processing to achieve safe and reliable operation of the nuclear reactors.

Isotope Enrichment

Isotope enrichment is a major activity of the nuclear industry. The naturally occurring chemical elements are composed of isotopes with varying nuclear properties. By enriching the chemical element in the nuclide with the desirable nuclear property, the performance of the nuclear processes is enhanced. In addition, the sensitivity of isotopic analysis permits the use of enriched chemical elements in place of radioisotopes as tracers in research, medicine, and industry.

The separation of chemical isotopes is based on small differences in their physical and chemical properties. For the lower-mass isotopes, chemical exchange, distillation, and electrolysis have been used. For the higher-mass isotopes, techniques based on mass have been used, including gaseous diffusion, centrifugation, thermal diffusion, and ion activation.[29,30] A newer method uses lasers that produce coherent light tuned to the specific wavelength of a vibration bond related to the desired isotope in an atom or molecule. This technique is still under development but promises much higher per stage separation factors resulting in lower energy consumption and capital costs. The main issue with this technology is the availability of low capital and operating cost lasers with sufficient wavelength selectivity.

The most universal of these techniques uses the Calutron, an electromagnetic accelerator employing the first arc of a cyclotron (Fig. 21.10). In 1945 it provided the enriched U-235 that was used in an early atomic bomb. Since then it has been used to enrich small quantities of most of the stable isotopes. The separation is achieved by ionizing the feed material and then accelerating the ions through the field of a bending magnet. The differences in mass causes the heavier elements to pass through a larger arc with the ions trapped in carbon pockets located 180° from the feed point. The accelerating voltage and magnetic field are controlled to optimize performance. Separation factors of 10 to 100 are obtained, but the yields are only 1 to 10 percent.

Large-scale production has been applied to enrich U-235, deuterium, and lithium-6. The original gaseous diffusion technology is now being replaced by high-speed centrifuges as the principal technology for U-235 and is discussed further as part of the fuel cycle. Dual-temperature chemical exchange processes are used to enrich hydrogen and lithium isotopes.

Several methods can be used to obtain high-purity deuterium for use as a coolant in heavy water reactors. Natural hydrogen contains 0.16 percent deuterium and is enriched to 10 to 20 percent deuterium in pairs of two hundred plate columns.[31] The chemical exchange system is hydrogen sulfide gas and water. One column operates at 27°C and the second at 220°C, with the enriched product removed from the bottom of the cold column that flows to the second set of columns. From the second set it goes to distillation and then to electrolysis to yield 99.8 percent deuterium. The specific enrichments at the crossovers between processes are controlled by energy considerations. Other methods in use are the chemical exchange reaction between liquid ammonia and hydrogen and nitrogen gas

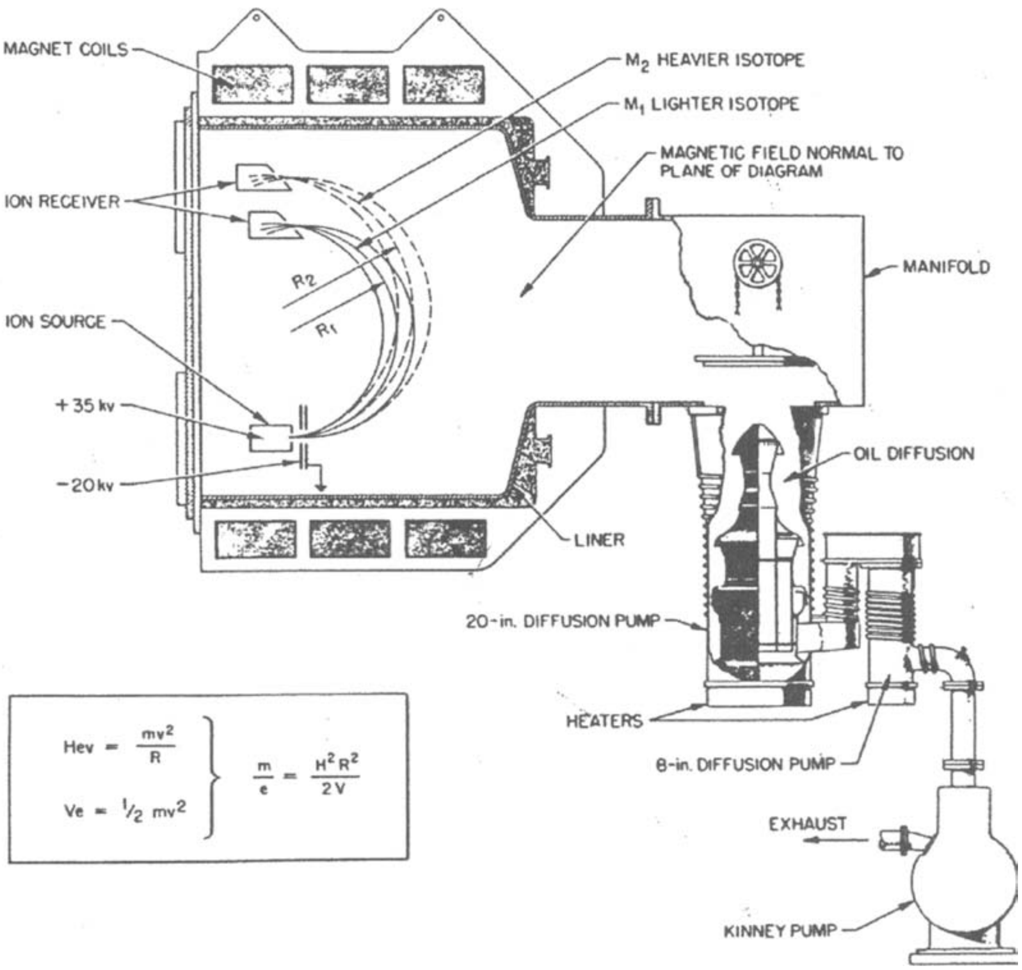

Fig. 21.10. Oak Ridge Calutron for separation of isotopes. The first method to achieve large-scale separation of U-235 and today the source of research quantities of most stable and a few radioactive nuclides. *(Courtesy USDOE.)*

using potassium amide as a catalyst. At least two plants are in commercial operation: one in Argentina and one in India.

The chemical exchange system employed for lithium-6 enrichment is lithium amalgam and aqueous lithium hydroxide. It also employs paired dual-temperature columns.

Cryogenic distillation is used to enrich carbon-13, nitrogen-15, oxygen-17, and oxygen-18. Typical of these processes is carbon monoxide distillation, which has a C-13/C-12 separation factor of 1.008 between the vapor and the liquid. The initial 20-meter packed column is tapered from 2.5 cm at the boiler to 10 cm at the reflux condenser. This brings the carbon-13 from 1.1 to 12 percent. In a second column it is brought to 93 percent.

Zirconium Production

Nuclear-grade zirconium alloys are used for the manufacture of nuclear fuel rod tubes and structural components. Processing begins with zircon sand ($ZrSiO_4$) obtained from Australia, India, or a variety of locations (see Figure 21.11). Zircon sand is co-milled with coke, and injected into a large fluidized bed reactor (crude chlorination). The reactor is heated to approximately 1000°C by induction heating of the graphite liner, and the bed is

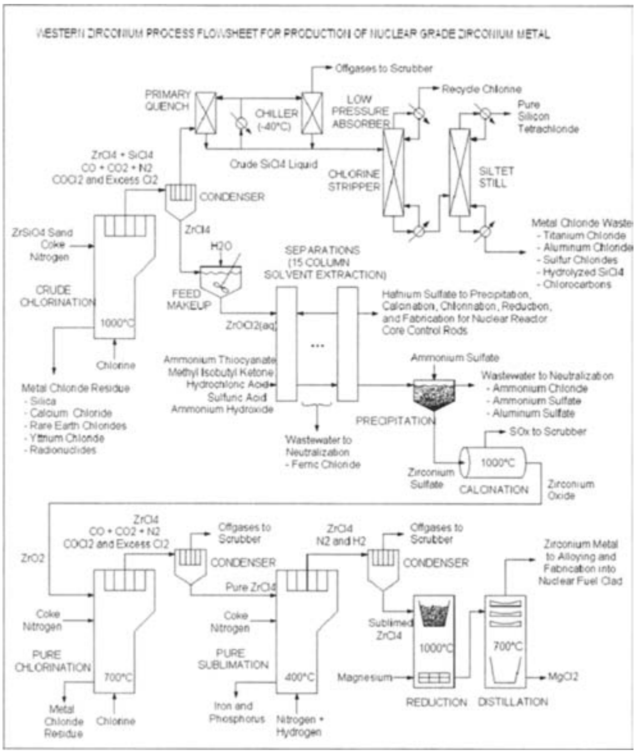

Fig. 21.11. Schematic of Western zirconium plant in Ogden, Utah.

fluidized with chlorine gas. Zircon is converted to zirconium tetrachloride by the following reactions.

$$\mathrm{ZrSiO_4 + 4\,Cl_2 + 4\,C \rightarrow ZrCl4_4 + SiCl_4 + 4\,CO\,;}$$

$$\mathrm{ZrSiO_4 + 4\,Cl_2 + 2\,C \rightarrow ZrCl4_4 + SiCl_4 + 2\,CO_2\,.}$$

The process gases pass through two stages of condensation, separating low-volatility $ZrCl_4$ product from the remaining offgases. Metal chloride impurities that escape from the chlorinator also condense with the zirconium product. The offgases pass through several stages of cryogenic quenching with liquid $SiCl_4$, recovering $SiCl_4$, Cl_2, and $COCl_2$. Distillation operations then separate silicon tetrachloride from chlorine and phosgene. The latter two species are recycled back to the crude chlorinator, and $SiCl_4$ is purified further by activated charcoal absorption, and sold as a valuable byproduct. Remaining offgases are scrubbed with aqueous NaOH to remove traces of chlorine and phosgene, and are then vented; salt solutions resulting from scrubbing operations are sent to evaporation ponds.

The zirconium tetrachloride product must then be purified before reduction to metal. In particular, hafnium must be removed to less than 100 ppm Hf:Zr because of the high neutron absorption cross-section it exhibits, and phosphorus and aluminum must be removed to even lower specifications due to their deleterious metallurgical impact on the final zirconium alloys. The tetrachloride product is first dissolved in water under carefully controlled conditions to produce an acidic $ZrOCl_2$ solution. This solution is complexed with ammonium thiocyanate, and contacted with methylisobutyl ketone (MIBK) solvent in a series of solvent extraction columns. Advantage is taken of the relative solubilities of Zr, Hf, and Fe thiocyanate complexes to accomplish a high degree of separation of hafnium and iron from the zirconium.

Purified zirconyl chloride solution is then reacted sequentially with surfuric acid and ammonium hydroxide to precipitate a complex zirconium oxysulfate. The precipitate is washed, filtered, and stripped to remove traces of MIBK, and then calcined to drive off sulfur and convert the product to ZrO_2. The precipitation process leaves behind most of the aluminum and phosphorus. The hafnium stream leaving solvent extraction is treated similarly, producing a HfO_2 byproduct.

Zirconium dioxide is then carbochlorinated a second time (pure chlorination) and converted once again to $ZrCl_4$. This operation is very similar to crude chlorination, but takes place at lower temperature due to more favorable thermodynamics. The purified $ZrCl_4$ product is then reduced to zirconium metal by direct reaction with molten magnesium metal, according to

$$ZrCl_4 + 2\ Mg \rightarrow Zr + 2\ MgCl_2\ .$$

This reaction (known as Kroll Reduction) is carried out in sealed vessels at approximately 1000°C, producing a mixture of $MgCl_2$ and a porous zirconium metal morphology known as sponge. Because of the large density difference, the molten $MgCl_2$ floats above the sponge; most of it is recovered by mechanical separation, and sold as a high-purity byproduct.

The purified sponge is then crushed, combined with alloying elements, pressed into large cylindrical ingots, and electron-beam-welded into a solid cylinder. This cylinder is then remelted by vacuum arc melting to obtain the proper metallurgical properties. All of these operations must be done with great care to avoid introduction of oxides and nitrides into the final metal product, both of which will result in embrittlement. From here, the ingot is heated, forged, and formed into either flat plate or heavy walled tubing known as Trex. The Trex is then extruded and pilgered into the final nuclear fuel to form, and the plate is formed into other structural internal components. A variety of different zirconium alloys are produced, having specific applications for different reactor designs.

THE URANIUM FUEL CYCLE

The uranium fuel cycle for the uranium–plutonium system is a multicomponent system of chemical process operations that begins with mining uranium ore from the earth as the starting material and ends with the radioactive waste products in above-ground or below-ground terminal repositories. Some radioactivity is released in the air and water discharged to the environment at concentrations and quantities below those specified by federal regulations. The topics to be considered in this section are mining, milling the ore, uranium fuel preparation, and reprocessing.[32,33] The following section discusses radioactive waste management.

A similar set of processes has been partially developed for the thorium–uranium system but is not discussed here because it is not expected to be employed in the next several decades. The important feature of the thorium cycle is that it could be used to achieve breeding (to produce more fissionable material than is consumed) in thermal reactors, but nuclear as well as chemical factors have frustrated this development (for more information, see Reference 22). The increasing cost of the natural uranium supply for the uranium/plutonium cycle may, several decades in the future, justify development of the thorium cycle.

It should be noted that breeders would not reduce the demand for uranium ore for many decades because several LWR and/or HWR converters (which produce fissionable material, but less than consumption) are required during the run-in of a breeder cycle to equilibrium. The doubling time of a breeder (the time required for the breeder to produce sufficient fissionable material to start up a second breeder reactor) might be a significant part of its operating life. Furthermore, natural uranium will be required for the thorium cycle, if it is used, and for startup of the fusion cycle. The tritium for the fusion cycle will be made in nuclear reactors, as it now is for nuclear weapons. The nuclear industry will always be dependent on a continuing supply of uranium from ore.

Mining

Uranium mines are primarily the open pit type, but there is significant production from deep mines as well as from solution mining. Sometimes uranium is produced as a byproduct of mining operations for vanadium, phosphate, and gold.

Where uranium ore deposits occur in permeable aquifers with low-permeability geologic formations above and below them, the uranium is extracted by circulating a carbonate leaching solution. The leach solutions are sulfuric acid, ammonium carbonate, or sodium carbonate–bicarbonate along with air, peroxide, or sodium chlorate as the oxidizing agent. Holes are drilled to the bottom of the ore body on a grid pattern, 50 to 200 feet apart. The holes are cased with screened sections through the ore body. Varying combinations of injection and withdrawal of the leaching agents are applied to recover the uranium. The uranium is recovered from the leaching agent by ion exchange. The leachate is adjusted and recycled.

Milling

In milling, the uranium is leached from the ore, separated from major contaminants, and converted to yellow cake [or sodium diuranate ($Na_2U_2O_7$)_]. The uranium ores milled in the United States generally contain 0.05 to 0.2 percent U_3O_8, with an average of 0.1 percent. To save transport costs, the mills are located near the mines. There are two major processes, one based on a carbonate leachant and the other on sulfuric acid (see Fig. 21.12.)

Carbonate Process. In this process the ores are leached with hot sodium carbonate for 24 hours, with sparging with air to provide oxidation. The leachate is cooled in countercurrent heat exchangers, heating the carbonate solution for the next batch. The carbonate leachate is filtered on rotary drums, and the uranium is precipitated with sodium hydroxide and filtered. The filtrate is converted back to carbonate by sparging with carbon dioxide, usually from a boiler flue gas, and recycled. The dried precipitate in the form of sodium diuranate is about 90 percent U_3O_8. Carbonate leaching is fairly specific for uranium, and the product is fairly low in contaminants.

Sulfuric Acid Process. In the sulfuric process, the pulverized rock is leached for about 8 hours with sodium chlorate or manganese dioxide added for oxidation. The leachate passes through a series of thickeners countercurrent to the flow of the wash water. The liquid (leachate) is then separated from the solids using filtration. The acid leachate is not very specific for uranium and therefore goes through ion exchange or solvent extraction for purification. The solvent is a hydrocarbon-diluted 2-diethyl-hexyl phosphoric acid applicable to the recovery of both vanadium and uranium when both are present in the ore. A variation is the RIP (resin-in-pulp) process. After thickening, to avoid the filtration step, the slime-containing leachate goes into tanks where screen baskets of anion exchange resins, mechanically sloshed up and down, absorb the uranium. After loading, the uranium is stripped from the resin with an acidified chloride or nitrate solution. These product streams are precipitated with sodium hydroxide and the resulting sodium diuranate is dried. An additional cycle of ion exchange may be required to achieve the desired product quality.

Fuel Preparation

There are two general processing routes for making feed for the nuclear fuel manufacturing plants. The primary route is through solvent extraction for purification, denitration to UO_3, and reduction with hydrogen to UO_2, followed by either hydrofluorination to UF_4 and reduction to metal, or fluorination to UF_6 followed by isotopic separation and reduction to UO_2. The other route starts with higher-grade yellow cake, skips solvent extraction, and goes directly to hydrofluorination, with distillation of the final hexafluoride added for purification.

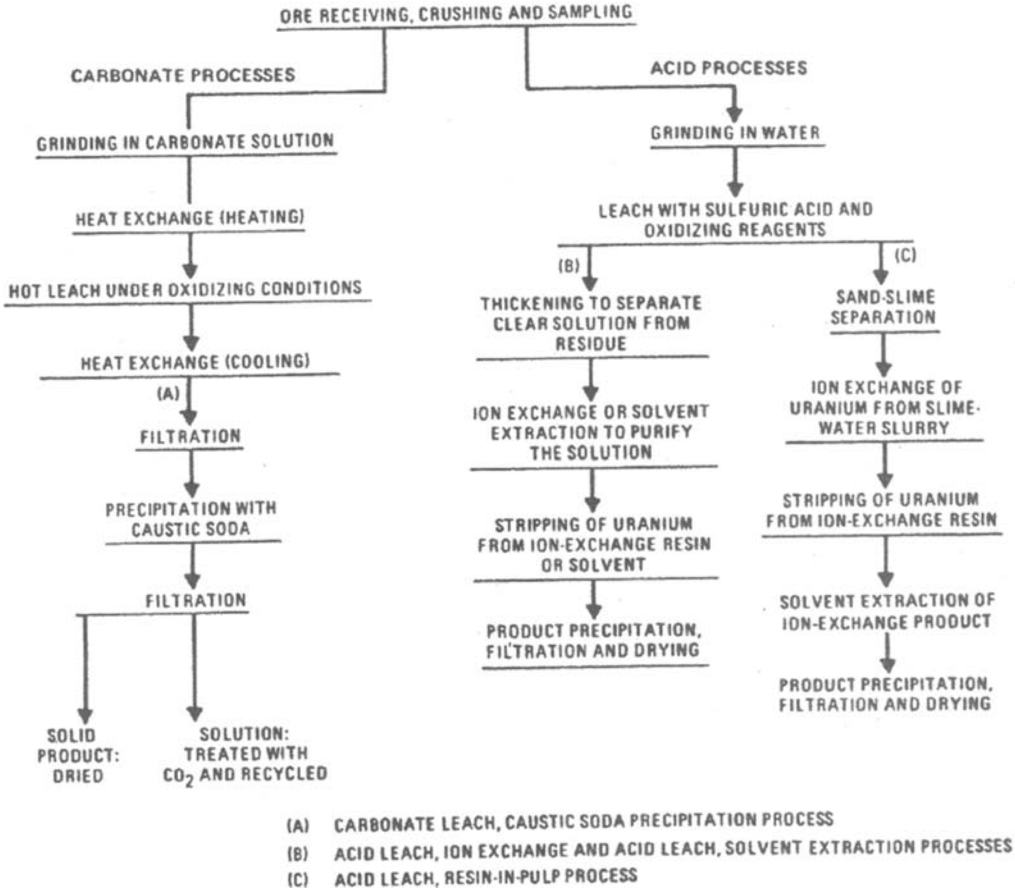

Fig. 21.12. Uranium ore mill processes: (A) carbonate leach, caustic precipitation process; (B) acid leach, ion exchange and acid leach, solvent extraction processes; (C) acid leach, resin-in-pulp process.

Uranium fuel preparation takes the UF_6 and is converted to either (a) aluminum-clad uranium metal for the weapons plutonium production reactors or (b) to Zirconium-clad UO_2 for electricity production in the light and heavy water power reactor (see Fig. 21.13).

Solvent Extraction. The yellow cake is dissolved in nitric acid and extracted from this aqueous phase by 5 percent tributyl phosphate (TBP) in a hydrocarbon diluent. The diluent reduces the density and viscosity of the TBP, enhancing the aqueous/solvent phase separation. The extraction is very specific for uranium, with separation factors of 10^3 to 10^5. The product thus obtained is an aqueous uranyl nitrate solution (Fig. 21.14).

Denitration. The uranyl nitrate solution from solvent extraction is converted to UO_3 by evaporating the solution to a final boiling point of 120 to 140°C, followed by calcination at 620°C. The product characteristics are dependent on the type of calciner (pot, trough, or fluid bed) and significantly affect the subsequent steps (see Fig. 21.15).

Conversion to Uranium Hexafluoride. The calcined UO_3 is reduced to UO_2 with hydrogen, then converted first to UF_4 with hydrogen fluoride and finally to UF_6 using fluorine. These steps usually are carried out in fluidized-bed reactors, but the conversion to UF_4 sometimes is done in a stirred trough reactor.[34]

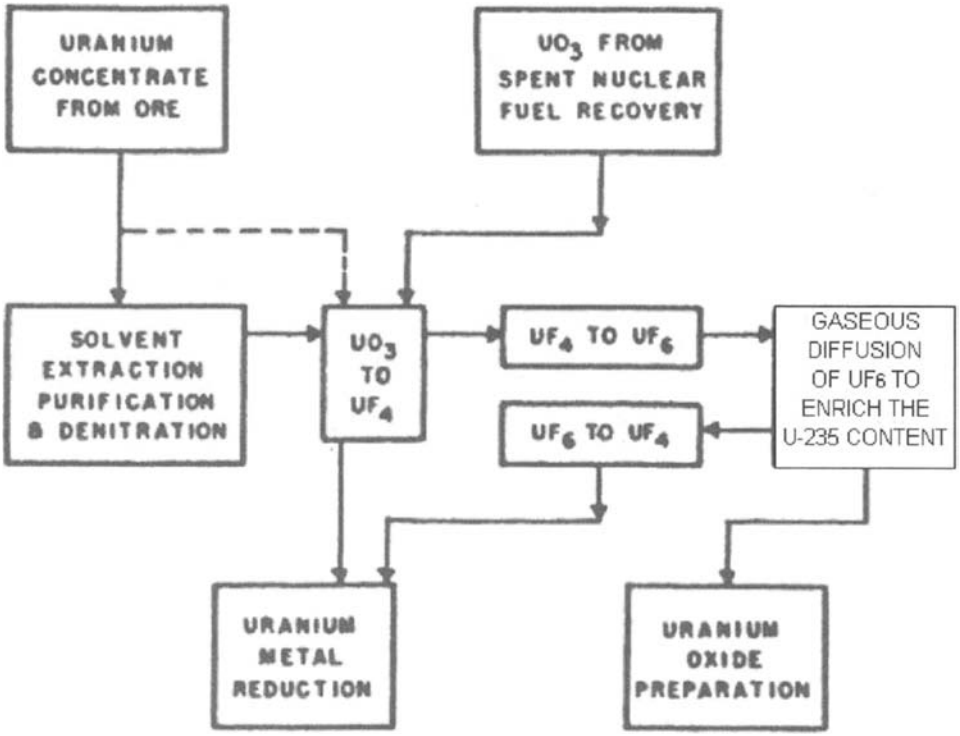

Fig. 21.13. Uranium feed materials flow sheet.

The fluid-bed process at the Honeywell Metropolis Plant uses a series of three sets of fluidized beds (see Fig. 21.16). The first bed is fluidized with hydrogen and reduces the UO_3 to granular UO_2. Then it is hydrofluorinated to a granular UF_4 in a two-stage fluid bed to achieve efficient consumption of the hydrofluoric acid. Finally, in two parallel fluidized bed reactors, the granular UF_4 burns in a fluorine atmosphere to UF_6 vapor. The fluorine is fed in through the ash at the bottom of the reactor to maximize the uranium yield, and then exits through the UF_4 screw feeder to minimize the loss of the high-cost fluorine.

UF_6 Purification. Distillation of the UF_6 is required for purification when the yellow cake does not go through solvent extraction. At Metropolis, the molybdenum, vanadium, and other impurities are removed in a pair of columns, one a 120-foot, 100-plate column operating at 200°F and 85 psia to remove the high volatiles and the second a 45-plate column operating at 240°F and 95 psia to remove the low volatiles. The UF_6 is condensed into 10-ton (48b) cylinders and delivered to the enrichment plant.

Reduction to Metal. Uranium metal is produced by bomb (high-temperature chemical reactor) reduction of UF_4 with magnesium metal. In this process, granular UF_4 is blended with magnesium metal pellets and tamped into a steel reactor lined with the reaction byproduct, magnesium fluoride (see Fig. 21.17). After the container is capped, it is placed in a furnace where the temperature is raised to the ignition temperature at which the magnesium and UF_4 react spontaneously. The reaction mass reaches a temperature sufficiently high for the liquid uranium metal to form a puddle within the bomb. This is a very empirical technology, dependent on the control of many physical factors. Failure to maintain control of the process parameters will

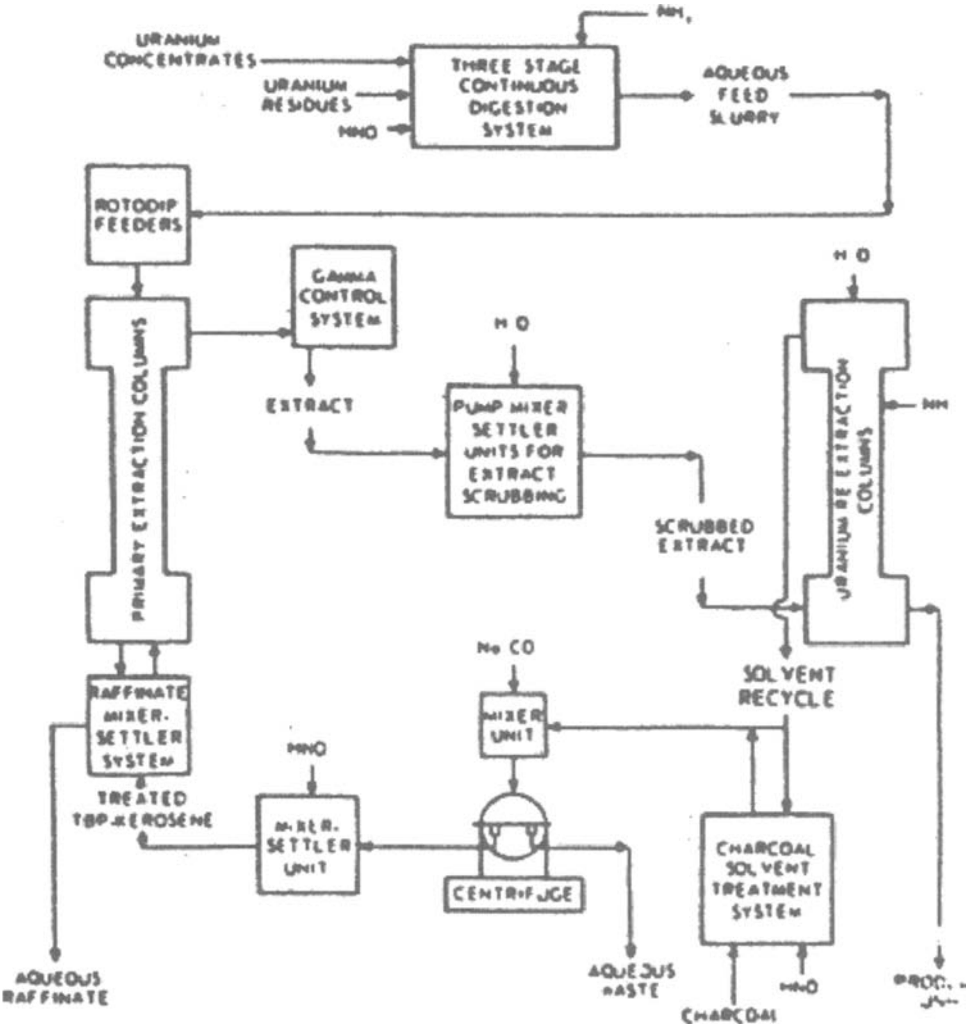

Fig. 21.14. Uranium solvent extraction process for purification of ore concentrate and scrap; slurry feed eliminates clarification cost and losses. *(Courtesy USDOE.)*

produce dispersed uranium shot instead of the desired regulus.

The resulting 350-lb uranium regulus, called the derby, is broken out, remelted in a vacuum furnace, and held at 1454°C to volatilize and remove the impurities. It then is recast in graphite molds to produce the ingot. This is formed into 1- to 2-inch-diameter rods by extrusion and rolling-mill operations followed by machining and cladding with aluminum. Its primary use is for plutonium production; however, some of the depleted metal is used for shipping-cask shields, military projectiles, and counterweights. The magnesium fluoride byproduct is ground and screened to provide material for lining the metal reduction bombs. Excess MgF_2 is disposed of as a low-level waste.

Uranium Enrichment. Enrichment of uranium-235, from 0.711 percent as present in natural uranium, is essential to the economical operation of light water reactors where the fuel life is a function of the enrichment. With approximately 4.95 percent U-235 fuel, the pressurized light water reactors

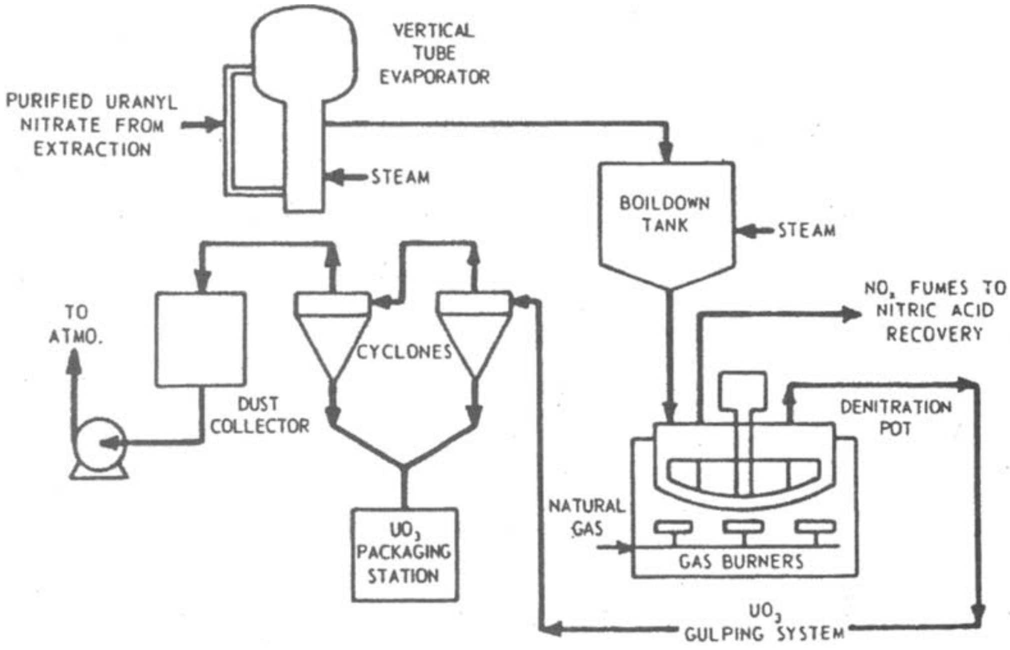

Fig. 21.15. Uranium denigration process prepares UO_3 for conversion to metal and UF_6. A continuous fluid-bed process has been developed. *(Courtesy USDOE.)*

produce an average of about 55,000 thermal megawatt-days of energy/metric ton of uranium during the 4.5 years the fuel is in the reactors. In the naval submarine and ship reactors using highly enriched U-235, the fuel life exceeds ten years. During the original development, gaseous diffusion was selected over electromagnetic separation and thermal diffusion to separate uranium isotopes. Enrichment represents about 15 to 25 percent of the production cost (fuel plus operations and maintenance cost) of nuclear-generated electricity. Gas centrifugal separators are now used in newer enrichment plants. Two other technologies are now in various stages of development that might reduce this cost by a factor of four; they are based on jet nozzles and lasers. The relative capabilities of the different methods are shown in Table 21.11. Separation in the ultra-high-speed centrifuge depends on isotopic mass difference, as does the jet nozzle.[35] The last-named device is a pneumatic cyclone with a high-velocity gas flow induced by a high differential pressure.

Laser-induced separation is being developed, based on differential activation energies of the uranium hexafluorides. This latest enrichment technology uses laser irradiation of the hexafluoride vapor to selectively decompose the U-235F_6 and precipitate U-235F_5. This process promises to recover much of the U-235 that remains in the tails from gaseous diffusion plants. Although this process is not yet commercial (as of June 2006), it is very advanced in its development stage. The only process that is currently developing this technique is the SILEX process in Australia.

TABLE 21.11 Capacities of Major Enrichment Plants (million SWU/year)

	Gaseous Diffusion	*Gas Centrifuges*
USA	11.3	—
France	10.8	—
Russia	—	15–20
Japan	—	1.1
Urenco	—	5.9–6.6
China	—	0.4–1.9

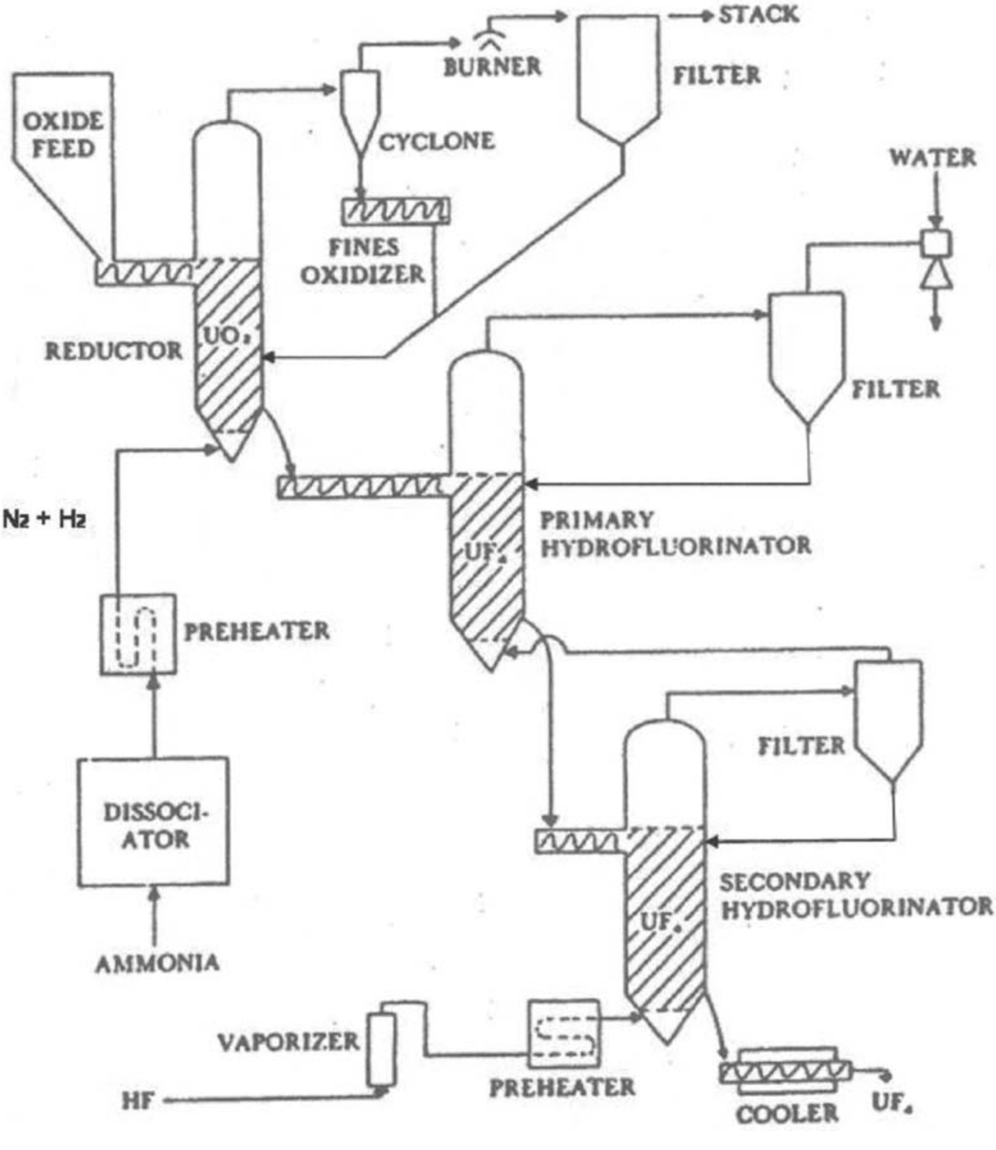

Fig. 21.16. Fluid-bed system for UO$_2$ conversion to UF$_4$. *(Metropolis process.)*

Gaseous Diffusion. In the gaseous diffusion process, the UF$_6$ flows through a porous nickel membrane called the barrier. The heavier U-238F$_6$ flows more slowly than the U-235F$_6$, and the theoretical separation factor for an equilibrium stage is:

$$\alpha = \sqrt{\frac{^{238}\mathrm{UF}_6(352)}{^{235}\mathrm{UF}_6(349)}} = 1.0043$$

The optimum conditions require elevated temperature and reduced pressure with a pressure differential across the barrier sufficient to transport half the UF$_6$ flowing through that stage. However, by varying this flow ratio, the stages are reduced in size as the enrichment of the U-235 is increased. This practice, called tapering, is very important because a large number of stages are required. In the largest stages, axial flow compressors driven by electric motors rated to 3300 horsepower transport the UF$_6$, and 640,000 kg of this material is circulated to produce one separation work unit (SWU).

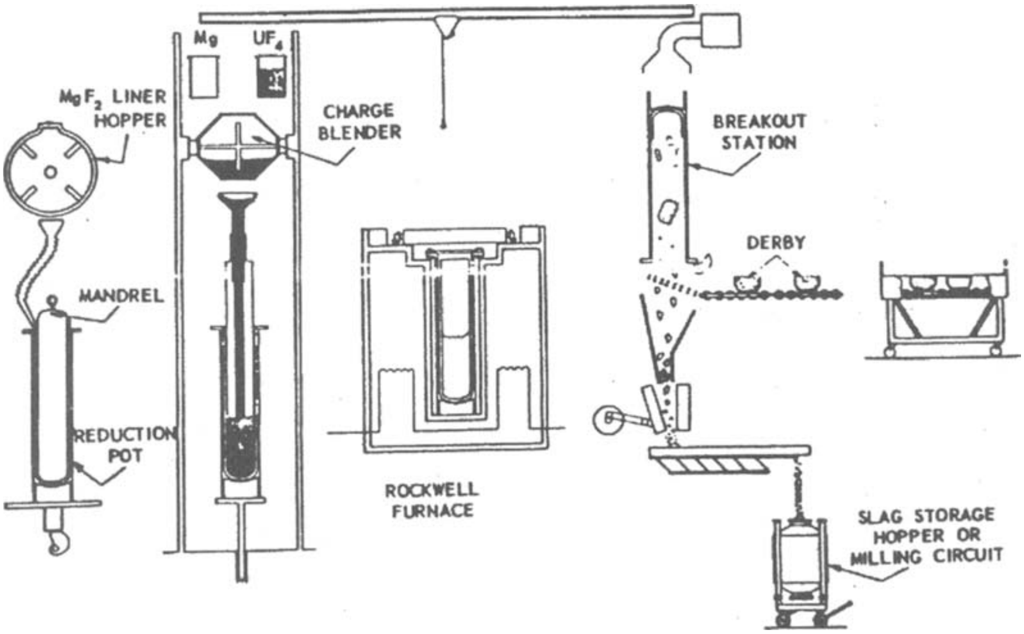

Fig. 21.17. Uranium metal reduction process; a similar process is used for plutonium metal production. *(Courtesy USDOE.)*

When it was operating at full capacity, about 150 metric tons of UF_6 was fed to the plant daily in the process described here. Tapering reduces the time required for the plant to come to equilibrium.

The Department of Energy's gaseous diffusion plant in 1980 had 10,812 stages, consumed 6000 MW of electric power and 1350 million gallons of water a day, and took months to come to equilibrium. The building housing the plant had a combined floor area of a square mile. Plans to further expand the plant using gas centrifuges were scrapped in the 1980s when the expected growth of nuclear power was not realized. In addition, other countries and companies, including Russia and URENCO (which services such countries as the United Kingdom, Holland, and Germany), now offer such services.[36]

The capacity of processes employing diffusion and mass separation is reported on the basis of the work required, and is expressed as separation work units. (The SWU is a measure of the work required to separate uranium of a given U-235 content into two components, one having a higher and the other a lower U-235 content.) It takes 5.3 SWU to enrich natural uranium to 3.44 percent U-235 (see Table 21.12). The Department of Energy facility has the capacity of 11.3 million SWU, and there is a world capacity of about 48 million SWU (Table 21.11.)

TABLE 21.12 Separation Work Unit Requirements (SWU/kg product)[a]

Percent U-235	*SWU*
0.5	0.17[b]
0.6	0.11[b]
0.711	0.00
1.0	0.4
2.6	3.4
3.4	5.3
4.0	6.5
5.0	8.9
20.0	45.7
98.0	270

[a] Tails 0.2 percent U-235.
[b] Added for depleted fuel. Reference cost $112/SWU-kg.

Uranium Hexafluoride Conversion to Oxide. There are several processes used commercially to make nuclear fuel. A diagram of the conversion and fuel manufacturing process is shown in Figure 21.18. The oldest is called the ADU process. In this process, the UF_6 is hydrolyzed in an aqueous ammonia solution to precipitate ammonium diuranate and then calcined in a hydrogen and steam atmosphere to UO_2. Another process is called the AUC process. In this process, UF_6 is injected into a solution of ammonium carbonate to form an uranyl tricarbonate precipitate. This precipitate is then calcined in a fluidized bed using steam and hydrogen to UO_2 powder. In both the ADU and AUC processes, a liquid waste of ammonium fluoride is produced that is treated using lime to produce a calcium fluoride waste and recycle ammonia. Disposal of the calcium fluoride is sometimes difficult depending on how contaminated with uranium it is. In order to get around producing a solid waste that is hard to decontaminate, the IDR process was developed. In this process, UF_6 and steam are combined in a nozzle to form UO_2F_2 powder. This UO_2F_2 powder then drops into a calciner where it is calcined to UO_2. The advantage of this process is that the waste stream is only HF which can be disposed of (or sold) much more readily than the wastes from either the AUC or ADU processes. This is because the HF can be easily decontaminated (separated from any residual uranium).

The UO_2 powder produced from these conversion processes is then mixed with U_3O_8 recycle material, die lubricants, and other materials and then granulated, pelletized, and fired in a hydrogen furnace to achieve high density (see Fig. 21.18.). Typical densities are >95.5 percent of the theoretical UO_2 density (10.96 grams/cm^3). The pellets are then ground to size in wet centerless griders and inspected for chips, cracks, etc. Rejected pellets and grider sludge are oxidized in air furnaces at about 400°C to U_3O_8 which is then recycled to the pelleting operation.

As with most industrial operations, uranium-bearing wastes as well as excess uranium scrap is produced during the course of normal operations. Because of the value of the uranium in this waste and scrap, it is processed to recover the uranium. The scrap and waste is treated with nitric acid to dissolve the uranium. This uranium nitrate solution is then treated using solvent extraction to recover the uranium from the other waste components. The raffinate from this waste is then neutralized with lime to precipitate any metals and then discharged. The resulting uranyl nitrate is processed in either a ADU or AUC process.

The oxide pellets are then ground to size and loaded into zirconium tubing to form fuel rods. There are approximately 80 miles of these rods in a nuclear power reactor. This zirconium is made by using a Kroll reduction process to recover zirconium metal from zirconium oxide using magnesium metal. The zirconium oxide is made by several routes that include converting zircon sand (a zirconium-silicate mineral) to an intermediate compound that is then treated to remove hafnium. Hafnium is an element that chemically behaves very much like zirconium but has a very high cross-section for neutron capture. Because light water and heavy water reactors must use very low cross-section materials in order to achieve good use of their fuel, this halfnium must be removed. Two methods that are currently used are solvent extraction of the zirconium chloride solution or distillation. This latter method, called the Cezus Process, involves dissolving purified (Zr, HF)Cl_4 in $KAlCl_4$, which is then distilled to separate the zirconium from the halfnium. The zirconium metal is then alloyed using vacuum arc remelting. These resulting ingots are then forged into a TREX (tube reduced extrusion). This TREX is then processed into tubing using a pilgering process, heat treated, pickled, and cleaned.

Spent Fuel Reprocessing

It is expected now (2006) that after burnup in the reactors, these rods will go to final disposal. However, the spent fuel still contains a large percentage of fissionable material (see

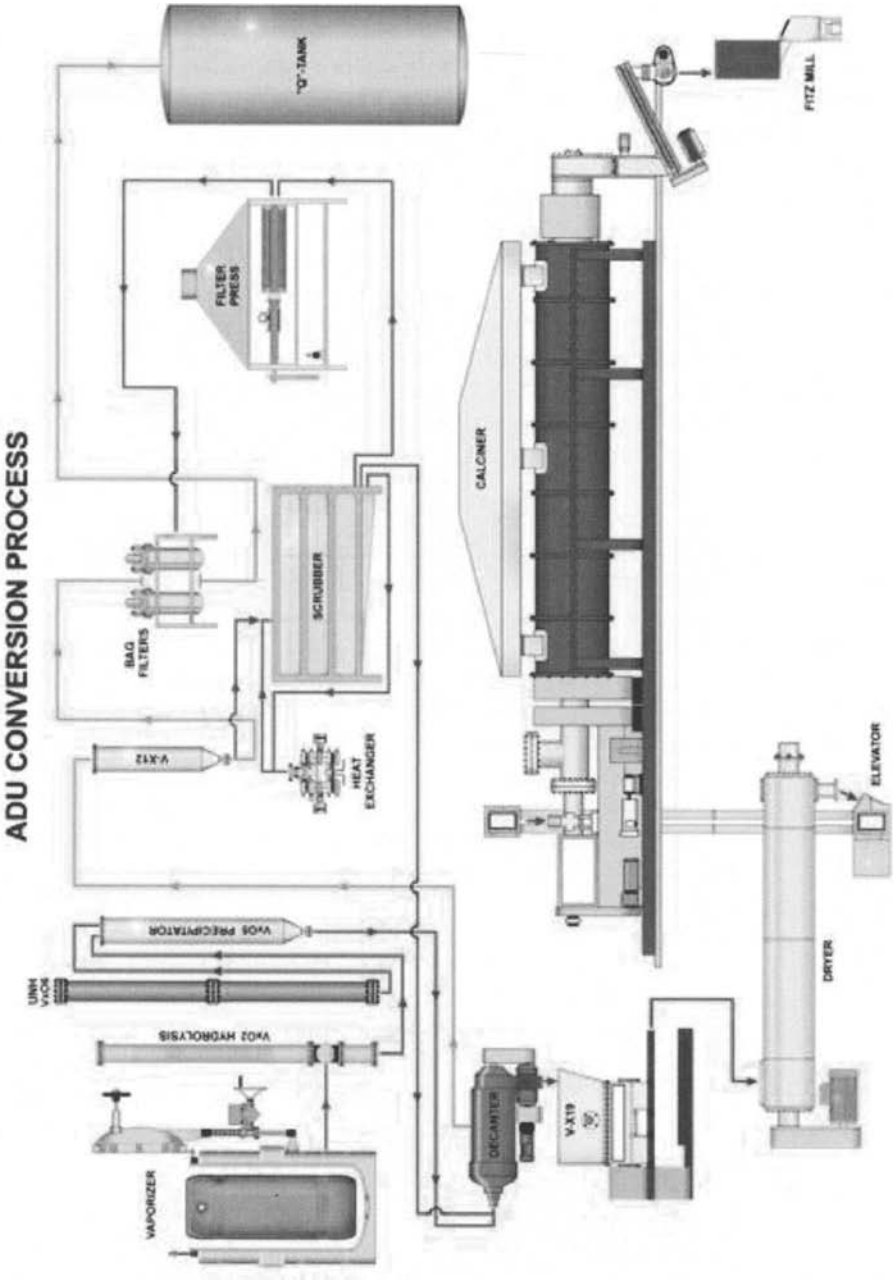

Fig. 21.18. UF_6 conversion to commercial UO_2 nuclear fuel. (Courtesy Westinghouse Electric, LLC, Nuclear Fuels Division.)

TABLE 21.13 PWR and BWR Fuel Burn-Up (late 1980s)

	Fuel Burn-up (Mwd/MT)	*Fuel Composition (g/MT)*		
		U-235	*Pu-239*	*Pu-241*
	PWR–Plutonium Withdrawal			
Charge	—	32,500	0	0
Discharge	32,000	8,359	5,327	1,213
	PWR–Plutonium Recycle			
Charge	—	6,869	19,849	4,082
Discharge	32,000	2,919	8,765	4,647
	BWR–Plutonium Withdrawal			
Charge	—	25,000	0	0
Discharge	27,000	6,403	4,808	1,034

Table 21.13 for an example from a 32,000 MWdays/ton fuel) because the buildup of fission products has effectively poisoned the fuel by capturing too many neutrons. Another option instead of disposal is to reprocess the spent nuclear fuel. Most of the reprocessing to date has been performed for the weapons program. Currently, only in England and France are there plants for commercial reprocessing. Japan is building a plant, but for the present is shipping its spent fuel to England and France for reprocessing. Reprocessing provides more efficient use of natural uranium for both the uranium and the thorium cycles.

High-level waste reprocessing is the most hazardous operation in the nuclear industry. It is there that the largest quantities of fissionable and radioactive nuclides are handled in aqueous solution. These large-scale operations require both remote control and remote maintenance of the plant to protect the workers from radiation. In addition, the air and water effluents along with the solid refuse must be closely monitored to assure that the public is protected. Finally, the fissionable material requires strict accountability to ensure that it is not diverted to unauthorized uses.

As nuclear safety is a foremost public concern, it is well to note that the operation of the plutonium recovery plants since 1945 has met design specifications. There have been no significant health effects on either the workers or the public. The recovery plants were designed with large underground tanks for interim storage of the aqueous waste.. This waste is then processed to an oxide, blended with glass-forming additives, and then melted into a glass mass (log) within a metal canister. The use of glass to encase the high activity waste has been chosen for defense waste in the United States and for defense and commercial waste in the rest of the world. Cement/grout or glass has been picked for stabilizing the mid- and low-activity waste that results from this processing. The canisters containing the high-level waste are then disposed of for the long term (hundreds or thousand to millions of years) deep underground in geologic formations. In the United States, the proposed geologic formation is at Yucca Mountain in Nevada.

The main separation process in spent fuel reprocessing is the Purex process (plutonium-uranium-extraction). Fig. 21.19 shows a generalized flow diagram for nuclear fuel recovery via solvent extraction.[37] The solvent is tributyl phosphate in a hydrocarbon diluent. The process was first used at the Ames Laboratory for uranium purification, then at Oak Ridge National Laboratory for spent fuel. Although other processes were used in earlier days, the Purex process, with various modifications, is now used for many chemical separations in the uranium fuel cycle. It first was used on a large scale to recover uranium from the bismuth phosphate waste generated by the original plutonium recovery operation. This technology was shared with the world at the first Atoms for Peace Conference in Geneva in 1955.

A typical Purex process includes the following.

1. Aging the spent fuel to reduce by radioactive decay the 8-day iodine-131 that would be released to the atmospheric during dissolution
2. Shearing the Zircaloy-clad fuel rods into approximately one-inch lengths so the UO_2 can be dissolved
3. Dissolving the UO_2 in nitric acid
4. Solvent extraction to separate the plutonium, uranium, and fission products

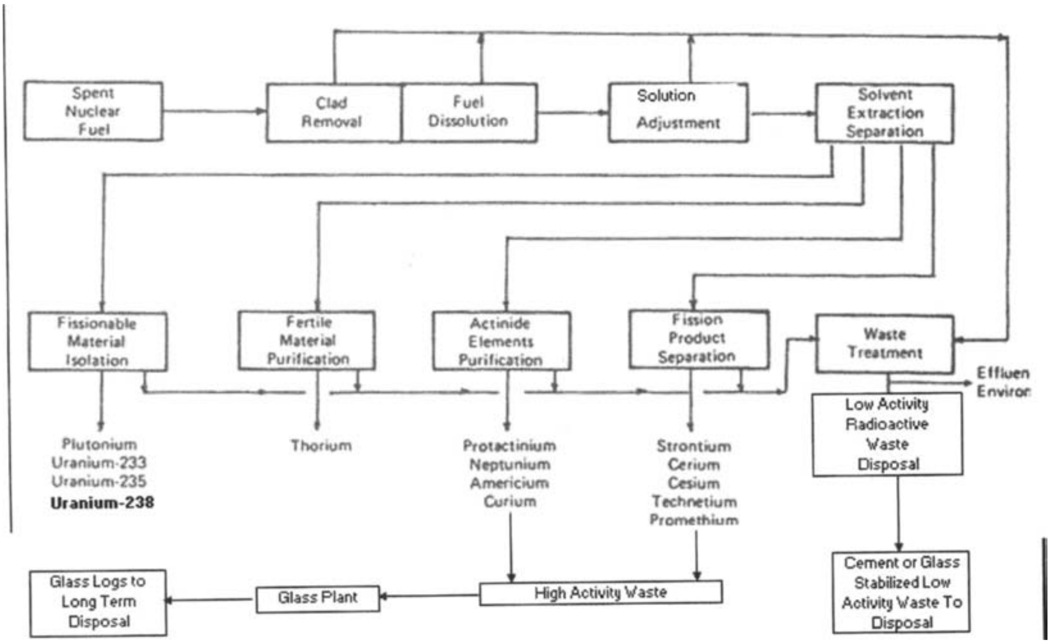

Fig. 21.19. General solvent extraction process for spent fuel recovery.

5. Anion exchange to clean up the plutonium
6. Sorption to clean up the uranium

Modifications to this process can be made to effect recovery of neptunium, americium, curium, californium, strontium, cesium, technetium, and other nuclides. The efficient production of specific transuranic products requires consideration of the irradiation cycle in the reactor and separation of intermediate products for further irradiation.

The facilities and equipment are designed for worker and public protection and for accident avoidance. Also, provision is made to allow remote replacement, decontamination, repair, and disposal. The instrumentation used in the process is designed to measure and report all the independent and dependent parameters essential to controlling and monitoring the operation. Management of the defined standard operating procedures, including quality control and quality assurance, is dictated by international standards and safety regulations.

Dissolution. The spent fuel is dissolved in nitric acid to prepare it for solvent extraction. This is a batch process that is made complex by the fuel cladding and radioactivity. A typical fuel assembly for a PWR contains about 250, 0.37-inch zirconium-clad rods 12-feet long containing in total about 1200 lb of UO_2. The assembled rods are mechanically sheared into 1- to 2-inch lengths and dropped into a perforated basket. This is placed in the dissolver tank where the uranium oxide is dissolved as discussed above. The basket then is removed from the dissolver, and the zirconium hulls and associated hardware therein are dumped into containers, compressed, sealed, and sent to transuranic waste storage. Ultimately, they will go to a disposal facility.

Separation and Purification. In the Purex process discussed here, the uranium, plutonium, and fission products are separated by solvent extraction into three different streams (Fig. 21.20). The plutonium stream goes through anion exchange (discussed later) to reduce traces of ruthenium, and the uranium stream goes through silica gel sorption to reduce traces of zirconium. The fission-product stream, which contains the fission products

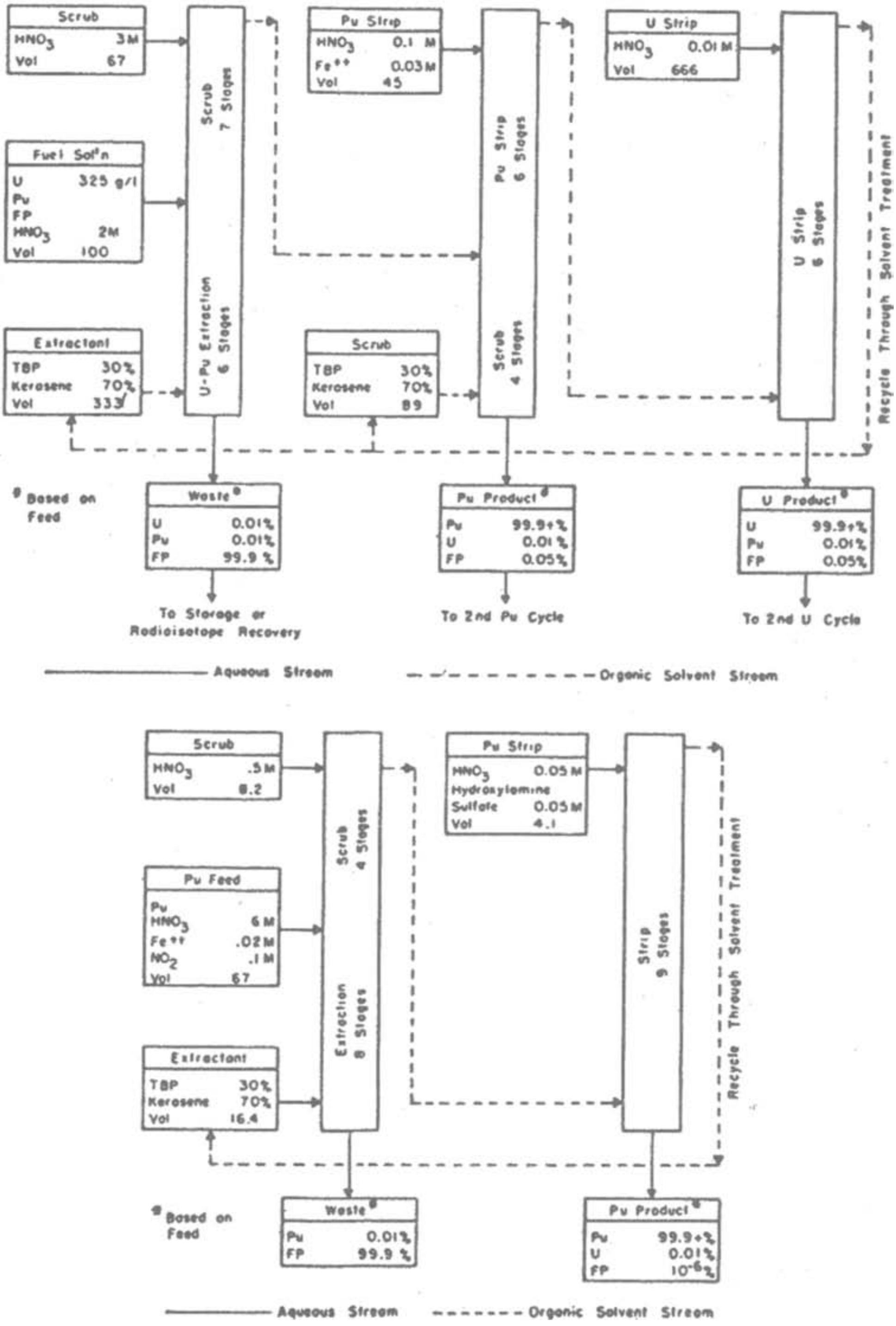

Fig. 21.20. Purex process showing (top) first solvent extraction cycle, and (bottom) second plutonium solvent extraction cycle.

with the transplutonium products, is sent to waste treatment. About 99 percent of the uranium and plutonium is recovered in separate product streams, and decontamination from fission products by a factor greater than 10^7 is effected.[38] The spent fuel solution from the dissolver, adjusted to 2 molar nitric acid, flows into the middle of the first column. From there it flows downward countercurrent to a 5 percent tributyl phosphate/95 percent hydrocarbon solvent, which is introduced into the bottom section. The uranium and plutonium transfer to the solvent, leaving most of the fission products in the acid phase, which pass out the bottom of the column. The U/Pu solvent solution is scrubbed in the top half of the column with 3 molar nitric acid to wash out additional fission products.

The valence of the plutonium in the U/Pu solvent stream is reduced from +6 to +4 with hydrazine to lower its extractability, and the stream flows to the middle of the second column, where the plutonium is stripped out of the solvent with mild acid. The uranium passes on in the solvent stream to the third column, where it is washed from the solvent with a weak acid. The fission product stream flowing from the bottom of the first column, containing less than 1 percent of the U and the Pu, is the high-level waste. It is evaporated to remove most of the acid, and neutralized with sodium hydroxide before going to earth-covered storage tanks in the United States. In Europe, the high-level waste steam is oxidized, mixed with glass formers, and disposed of in glass logs.

The uranium and plutonium streams flow separately through second sets of two columns for re-extraction and stripping for further separation from fission products, according to the scheme shown for Pu in the lower portion of Fig. 21.20. The acid waste streams from the second cycles may be recycled to the first to eliminate the second cycle losses.

Centrifugal contraction or pulsed columns are used for these solvent extraction operations in preference to longer packed columns because the latter would complicate the shielding problem. Mixer-settlers also can be used. The solvent is recycled after treatment to remove decomposition products caused by radiation and chemical effects. This treatment may include a combination of distillation with acid and caustic washes.

From the second cycle the plutonium goes through anion exchange for final purification (Fig. 21.21). The principal problem here is due to ruthenium, which is difficult to remove because of its many valence states. The uranium stream goes through silica sorption primarily to remove zirconium, which seems to be carried along as a colloid.

The process is installed in buildings called canyons, where there are rows of concrete shielded cells serviced by overhead gantry cranes for remote replacement and servicing of valves, pumps, piping, and other equipment. Penetrations through the cell walls are offset to prevent radiation from streaming through. Some equipment is located in cells equipped with windows, TV cameras, and manipulators. Cells for remote repair of the equipment may be similarly equipped.

It is occasionally necessary for personnel to enter some of these facilities. This requires considerable time for decontamination and placement of local shielding. The operations that will be performed are carefully planned and rehearsed in order to get the entry over with as swiftly as possible. A special group of employees, the health physicists, are responsible for radiation safety throughout the plant, and they provide close monitoring of these entries.

Another unique consideration is the prevention of nuclear criticality within the cells. In the dissolver and first cycle, criticality is prevented by the presence of the uranium-238, which absorbs neutrons. Later in the process, the plutonium is separated from the uranium. Criticality is prevented by proper design of the vessels and piping. This includes the cell floor and sumps, where materials would collect in case of leakage from the equipment. To prevent criticality, the vessels are limited in either diameter or thickness. Vessels and piping are placed in arrangements designed to avoid a critical array.

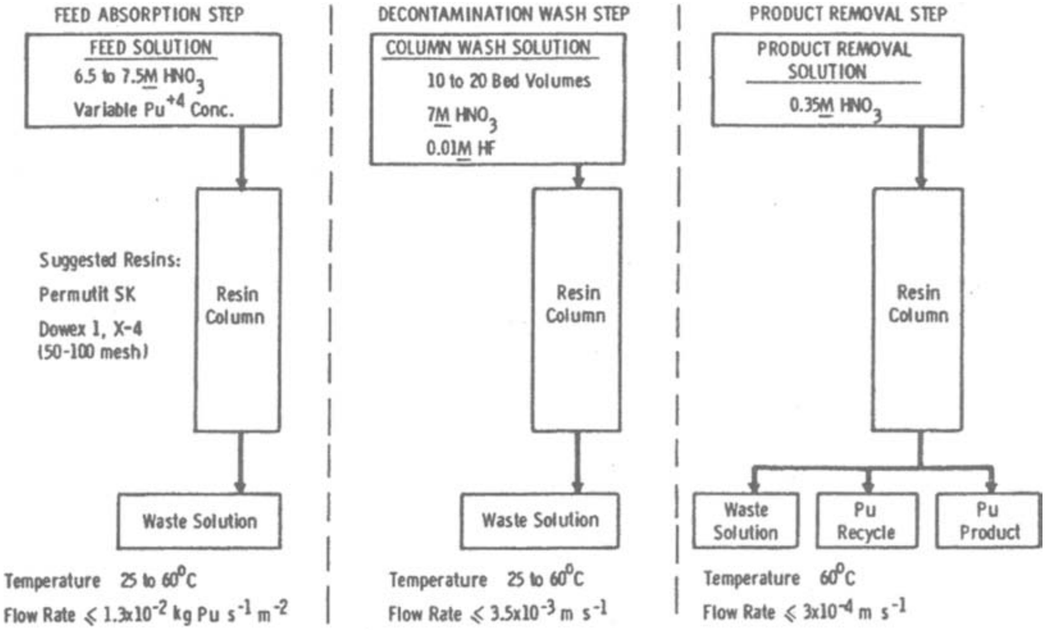

Fig. 21.21. Plutonium ion exchange flow sheet.

In one modification of the Purex process, the plutonium is not separated from the uranium. In this version, the first cycle has only two columns instead of three. In addition to reducing the criticality risk, this modification reduces the risk of unauthorized diversion of the plutonium.

RADIOACTIVE WASTE MANAGEMENT

Radioactive waste management involves the treatment, storage, and disposal of liquid, airborne, and solid effluents from the nuclear industry's operations, along with those from other activities that employ the radioactive products. Its strategy involves four approaches: limit generation, delay and decay, concentrate and contain, and dilute and disperse. Combinations of all four of these usually are employed to manage each waste stream.[39]

There are three types of nuclear wastes, based on their radionuclide characteristics:

- Uranium-contaminated waste, principally from mining, milling, and enrichment. Under some circumstances uranium's 15-hour radon decay product requires additional control. The uranium tails from enrichment generally are not included in waste tabulations because in time they may be incorporated as a fertile material in breeding.
- Plutonium-contaminated waste, principally from weapons materials processing. There are two classes of plutonium wastes: the transuranic, containing mostly plutonium and other transuranics, and the high-level wastes that contain significant heat-generating products, strontium, and cesium.
- Other radionuclide-contaminated waste, generally called low-level waste, as generated in the reactor and fuel cycle, as well as in the radionuclide applications. (See Tables 21.14 and 21.15.)

Limiting the generation of waste is the first and most important consideration in managing radioactive wastes. The Purex process was developed to eliminate the solids additions to the high-level wastes in earlier technology. Improved housekeeping procedures have greatly reduced the low-level and transuranic

TABLE 21.14 Radionuclides in a PWR Spent Fuel Assembly[a]

			(Curies: Years After Discharge)			
Nuclide	*Half Life (y)*	*mpc (Ci/l)*[b]	*10*	*100*	*1000*	*10,000*[c]
		Zr Clad				
Fe 55	2.7	8E-7	180	10^{-8}	—	—
Co 60	5.3	5E-8	970	0.01	—	—
ZR 93	9E5	6E-8	0.10	0.10	0.10	0.10
		UO_2 Fuel—fission products				
Kr 85	10.4	3E-10	2300	6.8	10^{-25}	—
Rh 106	2.2 hr	—	270	10^{-25}	—	—
Pd 107	7E6	—	0.05	0.05	0.05	0.05
I 129	2E7	6E-11	0.01	0.01	0.01	0.01
Xe 133	0.014	3E-10	—	—	—	—
Cs/Ba 137m	30	2E-8	74,000	9,300	10^{-5}	—
		UO_2 Fuel–transuranium products				
Ra 226	1620	3E-11	10^{-7}	10^{-5}	10^{-3}	10^{-1}
Np 237	2E6	3E-9	7.6	7.6	7.0	3.1
Pu 238	89	5E-8	1000	500	0.5	0.06
Pu 239	24,360	5E-8	142	142	139	108
Pu 240	6580	5E-8	236	235	214	85
Pu 241	13	2E-7	36,000	500	0.01	0.003
Am 241	458	4E-9	770	1700	410	0.005
Cm 244	17.6	7E-9	480	15	10^{-14}	—

[a] Characteristics: 33,000 MWd/tU burnup, 14.7 kg U^{235}, 446 kg U^{238}, 108 kg Zr, 10 kg steel and Inconel.

[b] Normal human consumption in water would be equivalent to a radiation dose of 500 mrem/year, approximately 2 to 5 times the natural radiation background.

[c] After 10,000 years the radioactive toxicity is approximately constant for several million years.

TABLE 21.15 Estimated Cumulative Waste Quantities and Land Requirements: United States[a]

	Volume (10^6 ft^3)			*Land (acres)*		
	1980	*1990*	*2000*	*1980*	*1990*	*2000*
Low Level						
Defense	60	80	90	540	650	680
Commercial	15	45	83	150	300	420
TRU[b]						
Defense	0.05	0.05	0.05	20	100	200
Commercial	0.01	0.01	0.01	nil	nil	nil
High-Level[d]						
Defense (sludge & calcine)[c]	—	—	0.004	0	0	900
Defense (LLW salt cake)	—	—	0.008	nil	nil	15
Commercial spent fuel[d]	0.014	0.7	1.4	20	50	2200
Disposal facilities[d]						
Near surface burial grounds[e]	—	—	—	690	950	1100
Deep geologic vaults	—	—	—	nil	nil	2700

[a] IRG Appendix D, assuming 148 GWe in 2000 and commercial fuel not processed.

[b] TRU in interim surface storage until decision made regarding disposal.

[c] Packaging of defense wastes started in early 1990s.

[d] Spent fuel and defense high-level waste in surface storage until decision made regarding disposal.

[e] Includes scrap from decommissioned facilities.

wastes generated in the power plants, defense plants, and laboratories. Delay and decay is frequently an important strategy because much of the radioactivity in nuclear reactors and accelerators is very short-lived, a few minutes to a few days. Concentrating and containing is the objective of the treatment activities for the longer-lived radioactivity. The concentrate and contain strategy generally involves converting the airborne, liquid, and solid waste to stable solids in corrosion-resistant containers for storage and for transport to and emplacement in carefully selected disposal sites.

Finally, both dilution and dispersion of large volumes of air and water effluents containing very low quantities of radioactivity generally are necessary. The concentration of radioactivity in these effluents is controlled by federal and international regulations, and such effluents are continuously monitored before release to the environment from the waste treatment and other activities. In a particular operation, the regulations may be reflected by a set of actions that are triggered by successively higher levels of radioactivity.

The first action, where correction by the plant operator is called for, occurs at a small fraction of the permissible level. A somewhat higher level of radioactivity requires both operator action and regulatory notification. The next higher level requires regulatory participation, and if the final level is reached, the operation must be shut down.

Liquid Waste Treatment

Liquid waste usually arrives for treatment as an aqueous solution, and the treatment is primarily concerned with conversion of this solution to a solid form suitable for storage and disposal. Evaporation is the most effective process and achieves decontamination factors for the evaporate of 100 to 100,000 per cycle.

Ion exchange and scavenging precipitation can be employed at a lower cost than that of evaporation when decontamination factors of 5 to 100 are adequate.

As an example, in the U.S. defense establishment most of the radioactivity from plutonium production is found in the liquid high-level waste from the first cycle of the Purex process. This liquid is neutralized with sodium hydroxide and stored in earth-shielded tanks. There a sludge settles out that contains most of the radioactivity. The residual liquor is partially evaporated to decrease the volume of the waste, and sodium nitrite crystallizes on top of the sludge. In a process to be used at the Savannah River plant, the sodium liquor fraction, containing most of the cesium fission product, is pumped from the tank and treated using solvent extraction. The concentrated cesium and strontium stream is then combined with the sludge that was slurried from the tank and washed to remove the salt liquor. The resulting mixture is then mixed with a pulverized glass frit and converted to a boro-aluminum silrate glass in a vitrification furnace, cast in a canister, and sealed. (See Fig. 21.22.) This high-level waste form is then sent to a high-level repository (Yucca Mountain is the designated site in the United States, but is not yet operational). The sodium nitrate and the residual liquor will be combined with cement and other components to make a "Saltcrete" for long-term disposal in above-ground storage vaults.

Airborne Waste Treatment

The largest volume of radioactive waste is made up of the ventilation air from processes, plants, and laboratories. However, the quantity of radioactivity is relatively low. It generally occurs in a particulate form readily removed by dry filters. A special class of paper filters developed for this purpose (HEPA or High Efficiency Particulate Air) removes 99.97 percent of 0.3-micron particles. These filters frequently are preceded by scrubbers, sorbers, and roughing filters to extend the life of the paper filter.

A notable exception to the above is the airborne effluent from spent nuclear fuel reprocessing and from tritium production. In this case, iodine-131 volatilizes from fuel reprocessing and must be removed by passing the

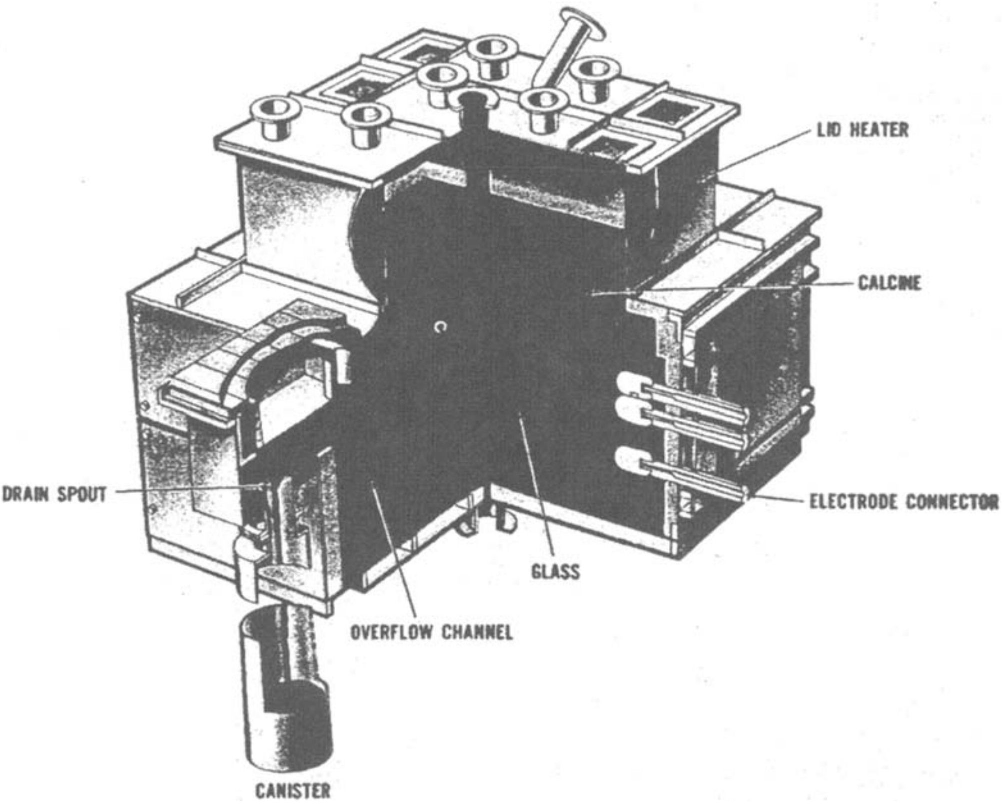

Fig. 21.22. Slurry-fed vitrification furnace converts aqueous high-level waste into a boro-aluminum silicate glass form. (*Courtesy USDOE.*)

airborne effluent through a silver catalyst bed, which provides a decontamination factor of about 100,000. Prior to reprocessing, the spent fuel is stored for a sufficient time for decay of the 8-day iodine-131, usually 10 half-lives, to make this treatment adequate. The processing effluent also carries increased levels of krypton85, xenon and tritium, gases, and solid radioactive particulates. The particulates are removed by filtration, but the other substances generally can be released. In tritium production, cold traps and sorbers are used to remove tritium from the airborne effluents.

Solid Waste Treatment

The solid wastes from nuclear operations include the concentrates from liquid and airborne treatment along with paper, clothing, lab glassware, and scrap equipment. The transuranic and low-level wastes both receive the same but separate treatment. In plutonium operations the first step frequently is monitoring to separate the transuranic from the low-level wastes. Some of these are incinerated to reduce their volume, but this process produces additional airborne and liquid waste. Physical compaction frequently is more useful. Equipment items, including large reactor components, are decontaminated by using a series of acid, chelating, and caustic washes. Finally, the solids may be incorporated in asphalt or concrete to reduce their leachability and then are drummed. Most of these solid wastes go to low-level disposal.

From power reactors with no reprocessing, the high-level waste consists of assemblies of zirconium-clad spent fuel rods to be packaged in stainless steel canisters. If the spent fuel is reprocessed, then the high-level waste will be converted to a silicate glass form similar to that from defense operations. The uranium and plutonium content in the waste, and also possibly the strontium-90 and cesium-137, would be reduced by 90 to 99 percent. The nuclides are useful, and their removal reduces the waste disposal problem in several ways: it reduces the toxicity and heat load of the waste and also reduces the incentive for future generations to undertake the very costly recovery of what may become valuable energy-producing resources.

Storage of Spent Fuel

Spent fuel originally was originally supposed to be stored at each nuclear power reactor in water pools designed to accommodate a 5-year inventory on the assumption it would be reprocessed later. However, reprocessing did not develop, and there has been a delay in identifying the ultimate repository. The storage pools were reracked to increase their capacity, and now the older reactors also are installing dry storage capacity to accommodate the life of the reactor, approximately 40 years or even 60 years, as power plant license extensions are granted.[40]

The federal government was committed to begin accepting spent fuel in the late 1990s and was planning to provide away-from-reactor storage that would have included a fuel rod consolidation facility. The nuclear utilities have been assessed at 1.0 mil per kWhr electric that is expected to pay for disposal of their high-level spent fuel wastes. As is usual for all new nuclear operations, the site and the schedule for this facility was dependent on public acceptance. Because this public acceptance was not forthcoming, the political support was not available in the U.S. Congress to first of all make a decision on an ultimate final burial site for high-level waste. Many options have been considered (see Table 21.16). The Yucca Mountain site in Nevada (a deep mined vault for the transuranic wastes, including the high-level wastes) was declared the sole repository site to be characterized. Although this site has been studied for almost 20 years, there is still a considerable amount of work to be done before any high-level waste can be accepted. It is very uncertain when the first repository will be receiving any transuranic waste, and it probably will not be before 2010.[41]

It appears that the repository capacity for high-level wastes will be heat-limited to one kilowatt/acre. This is equivalent to one ton of fuel after storage for ten years. The spent fuel discharged by the year 2000 will require about 2700 subterranean acres. This would be reduced to 1000 acres if the uranium and the plutonium were removed, and considerably less than that if the strontium and the cesium also were removed. Although reprocessing would reduce the transuranic content by a factor of 10 to 50, this amount would be an insignificant fraction of the transuranic hazard in the waste.

The most restrictive of the current regulations require this waste to be contained for about 10,000 years. Intensive international R&D has been conducted, along with extensive geologic surveys by responsible scientists. A 1990 National Academy of Sciences study concluded that this requirement is beyond the possibility of proof. Extensive experimental work has been carried out, and perhaps the most interesting study is related to a group of natural nuclear reactors found by the French in Gabon, Africa. During the Neolithic time, while uranium was being deposited in a swamp, the developing ore body repeatedly achieved criticality. This

TABLE 21.16 Conceptual Methods for Radioactive Waste Disposal

Terrestrial	Shallow land burial
	Deep geological vaults
	–Continental sites
	– Island sites
	Caves
	Deep hole
	Sub seabed
	Ice cap
	Hydrofracturing
	In situ rock melting
Extraterrestrial	Solar orbit–Space shuttle
	Moon crater–Rocket/soft lander
	Solar escape–Electric cannon
Transmutation	Fission reactors
	Fusion reactors
	Electromagnetic accelerators

occurred over a period of centuries at a series of interconnected sites. It appears that the long-lived products were closely held within the ore body. Unfortunately, the translation of this information to other geologic environments poses many uncertainties.

In 1989 a deep mined repository, The Waste Isolation Pilot Plant, was opened in a bedded salt formation near Carlsbad, New Mexico. The site has been receiving transuranic wastes since and has now disposed hundreds of tons.

Low-Level Waste Disposal

The solidified low level radioactive wastes for which ultimate disposal must be provided fall into two broad categories: the uranium-containing wastes from milling and the front end of the fuel cycle, including enrichment; and the radionuclides from the nuclear reactors and spent fuel operations, together with the radionuclides from applications in research, medicine, and other industries.[42]

The proposed technology for disposing of the second category of wastes consists of near-surface burial for low-level wastes.

Mill Wastes. Agreement has been reached with the public regarding mill waste, and disposal of the accumulated backlog is nearly complete via monitored surface repositories. The uranium-containing wastes from milling are mounded and covered with earth. This earth cover prevents erosion and delays escape of the 14-hour half-life radon gas, the gaseous decay product of uranium, long enough to significantly reduce released levels. These mill waste repositories are located near the mines and mills and are not a very different hazard from the original naturally occurring uranium deposits.[43,44] The depleted uranium from the enrichment operations is stored in cylinders as uranium hexafluoride for possible future use in the uranium-plutonium breeding cycle. Other uranium containing wastes from enrichment and fuel fabrication go to the low-level repositories.

Low-Level Wastes. Low-level wastes presently are being entombed in near-surface, earth-covered trenches. These trenches are designed to minimize the inflow of water and drain to a sump that is constantly monitored. Treatment facilities are maintained to process drainage that exceeds licensed levels of radioactivity for release in the groundwater.

One of the major repositories for commercial low-level waste is near Barnwell, South Carolina and is nearing its capacity. Other repositories around the country were closed when full or when required to be closed by state regulatory action. The major repository for very low-level or incidental nuclear waste is near Salt Lake City, Utah and there is another located in Texas. There have been no health effects or releases of radioactivity exceeding licensed limits from these low-level repositories. The schedule for opening additional low-level repositories has been delayed for several years by the same type of public opposition encountered by proposals relating to treatment and disposal of municipal and other wastes.

TRANSPORTATION OF NUCLEAR MATERIALS

The transport of nuclear materials reflects many years of experience based on millions of shipments. These shipments range from microcuries in liquid and gaseous forms in glass vials carefully packaged in sealed cans inside cardboard boxes to megacurie quantities contained in spent fuel packaged in hundred-ton shipping casks. They are shipped by both private and public carriers, by car, bus, truck, railroad, airplane, barge, and ship.

During these years there have been many incidents involving all forms of transportation, both commercial and military, and a few of these events resulted in some contamination of facilities. However, there have been no health effects to any individual transport worker or the general public.

Shipments of radioactive materials, originated by many different shippers throughout the world, are controlled by an established code of national and international regulations. Although most nuclear operations relate to a localized population, transport potentially affects large numbers of people. Many communities have attempted unsuccessfully to prohibit nuclear transport through their jurisdiction. Some, notably New York City, have succeeded.

Shipments of spent nuclear fuel constitute the largest quantity of nuclear materials being transported. These shipments involve 50-ton truck casks with the capacity for one to several fuel assemblies. Rail and ship casks weighing about 100 tons carry many assemblies. The design of these casks provides:

1. Radiation shielding to limit radiation to the workers and the public
2. Passive cooling for the heat-generating spent fuel, up to several kW per assembly
3. Assured containment in the event of the maximum accident

Current casks are designed to handle five-year aged spent fuel that travels by truck, rail, and ship. There are frequent ocean shipments from Japan to England and considerable rail shipment in Europe. Most of the spent fuel shipments in the United States are by truck. No cask failure has been experienced in any accident to date.

The transport of plutonium wastes to the transuranic and high-level repositories will represent a major increase in the quantities of radioactive materials shipped; therefore an extensive demographic study has been made to examine potential exposures of the public and to select routes to minimize such exposure.[45,46]

New casks are being developed to maximize the efficiency of the longer-aged spent fuel shipments and to reduce the exposure of the workers involved in loading and unloading the casks. There are expected to be two families of casks, those transporting the spent fuel from the reactors to a temporary storage facility, primarily truck casks, and those transporting the spent fuel from the storage facility to the repository, primarily rail casks. Another set of casks is being developed for the high-level defense waste glass at the Hanford and Savannah River facilities.

THE NUCLEAR REACTOR

Development of the commercial nuclear power industry for electricity generation began in the early 1950s. The first commercial electric power was produced using a light water reactor (LWR) at Shippingport, PA in 1957. By the 1970s, large numbers of power reactors had come online. In the United States all but one were LWRs, but in Canada the preference was for heavy water reactors (HWRs). There were many gas-cooled graphite reactors (GCRs) in England, France, and Russia and one in the United States. France and Russia had the first liquid metal fast breeder reactors (LMFBRs) operating, with several engineering test reactors in the United States. The characteristics of typical units are presented in Table 21.17. Nuclear reactors can be classified in several ways:

- *Burners vs. breeders:* Burners depend on uranium from ore but may recycle the residual fissionable material from the spent fuel to reduce the ore requirement. Breeders produce more fissionable material than they consume by converting either uranium-238 into plutonium-239 or thorium-232 into uranium-233. The breeders depend on the plutonium-239 or uranium-233 from the burners to bring them into equilibrium, and this may take 30 years.
- *Choice of neutron moderator-light water (protium), heavy water (deuterium), liquid (sodium metal), or solid (graphite):* Except in the case of graphite, the moderator also serves as the coolant. In the case of a graphite moderator, gas (He or CO_2) is used as a coolant.
- *Neutron energy (thermal vs. fast):* The sodium-moderated reactor operates with fast neutrons to breed using the uranium cycle. The water and graphite reactors operate with thermalized neutrons to more effectively burn the fissile material.
- *Pressurized vs. boiling LWRs:* The pressurized water reactor (PWR) transfers its energy from the fuel to an intermediate heat exchanger to generate the steam that

TABLE 21.17 Characteristics of Nuclear Power Reactors

	PWR[a]	*BWR*[b]	*Candu*[c]	*LMFBR*[d]	*HTGR*[e]	*AGR*[f]
Electric power (GWe)	1.1	1.06	0.74	0.23	0.33	0.63
Plant efficiency (%)	33	33	30	40	42	42
Fuel	UO_2	UO_2	UO_2	UO_2	UO_2ThO_2	UO_2
Clad	Zr	Zr	Zr	SS	C	Mg
Uranium loading (t)	99	155	130	—	95	—
U-235(5)	3.0	2.9	0.81	12[g]	40	0.75
Burnup (GWd/t)	30	28	7.3	100	110	—
SWU(t/GWey)	109	114	0	0,03	130	—
Fueling internal (y)	1.1	1	0.003	0.5	1	—
Fuel fraction	0.3	0.2	0.003	0.3	0.3	—
Power density (kW/kgU)	38	25	26	160	115	38
Coolant/Moderator	H_2O	H_2O	D_2O	Sodium	He/C	CO_2
Pressure (bars)	155	73	100	2	45	40
Temp (°C)	327	—	310	560	750	670
Flowrate (t/s)	21	15	12	3	0.4	—
Conversion ratio	0.6	0.6	0.6	1.2	0.8	0.6

[a] Zion 1 1973
[b] Peach Bottom 2 1974
[c] Bruce 1 1977
[d] Phenix 1973
[e] Fort St. Varain 1979
[f] Hinkley Point B1 1976
[g] Includes Pu-239

drives the turbine. The boiling reactor generates steam within the reactor that goes directly to the turbine.

The LWR, CANDU, and graphite reactors are the major producers of electricity from nuclear energy. When compared on the basis of cost, reliability, and safety, they are competitive. The PWR is the most widely used LWR, but there are also many BWRs in use. In 1990, there were 16 CANDUs operating in Canada where they were developed, with several in other countries as well.

When PWRs and CANDUs are compared on the basis of uranium utilization, the CANDU requires 19 percent less uranium from ore on a per kWhr basis (see Table 21.18). As there is now an abundance of low-cost uranium from ore and enrichment service, the electric utilities continue to favor the PWR and BWR types. Operating experience with graphite reactors in England, France, and Russia also has been generally satisfactory. However, the graphite moderator represents a large inventory of combustible material, which contributed significantly to the severity of the Chernobyl accident. Further development of the high-temperature, gas-cooled, graphite-moderated reactor is being considered for the advanced Generation IV reactors to be deployed from 2015 and beyond. Due to its much higher operating temperature (~900°C), this type of reactor can be used to provide process heat for a variety of industries.

A new family of advanced nuclear reactors has been designed by the PWR, BWR, and CANDU suppliers, which are now (2006)

TABLE 21.18 PWR and CANDU Requirements for Uranium from Ore

	Metric Tons/GWy-electric	
	PWR	*CANDU*
Once-through cycle	217[a]	175[b]
Pu/U recycle	157	129[c]

[a] 3.25 percent U-235 fuel
[b] 0.71 percent U-235 fuel (natural uranium)
[c] Pu/U recycled to CANDUs, from PWR once-through spent fuel (PWR/TANDEM)

being licensed. It is the industry's objective to use proven technology to provide reactors that are simpler and less costly to build than older designs. These reactors would have wider safety margins through the increased use of passive safety systems, relying more on reactor design and less on operator action to prevent accidents. They would be made less complex by drastically reducing the number of components necessary for operation. This design also would facilitate plant construction by requiring less concrete, fewer valves and pumps, and less piping. Most likely the future will see introduction of modular built systems that will be prefabricated at the factory. Safety considerations always will be paramount.[47,48]

Light Water Reactors

Pressurized Water Reactor. The PWR contains three coolant systems: the primary system, which removes heat from the reactor and partially controls nuclear criticality; the secondary system, which transfers the heat from the primary system via the steam generator to the turbine-driven electrical generator; and the service water system (the heat sink), which dumps the residual coolant energy from the turbine condenser to the environment. The service water is recirculated from a river, lake, ocean, or cooling tower. In the primary system (Fig. 21.23), dissolved boron is present to control nuclear criticality. Fixed-bed ion exchange units are used to maintain the water quality in both the primary and the secondary systems. The chemical and volume control system reduces boron concentration during the power cycle to compensate for fuel burnup. These operations are carried out continuously through bypass systems.[49] A more complete view of the current technology is presented in Table 21.17.

Boiling Water Reactor. In the BWR, steam is generated in the reactor and goes directly to

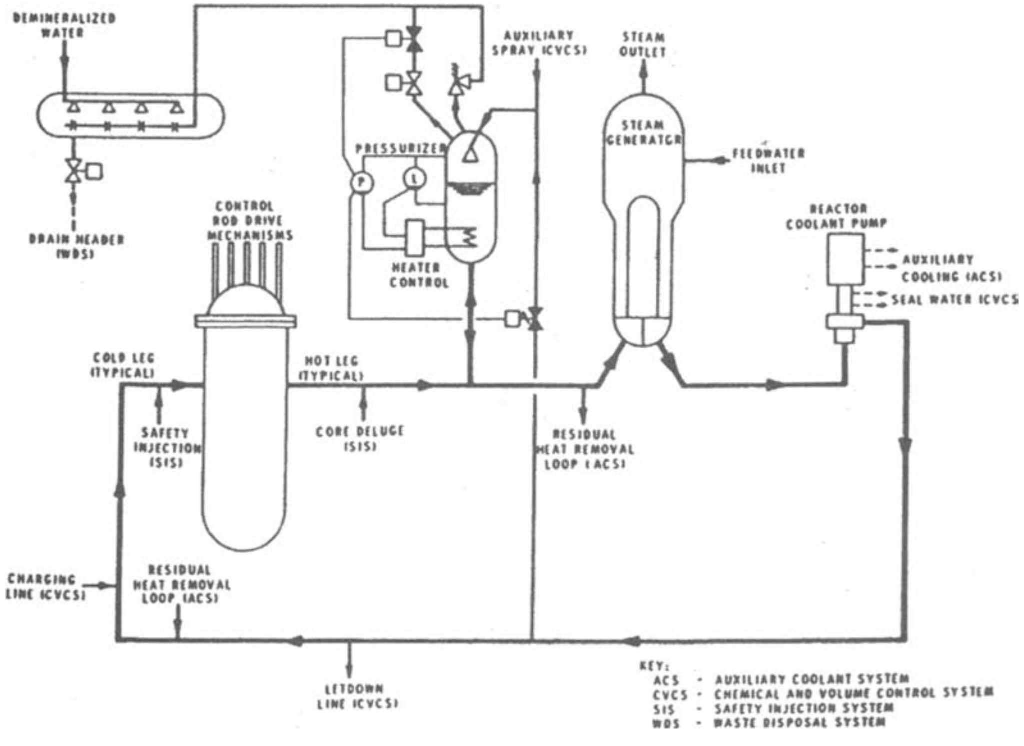

Fig. 21.23. Pressurized water reactor (PWR) for power generation. (*Courtesy USDOE.*)

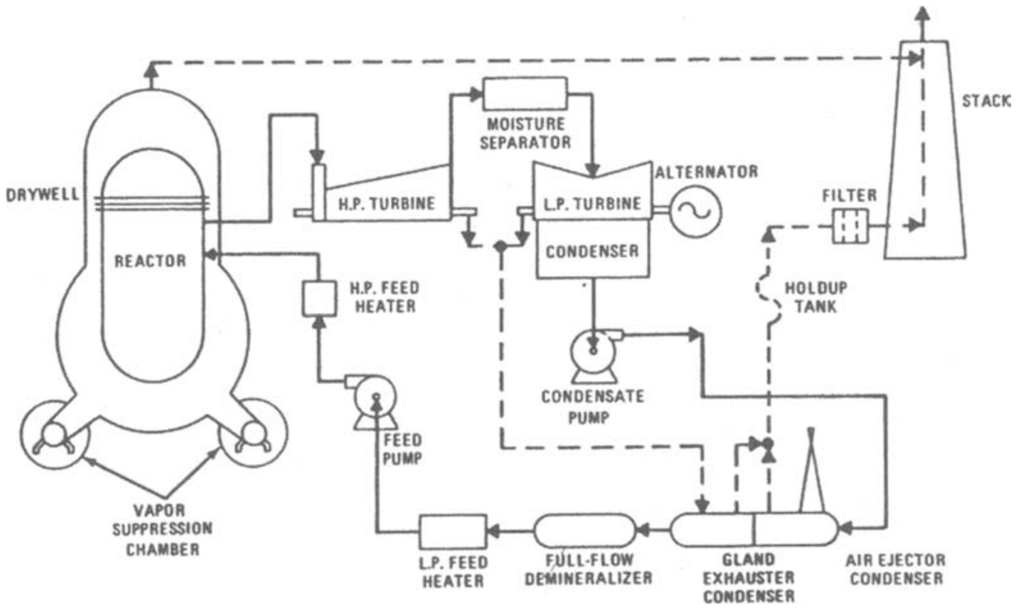

Fig. 21.24. Boiling water reactor (BWR) for power generation. (*Courtesy USDOE.*)

the turbine (Fig. 21.24). Although the BWR eliminates the intermediate heat exchanger, it places the steam turbine in a radioactive environment. Here in the primary system, there are full-flow ion exchange units treating about 30,000 gal of water/min, which serves two objectives: removal of radioactivity resulting from neutron-activated corrosion products and fuel cladding leaks; and corrosion protection of the reactor, fuel, and turbine from possible leakage of service water to the primary coolant from the turbine condenser.

In the following paragraphs, three important aspects of LWRs are discussed: fuel elements, the primary coolant system, and reactor containment. They each play an important role in providing multiple barriers that prevent radioactive fuel and fission products from being released to the environment.

Fuel Elements. The fuel element is designed to provide the primary containment for the radioactive fuel and fission products over the three- to four-and-one-half-year operating life of the PWR and BWR fuel. In addition, without significant problems, it has provided the containment of the spent fuel after discharge from the reactors for more than 30 years under water in the storage pools at some of the reactors. At some of the older reactors it is now being packaged in stainless steel canisters and placed in dry storage vaults at the reactor site.[50]

The quality of the reactor coolant depends on the integrity of about 80 to 110 miles of 22-mil Zirconium alloy tubing operating at temperatures up to 350°C containing uranium oxide pellets. The PWR fuel assembly, which is about 14 feet long, is made up of up to about 264 fuel pins (Fig. 21.25.). A typical PWR contains about 110 metric tons of fuel charged with up to 4.95 percent U-235. The economics of the fuel cycle require this degree of enrichment to achieve a reasonably long operating life and high energy yield from each assembly. Because these nuclear power stations represent a significant part of the total power system capacity of the United States (about 20% in 2005), frequent shutdowns are undesirable. With the increased experience and expectation of a once-through fuel cycle, the enrichment has been increased to raise the energy yield to an average of 50,000 MW days/metric ton and to extend the time between refueling shutdowns to 18 months or longer. This reduces the amount of

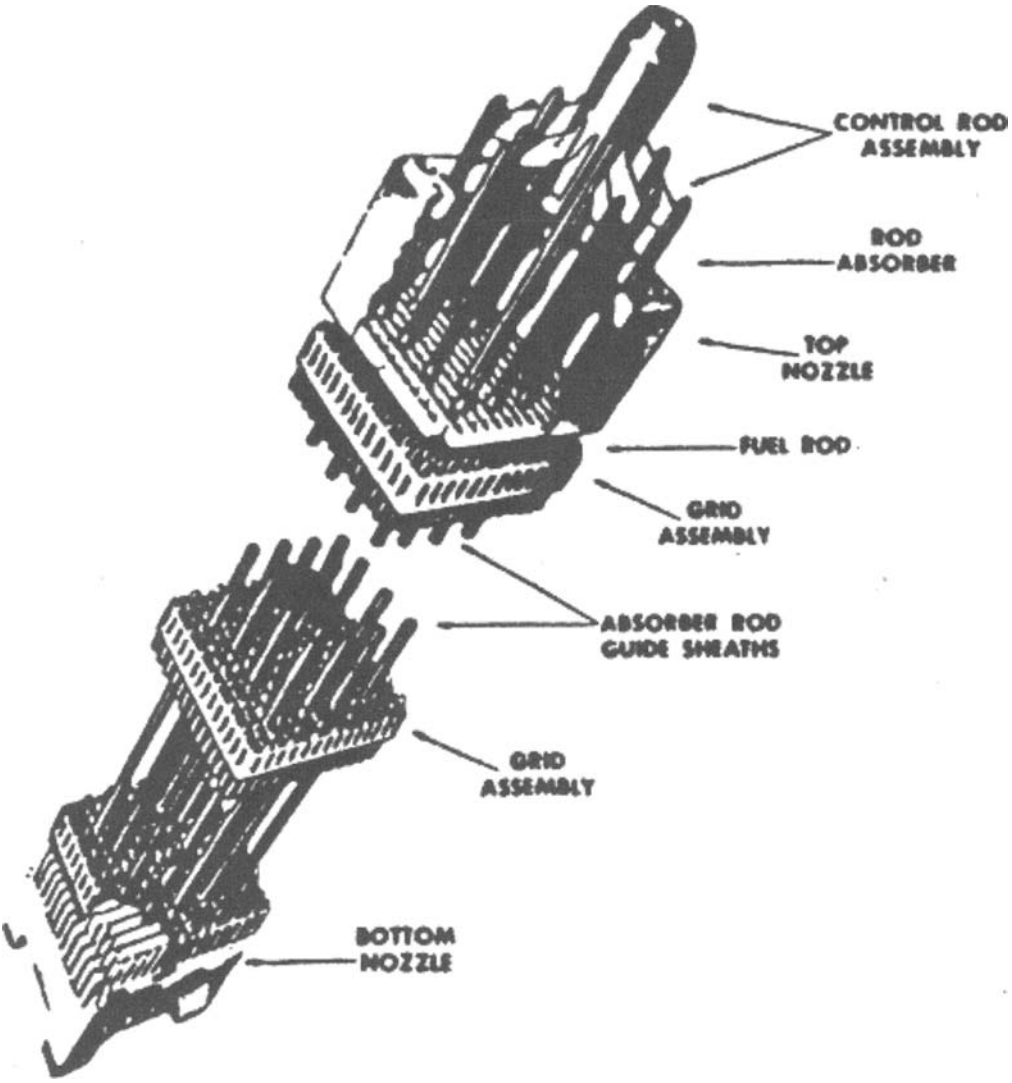

Fig. 21.25. Cutaway of PWR fuel element with the control rod cluster assembly. Element contains about 1200 lb UO_2 in up to about 264 rods. (*Courtesy USDOE.*)

replacement power needed during shutdowns, which generally comes from the older, more costly, fossil-fuel-powered plants or must be purchased from outside the system. Approximately one-third of the fuel is replaced in the core of the reactor during each refueling shutdown. In order to improve fuel cycle economics and protect the reactor vessel from radiation-induced embrittlement, partially burned fuel is often loaded in the core periphery. Appropriate placement of the new fuel and shuffling of other partially burned fuel ensures satisfactorily flat power distributions.

A 1000 MWe BWR core contains about 165 tons of UO_2, which is charged up to 5 percent U-235 (Fig. 21.26). Here the integrity of the 32-mil fuel cladding (Fig. 21.26) is even more important than in the PWR because the steam is generated in the reactor core and goes directly to the turbine. This has proved to be a feasible design and eliminates the PWR expense of replacing steam generators.

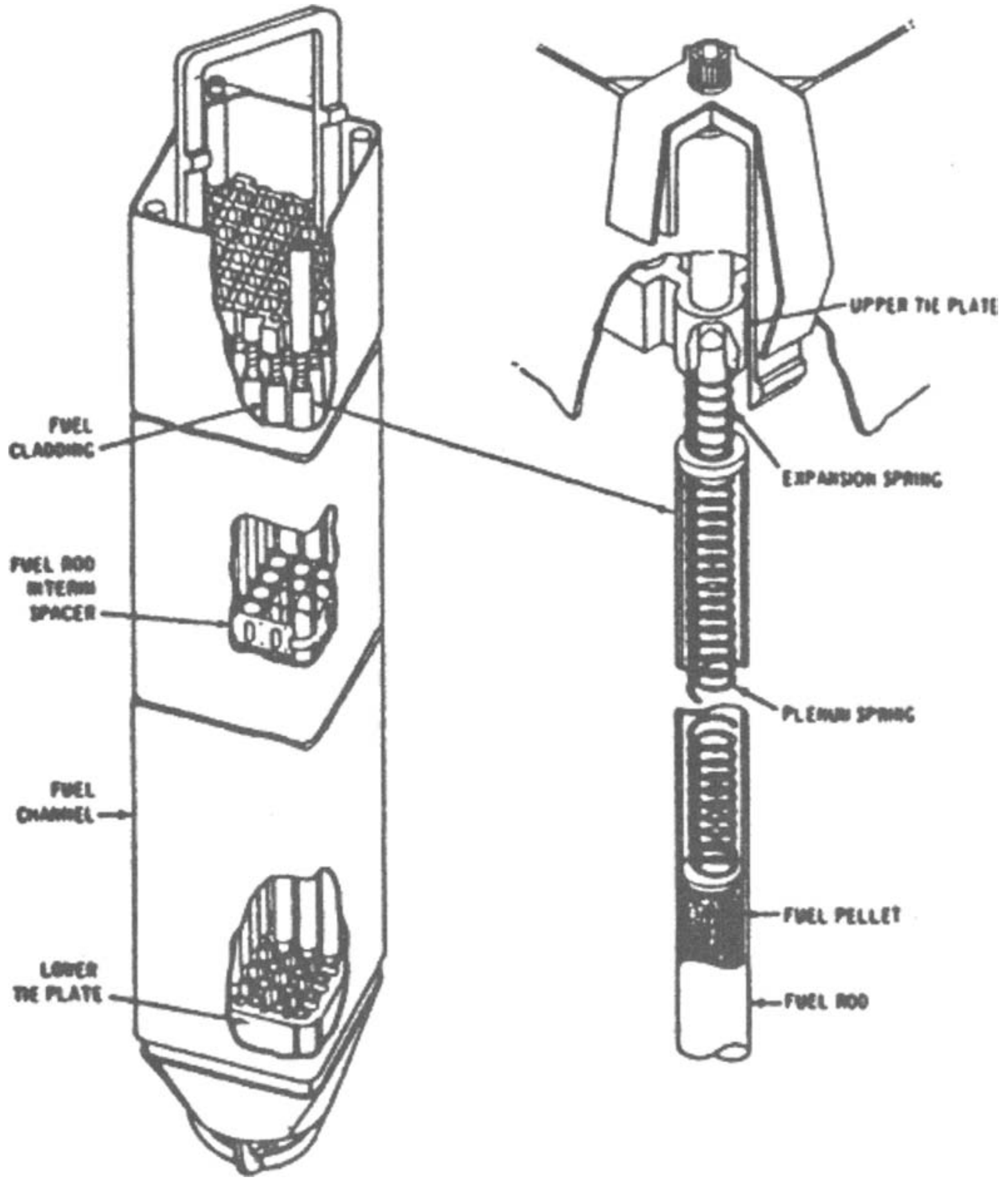

Fig. 21.26. Cutaway of BWR fuel element containing 488 lb UO_2 in 49 rods. (*Courtesy USDOE.*)

Two engineering system demonstrations were performed to reduce the uranium-from-ore requirements of LWRs: recycle of the plutonium and conversion to the thorium–uranium cycle to achieve thermal breeding. The demonstration phase of the plutonium recycle development was carried out in seven power reactors. Several LWRs originally were started up on the thorium–uranium cycle, and a light water breeder reactor core using the thorium cycle was tested in the Shippingport reactor. Although these tests did not resolve all the technology questions, no major problems were uncovered. In the United States, plans for fuel recycling and the use of the thorium cycle have been postponed because of the low cost of uranium from ore as well as the perceived danger of diversion of the fissionable materials.

Primary Coolant System. Nuclear technology faces challenges on most of the frontiers of engineering, with design and fabrication of the primary coolant system being one of the greatest. The reactor vessel and associated piping and heat exchangers comprise the second barrier to prevent the release of radioactive materials from the fuel to the environment. The steel for the reactor vessels must meet metallurgical standards developed for them to withstand the temperature and pressure cycles as well as neutron irradiation, which in time reduces the ductility of the metal. Carbides and phosphides precipitated in the grain boundaries prevent slippage along the crystal planes. The design criteria for optimum performance require operating pressures of 1050 psia for BWRs and 2250 psia for PWRs. Typical dimensions for vessels serving 1000 MWe stations range from 21 feet in diameter by 70 feet in height for BWRs, to 14 by 42 feet for PWRs, with vessel weights of 782 and 459 tons, respectively. These vessels are shop-fabricated, and their transport to the reactor site tests the ingenuity of the carriers.[51,52]

As anticipated, there have been occasional equipment failures involving reactors, but the safety systems have been sufficiently redundant so that one or more have always worked. Even in the Three Mile Island accident in 1979, the safety systems worked as designed. Much of the damage resulted from operator actions to override the safety systems. As concluded in the Reactor Safety Report, the limitations of the operator created and then seriously aggravated the Three Mile Island incident. Nevertheless, the features of the containment system prevented significant exposure to the plant personnel or any off-site individual, this despite failure of the barriers provided by the fuel cladding and the primary coolant system.

LWR tests-to-failure had been performed to evaluate accident scenarios involving loss of coolant accident (LOCA) events such as occurred in the Three Mile Island incident. The power burst tests in a 20 MWt PWR have created fuel failures and defined the initiating conditions. The LOCA tests with a 50 MWt PWR have demonstrated recovery from catastrophic major feedwater and steam line breaks without fuel damage.

Advanced reactors such as the Westinghouse AP600, AP1000, and General Electric ESBWR have been developed that produce the same amount of power but with many fewer components. These reactors will have increased reliability and greater safety due to the reduced component count that reduces both the cost and the number of points at which failures can occur. In addition, these newer reactor designs are designed to be passively safe. That is, active involvement by the reactor operators is not required to maintain the reactor in a safe state. These types of designs were developed to allow time for reasoned decisions to be made in case of an accident or major equipment failure while still keeping the reactor in a safe state.

CANDU Heavy Water Reactor

This Canadian reactor design originally utilized (1) natural uranium as the fuel, thus avoiding the necessity for uranium enrichment, and (2) heavy water as both coolant and moderator, to maximize neutron utilization. Current designs on the drawing boards plan to use slightly enriched fuel and light water as a coolant. In addition to electrical power, this reactor can efficiently produce cobalt-60, which has commercial markets for product irradiation. Each reactor can produce 30 to 50 megacuries annually, and in 1999, the catalogue price was $1.20/curie. The CANDU requires 18 percent less uranium from ore than the LWR once through cycles need. The spent fuel contains 0.23 percent U-235, roughly equivalent to the tails from the gaseous diffusion enrichment operation. It also contains 0.27 percent fissionable plutonium. The average residence time of a fuel is about one year, with approximately 0.3 percent replaced each day. This requires refueling during operation. The CANDU fuel bundles contain 37 Zircaloy-clad UO slugs, a less complex design than that of the LWR assemblies. Many alternative fuel cycles are being considered for the CANDU to further decrease the requirements of uranium from ore, but they will require enrichment or reprocessing.[53]

Liquid Metal Fast Breeder Reactor

The salient features of the liquid metal fast breeder reactor include a fuel doubling time of 10 to 30 years, along with a high coolant temperature for more efficient energy conversion and low operating pressure through the use of liquid sodium as the coolant. Operation of the reactor with fast neutrons increases the neutron multiplication factor for the mixed uranium–plutonium oxide fuel. The power density in the LMFBR is about five times greater than that in the LWR cores; so the reactor vessel is much smaller. This core is surrounded with radial and axial blankets of fertile material. These blanket regions, which capture the leakage neutrons, must be used to achieve breeding, which consists of the production of fissile material from fertile material. Finally, the use of the uranium–plutonium fuel cycle makes optimum use of the plutonium produced in the LWRs.

All major nuclear countries have participated in LMFBR demonstration plants, ranging from about 200 to 500 MWe. In the United States, the Experimental Breeder Reactor I (EBR I), the starting point of this program, went critical in 1951. LMFBRs were operated in the United States, England, France, Japan, and Russia. In France, the Phenix, a 590 MWt/230 MWe reactor, was operated starting in 1973, and the Super Phenix, a 1200 MWe reactor, first generated power in 1987 and was in commercial operation. Although the EBR I and Super Phenix have been shut down, the Phenix is currently operational. In the former Soviet Union, the BN-350 MWe reactor produced power and steam heat for Shevchenko on the shore of the Caspian Sea from 1973 to 1999; and another reactor, the BN-600, has been operating since 1980. In the United States, the 400 MWt Fast Flux Test Facility started operation in 1980 to test fuels and, materials. The objectives of this program were satisfied in 1989, and it was shut down.

During the 1980s, the Argonne National Laboratory was developing a metal-fueled LMFBR in contrast to the oxide-fueled version described above. The principal advantage of the metal fuel was to be the ease of recycle using electrometallurgical technology. The program was called the Integral Fast Reactor with emphasis on high burnup instead of breeding, possibly 185,000 MW-days/metric ton versus 50,000 for the present-day PWRs (Fig. 21.27) Early work with metal fuel showed that it expanded because of the presence of the xenon and ruptured the cladding. The fuel rods have been redesigned to accommodate this phenomenon. Finally, each reactor site would have its own reprocessing unit, and this would greatly reduce concern about proliferation. Cost estimates indicate that the cost of the electricity generated might be equivalent to that from LWR and HWR systems. Much work remains to be done to confirm all these attractive indications.[54]

Other Nuclear Reactors

Many other nuclear reactors have been developed for research, engineering development, nuclide production, and mobile power. Must noteworthy are the nuclear propulsion systems for naval applications. In the United States, the PWR technology for electric power generation is an outgrowth of the nuclear naval development program. The first nuclear submarine, the *Nautilus*, was commissioned in 1954. Today there are more than 100 nuclear submarines, along with a number of aircraft carriers and other surface vessels. Other nuclear propulsion systems have been studied for arctic tractor trains, aircraft, and rockets for space propulsion, but these studies were terminated before completion owing to insufficient benefit or other funding priorities and environmental concerns. Recently, nuclear reactor development was started again to provide power on orbiting space stations in connection with the United States Strategic Defense Initiative program and for propulsion as part of the NASA manned and unmanned planetary explorer programs.[55] As part of this

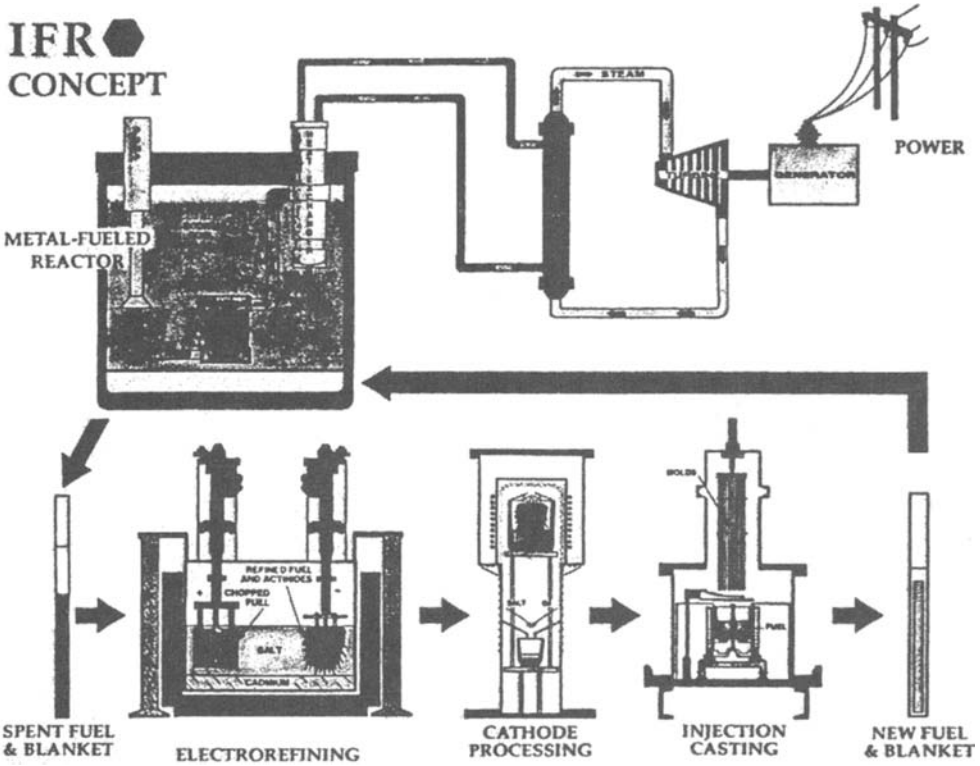

Fig. 21.27. Schematic of integral fast reactor and fuel cycle concept (IFR). *(Courtesy USDOE.)*

activity, a "space" reactor was purchased from the Russians who have been employing them for many years.

The remaining classes of nuclear reactors range from zero-power, subcritical neutron sources for university training to large-scale reactor systems for plutonium-239 production. Portable reactors have provided heat, power, and water to U.S. bases in Alaska, Antarctica, and Panama. Private industry has operated various test reactors for reactor studies and radioisotope production.

RADIATION PROCESSING

Next to the generation of electric power, radiation processing is potentially the most important commercial application of nuclear energy. Radiation processes have been developed for treating food and medical supplies to inhibit growth of bacteria, viruses, fungi, and insects, and for polymerization of plastics and rubber. They take the place of thermal and chemical treatment to achieve these objectives.

Radiation also is an important tool in the medical treatment of cancer. However, what may be one of its most important applications, extending the storage life of foods, has been seriously delayed by public concern, in spite of extensive tests showing that irradiated foods meet all international safety standards. Currently, the United States irradiates spices for the main purpose of bug removal.

In early radiation processing work, cobalt-60 was the principal source of radiation, but now it has been displaced by machine-produced electrons wherever possible. Cobalt-60 is commercially available from the Canadian CANDU power reactors.

Commercial information regarding the application of machine-produced electrons is closely held, but the following are a few examples of their use:

1. Low-energy electrons, up to 0.5 MeV, are in general use to polymerize thin

plastic sheeting and coatings. Fixed beams up to 2.5 meters wide irradiate 0.4-mm-thick products with 10 kiloGray (kGy) at 1600 meters/hour.

2. Electrons with energies up to 5 MeV are widely used in scanned beam machines. The typical scan width is 1 to 2 meters when applied to the polymerization of thicker materials. These machines also are being used for irradiating grain and chicken feed. The Russians are treating 200 tons/hour of grain with a dose of 0.4 kGy, and the Israelis treat chicken feed with 3.8 kGy at 15 tons/hour. In both cases the flowing-bed depth is about 6 mm.
3. A 5-kW beam of 7 MeV electrons is delivering a 3 kGy dose to flat cakes of deep frozen cakes of chicken meat at a rate of 50 tons/day; disposable medical supplies are being treated at the rate of 100 cubic meters/day.[56]

RADIOISOTOPE APPLICATIONS

The tremendous release of energy from nuclear reactions makes possible a unique family of applications for long-lived radioisotopes that are important to health, science, and industry. Whereas fission and fusion occur almost instantaneously, other radioactive decay processes occur in times ranging from a few minutes to thousands of years. The general areas of application may be grouped into irradiation, thermal energy generation, and tracer applications.[57]

Radiation Sources

Radiation from radioactive nuclides is used to detect changes in density or other characteristics of materials, to promote chemical, physical, or biological changes, and to provide a source of thermal energy. Some radioactive materials find use in industry, research, and medicine as tracers for physical, chemical, and biological processes.[58]

The very important radioisotope cobalt-60 has a 5-year half-life and emits 1.17 and 1.33 MeV gamma rays. Co-60 is made by irradiating naturally occurring Co-59 with neutrons in a reactor. It is the major radioisotope in a number of applications including the following.

1. Destruction of malignant tumors.
2. Sterilization of prepackaged medical supplies, particularly those that are heat-sensitive or can be contaminated by chemical agents.
3. Food irradiation to extend shelf life.
4. Radiographing metals to determine weld quality. (Iridium-192, with a 0.3 MeV gamma ray, provides better definition than Co-60 in some cases but has a shorter half-life.)
5. Polymerization of coatings on electrical conductors and paper and monomers impregnated into wood and concrete. (Machine radiation generally is preferred in applications involving thin materials.)
6. Density measurements for level controllers in silos and other vessels bearing solids.

There also are many applications for alpha and beta radiation sources, including:

1. Polonium-210 alpha activation of beryllium to yield neutrons that in turn are used to start up nuclear reactors
2. Plutonium-238 activation of beryllium to produce neutrons for logging drill holes to detect hydrogenous materials in geologic formations
3. Strontium-90 beta sources to measure the thickness of paper in paper manufacture and also to discharge static electricity by ionizing the air in high-speed printing operations

Radioisotope Thermoelectric Generators

These devices (RTGs) provide reliable long-life sources of electrical energy. The thermal energy required to drive the thermoelectric element is provided by a long-lived radionuclide. The electricity is generated by passing the thermal energy through the thermoelectric element from the heat source at one end (the hot foot) to a heat sink at the other end (the cold foot). Germanium silicide is now the thermoelectric material of choice; because of its higher thermal stability, it can be driven with larger temperature differentials. Lead telluride is more efficient per degree of temperature difference but has lower thermal stability.[59]

Plutonium-238 is the most important heat source; it is an alpha emitter whose radioactive decay energy is easily absorbed and converted into thermal energy. It also is easily shielded. In addition, its long half-life, 90 years, makes possible hundred-watt power sources with a design life exceeding ten years. Although solar cells generally comprise the technology of choice to power extraterrestrial missions, a Pu-238 RTG was used for the Apollo moon landings. Such devices are used when the mission path is shielded from the sun, and they continue to be used for deep-space missions. (See Fig. 21.28.)

Terrestrial RTGs have also been developed and are commercially available. A beta emitter, 30-year strontium-90, has been the major radionuclide used for terrestrial applications. Although beta particles (negative electrons) are easily sorbed and converted to thermal energy, the sorption process is accompanied by the emission of X-rays that require shielding. This necessitates a heavy device, about 300 lb for a 5-watt source. Because strontium-90 is a fission product, the supply now is limited to 100 megacuries recovered in the 1960s from the Hanford defense waste. This is equivalent to about 10 kW of electrical energy, assuming a 5 percent conversion efficiency.

Tracers

The radiation emitted from decay of single atoms can be easily measured. Thus, small amounts of radioisotopes can be incorporated into systems to study, for example, chemical reactions and the flow of fluids through complex systems such as organs of the human body. They also can be used in more mundane applications such as locating leaks in buried piping. Many organic compounds tagged with carbon-14 in specific positions are commercially available.

Because C-14 with a half-life of 5700 years occurs in nature, it can be used to determine the age of carbon-containing artifacts. The basis for such determinations is the loss of C-14 that was trapped in the artifact (animal, human, fired pottery, and so on). The related art and science of carbon dating are fairly complex and thus subject to significant uncertainties.

Nuclear Medicine

Nuclear medicine is one part of nuclear utilization readily accepted by the public. However, there is strong opposition to the siting of the low-level waste repositories needed for disposal of the wastes that result from the practice of nuclear medicine.

The medical applications of nuclear technology range from in vitro and in vivo injections for diagnostic tests to cobalt radiation for cancer therapy. A new medical specialty was created, a family of compact cyclotrons was developed to provide short-lived nuclides, and a sizable industry evolved to produce technetium. Until the nuclear industry was created, technetium had been missing from the chart of chemical elements because the half-life of the most stable member was too short, 21,000 years. Technetium and several other nuclides of importance here are discussed elsewhere in the chapter in connection with their production (see Table 21.19).[60,61]

TABLE 21.19 Radionuclides Used in Nuclear Medicine

For organ function and tumor visualization:	
Technetium-99m	Carbon-11
Iodine-131	Oxygen-15
Gallium-67	Nitrogen-13
Strontium-85	Fluorine-18
Indium-111	Iodine-123
Xenon-133	
For irradiation of tumors:	
Cobalt-60	Cesium-137
Radium-226	Iodine-131

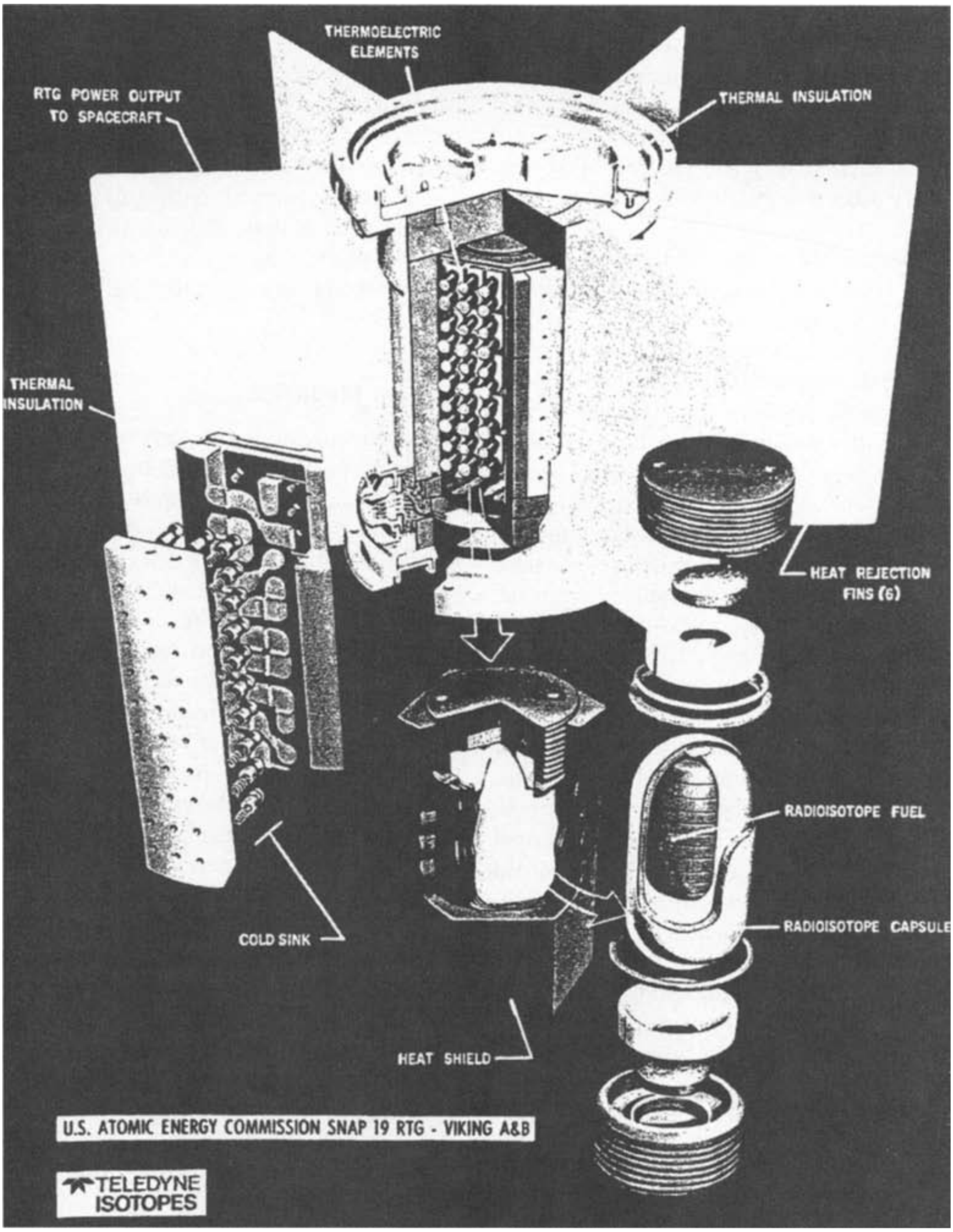

Fig. 21.28. The multi-hundred-watt electric generator. Three units on *Voyager* generated 475 watts from the decay heat of plutonium-238 acting on thermoelectric couples. *(Courtesy USDOE.)*

The two principal applications of nuclear medicine are for radiation treatment of tumors and for visualizing organ functions and tumors. These nuclear-medical procedures supplement other medical treatment and diagnostic procedures, and are available in most hospitals throughout the industrialized countries and in the major hospitals in other countries.

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