Geological Media and Factors for the Long-Term Emplacement and Isolation of Carbon Dioxide and Radioactive Waste

Stefan Bachu and Tim McEwen

Abstract A review is presented of the factors considered important in the selection of environments and sites for the geological storage of carbon dioxide (CO_2) and the disposal of radioactive waste (RW)—with a focus on those of a geological nature. The distinction between the terms *storage* for CO_2 and *disposal* for RW is not significant in this regard. The relevant properties of the two product types are presented, as are the desirable characteristics and types of geological environments that are considered suitable for disposal purposes. The role that the geological barrier plays in trapping the disposed substance, in the case of CO_2, and in containing and slowly releasing the waste, in the case of RW, is explained. The comparative roles played by the geological barrier and the engineered barrier system of a repository for RW is also outlined—although the emphasis of the discussion is on the geological barrier itself. The status and challenges associated with the storage of CO_2 are presented, together with a discussion of the geographic distribution of areas of the world potentially suitable for its storage and the criteria for site selection that could be applied. A discussion is also presented of the geological environments that are most likely to be used for the disposal of RW.

A considerable part of the chapter presents a comparison between the storage or disposal of the two types of disposed substances, discussing their similarities and differences. This comparison is considered under the four subject headings: Characteristics of the Geological Media, Emplacement Characteristics, Effects of Emplacement and Potential Migration from the Disposal Site, and Site Activities.

Keywords Radioactive waste disposal • CO_2 storage • Trapping mechanisms • Migration mechanisms and pathways • Repository • Geological/natural barrier • Engineered barrier system

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1 Why Geological Storage of CO_2 and Disposal of Radioactive Waste?

1.1 Introduction

The emplacement in geological media of radioactive waste (RW) and carbon dioxide (CO_2) is considered to be a safe method for isolating these substances from the hydrosphere, the atmosphere and the biosphere. The disposal of long-lived RW, e.g. spent fuel (SF), long-lived intermediate-level waste (ILW-LL), etc., currently takes place at only one location, at the Waste Isolation Pilot Plant (WIPP) in the USA, although plans to dispose of SF are well advanced in several countries and the disposal of this type of waste is likely to be taking place at several sites over the next few decades. Investigations and research programmes concerning the disposal of RW have, however, been taking place since the 1970s or 1980s in many countries. The recent increased interest in the use of nuclear power for electricity generation has provided a greater focus on developing long-term management solutions for the waste that is inevitably produced.

In contrast, the storage of CO_2 is a relatively recent consideration, although there has been injection of approximately one million tonnes per year of CO_2 at Sleipner in the North Sea since the mid-1990s and, similarly, at In Salah in Algeria since the mid-2000s. The importance of CO_2 storage has risen rapidly up the political agenda over the last decade as representing a climate change mitigation strategy with significant potential, in particular as the significance of the effects of global warming has been appreciated.

1.2 Carbon Dioxide

The widening gap, on the one hand, between the increase in CO_2 emissions due to the expected increase in population, global standards of living and carbon intensity of the energy system, and, on the other hand, the decrease in CO_2 emissions due to the increase in energy efficiency and conservation, can be partially or totally covered by artificially increasing the capacity and uptake rate of CO_2 sinks through CO_2 storage or sequestration. This involves either the diffuse removal of CO_2 from the atmosphere after its release through terrestrial and marine photosynthesis, with subsequent storage of the carbon-rich biomass (natural sinks), or the capture of CO_2 emissions prior to their potential release and their storage in deep oceans or geological media, or through surface mineral carbonation (known collectively as carbon capture and storage, or CCS).

In contrast to natural sinks, CCS is a process that consists of separating and capturing CO_2 from large stationary sources, transporting it to a storage site, and isolating it from the atmosphere for very long periods of time, in the order of several centuries to millions of years. Three processes have been considered: surface mineral carbonation, ocean storage and geological storage (IPCC 2005). Surface mineral carbonation consists of converting CO_2 into solid, inorganic carbonates by chemical reactions, but requires the use of certain minerals such as olivine and serpentine, mining on a large scale, large amounts of energy for crushing, milling and heating the minerals, and the transportation and disposal of very large amounts of the resulting carbonate rock, thus excluding this process as a viable option for reducing atmospheric CO_2 emissions (IPCC 2005). Ocean storage consists of injecting CO_2 at great depths, where it will dissolve or form hydrates or heavier-than-water plumes that will sink to the bottom of the ocean (Aya et al. 1999), thus removing CO_2 from the atmosphere for several hundreds of years. However, ocean CO_2 storage would result in a measurable change in ocean chemistry, with corresponding consequences for marine life (IPCC 2005), notwithstanding issues of ocean circulation, storage efficiency, technology, cost, technical feasibility, international limitations regarding dumping at sea, and strong public opposition.

Geological storage of CO_2 thus currently represents the best and likely only short- to medium-term option for significantly enhancing CO_2 sinks. The technology exists today and can be applied immediately, being based on experience to date from the oil and gas industry, from the deep disposal of liquid wastes and from water resources management (IPCC 2005), and is forecasted to play an important role in reducing anthropogenic CO_2 emissions into the atmosphere in the first part of this century and beyond (IEA 2004, 2006). The storage of CO_2 in geological media shares many similar features with oil and gas accumulations in hydrocarbon reservoirs and methane in coalbeds, whilst the capture, transportation, injection and monitoring of CO_2 in the subsurface has already been practised for a few decades in enhanced oil recovery, acid gas disposal and CO_2 storage (IPCC 2005). However, although the individual components of this technology all exist separately, they have not yet been implemented on a large scale in an integrated system because of significant challenges and barriers of an economic, legal and regulatory nature and due to public attitudes to large-scale deployment (Bachu 2008a).

Although various climate change mitigation options have different spatial and temporal ranges of applicability and timing of deployment, it is clear that the reduction in atmospheric CO_2 emissions needed for stabilizing the climate can be achieved through the application of a portfolio of measures, which includes energy efficiency and conservation, increasing the share of non-fossil fuel energy sources and carbon capture and storage (Pacala and Socolow 2004; Socolow 2005). The latter could provide 15–43% of the emissions reduction needed to stabilize atmospheric greenhouse gas levels at 550 ppm CO_2 equivalent (Pacala and Socolow 2004), compared to 380 ppm today and 280 ppm in the mid-nineteenth century.

In this context, carbon capture and storage means the removal of CO_2 directly from anthropogenic sources and its emplacement in geological media for long periods of time. From an engineering point of view, this is a geological disposal operation, similar to acid gas (CO_2 and hydrogen sulphide (H_2S)) disposal at more than 70 sites in North America (e.g. Bachu and Gunter 2005) and to other fluid-waste disposal operations, albeit on a much larger scale. However, for various reasons the term CO_2 disposal has been avoided, and various terms have been used historically such as CO_2 removal, CO_2 sequestration and CO_2 storage. The term *CO_2 sequestration* continues to be used preferentially in the USA, where it is defined as the long-term isolation of CO_2 from the atmosphere through physical, chemical, biological or engineered processes. Geological CO_2 sequestration refers specifically to the emplacement of CO_2 deep underground. The term *CO_2 storage* is sanctioned by UN agencies and is used, particularly in Europe, to indicate CO_2 underground emplacement, the term CO_2 sequestration in these countries being reserved for other processes that reduce atmospheric CO_2 emissions. For consistency with the purpose of this book and with other chapters, the term *CO_2 disposal* will be used from now on in this chapter, the meaning, nevertheless, being the same as that of CO_2 sequestration and carbon capture and storage, or CCS, namely the injection of CO_2 into geological media in order to isolate it from the atmosphere and biosphere for long periods of time—at least several centuries to millennia.

1.3 Radioactive Waste

Deep geological disposal (generally at hundreds of metres depth) is the option favoured internationally for the long-term management of heat generating RWs (i.e. SF and high-level waste (HLW)) and RWs with a considerable content of long-lived radionuclides, such as ILW-LL, which produce only negligible amounts of heat. Countries that possess these waste types typically have significant active programmes aimed at developing suitable geological repositories.

Direct experience of the geological disposal of HLW does not yet exist, as the only operating repository is the WIPP in New Mexico, USA, which has been licensed to dispose of transuranic RW (i.e. intermediate-level waste (ILW)) derived from the research and production of nuclear weapons. Several countries' disposal programmes for SF and HLW are, however, nearing fruition: what will be the access route to a repository for SF at Olkiluoto, Finland, is currently under construction; Sweden has recently chosen a preferred site for an SF repository at Forsmark; and France is investigating a potential disposal area on the border of the Departments of Meuse and Haute Marne, around the Bure site where the Underground Research Laboratory is located, to take all wastes not acceptable for surface disposal. In addition to the waste disposal programmes in these and other countries, international organizations such as the International Atomic Energy Agency (IAEA) and the OECD Nuclear Energy Agency (NEA) are contributing towards developing confidence in relevant technologies, approaches and concepts for the geological disposal of RW. These same organizations, in addition to others, such as the European Union, are also supporting international projects on training and demonstration, in line with the general principles defined in the IAEA Safety Fundamentals (IAEA 1995) and with the principle of sustainability. This has been defined by the Brundtland Commission as: 'development that meets the needs of the present without compromising the ability of future generations to meet their own needs' (WCED 1987).

The fundamental principles involved in geological disposal are discussed in, for example, Chapman and McKinley (1987), Savage (1995), Chapman and McCombie (2003) and Alexander and McKinley (2007). A key concept in this disposal is the multi-barrier principle, in which long-term safety is assured by a series of engineered and natural barriers that act in tandem (Fig. 1)—geological repositories are designed to be passively safe. These barriers prevent or reduce the transport of radionuclides in groundwater, which is generally the most important transport mechanism. The barriers may also influence the migration of gas, which will be evolved in RW repositories by chemical and biochemical reactions and by radioactive decay (e.g. Rodwell et al. 2003). For example, some radionuclides (such as ^{14}C) may be transported in the gaseous phase, being subject to many of the same transport processes as CO_2.

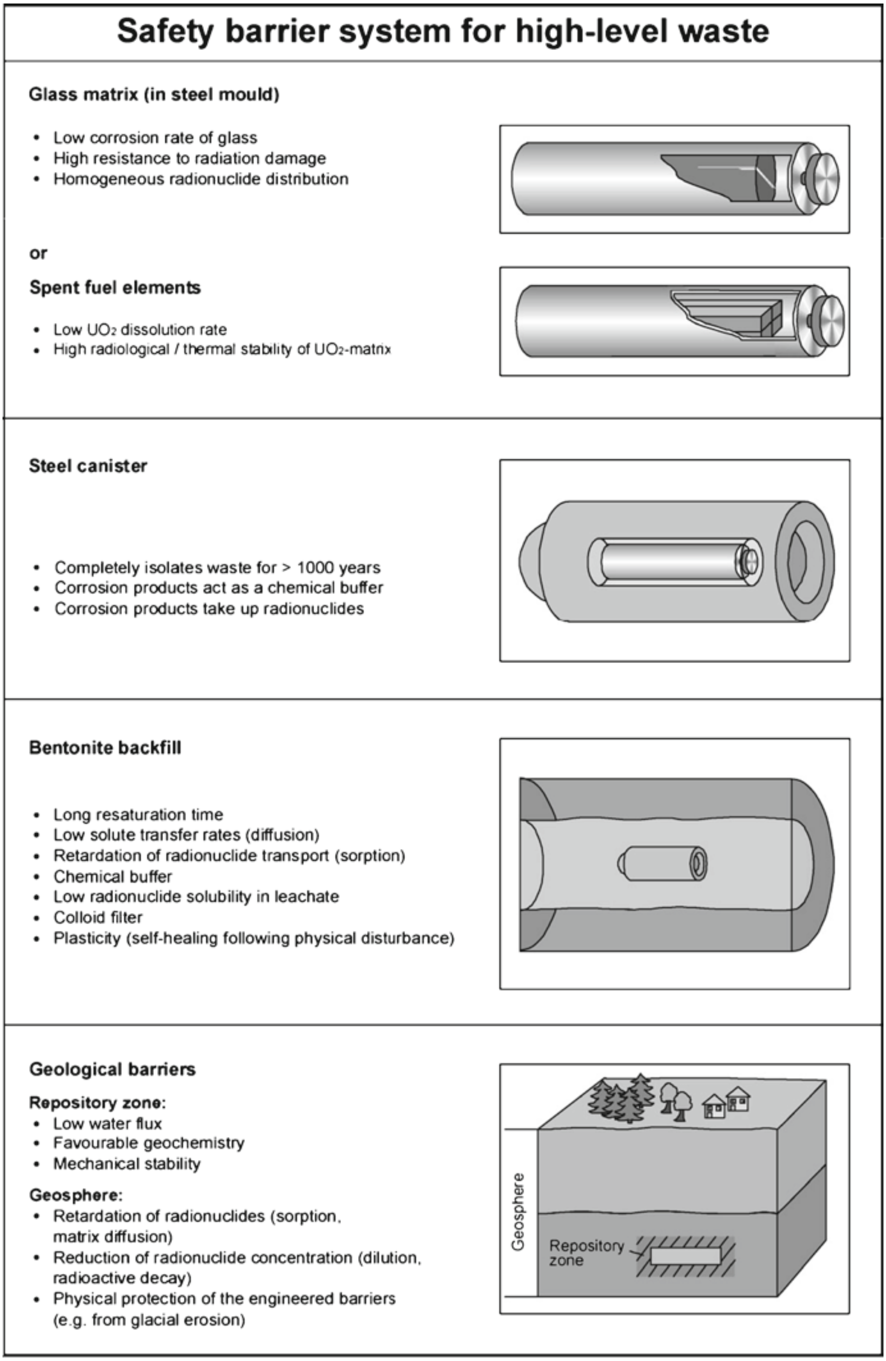
The long-term safety of a deep geological repository for RW will be strongly dependent on the performance of the geosphere. The geosphere potentially isolates the RW from possible future intrusions by humans; provides a stable physical and chemical environment for the engineered barriers within the repository, insulating against external perturbations such as earthquakes and climate change; and prevents, delays and attenuates radionuclide transport by virtue of its hydraulic and sorptive properties.

A safety case for a deep geological repository typically makes use of geoscientific information within a long-term safety assessment that evaluates potential impacts. These studies require a conceptual model of the geosphere that quantifies, for instance, groundwater flow rates and consequent radionuclide transport (as, eventually, the RW will come into contact with, and dissolve in, the groundwater—although this process may take place many thousands of years in the future). Geoscientific information can, however, play a larger role in the development of a safety case; in particular, geoscience can offer multiple and independent lines of evidence (both qualitative and quantitative) to support a safety case. Moreover, it can play an important role in other repository activities that bear on safety, such as site selection and repository design.

2 Current Status of CO_2 Disposal in Geological Media

2.1 Relevant CO_2 Properties

The concept of disposing of anthropogenic CO_2 by injecting it deep underground is based on the properties and behaviour of CO_2 at the conditions found at depth and on the physical and chemical properties of the rocks. At normal atmospheric conditions CO_2 is an odourless, colourless gas, slightly heavier than air, which is present in the atmosphere at concentrations of ~0.4%. Its density is 1.872 kg/m^3 at standard conditions of temperature and pressure. Like any substance, CO_2 changes phase from gaseous to liquid, solid or supercritical, depending on pressure and temperature (Fig. 2a). At very low temperatures CO_2 is a solid (dry ice), and is used as such



Safety barrier system for high-level waste

Glass matrix (in steel mould)

- Low corrosion rate of glass
- High resistance to radiation damage
- Homogeneous radionuclide distribution

or

Spent fuel elements

- Low UO_2 dissolution rate
- High radiological / thermal stability of UO_2-matrix

Steel canister

- Completely isolates waste for > 1000 years
- Corrosion products act as a chemical buffer
- Corrosion products take up radionuclides

Bentonite backfill

- Long resaturation time
- Low solute transfer rates (diffusion)
- Retardation of radionuclide transport (sorption)
- Chemical buffer
- Low radionuclide solubility in leachate
- Colloid filter
- Plasticity (self-healing following physical disturbance)

Geological barriers

Repository zone:
- Low water flux
- Favourable geochemistry
- Mechanical stability

Geosphere:
- Retardation of radionuclides (sorption, matrix diffusion)
- Reduction of radionuclide concentration (dilution, radioactive decay)
- Physical protection of the engineered barriers (e.g. from glacial erosion)

Fig. 1 The safety barriers for high-level waste, based on Nagra's disposal concept for use in Switzerland (From Nagra 2002)

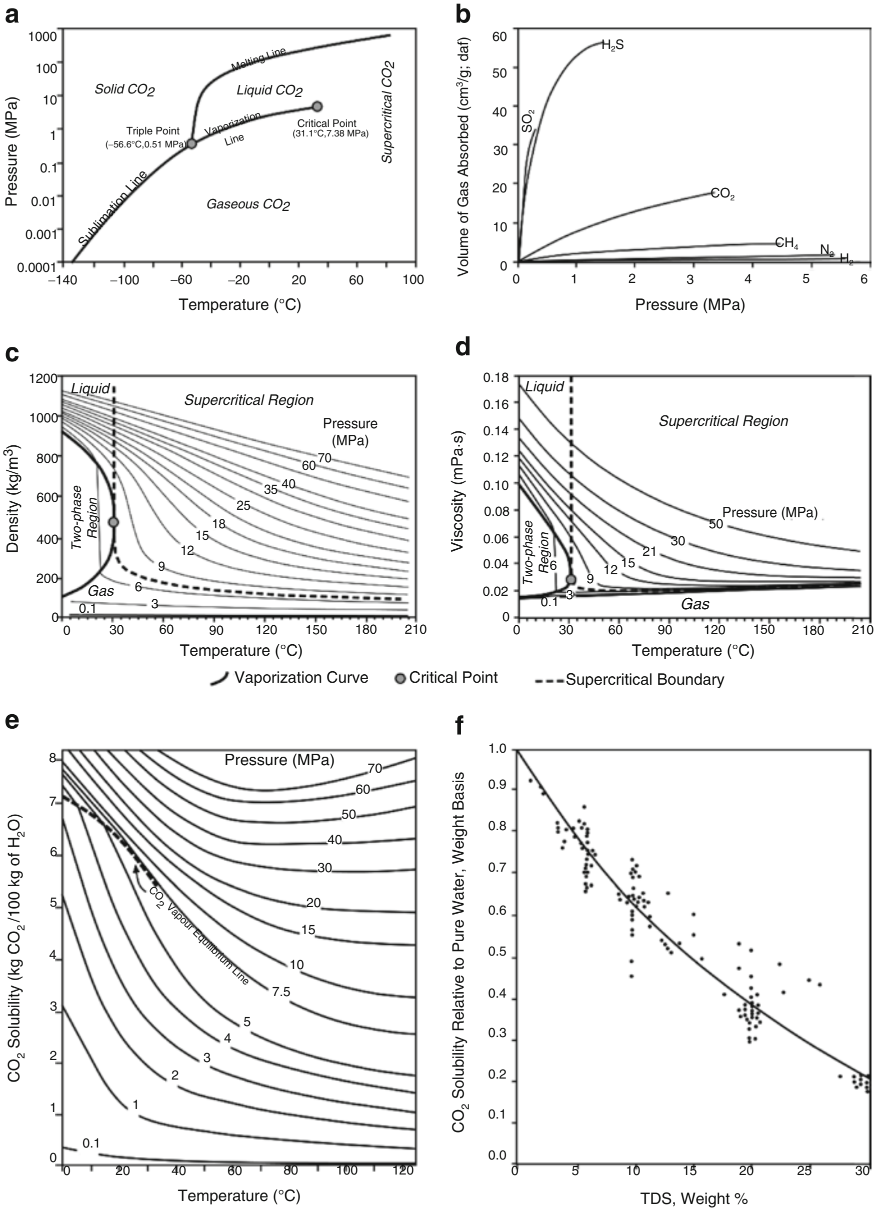


Fig. 2 Relevant CO_2 properties: (**a**) phase diagram; (**b**) adsorption capacity of various gases on coal (From Chikatamarla and Bustin 2003); (**c**) density variation with pressure and temperature (From IPCC 2005); (**d**) viscosity variation with pressure and temperature (From IPCC 2005); (**e**) solubility in water as a function of pressure and temperature (From Kohl and Nielsen 1997); and (**f**) decrease in solubility with increasing water salinity (From Enick and Klara 1990)

in industrial processes. However, except at shallow depths in Arctic and Antarctic regions and at high altitudes, where temperatures may be below 0°C, temperatures in the ground are always greater than zero and increase with depth according to the local geothermal gradient, whose global average is ~30°C/km, but which can vary widely, particularly in areas of active tectonics (e.g. volcanic regions and along the margins of tectonic plates).

At temperatures less than 31.1°C (the critical temperature, T_c) an increase in pressure will result in CO_2 changing phase from gaseous to liquid once it reaches the vaporization line (Fig. 2a). The pressure needed for CO_2 to change phase from gaseous to liquid increases with increasing temperature, reaching 7.38 MPa (the critical pressure, P_c) at the critical temperature, T_c (Fig. 2a). For reference, this pressure is equal to the hydrostatic pressure exerted at the bottom of a column of pure water at a depth of 738 m. For temperatures greater than the critical temperature, gaseous CO_2 becomes supercritical for pressures greater than the critical pressure. The characteristics of a supercritical fluid that are relevant for CO_2 disposal in geological media are that its density is comparable to that of the liquid phase (Fig. 2c) whereas it retains gas-like behaviour by filling the entire volume available and by mixing with other gases according to gas mixing rules. For temperatures below the critical point, CO_2 condensation from gas to liquid across the vaporization line (Fig. 2a) takes place gradually in the so-called ‘two-phase’ region (Fig. 2c) where the two phases coexist until all the gaseous CO_2 liquefies. The density difference at the vaporization line between gaseous and liquid CO_2 is sharp and significant, although decreasing along the vaporization line (Fig. 2c). For temperatures greater than the critical temperature, the transition from gaseous CO_2 to supercritical and the associated increase in density are gradual (Figs. 2a, c). The viscosity of CO_2, which depends strongly on its density (Fenghour et al. 1998), displays a similar behaviour (Fig. 2d). Notably, in the supercritical region CO_2 viscosity is closer to the viscosity of the gaseous phase than to that of the liquid phase (Fig. 2d).

The significance of this phase behaviour and of the variation of density and viscosity with temperature and pressure can be understood in the context of the increase with depth in the Earth’s crust of both pressure and temperature. Broadly, pressure increases hydrostatically with depth (i.e. with a gradient of ~10 kPa/m), although lower (sub-hydrostatic) gradients have been documented, and overpressurized zones have been identified, where pressure gradients approach lithostatic (20 kPa/m and higher). Thus, the increase in pressure with depth would normally lead to a continuous increase in CO_2 density. However, the corresponding increase in temperature associated with the same increase in depth leads to a decrease in density such that, after a significant increase in density with depth in the first few hundreds of metres, at a certain depth the two factors (pressure and temperature) balance each other, leading to a marginal increase in density, a constant value or even a decrease in density, depending on the interplay between mean long-term surface temperature, geothermal gradient and pressure (Bachu 2003). Assuming a hydrostatic pressure gradient, the density of CO_2 would be higher in regions characterized by a low mean long-term surface temperature and low geothermal gradient (up to 800 kg/m^3) than at the same depth (up to only 500 kg/m^3) in a region characterized by a high mean long-term surface temperature and/or high geothermal gradient (i.e. ‘cold basin’ versus ‘warm basin’ (Bachu 2003)). Correspondingly, the volume occupied by the same mass of CO_2 emplaced underground at the same depth will be smaller in the ‘cold basin’ than in the ‘warm basin’ case.

The void space in rocks at depth, in the form of pores or fractures, is saturated with fluids, the great majority of which is water, with oil and hydrocarbon gases accumulated in oil and gas reservoirs. Many gas reservoirs naturally contain CO_2 in various proportions, with several giant pure CO_2 reservoirs in the USA that are used to produce CO_2 for enhanced oil recovery (Stevens 2005). CO_2 dissolves in water, with its solubility increasing with increasing pressure and decreasing with increasing temperature (Kohl and Nielsen 1997; Fig. 2e); however, the presence of other dissolved substances reduces significantly the CO_2 solubility in water, by a factor of up to 5 (Enick and Klara 1990; Fig. 2f). Once dissolved in water, CO_2 forms a weak carbonic acid that, depending on the mineralogy of the rock, may lead to CO_2 precipitation in the form of carbonate minerals (Gunter et al. 2004). CO_2 has a greater solubility in oil and, depending on oil gravity and reservoir temperature, at pressures greater than a minimum miscibility pressure it mixes with oil (Holm and Josendal 1982). CO_2 mixes with other gases in gas reservoirs and with air in the unsaturated or vadose zone, although in the latter case, being heavier than air, it tends to accumulate at the bottom of the zone (Oldenburg and Unger 2003).

Finally, coal has variable adsorption affinity for various gases, including CO_2 (Chikatamarla and Bustin 2003; Fig. 2b). Coal has a higher affinity for CO_2 than for methane (CH_4) (also a greenhouse gas, which, for a given quantity, has 25 times greater global warming potential than CO_2 over a time horizon of 100 years) by a factor of 2–8, and for nitrogen (N_2), a gas that forms the majority of flue gases in fossil fuel power plants. Conversely, H_2S, found in gas reservoirs, and sulphur oxides (SO_x), found in flue gases, have greater affinities for coal than CO_2 (Fig. 2b). These adsorption properties are important because: (1) injecting CO_2 into coalbeds would replace methane, which should be recovered and used as a clean fossil fuel (it has the lowest carbon/hydrogen ratio), and (2) the CO_2 stream will most likely contain impurities in various proportions, and these, except for N_2, would preferentially adsorb onto the coal surface, with the advantage of retaining toxic substances such as H_2S and SO_x, but with the associated disadvantage of reducing the disposal capacity available for CO_2.

The properties of CO_2 on which its disposal is based are, therefore, its increased density with depth, its solubility in water and oil (with the associated potential mineral reactions) and its higher adsorption affinity onto coal than that of methane.

2.2 Geological Media for CO_2 Disposal

CO_2, being a fluid, will be disposed of at depth in rocks via well injection, and will retain its fluid characteristics and ability to flow as long as it does not precipitate as a carbonate mineral or adsorb onto coal. A decrease in pressure in coal will result in CO_2 being desorbed, with subsequent flow through any fractures present. Where it is dissolved in formation water or oil, CO_2 can be transported by the movement of the fluid and may exsolve when pressure and temperature conditions change, thereby regaining its free-phase form and its ability to flow. The geological disposal of CO_2 therefore needs to meet three requirements:

1. *Capacity*: the disposal unit has to have sufficient capacity to receive and retain the intended volume of CO_2;
2. *Injectivity*: which is the ability to inject CO_2 deep into the ground at the rate that it is supplied from the CO_2 source;
3. *Confinement*: if CO_2 is not confined, then, due to its buoyancy (being lighter than water, see Fig. 2c) it will flow upwards, ultimately entering the shallow hydrosphere (including potable groundwater), the biosphere and the atmosphere.

The first condition for CO_2 disposal requires the availability of large volumes of suitable rock (*capacity*). As an example, a coal-fired power plant that emits five million tonnes of CO_2 per year (Mt CO_2/year) would require a disposal volume of 10×10^6 m^3/year, or 0.4 km^3 over 40 years lifetime of emissions and an in situ CO_2 density of 500 kg/m^3. The volumes required for CO_2 disposal can be provided by the pore volume of the rocks or by mined caverns. At a porosity of 10%, the volume of rock needed to store the previously quoted storage volume is 4 km^3. Crystalline and metamorphic rocks have very low porosities unless they are fractured, and only sedimentary rocks, such as sandstones and carbonates, have generally sufficient connected porosity to provide the space needed for CO_2 disposal.

The second condition, *injectivity*, depends on the fluid viscosity and the permeability of the rock. CO_2 is less viscous than water by a factor of 10–20 and much less viscous than oil, which means that it is easier to inject CO_2 than water into the same rock, but, conversely, CO_2 is more mobile and may escape more easily than the other two fluids. Rocks that allow the production or injection of fluids (water, oil, gas) through wells are considered as permeable and, if they are saturated with water, are known as aquifers or, if they contain oil and/or gas, as reservoirs. Such rocks vary from unconsolidated gravel and sands to their lithified equivalents (conglomerates and sandstones) and also include carbonates. Other rocks, such as clays and shales and evaporites (such as halite), generally have such low permeabilities that they are referred to as aquitards or aquicludes in hydrogeology, and form caprocks, because they cap oil and gas reservoirs, impeding the flow of hydrocarbons out of the reservoir.

Capacity and injectivity are not completely independent of each other. Whilst the volumetric capacity, known also as static capacity (i.e. the necessary pore space), may exist, limitations in injection rates due to low injectivity (i.e. maintaining the maximum pressure below a certain limit imposed by safety measures) may reduce the amount of CO_2 that can be safely injected during the active injection period (this actual capacity is referred to as dynamic capacity).

The third condition for CO_2 disposal, *confinement*, requires the existence of impermeable rock units that would impede the upward migration and leakage of the injected CO_2. Sedimentary basins characterized by layered sequences of permeable and impermeable rocks, such as sandstone, carbonate, shale/claystone and evaporite, provide the type of geological environment that might prove suitable. In contrast, crystalline and metamorphic rocks do not meet any of the requirements for CO_2 disposal because of their lack of suitable porosity and permeability. Some volcanic rocks (e.g. basalts) may possess the required porosity and permeability, but generally they lack the necessary confinement properties. Mined caverns in soft or hard rock are also unsuitable for CO_2 disposal for a variety of reasons, including their low capacity (CO_2 would have low density because of the low pressures at the relatively shallow depths of such caverns) and the likely lack of confinement (which would have to be provided by engineered seals). Salt caverns formed via solution mining could allow the necessary pressurization through well injection, and confinement of the CO_2 would be ensured by the low permeability and plastic properties of the salt; however, such caverns would have only relatively low capacities (typically a fraction of 1 Mt CO_2 (Dusseault et al. 2004)), which would be insufficient for their use on the scale needed. Salt caverns mined through solution mining, not through regular shaft and tunnel systems, may, however, be used for the temporary disposal of CO_2, or as a buffer element in a CO_2 collection and distribution (i.e. transportation) system.

The conditions of capacity and injectivity are somewhat flexible, in the sense that some measures can be taken if any of these criteria are not being met. For example, injectivity can be increased by drilling more wells and/or drilling long horizontal wells and/or stimulating the wells whilst maintaining caprock integrity. Or, if capacity is insufficient, either several sites may be considered (e.g. store in the first site whilst the search and/or the preparation for another site is being pursued), or a smaller amount of CO_2 will ultimately be stored. But if the third condition, that of confinement, which basically relates to the safety and security of CO_2 disposal, is not being met, then that site will definitely not be considered and approved.

The above considerations indicate that the vast majority of crystalline, metamorphic and volcanic rocks are not suitable for large-scale CO_2 disposal; in addition, many sedimentary rocks also do not meet all three conditions. Sedimentary rocks that are faulted, folded and fractured generally do not meet the condition of confinement because CO_2 may escape along transmissive faults and fractures. For example, the Rocky Mountains in North America, which were formed by the compression and uplifting of sedimentary strata, are, generally, unsuitable because of their faulted and fractured nature, although storage structures can be found locally (e.g. oil and gas reservoirs in the foothills). Sedimentary basins, preferably with relatively simple geological histories and displaying minimal faulting and with successions containing at least one, if not several, low permeability confining units, are, thus, most likely to be suitable for CO_2 disposal (Bachu 2003, 2010; Bradshaw and Dance 2005; IPCC 2005).

Within sedimentary basins, aquitards and aquicludes (e.g. shales and evaporitic rocks such as salt and anhydrite) do not meet the requirement of injectivity and constitute barriers to the upward migration and leakage of CO_2. For reasons explained in more detail in the next section, the environments most suitable for CO_2 disposal are deep saline aquifers, oil and gas reservoirs and coalbeds.

In contrast to water supply aquifers that are normally relatively shallow, with low groundwater salinities (e.g. water with a salinity of less than 4,000 or 5,000 ppm for protected groundwater), deep saline aquifers are defined here as aquifers whose groundwater salinity makes them unfit for human consumption and that meet the necessary conditions for CO_2 disposal. Groundwater salinity may be in excess of 400,000 ppm, particularly in the vicinity of evaporitic beds (by comparison, seawater has a salinity of ~33,000 ppm), and in some places minerals dissolved in formation water are extracted for industrial purposes. In such cases, the respective aquifers constitute an economic resource that would be sterilized if used for CO_2 disposal.

Oil and gas reservoirs have properties similar to those of confined aquifers (i.e. permeable porous reservoir rocks capped by impermeable strata), but are saturated with hydrocarbon fluids (oil and/or gas) rather than water. The oil and gas would be produced first before any consideration could be given to the disposal of CO_2. In many cases oil and gas reservoirs are underlain by aquifers with which they are in hydraulic communication, and this factor would need to be taken into account. Coalbeds retain CO_2 as a result of a different process, but they too may constitute a resource that could be mined (or in which in situ combustion could be employed), or may represent aquifers by themselves due to their relatively high permeability, in which case they are not suitable for CO_2 disposal. Figure 3 diagrammatically illustrates the geological conditions and emplacement system for CO_2 disposal.

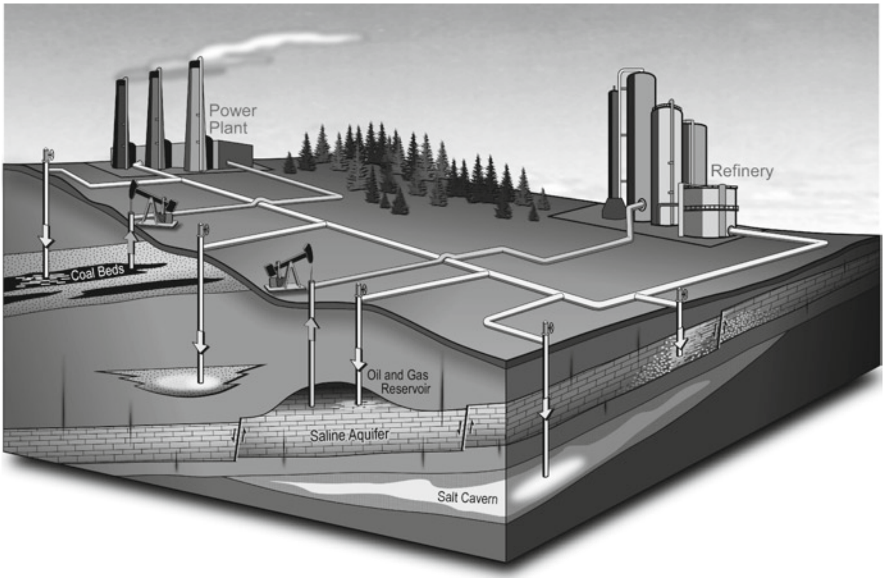


Fig. 3 Diagrammatic representation of the geological media and the transportation and injection system for onshore CO_2 disposal

2.3 Trapping Mechanisms for CO_2 in Geological Media

Long-term geological processes can result in the formation of oil and gas from organic rich shales, from which they are expulsed (primary migration) into adjacent aquifers. Once in aquifers, hydrocarbons flow updip along bedding and upwards, driven by their buoyancy (secondary migration), until they are trapped in geological regions in an aquifer, where changes in permeability impede any upward and lateral flow. This leads to oil and/or gas accumulation in what then become hydrocarbon reservoirs. The changes in permeability that form the trap for buoyant fluids (in this case oil or gas) are due to depositional and/or diagenetic changes (stratigraphic traps) or to the development of structural traps (due to folding and faulting) (Gunter et al. 2004). It is important to note that there are many such stratigraphic and structural traps in sedimentary basins that are not charged with oil or gas because they were not located along the hydrocarbon migration path. These stratigraphic and structural traps, saturated initially either with water (aquifers) or hydrocarbons (reservoirs), constitute the main targets for CO_2 disposal. Obviously oil and gas reservoirs, because of their economic value, may or will be used for CO_2 disposal only after their depletion. These traps can be very large in size (up to hundreds of square kilometers in areal extent and tens to hundreds of metres thick). CO_2 injected into these traps forms a continuous phase and can flow through the pore space, and actually will flow throughout the trap until steady state conditions are achieved, but it will not flow out of the trap. This type of trapping is called *stratigraphic and structural trapping*.

CO_2 is a non-wetting fluid that may flow through the rock pore space where it is continuous. However, when water (a wetting fluid) invades the rock previously saturated with CO_2, disconnected gas bubbles are caught in the pore space due to capillary snap-off, losing their ability to flow and becoming immobile at residual gas saturation. This is due to the hysteretic nature of the relative permeability of the two fluids, water and CO_2. Significant amounts of CO_2 can be trapped this way in the pore space in the wake of a migrating stream or plume of CO_2 (Kumar et al. 2005; Juanes et al. 2006; Ide et al. 2007). In this case there is no need for a stratigraphic or structural trap because the CO_2 is immobilized in the pore space. This type of trapping is called *residual gas trapping*.

As mentioned before, CO_2 in contact with water, either at the interface between a stream or plume of CO_2, or in each pore (non-wetting CO_2 against wetting water), will dissolve in water over a timescale of years to centuries (Gunter et al. 2004). Once dissolved, CO_2 loses its free-phase buoyant properties and will flow with the natural flow of water in the aquifer. Because CO_2-saturated water is heavier by approximately 1% than unsaturated water, if certain instability requirements are met, the heavier water will flow in a cellular pattern (free convection), dropping to the bottom of the aquifer, thus removing the CO_2-saturated water from the CO_2–water interface and moving it downwards whilst unsaturated water replaces it, in this way accelerating the process of dissolution (Ennis-King and Paterson 2003). This process is called *dissolution trapping*.

The weak carbonic acid formed by CO_2 dissolution reacts with rock minerals and may precipitate as carbonate rocks in what is called *mineral trapping*

(Bachu et al. 1994), in a process that usually takes centuries to millennia to deposit significant amounts of CO_2 as solid rock (Xu et al. 2003; Perkins et al. 2005).

If CO_2 is injected outside stratigraphic or structural traps in deep, regional-scale saline aquifers, whose size is in the order of tens to hundreds of kilometres and where formation water usually flows with velocities in the order of millimetres to centimetres per year, CO_2 will form a plume that will migrate updip along the strata but still below the caprock until it is immobilized through the combined effects of residual gas trapping, dissolution and mineral precipitation, regardless of the presence or absence of stratigraphic and/or structural traps along the migration pathway. This combined trapping mechanism is called *hydrodynamic trapping* (Bachu et al. 1994) lately known also as Migration Assisted Storage (MAS).

Finally, if injected into coalbeds, CO_2 will flow through the coal's natural system of fractures (cleats), diffuse through the coal's micropores, and adsorb onto the surface of the coal, displacing methane, in a process called *adsorption trapping*. It is desirable that the coalbeds into which CO_2 is injected be themselves overlain by impermeable strata to impede the upward flow of any excess CO_2 that is not adsorbed by the coal. Coal's permeability depends on the effective stress, which increases with depth and closes the coal cleats. Thus, coals tend to lose injectivity with increasing depth (McKee et al. 1988) such that coals at depths greater than 800–1,200 m cannot be used for CO_2 disposal because of lack of injectivity. In addition, CO_2 has the effect of swelling the coal (Cui et al. 2007), further closing the cleats and reducing permeability, and hence its injectivity.

The various CO_2 trapping mechanisms identified above can be variously classified as physical and chemical, or as primary and secondary. Physical trapping mechanisms are those where CO_2 retains its chemical composition: structural and stratigraphic trapping, and residual gas trapping. Dissolution, mineral and adsorption trapping are chemical trapping mechanisms. Hydrodynamic trapping is based on both physical and chemical trapping processes.

More important is the evaluation of CO_2 trapping mechanisms in relation to the duration of injection, which for a power plant or industrial process would be in the order of several decades (Fig. 4a). Primary trapping mechanisms are those whose timescale is comparable with that of the CO_2 injection, namely the emplacement of CO_2 in the trapping geological medium (Fig. 4a). These are structural and stratigraphic trapping, adsorption trapping and hydrodynamic trapping. A key characteristic of the disposal unit in all these cases is that it must have the necessary capacity to take all the CO_2 that is injected during the active disposal period. Residual gas trapping, dissolution and mineralization are secondary trapping mechanisms because they are dependent on the primary trapping (CO_2 emplacement) occurring first; they depend on CO_2 and water movement, and they operate on longer timescales, from centuries to millennia (Fig. 4a). On the other hand, the secondary trapping mechanisms contribute to increasing disposal security and a reduction in the risk with increase in time because, through CO_2 immobilization (residual gas trapping), dissolution and mineralization, less free-phase mobile CO_2 is left that may migrate and leak to the shallow hydrosphere, biosphere and atmosphere (Fig. 4b). The security of CO_2 disposal broadly increases, and hence the risk also

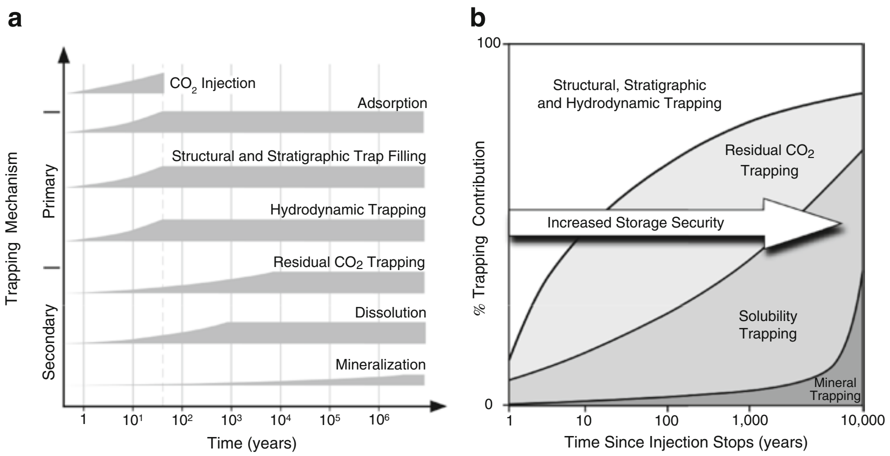
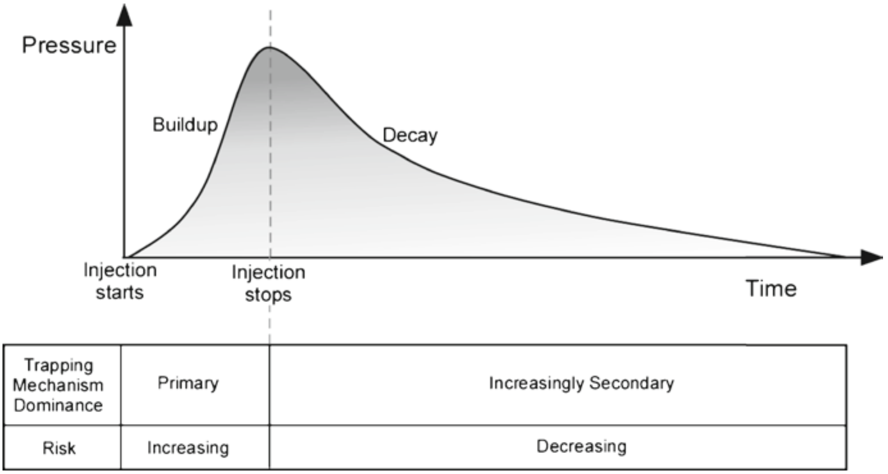


Fig. 4 Diagrammatic representation of the characteristics of CO_2 trapping mechanisms in geological media: (**a**) timescales for achieving full efficiency; and (**b**) variation in time of the amount of CO_2 trapped by various mechanisms when injected in deep saline aquifers (From IPCC 2005)



Trapping Mechanism Dominance	Primary	Increasingly Secondary
Risk	Increasing	Decreasing

Fig. 5 Diagrammatic representation of the pressure variation with time in a CO_2 disposal operation, of risk and of dominance of trapping mechanisms (After Bachu 2008a)

decreases, after cessation of CO_2 injection because, after injection ceases, the pressure, which increases continuously during injection, decays, thus reducing the driving force acting on the injected CO_2. The combination of pressure decay and the increasing role of secondary CO_2 trapping mechanisms leads to a decrease in the risk associated with CO_2 disposal after injection has ceased (Fig. 5). This scenario is generally true unless the plume of migrating CO_2 encounters a leaky well or an open fracture or fault, in which case the risk may locally increase as a result of leakage along this newly found leakage pathway.

2.4 Long-Term Fate and Potential Migration Mechanisms and Pathways

As discussed previously, CO_2 injected in deep saline aquifers or depleted oil and gas reservoirs may retain its form or may dissolve in aquifer brine or reservoir oil, or may precipitate as a carbonate mineral due to time-dependent processes. CO_2 injected into coalbeds will adsorb onto the coal surface. As long as CO_2 remains in, or, through exsolution or desorption, regains its original state, regardless of the phase (gaseous, liquid or supercritical), it will be subjected to hydrodynamic and buoyancy forces. The hydrodynamic forces are the result of injection (pressure forces) and of the natural flow systems in the injection aquifer. The buoyancy force is due to the in situ density difference between CO_2 and the groundwater or oil. If injected into porous rocks (deep saline aquifers or depleted gas reservoirs), CO_2 will, in addition, be subjected to viscous and capillary forces whereas, if injected into coalbeds, it will be subjected to molecular bonding forces. If the hydrodynamic and buoyancy forces are stronger than the capillary or adsorption forces, CO_2 will flow upwards if a pathway is available.

The transport mechanisms for free-phase CO_2 in porous media are diffusion and advection accompanied by dispersion. The former dominates in low permeability rocks such as shales, whilst the latter dominates in permeable aquifer and reservoir rocks and in fractures. Since the whole concept of CO_2 disposal is predicated on the existence of low permeability barriers that impede upward CO_2 flow, the issue is under what conditions these barriers could be breached, allowing upward CO_2 leakage. There are two possible mechanisms for the failure of the confining caprock caused by the injection of CO_2. Mechanical failure may take place due to hydraulic fracturing, the opening of pre-existing fractures or due to fault reactivation. This occurs when the injection pressure, which is highest at the injection well, exceeds a certain value P_m, equal to the minimum horizontal stress, if pre-existing fractures normal to the minimum stress direction are present or, in their absence, equal to the rock fracturing pressure. In general, mechanical failure is unlikely to occur because, during the injection stage, regulatory agencies limit the maximum bottom hole pressure at the injection well to values below the pressure corresponding to mechanical failure, and because of pressure decay in the post-injection stage (Fig. 5).

The other case of caprock failure occurs when the pressure at the interface between the CO_2 and the caprock exceeds the displacement pressure P_d (known also as the capillary entry pressure), above which water that saturates the caprock is displaced by the intruding gas (CO_2) phase. The capillary entry pressure depends on the interfacial tension (IFT) between CO_2 and water, which in turn depends on the in situ pressure, temperature and salinity conditions (Bachu and Bennion 2008) and is about half of that between methane and water (Chiquet et al. 2007). Usually P_m is smaller than P_d, particularly for low permeability rocks, whose capillary entry pressure is very high (Bennion and Bachu 2007), such that the integrity of the caprock is maintained by keeping the injection pressure below the threshold for mechanical failure. However, gas migration from gasfields has been documented (Gurevich et al. 1993). It is possible to have gas reservoirs that are overpressurized close to the displacement pressure

P_d corresponding to methane–water systems and, if these reservoirs are filled instead with CO_2 up to their initial pressure, it will exceed the displacement pressure for the CO_2–water system because of the lower IFT for the latter than for the former, resulting in CO_2 migration through the caprock. Even in such extreme cases, the timescale for leakage to occur will be very large (centuries to millennia and longer) because of the low permeability of the caprock and of relative permeability effects. The duration of CO_2 migration through the caprock depends not only on the caprock flow characteristics, but also on the caprock thickness.

Notwithstanding the possibility of CO_2 upward flow due to caprock failure, which has a very low probability, wells represent the most significant potential pathway for free-phase CO_2 leakage (Bachu and Celia 2009), as shown by documented natural gas leakage along wells in Alberta, Canada (Watson and Bachu 2007). The potential for leakage through wells is enhanced by the presence of CO_2, either in direct contact with well cement and casing, or dissolved in water, although under certain conditions well cement degradation is halted by the chemical reactions taking place in the presence of CO_2 (Scherer et al. 2005; Kutchko et al. 2007). Work to date seems to indicate that, depending on the type of cement used, if wells are properly drilled, constructed, completed and abandoned, the potential for leakage, including that of CO_2, is quite low due to the protective carbonate layer that forms when the CO_2-saturated brine reacts with well cement (Kutchko et al. 2007). However, preferential flow paths may be present due to pre-existing well defects, particularly in older wells, such as an annular space between the cement and the casing, poor bonding between the cement and the rock and cement fractures, which may be enhanced by the presence of CO_2 (Carey et al. 2007; Watson and Bachu 2009). Similarly, wells with cements that contain additives such as bentonite, or that have been stimulated through fracturing or acidizing, or that were abandoned with bridge plugs containing elastomers, will be more susceptible to CO_2 leakage (Watson and Bachu 2008).

In the case of free-phase CO_2 leakage through faults, fractures and wells, CO_2 will decompress relatively quickly as it flows upwards (due to the Joule–Thompson effect) and three-phase conditions will form, self-limiting the CO_2 flow rate due to three-phase relative permeability effects (Pruess 2004, 2005). On the other hand, in the case of diffusive transport across a caprock, or if CO_2 is dissolved in formation water that reaches the surface through faults, fractures and wells, the movement of CO_2 is extremely slow such that temperatures equalize and Joule–Thompson effects are avoided. In the case of CO_2 transport in solution, as the pressure decreases and the solubility drops, CO_2 will exsolve. The leakage rates in such degassing cases are very low and do not pose a significant risk (Shipton et al. 2005).

2.5 Geographic Distribution and Criteria for Site Selection

The selection of sites for CO_2 disposal has to consider the disposal requirements: confinement, capacity and injectivity. The confinement requirement implicitly includes an assessment of the long-term fate of the injected CO_2 and an assessment of the potential for leakage. From the analysis of the geological environments suitable

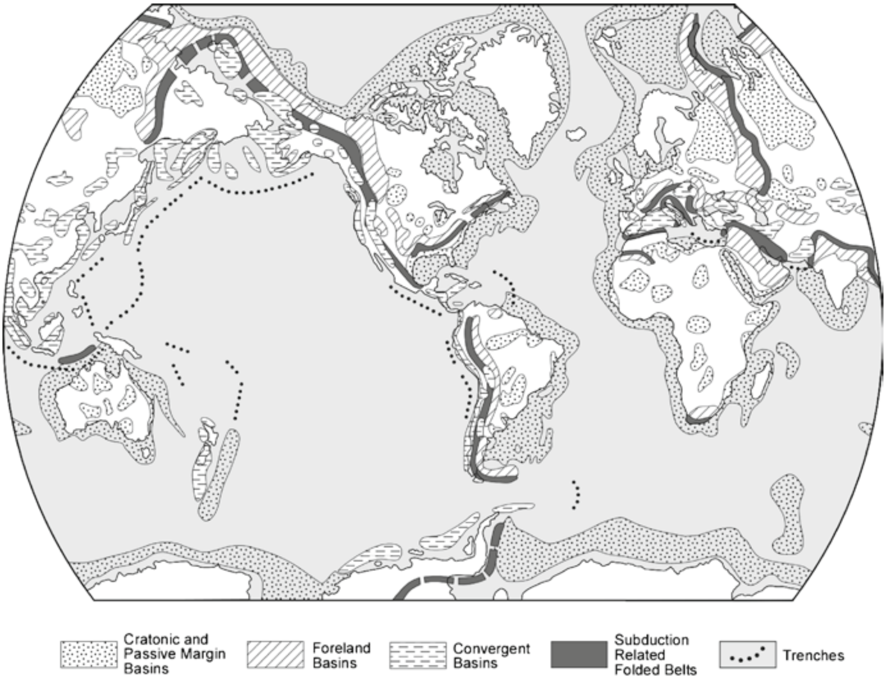


Fig. 6 Global distribution of sedimentary basins and their main types

for CO_2 disposal it is evident that only sedimentary basins could be considered, but even within these there are basins which are less favourable for CO_2 disposal, such as those located in areas of tectonic plate convergence, and basins better suited for CO_2 disposal, such as intracratonic and passive margin basins (Hitchon et al. 1999; Bachu 2003). Figure 6 shows the distribution and type of sedimentary basins around the world. It is instructive to note that circum-Pacific basins are of the convergent type, and hence are likely to be faulted and prone to tectonic activity and also tend to be comparatively small whereas circum-Atlantic basins and those around the Indian Ocean are large and of the passive margin type, which are more favourable to CO_2 disposal due to their simpler geological histories and more stable natures.

There are few sedimentary basins in Africa and Asia relative to their size, population and CO_2 emissions. In North America, foreland and intracratonic basins are found between the Rocky and Appalachian mountains, whilst in South America they are found east of the Andes Mountains. In Europe, foreland basins are found north of the Alps and the Carpathian Mountains and west of the Urals in Russia, but the sedimentary basin with the greatest potential is the prolific North Sea basin. Mediterranean basins are located in an area of plate convergence and possess all the associated unfavourable characteristics. Foreland basins in south-west and southern Asia are located south of the Zagros Mountains in Iran, where the major Middle East oil and gas resources are found, and south of the Himalayas in the Indian subcontinent. The main sedimentary basins in Australia with the largest CO_2 disposal potential are offshore.

Other criteria for assessing the suitability of a sedimentary basin for CO_2 disposal are its size, depth, geology and degree of faulting and fracturing, hydrogeological and geothermal regimes, and the presence of coals, oil and gas reservoirs, salt beds and deep saline aquifers (Bachu 2003, 2010). For example, a 'warm' sedimentary basin is less suited for CO_2 disposal than a 'cold' basin because, for the same depth, temperatures will be higher in the former, hence CO_2 density will be lower by a factor of up to two, leading to higher CO_2 buoyancy and lower efficiency in terms of the utilization of the pore volume. Other considerations are basin maturity (degree of exploration and production of oil and gas reservoirs, if present), accessibility and existence of infrastructure (e.g. roads, pipelines).

In terms of the potential for CO_2 disposal, another major element in site selection is the location of major stationary CO_2 sources (emitters) in relation to possible disposal sites (also known as source–sink matching). For example, there are many Arctic, sub-Arctic and Antarctic basins, many offshore basins, intracratonic basins in Africa or in the Amazon in Brazil (Fig. 6) that are too far from any significant CO_2 source amenable to capture and disposal; transportation of CO_2 by ship and/or pipeline to disposal sites in these basins would be uneconomic.

Even in countries that, overall, have sufficient CO_2 disposal potential, it may, in some cases, be located too far from large CO_2 sources. For example, in Australia the major CO_2 sources are located along the coast in the southeast (mainly coal-fired power plants), whilst the best sites for CO_2 disposal are offshore in the north-west (Bradshaw et al. 2002). In Canada, the capacity and potential for CO_2 disposal lies in the western provinces of Alberta and Saskatchewan, whilst the major sources of CO_2 in central Canada (Ontario) have no conveniently located disposal sites (Bachu 2003). In the USA, major CO_2 sources in the north-east and the Midwest (Ohio Valley) do not have sufficient CO_2 disposal capacity within an economic distance. Even if a sedimentary basin meets the criteria for CO_2 disposal in general terms, there will be regions within the basin that do not meet these criteria, particularly along the shallow edge of the basin or in faulted and folded regions. Such is the case of the Alberta Basin in western Canada—where major CO_2 sources related to the production of synthetic oil from tar sands are located in the north-east close to the basin edge, where there is no CO_2 disposal potential—or in south-western Ontario, where major coal-fired power plants and refineries are located on a sedimentary wedge less than 1,000 m deep that separates the Michigan and Appalachian Basins in the USA. In these cases, CO_2 captured at these large sources would have to be transported by pipeline, several hundred kilometres in length, to appropriate disposal sites.

Yet another consideration in the selection of CO_2 disposal sites is the type of geological medium. Countries with significant oil and gas reserves and in an advanced stage of exploration and production will most likely consider oil and gas reservoirs for CO_2 disposal, either at reservoir depletion or to increase production through CO_2 enhanced oil recovery (EOR). This is the case of countries in the Middle East and around the North Sea, and Indonesia and Mexico, but this is also a viable option in the USA and Canada. On the other hand, sedimentary basins in China and southern Africa are rich in coal, which puts them at a disadvantage because CO_2 disposal in coalbeds is an immature technology (IPCC 2005) and because coal is used for energy production and hence will not be available for disposal.

On a global basis and considering the major world CO_2 emitters, the distribution and type of sedimentary basins and the main disposal media, it seems that Asian countries along the Pacific Rim (i.e. Japan, South Korea, China) do not have sufficient CO_2 disposal capacity (Newlands et al. 2006), neither do India (Holloway et al. 2009) or South Africa. Middle Eastern countries (e.g. Saudi Arabia, United Arab Emirates) and European countries around the North Sea (e.g. Germany, UK, Norway) are likely to have sufficient CO_2 disposal capacity, although an extensive pipeline infrastructure would have to be built. Continental-size countries like the USA, Canada, Australia, Russia and Brazil appear likely to possess the necessary CO_2 disposal capacity, but in some cases there is a mismatch between the location of major CO_2 sources and disposal sites. In countries such as the USA, Canada and Russia and in the Middle East, CO_2 disposal will most likely be implemented onshore, whilst in northern Europe, Brazil and Mexico it is more likely to be implemented offshore.

On a local scale, site selection has to be based on the same criteria of confinement, capacity and injectivity. Additional criteria are protection from possible contamination of other energy and mineral resources and of groundwater, land ownership and rights of access, ownership of the ‘pore space’ (i.e. the right to inject CO_2), and infrastructure (roads, pipelines and wells). In some countries the subsurface is owned by the state, in others by both freeholders (individuals or private companies) and the state. Specific selection criteria for the case of oil and gas reservoirs are the degree of depletion, their suitability for EOR, reservoir heterogeneity, and the individual reservoir capacity (i.e. it is not economic to build the necessary CO_2 disposal infrastructure for reservoirs that will be quickly filled up). In the case of coalbeds, in addition to the standard criteria, the lack of any economic potential for the coal, now and in the foreseeable future, is a major consideration in site selection. If the coal is likely to be mined for power generation or for industrial use (e.g. steel making), or could be used for gasification or coal liquefaction to increase energy security and sustainability, then the coalbeds will not be used for CO_2 disposal. This is particularly important for countries endowed with large coal resources and with major energy needs such as the USA, China and India. Also, unlike deep saline aquifers and oil and gas reservoirs, the use of coalbeds for CO_2 disposal is limited to a narrow depth range because of their loss of permeability with increasing depth and in the presence of CO_2 and because shallow coalbeds are likely to have already been mined or lie in the depth range where the protection of groundwater resources is an issue.

A very preliminary estimate of the worldwide capacity for CO_2 disposal suggests that coals have the lowest potential at 15–200 gigatonnes of CO_2 (Gt CO_2), oil and gas reservoirs have ultimately a capacity of 675–900 Gt CO_2, and deep saline aquifers have the largest capacity at more than 1,000 Gt CO_2 (IPCC 2005). This should be compared with global annual emissions from fossil fuel use of approximately

25 Gt CO_2/year, of which emissions from large stationary sources (each greater than 0.1 Mt CO_2/year) constitute approximately 60%, or 15 Gt CO_2/year. The latter are clustered mainly in the midwestern and eastern USA, in central and northern Europe, eastern Asia (China, Korea and Japan), India and South Africa. If, in addition to the criteria of confinement, capacity and injectivity, other considerations for site selection (such as individual site size, access, economics, land ownership and use, and population distribution) are applied, the worldwide CO_2 disposal capacity is likely to become smaller by probably an order of magnitude.

2.6 Status and Challenges

CO_2 disposal in geological media has not yet been implemented as a mitigation measure for climate change, although CO_2 injection and disposal has occurred for different reasons in the last 3 decades.

The most significant experience with CO_2 transportation and injection exists in the Permian basin in west Texas, USA, where there are more than 90 CO_2 EOR projects, injecting approximately 30 Mt CO_2/year (Moritis 2006). Of the amount injected, approximately 60% is produced together with oil, and is captured and recirculated, whilst the other 40% remains in the ground. The oldest CO_2 EOR scheme in west Texas has been in operation since 1974. There are a few other CO_2 EOR operations in the world, the most notable being at Weyburn in south-eastern Saskatchewan, where approximately 5,000 t CO_2/day are injected. The Weyburn operation is a CO_2 EOR scheme, like all the others, except that it has been accompanied by a monitoring research programme (Wilson and Monea 2004).

The other important experience with CO_2 disposal has occurred in conjunction with the production of sour natural gas, which is natural gas that contains CO_2 and/or H_2S (both CO_2 and H_2S form a corrosive acid in the presence of water, hence the industry designation as ‘acid gas’ once these are stripped of the natural gas to meet pipeline and market specifications). As a result of regulatory requirements in western Canada that do not allow venting and/or flaring of H_2S, and because incineration or desulphurization of the acid gas are uneconomic, operators are increasingly turning to the geological disposal of acid gas in depleted hydrocarbon reservoirs and deep saline aquifers. Consequently, in 2007 there were close to 50 such operations in western Canada that have injected more than 6 Mt of acid gas since 1990, approximately half of which is CO_2 (Bachu and Gunter 2005). There are more than 20 such operations in the USA, mostly in Texas, Oklahoma and Wyoming, and new operations are currently being built in Iran and Kazakhstan. The main driver for these disposal operations is the need to deal with H_2S, which is a toxic hazardous substance.

Also worthy of note are two CO_2 disposal operations where CO_2 is stripped of natural gas that contains approximately 9–10% CO_2 and is injected on site into deep saline aquifers, with the gas being sent to markets in Europe. Both operations inject in the order of 1 Mt CO_2/year. The first one is at Sleipner in the North Sea, where the CO_2 has been injected into the Utsira formation since the mid-1990s, approximately 800 m below the seabed, and where a project for monitoring the fate of the injected CO_2 has been in operation (Torp and Gale 2003). The driver for the Sleipner operation is a carbon tax imposed by the Norwegian government on CO_2 emissions from offshore gas production, and in this regard this project can be considered as being a mitigation measure for climate change. The second operation is at In Salah in Algeria, which started in the mid-2000s and where the CO_2 is injected in the downdip water leg of the gas reservoirs that produce the gas containing CO_2 (Riddiford et al. 2003). A third operation started in 2008 at Mongstad, offshore Norway in the Norwegian Sea.

With regard to the injection of CO_2 into coalbeds, the only successful operation to date was run between 1995 and 2001 at the Allison Unit in the San Juan Basin, New Mexico, USA, as a pilot for enhanced coalbed methane (ECBM) production (Reeves 2003); however, no monitoring project was run in conjunction.

In addition to these commercial scale projects, there are a number of demonstration and pilot operations, mostly funded by governments, mainly for testing and developing technology for monitoring the fate of the injected CO_2 and developing monitoring techniques in the case of CO_2 injected into deep saline aquifers and depleted gas reservoirs (e.g. van der Meer et al. 2005; Hovorka et al. 2006; Förster et al. 2006). Pilot operations run to test CO_2 disposal in coalbeds in Canada, Poland, China and Japan have been less successful, mainly because of coal swelling in the presence of CO_2 (e.g. van Bergen et al. 2006; Yamaguchi et al. 2008; Wong et al. 2007).

These commercial and pilot scale operations indicate that CO_2 injection through wells does not pose any particular technological challenge. Generally, except for CO_2 disposal in coalbeds, the technology is mature and can be deployed immediately, at least on a demonstration scale (i.e. several large-scale operations, greater than 1 Mt CO_2/year each). However, there are still a few geoscientific and technical challenges that need addressing before the large-scale deployment of CO_2 disposal as a mitigation measure for climate change. These are:

1. *Resource mapping*: If the disposal volume that would be required for large-scale deployment is defined as a resource, there is a need to implement a sustained geoscience programme for the definition, identification, mapping and characterization of this resource.
2. *Timescale and effect of geochemical reactions*: If geochemical reactions between CO_2 and in situ fluids and rocks are likely to have a discernible effect over a time frame of millennia, then it may be possible to neglect them from a disposal point of view (where time frames of the order of a few centuries are likely to be more significant). Currently there is a divergence of opinion with regard to the geochemical effects associated with CO_2 disposal, particularly with respect to mineral trapping.
3. *Predictive modelling*: In order to properly predict the fate of the injected CO_2 over periods of time measured in centuries to millennia, there is a need to develop comprehensive mathematical and numerical models that couple multi-phase fluid flow, heat transfer and phase change(s), reactive geochemistry and geomechanical

effects of CO_2 disposal. Currently there are sophisticated models that treat one or two of these processes (e.g. flow and geomechanical, flow and geochemistry, flow and heat transfer, geomechanical and heat transfer), but there are no models that can treat three or more of these processes, because of the complexities involved, the nonlinearity of the system, and limitations in computing capabilities.

4. *Data collection*: There are insufficient physical and geochemical data, such as relative permeability and reaction kinetics, to characterize and model the fate of the injected CO_2 for the pressure, temperature and salinity conditions found at the disposal depths in various geological environments.
5. *Fate of wells*: Although wells have been drilled for more than 100 years with improving technology, there is no experience with the 'thousand year well', i.e. there is no experience with wells that should last as long as the CO_2 disposal operations should retain their effectiveness. This concerns existing wells, some from the nineteenth century, and new wells, both for CO_2 disposal and for other uses, mainly oil and gas exploration and production. This is essential for maintaining disposal efficacy (i.e. avoiding or minimizing CO_2 leakage). The magnitude of the problem is best illustrated by the following facts: there are more than 1,000,000 wells in Texas alone; there are more than 350,000 wells in Alberta, Canada, and new wells are being drilled at a rate of approximately 20,000 per year; generally there are no records about the completion and abandonment of old wells, particularly those drilled in the nineteenth century and early in the twentieth century. The fate of cement and casing in a CO_2-rich environment has to be understood and remediation measures have to be developed.
6. *Applicability of CO_2 disposal in coalbeds*: The loss of permeability due to coal swelling, and coal plasticization in the presence of CO_2 under certain conditions of temperature and pressure, severely limit the potential of coal to be used as a medium for CO_2 disposal. Coal is a brittle (glassy) material that becomes plastic at high temperatures and pressures. In the presence of CO_2 the temperature at which coal becomes plastic drops dramatically to around 30°C for pressures above 5 MPa (Larsen 2003).
7. *Effect of impurities*: CO_2 streams from power generation, energy production and industrial processes will contain various impurities, such as H_2S, SO_x and nitrogen oxides (NO_x), whose effects in the long term are not well understood. There is a trade-off between the increasing cost of purification and the fact that these impurities reduce the available disposal volume and may have a negative effect in the long term.
8. *Fate of displaced water*: Injecting such large volumes of fluid (liquid or supercritical CO_2) which are required to achieve climate stabilization targets would displace very large volumes of saline water, whose fate needs to be determined because they may have adverse impacts on potable groundwaters and the surface ecology if they migrate into shallow aquifers or to the surface.

There are other challenges facing the large-scale development of the geological disposal of CO_2, but they are of an economic, financial, legal and regulatory nature and are also likely to be linked to the attitude of the public to such developments (Bachu 2008a). These subjects are considered in other chapters of this volume.

3 Current Status of Radioactive Waste Disposal

3.1 What Are Long-Lived Radioactive Wastes?

Radioactive waste is defined by the IAEA (1994) as ‘material that contains or is contaminated with radionuclides at concentrations or activities greater than the clearance levels as established by the regulatory body, and for which no use is foreseen.’ National policy may consider some of the potential RW to be a resource, but this is likely to apply only to SF, which can be recycled to produce reusable plutonium and uranium for possible reuse in nuclear reactors. In other countries SF is considered a waste and is disposed of directly, although whether the SF is considered a resource or a waste is not necessarily based on an economic assessment, but often on political considerations. In this respect, RW is treated differently from other forms of hazardous and/or toxic waste.

RW is classified so as to determine how it should be handled and how suitable disposal options can be identified. The classification of the different types of RW varies from country to country and, as such, makes comparison difficult (see Vankerckhoven and Mitchel 1998). The IAEA has, however, implemented the Net Enabled Waste Management Database (NEWMDB) (www-newmdb.iaea.org), which attempts to harmonize waste definitions (Table 1) and these are used in this chapter.

The RW that is of interest here is the long-lived waste derived from the following sources that will require disposal in a geological disposal facility or repository:

- SF from reactors (which is heat emitting);
- Reprocessed SF, which results in the formation of HLW (which is also heat emitting) and other by-products, which are classified mainly as long-lived low- and intermediate-level waste (LILW-LL);

Table 1 Details of the waste classes defined by the IAEA

Waste class	Typical characteristics	Possible disposal options
Short-lived (L/ILW-SL)	Restricted long-lived radionuclide concentrations, e.g. long-lived α-emitters average <400 Bq/g or 4000 Bq/g maximum per package	Near-surface or (in some countries) geological disposal facility
Long-lived (L/ILW-LL)	Long-lived radionuclide concentrations exceeding limitations for short-lived wastes	Geological disposal facility
High-level waste (HLW)	Thermal power greater than about 2 kW/m^3 and long-lived radionuclide concentrations exceeding limitations for short-lived wastes (includes SF and HLW)	Geological disposal facility

L/ILW-LL long-lived low/intermediate-level waste, *L/ILW-SL* short-lived low/intermediate-level waste, *SF* spent fuel

- ILW from other sources such as reactor operations and decommissioning;
- Some countries, such as the UK, may also require the disposal of some long-lived low-level waste in a geological facility;
- Waste derived from military sources in countries that have nuclear weapons (this waste can be of a variety of types);
- Medicine and industry (although, again, the majority of this waste is not long-lived).

RWs need to be treated and conditioned to convert the waste materials into a form that is suitable for subsequent management, such as transportation, storage and disposal. The principal aims are to minimize the volumes requiring management via optimized treatment processes and to reduce the potential hazard of the waste by conditioning it into a stable, solid form that immobilizes it and provides containment. This is to ensure that the waste can be safely handled during its management. The processes used in this treatment and containment depend on the level of activity of the waste, with each country having its own waste management policy that influences the approach taken.

Many of the treatment methods, such as compaction and incineration, are applicable only to the shorter-lived wastes. Conditioning methods include cementation, bituminization and vitrification. Whilst the first two of these are applicable to ILW, vitrification is most commonly used for conditioning the highly radioactive liquors that result from reprocessing (where SF is dissolved in concentrated nitric acid to recover the uranium and plutonium, which can be reused), with the resulting glass being cast into stainless steel containers and then stored. SF is already in a reasonably stable waste form and its conditioning consists of placing it inside a metal canister. Canister designs vary, with existing designs including a copper canister with a cast iron insert (to be used in Sweden and Finland, e.g. SKB (2004) and Fig. 7) and a titanium–carbon steel equivalent, e.g. JNC (2000). Further information on waste sources and classification can be found in McGinnes (2007).

After nuclear fuel has been involved in the nuclear fission process, the fuel becomes intensely radioactive, largely as a result of the formation of new radionuclides, known as fission products, which reduces the efficiency of the reactor. After a few years the fuel needs to be removed from the reactor and becomes SF and, after some period of surface storage so as to reduce its heat output, it is normally placed in canisters. The storage time depends on the disposal concept considered, which in turn will determine the maximum acceptable temperature in the near field of a repository. It may also depend on other factors such as the regulations in the country in question.

HLW originates as a liquid residue from reprocessing SF to extract the uranium and plutonium for reuse, with the liquid containing most of the radioactivity from the original SF. It is commonly then evaporated to dryness and the residue containing the radionuclides then melted with a much larger volume of inert borosilicate glass-forming material to produce a homogeneous, solid, vitreous waste form. The glass is cast into stainless steel containers that are sealed and may be placed in an additional metal container for emplacement in a repository.

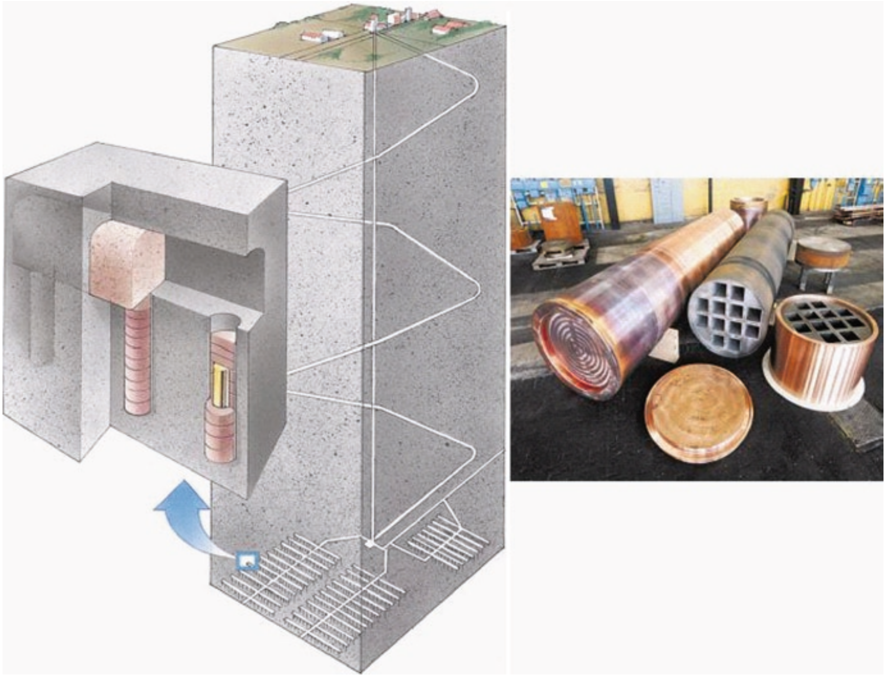


Fig. 7 The repository design proposed by SKB (Sweden) for the disposal of spent fuel in steel canisters sheathed with copper and emplaced within a bentonite buffer in disposal holes drilled into the floor of horizontal disposal tunnels (an alternative, but similar system has the waste canisters emplaced in horizontal disposal holes). The repository would be located at a depth of approximately 500 m in hard, fractured rock. A similar repository concept is being developed in Finland (Picture courtesy of SKB)

ILW can come in many forms. It arises principally from reactor operations, from reprocessing of SF and from decommissioning nuclear facilities. It is also derived from the production and decommissioning of nuclear weapons—and this is the primary source of wastes that are being disposed of in the WIPP repository in New Mexico, USA.

The volume of RW produced by the nuclear industry is very small compared with the other wastes generated. For example, in the OECD countries some 300 million tonnes of toxic wastes are produced each year, compared with 81,000 m^3 of conditioned RWs. In countries with nuclear reactors, RWs comprise less than 1% of total industrial toxic wastes. The volumes of RWs worldwide, as taken from the NEWMDB database (which includes the majority of the installed nuclear power capacity worldwide) were last updated in 2007, and are listed in Table 2.

Figure 8 shows a curve of relative radioactivity (compared with the radioactivity of the mined uranium ore) for typical SF (Swedish boiling water reactor fuel) as a function of time after discharge from the reactor, showing the early contribution of

Table 2 Volumes of declared waste arising worldwide

Class	In storage (m^3)	Disposed wastes (m^3)
L/ILW-SL	2,222,980	23,777,710
L/ILW-LL	3,127,681	10
HLW	365,404	0

From the NEWMDB database (www-newmdb.iaea.org) as of 2007. A much more detailed breakdown of the available data can be found on the NEWMDB website
L/ILW-LL long-lived low/intermediate-level waste, *L/ILW-SL* short-lived low/intermediate-level waste, *HLW* high-level waste

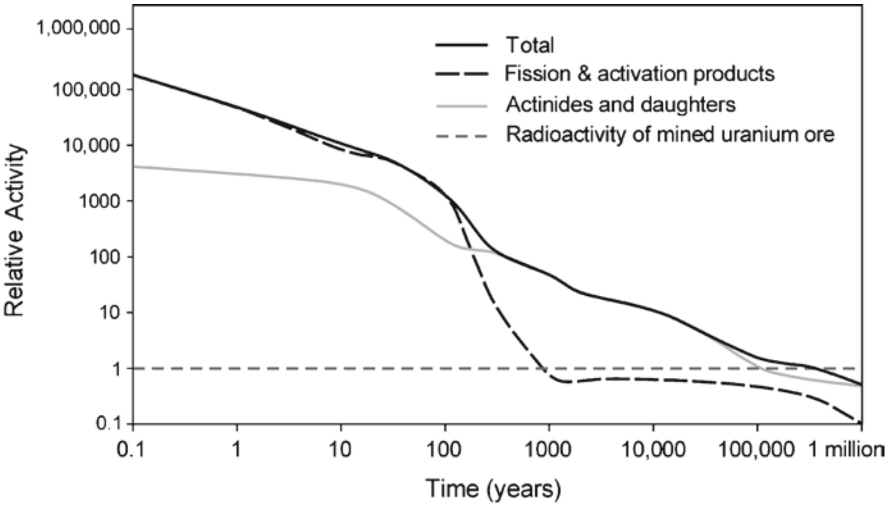


Fig. 8 The relative activity of spent fuel over time for SKB spent fuel, compared with the radioactivity of the mined uranium ore. (After Hedin 1997)

the fission and activation products. The sharp decline in fission product activity between 100 and 1,000 years is largely a result of the decay of ^{90}Sr and ^{137}Cs, both with half lives of about 30 years. After a few hundred years the actinide elements become dominant. After a few hundred thousand years the total activity of the fuel is similar to that of the uranium ore from which the fuel was produced. Other waste types will have different activity-time curves.

3.2 Geological Disposal of Long-Lived Wastes

The discussion below refers to the common form of the geological disposal of RW, in what is often referred to as a *mined repository*, or *disposal facility*, located at depth in water-saturated rocks. The site originally proposed for a repository for

HLW in the USA, at Yucca Mountain, is different in that it is located in the unsaturated zone and so some of the statements below, for example in relation to reducing conditions at depth, are not applicable. (Following an announcement in March 2009 regarding funding for the Yucca Mountain project, it is now certain that this will not be the site of an HLW repository.) In addition to a mined repository, there are other disposal concepts for geological disposal, such as deep borehole disposal, which are potentially suitable for only certain types of waste and which are different in certain specific regards from mined repositories (see McEwen 2004).

The multi-barrier system, introduced above, consists of two main elements:

- The engineered barrier system (EBS), which comprises the solid waste matrix and the various containers and backfills used to immobilize the waste inside the repository;
- The natural barrier (also referred to as the geosphere), which is principally the rock and groundwater system that isolates the repository and the EBS from the biosphere. The host rock is the part of the natural barrier in which the repository is located. In some cases the host rock is effectively equivalent to the geosphere, e.g. in the situation where the crystalline rock, in which the repository is located, extends to the surface.

The extent to which these two principal components act to provide containment, the way in which the different parts of the EBS control the behaviour of individual radionuclides, and the relative importance of the natural and engineered barriers at different times in the future evolution of the disposal system, constitute what is known as the *safety concept*, with what are referred as *safety functions* allocated to the different components of the system. The safety functions of the host rock are, according to Posiva (2008): (a) to isolate the repository from the biosphere and normal human habitat, (b) to provide favourable and predictable mechanical, geochemical and hydrogeological conditions for the engineered barriers, protecting them from potentially detrimental processes taking place above and near the ground surface, such that they contain the SF, and (c) to limit and retard inflow to and release of harmful substances from the repository. Similarly worded descriptions of the safety functions of the host rock or geosphere have been developed by other waste management agencies. Other safety functions are associated with the EBS (see below).

The safety concept can be different for each disposal system. Thus Fig. 7 provides a contrast with Nagra's disposal concept shown in Fig. 1.

3.2.1 The Natural Barrier or Geosphere

The natural barrier, or geosphere, is the rock that surrounds the disposal facility. As indicated in Table 1 and in the Introduction, there are certain requirements placed on the geosphere which will vary with the disposal concept considered, the geological environment chosen and with the time after waste emplacement. Emplacement of the waste in carefully engineered structures placed at depth in suitable rocks is chosen principally for the long-term stability that the geological environment provides (see item 1 in Sect. 4.1). At depths of several hundred metres in a tectonically stable environment, processes that could disrupt the repository are so slow that the rock and groundwater systems at depth will remain almost unchanged for perhaps hundreds of thousands of years, and possibly longer.

There is considerable flexibility in selecting a suitable geological environment for hosting a repository, as can be seen from the list of environments given in Table 3 for existing and proposed disposal facilities. The host rocks for disposal can vary quite widely, from hard, fractured rocks such as granite and gneiss through argillaceous rocks, mainly mudstones and clays, to evaporites, normally halite—and these rocks can be present in a variety of geological environments, from ancient basement terrains through to relatively young sedimentary basins. The argillaceous rocks and the evaporites, in particular, are chosen for their very low hydraulic conductivities, normally $<10^{-11}$ m/s (equivalent permeability in the range of 10^{-20} to 10^{-18} m^2), so that diffusive transport processes tend to dominate. Hard, fractured rocks are unlikely to have such low hydraulic conductivities but, even so, values at depths of several hundred metres and on the scale of tens of metres in suitable environments are likely to be $<10^{-10}$ to 10^{-9} m/s (equivalent permeability less than 10^{-17} to 10^{-16} m^2). All suitable disposal environments also need to possess chemically reducing conditions at depth (indicated by factors such as negative Eh and the presence of sulphides and Fe(II)). (See comment at the beginning of Sect. 3.2 with reference to Yucca Mountain, where conditions may be only locally reducing.) A useful discussion of the factors that are of greatest interest and concern regarding the properties of the rock mass and the hydrogeological and hydrogeochemical environment at depth is provided, for the case of hard, fractured rocks in Sweden, by Andersson et al. (2000). Similar considerations are likely to apply to any host rock although, of course, the strength of the sedimentary rocks, including evaporites, is considerably lower than hard, fractured rock, with the result that there will be notable differences in the repository concepts, depending on the type of host rock.

The disposal concept will, thus, vary with the type of geological environment under consideration, specifically the host rock, and also the waste forms for disposal. The relative importance of the natural barrier compared with the EBS will also vary, with host rocks in which solute transport is determined by diffusive processes (e.g. mudstones and halite) allowing the EBS to provide a more secondary, but nevertheless complementary, role (there is transport of solutes through all rocks, even halite, although at very low rates). This is in comparison with disposal in hard, fractured rocks, where the EBS, in the form of the bentonite buffer and the long-lasting canister, provides the dominant barrier to radionuclide migration (see, for example, Fig. 7).

There are important interactions between the natural and engineered barriers that lie at the heart of the multi-barrier principle. These are illustrated in Fig. 9 for the case of a KBS-3 type repository concept (as shown in Fig. 7) that is to be employed in Sweden and Finland. (KBS is an abbreviation for *Kärnbränslesäkerhet*, a Swedish term which means 'nuclear fuel safety'.) Similar interactions would exist for other disposal concepts for spent fuel or HLW. For the first 1,000 years, the EBS provides complete containment;

Table 3 Examples of the geological environments considered for hosting repositories for long-lived radioactive wastes

Geological environment	Location	Comments
Hard, fractured rocks (the geological environments of these three sites are similar –they all consist of old basement crystalline rocks)	Olkiluoto, Finland	Site of Finland's proposed spent fuel repository. Access ramp and shafts to what is planned to be the repository are currently under construction
	Forsmark or Laxemar, Sweden	Forsmark was chosen ahead of Laxemar in 2009 as the location of Sweden's spent fuel repository. Site investigations are complete at both sites
Mudstones (both the mudstones in France and Switzerland are Jurassic in age and are in structurally relatively simple geological environments)	Bure, France	A URL has been constructed and further work over the next decade is likely to result in the development of a repository for HLW and ILW-LL close, or quite close, to the URL
	Northern Switzerland	Investigations of the Opalinus Clay took place (see Nagra 2002); a site selection programme has been developed (which is currently under review by the regulatory authorities), and this formation may be chosen to host a repository for HLW and some ILW-LL
Evaporites (the host horizon at the WIPP is halite and other countries that have considered evaporites for disposal, such as Germany, have also chosen halite)	New Mexico, USA	The WIPP facility for military-derived ILW-LL (referred to as transuranic waste) has been operating for several years
		Gorleben, Germany, was for many years the proposed location for HLW and ILW-LL disposal. The German disposal programme was in abeyance for several years for political reasons, but is now active again, and the programme at Gorleben may be restarted

(continued)

Table 3 (continued)

Geological environment	Location	Comments
Volcanic tuffs (this geological environment is different from any other in the world, as the originally proposed repository was located in the unsaturated zone)	Yucca Mountain, USA	An extensive investigation programme was carried out at Yucca Mountain for many years, together with considerable safety case development and the construction of many kilometers of exploratory tunnels. It is now known that the site will not be developed as a repository

There are other geological environments being considered, such as plastic clay in Belgium—but these can be considered as subsets of the environments listed here

HLW high-level waste, *ILW-LL* long-lived intermediate-level waste, *URL* underground research laboratory, *WIPP* waste isolation pilot plant

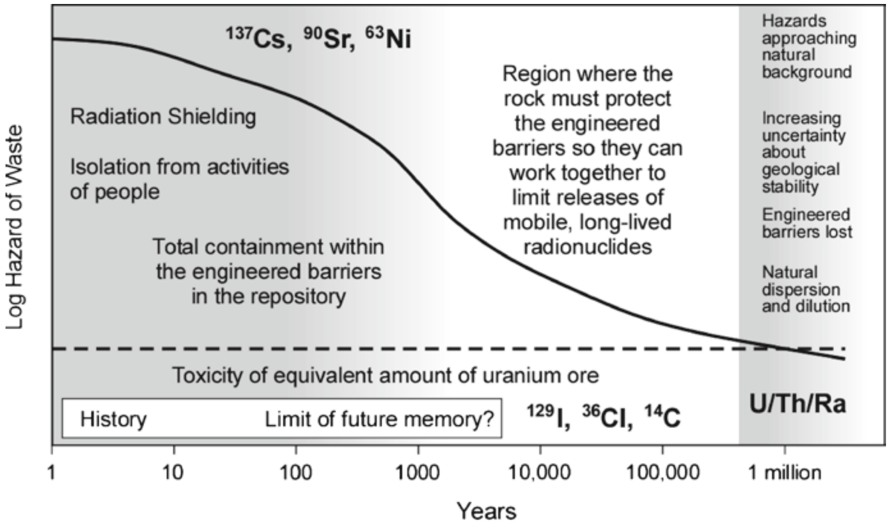


Fig. 9 The log of the hazard (relative to the hazard from the original uranium ore used to make the fuel elements) from spent fuel or high-level waste against time, illustrating the achievement of geological disposal in isolating the waste from the surface environment. The radionuclides which are of greatest significance in determining the hazard at different times in the future are also shown (From Chapman and McCombie 2003)

this period ends at approximately one to a few thousand years, following which, up to perhaps 100,000 years, the host rock is required to protect the EBS so that it can operate as planned and limit the release of the mobile, long-lived radionuclides. After this period, as discussed below, there is a gradual loss in the efficacy, or performance, of the EBS to limit the release of radionuclides.

After this period of up to 100,000 years, there are several factors that need to be taken into account when developing a safety case (see Fig. 9), as discussed below.

- *Geological stability can no longer be guaranteed*: at least not to the same extent that is possible up to this period. There are two separate components to this stability—the effects of climate change and tectonically related processes. For the first of these, regardless of the extent of future global warming, the Milankovitch forcing factors will ensure that glacial–interglacial cycling will reassert itself, with the result that ice sheets will advance and sea levels will change considerably, with a periodicity of approximately 100,000 years. For the second, it is more the effects of uncertainty as to what may take place that is important—for many geological environments, such as in the majority of Europe, tectonically related processes are unlikely to be significant for the next few million years as geological activity is relatively benign. The most important process over this period of time in Europe is likely to be uplift and erosion. For other, more tectonically active regions of the world, such as Japan, tectonic activity is likely to play a more significant role in locating a site for a nuclear waste repository and in developing a safety case. In such countries processes such as uplift and erosion, earthquakes, fault movement and volcanic activity may need to be an integral part of a safety case.
- *The hazard due to the waste is approaching the natural background*: it is also, by this period, likely to be below the toxicity of the uranium ore used to produce the fuel rods. In fact, the time after closure of the repository, when the crossover takes place with respect to the toxicity of the uranium ore, may be as little as 10,000 years. It could, therefore, be argued that it is necessary to demonstrate safety only up to this time following closure.
- *Engineered barriers are lost*: there can be no guarantee for times in excess of approximately 100,000 years that the EBS will maintain its essential functions. For example, the waste canisters will eventually degrade and allow the release of radionuclides; the compressed bentonite that might be surrounding an HLW waste canister is relatively thin and cannot be assumed to provide its diffusive barrier for ever—for example, it may be degraded by erosive processes or could undergo mineral transformations. Again, all these processes will need to be considered in a safety case.
- *Natural dispersion and dilution*: radionuclides released from the waste will be transported by flowing groundwater or diffuse away from the repository and be dispersed and diluted in the geosphere. The extent of this dispersion and dilution will depend on the types of rocks surrounding the host rock or the repository—on their porosities and permeabilities and on the hydrogeological environment in which the site lies, e.g. the hydraulic gradients and groundwater fluxes, etc.

The discussion above relates to the disposal of SF. For other waste types, in particular ILW-LL, which are likely to be surrounded by an EBS that will not restrict the release of radionuclides to the same extent, releases may occur earlier. The most likely conditioning methods for ILW-LL are cementation or bituminization, and such waste may be placed in vaults with additional cementitious backfill. In fact, it is likely to be more difficult to make a safety case for this type of waste than for the higher activity wastes, which may be present in considerably smaller volumes. This will be particularly significant in countries that carry out reprocessing of SF, such as the UK and France, where the volumes of ILW-LL are considerably in excess of those of HLW.

3.2.2 Engineered Barrier Systems

The type of EBS is linked to the type of waste, its conditioning and the disposal concept being considered. As indicated in the Introduction, this chapter concentrates on the geological aspects of waste disposal so that the discussion of the EBS is purposely limited in its extent. Examples of key engineered components of disposal systems currently being considered by waste management organizations (see, for example, Figs. 1 and 7) include:

- *Concrete or metal waste containers*: concrete and steel containers, although they may actually last for thousands of years, are generally conservatively assumed in safety analyses not to have any physical containment function after about 1,000 years. They can, however, buffer chemical conditions in the repository so as to limit the release and transport of radionuclides for very much longer times. Copper and titanium waste containers are expected to have a containment function for up to 100,000 years, although their corrosion may take even longer.
- *Backfill and buffer (around the waste)*: concretes can limit transport of radionuclides by diffusion for a long period and can also buffer the chemistry of the pore water and act as a sorbing medium for radionuclides; clays, such as bentonite, are naturally occurring materials which can provide a diffusion barrier for extremely long times.

Some disposal concepts place great emphasis on the protective roles of these EBS materials for protracted periods of time, the longest being the Scandinavian concepts for SF disposal in thick copper containers surrounded by a bentonite buffer (Fig. 7). Others rely more on the geochemical barriers in the near field of the repository and on dispersion and dilution in regions of the natural barrier system for some of the radionuclides. An example of this more chemically based approach is the phased geological repository concept developed by Nirex in the UK for the geological disposal of LILW-LL (Nirex 2005). There can be significant differences from one national programme to another, from site to site, and from one repository concept to another so that the role and the relative importance ascribed to each part of the multi-barrier system is very variable.

3.3 Implementation of Disposal Facilities

The development of a geological disposal facility, its operation and its final closure will take many decades. A proper legal and organizational framework must be established and a disposal strategy agreed with the various stakeholders before much progress can be made. The long timescales to implementation and the novel structure of the task mean that the activities themselves need to be carried out in a staged or stepwise manner.

The allocation of the functions for waste management and regulatory control is an important first step. In the majority of countries the regulatory task is left to the government and the implementation to those responsible for producing the waste, although there are exceptions regarding who is responsible for implementation as in some countries (e.g. the USA) this is also the responsibility of a government department. The nuclear power plant owners can join forces to form dedicated waste management organizations and there are many examples of these, e.g. Posiva (Finland), SKB (Sweden), Nagra (Switzerland), etc.

Following the establishment of an organizational structure within any country, it is necessary to formulate an overall waste management strategy. Such a strategy needs to include the key decision points, to decide how decisions will be taken and to ensure that sufficient resources will be available. Extensive guidance is available on such matters in international consensus documents produced by the IAEA. Any strategy, as indicated above, will need to be phased or staged: SF and HLW, for example, may need to be stored for several decades to reduce their heat outputs; waste repositories take several decades to develop (the combined effects of site selection, site characterization and construction); the repository operation is also likely to last several decades; and post-closure safety needs to be assured for many thousands of years. These extensive times have resulted in the development of a proposal for ‘adaptively managing’ such a staged development (National Research Council 2003). This implies adopting a flexible process in which the new knowledge gained at each stage is used to plan the content and duration of subsequent stages as opposed to defining in advance all the deadlines and milestones at the beginning of the programme. A useful review of repository implementation by McCombie (2007) includes a description of all the stages of the process, including the cost implications and the status of the disposal programmes in selected countries.

4 Comparison Between the Disposal of CO_2 and Radioactive Waste

A comparison between the disposal of CO_2 and RW is shown in Table 4, which is used to guide the discussion below regarding the similarities and differences between the disposal of the two types of substances.

4.1 Characteristics of the Geological Media

Four characteristics are discussed below: tectonic stability, the past stability of the site and the area in which it is located, the geological environment and the host rock type for disposal. These cover the main geological aspects considered in this chapter.

Table 4 Comparison of CO_2 and radioactive waste disposal, concentrating only on the geologically related issues considered in this chapter

Characteristic or attribute	CO_2 disposal	Radioactive waste disposal
Characteristics of the geological media		
1. Tectonic stability	Tectonically stable region preferred	Tectonic stability preferable, but still possible in tectonically active areas, such as Japan. Limitations regarding features such as active faults, Quaternary volcanoes, uplift rates, etc.
2. Past stability	Currently is not considered, or very little consideration is being given in a few projects	Important to understand and demonstrate past physical and chemical stability to increase confidence that such stability will continue into the future
3. Geological environment	In sedimentary basins in strata that • Have sufficient porosity (for capacity) and permeability (for injectivity) • Are confined by low permeability caprock (shales and/or evaporites) • Are minimally fractured, faulted or discontinuous	One in which • Groundwater fluxes at depth are sufficiently low • Reducing conditions exist within the disposal zone • Sufficient volume of host rock exists to house repository • Host rock has suitable geotechnical properties for underground construction • Geological complexity is acceptably low so that site can be adequately investigated and a convincing safety case developed
4. Rock type	Sedimentary rocks (sandstone, carbonate)	Hard, fractured (crystalline) rock, sedimentary rocks of various types (most probably mudstones and clays) and evaporites (most probably halite) (see Table 3)
Emplacement characteristics		
5. Mode of disposal	Injection through wells	Emplacement in (and from) tunnel and/or vault systems, i.e. in-tunnel, borehole/hole (both vertical and horizontal and both long and short) and vault emplacement. Considerable use of EBS, which can take a variety of forms
6. Volume	Very large (Gigatonnes, or 10^{10} m^3/year)	Comparatively very small (see Table 2) (total volume of long-lived wastes generated to date is approximately 4×10^6 m^3). A typical reactor generates approximately 30 t of packaged HLW per year

(continued)

Table 4 (continued)

Characteristic or attribute	CO_2 disposal	Radioactive waste disposal
7. Depth	>800 m up to >5,000 m	Relatively shallow: >200 m and probably <1,000 m (for the great majority of disposal concepts). One concept, deep borehole disposal, would require depths of up to 5,000 m approx.
8. Physical state	Fluid (mostly supercritical)	Solid
9. Containment mode	Natural barriers (shale and/or evaporitic layers) Most likely to have multiple natural barriers (confining strata)	Both natural and engineered barriers. Always a geological barrier acting in tandem with an EBS
10. Timescale of interest	Two timescales • Associated with global warming (greater than centuries) • Associated with local risks posed by injection and possible leakage (decades to centuries)	Widely discussed in radwaste community. Detailed, quantitative calculations required probably for at least 100,000 years, less quantitative for longer, possibly up to one million years (and possibly beyond)
11. Containment period	At least several centuries, up to millennia	Depends on disposal concept and waste types. Absolute containment, for some disposal concepts, could be >10^4 years and possibly as much as 10^5 years. For some waste types and disposal concepts (most likely for ILW-LL) absolute containment cannot be guaranteed for these periods. Releases are treated in a probabilistic manner and are acceptable if below the dose or risk target
Effects of emplacement and potential migration from the disposal site		
12. Direct effects of disposal	• Pressure increase	• Thermal effects due to radioactive decay (for heat emitting wastes)
	• Thermal effects due to cooling	• Geochemical reactions and processes in both the near and far fields
	• Geochemical reactions in the presence of formation water in a weak acidic environment	• Biochemical processes in both the near and far fields
	• Geomechanical effects as a result of pressure increase and stresses	• Geomechanical and hydrogeological effects due to repository construction and operation
	• Brine displacement	

(continued)

Table 4 (continued)

Characteristic or attribute	CO_2 disposal	Radioactive waste disposal
13. Effects on the natural barrier	No significant structural modifications to the geological environment caused by the engineered systems (wells), but the CO_2 itself may have effects on barrier integrity	The construction of the repository and the EBS employed will directly affect the natural barrier (although probably only locally). Also, heat emitting wastes will directly affect the natural barrier, although any effects will be limited to a few thousand years, at most
14. Transport mechanisms of CO_2 or radionuclides	The CO_2 itself (excess pressure due to its injection and also its buoyancy)	Dominantly via groundwater (advective and diffusional transport), but to a lesser extent via gas (produced by a variety of geochemical and biochemical processes in the near field and by radioactive decay). Transport of radionuclides can also take place in colloidal form
15. Return to the biosphere, hydrosphere and atmosphere	There are, in effect, no engineered barriers, and leaky wells, fractures and other local geological features may provide a pathway for the return of CO_2. Evaluating the potential impact of wells may be one of the key issues of assessing the performance of the disposal system	Considerable proportions of the long-lived wastes are encapsulated within containers that will remain intact for considerable times. Even after canister failure, the EBS will delay the release of radionuclides for further times (see Fig. 9)
Site activities		
16. Site characterization	Considerably simpler and shorter investigation programme. Considerably sparser information, based only on limited boreholes and seismic imaging	Very comprehensive and lengthy investigation programme. Eventual underground access allows considerably greater level of detail and certainty regarding the near field (a prerequisite for the development of a final safety case)
17. Monitoring	Monitoring required for baseline conditions (site selection), during injection, and decreasingly after cessation of injection for site closure and ensuring long-term safety of the system	All disposal concepts have extensive barrier systems, so, although monitoring will be a requirement, no releases are likely to be detected for a considerable time (i.e. several millennia) after closure. Monitoring is obviously linked to the possibility of waste retrieval and/or the reversibility of the disposal process

(continued)

Table 4 (continued)

Characteristic or attribute	CO_2 disposal	Radioactive waste disposal
18. Future access, intrusion or penetration	Penetration by future wells drilled for other purposes (e.g. exploration, production) is quite possible. Mining of coal is also possible	Sites will be selected only in areas where the intrusion risk is considered low (i.e. no mineral resources)
19. Retrievability of the waste	This is not an issue, except with regard to specific cases mentioned in Sect. 6	Retrievability of the waste has implications for repository operation and for the design of the monitoring programme. It does not necessarily influence the type of disposal environment selected nor does it necessarily exclude specific host rock types from consideration

EBS engineered barrier system, *HLW* high-level waste, *ILW-LL* long-lived intermediate-level waste

1. *Tectonic stability*: Relatively benign and stable tectonic conditions are preferred for both forms of disposal. (It is important to appreciate what is meant by the term *stable* in this regard. It does not imply that conditions at depth are unchanging, but that they change only slowly or, in most cases, extremely slowly. In fact, what is most significant is that they can be shown to change *sufficiently slowly* to ensure the long-term safety of the repository; see NEA 2005, 2009.) The presence of stable conditions is likely to provide an intrinsically safer disposal environment (and is also likely to make the demonstration of long-term safety an easier task); however, it has been shown that the presence of active tectonics does not preclude the possibility of disposing of RW (see also item 5 below in relation to the mode of disposal and item 10 in relation to the timescales involved of interest with regard to tectonic stability). An interesting discussion of this subject in relation to the siting of nuclear facilities (including waste repositories) is provided in Connor et al. (2009). It is not normally possible, especially in respect of waste repositories, to make specific statements regarding the required level of tectonic stability, except with regard to specific features, such as active volcanoes. It is probably better to develop preferences in this regard, for example the separation of any repository from an active fault, with any such preferences being related to the implications for the long-term safety of the facility. Japan provides an example of a country where the level of tectonic activity is of particular interest for the disposal of RW. Japan has had an RW disposal programme that has been operating for many years and the Nuclear Waste Management Organization of Japan (NUMO) is confident that it will be possible to dispose of long-lived RW in Japan, even though the entire country is tectonically active, with numerous active faults, areas of active volcanoes and areas with geologically rapid uplift rates. A series of reports produced as part of the H12 project in Japan (JNC 2000), combined with a large R&D programme, have led to the development of a structured approach to the

development of a repository for long-lived wastes (NUMO 2007), in which it is envisaged that a repository for HLW will be available for use in 2035. (The H12 project, whose full name is the Project to Establish the Scientific and Technical Basis for HLW Disposal in Japan, is named after the 12th year of the Heisei era—related to the current Emperor.) As a contrast, reference can be made to Posiva's proposed repository site in Finland, which is a site in a tectonically stable environment (see McEwen and Andersson 2009). Even in this stable environment, the possibility of future rock movements needs to be taken into account—in this case it is the effect of future glaciations and their impact on fracture displacements that is potentially important in locating waste canisters. Two recent reports on the significance of geological stability (which is a broader subject than just tectonic stability) in the context of the disposal of RW are of interest here (NEA 2005, 2009). These reports, which represent the outcomes of two workshops, discuss the significance of geological stability in all its forms, i.e. mechanical, hydrogeological, hydrogeochemical, etc., with reference to the use of argillaceous and crystalline rocks for disposal purposes. One of the conclusions of these reports is that, as long as disposal sites are selected with care, there would appear to be no significant reasons why RW cannot be disposed of safely at depth, even in areas with relatively high levels of tectonic activity. With regard to CO_2 disposal, although to date most existing pilot and commercial scale operations are in tectonically stable regions, such as the North Sea, Sahara and the Williston Basin in Canada, the Westcarb Regional Partnership on CO_2 Sequestration in the USA is looking at identifying and piloting CO_2 injection sites in California, and in Japan pilot demonstrations took place in a coalfield at Ishikari (Shi et al. 2008) and in a deep saline aquifer at Nagoaka, where an earthquake of magnitude 6.8 with an epicentre distance of 20 km occurred without adverse effects on the injected CO_2. Both California and Japan are located in tectonically active regions around the Pacific Rim. Nevertheless, as in the case of RW, stable areas are preferred and volcanic areas and sites close to active faults should be avoided.

2. *Past stability*: This is currently not explicitly considered in the disposal of CO_2, although it is suggested that this subject may become part of the assessment of a proposed CO_2 disposal facility (Maul et al. 2007; Maul 2011). Site stability in CO_2 disposal is implicitly considered under tectonic stability (see point 1 above). In the disposal of RW it is an important requirement to demonstrate such past stability, and in fact to understand the evolution, over perhaps the last 100,000–1,000,000 years, of the site and the area in which it is located, in order that a convincing account can be developed of its likely evolution, and hence its continuing stability, in the future. A very considerable amount of work is required to produce such an account. One field in particular is of interest in this regard, that of the use of palaeohydrogeology, where studies are made of the past hydrogeological evolution of a site. A useful review of the use of such data is presented in the conclusions of the PADAMOT (Palaeohydrogeological Data Analysis and Model Testing) programme in Degnan et al. (2005). The two NEA reports on geological stability (NEA 2005, 2009), referred to in item 1 above, are also of relevance here, and several of the presentations given at these workshops concern the past stability of a site.

3. *Geological environment*: The range of geological environments currently considered suitable for the disposal of CO_2 is smaller than those considered suitable for the disposal of RW. The disposal of CO_2, as currently accepted, can take place only in sedimentary basins, in environments that have the characteristics listed in Table 4, e.g. have sufficient porosity (for capacity) and permeability (for injectivity), are minimally faulted and/or discontinuous (see Sects. 2.2 and 2.3), and are confined by low permeability caprock (shales and/or evaporites) that is not fractured. Lately there has been interest in exploring the potential for CO_2 storage in basalts due to the fact that rapid geochemical reactions are likely between the injected CO_2 and the basalt, and two test sites are being pursued to explore this concept, one in the USA and another in Iceland in the Hellisheidi hydrothermal field. However, it is worth mentioning that in Iceland the concept consists of dissolving the CO_2 in water at the surface and injecting the CO_2-saturated water, rather than injecting free-phase, high density CO_2 which otherwise will most likely leak due to the characteristics of the basalt. RW can be safely disposed of in a variety of geological environments, i.e. a repository can be located in an argillaceous rock, in an evaporite (either bedded or domal in form) and in a wide range of hard, fractured rocks, any of these existing in a wide range of geological environments, from Archean basement terrains to relatively recent sedimentary basins. As long as the environments have the characteristics and properties listed in Table 4, they are likely to be suitable. Probably the most significant of these are that groundwater fluxes at depth are sufficiently low and that chemically reducing conditions are present (and that they remain reducing) (see Sect. 3.2). Another important factor is the requirement for there to be sufficient volume of suitable rock to house the repository. The location and layout of disposal tunnels or vaults may be most constrained in crystalline rock, due to the ubiquitous presence of fracture zones, which need to be avoided. The design and layout of the repository is likely to vary considerably between different types of geological environments and can be modified to take into account the specific characteristics of the site in question.
4. *Rock type*: The disposal of CO_2 is likely only in sedimentary rocks with sufficient porosity and permeability, such as sandstones and carbonates, although disposal in salt caverns is also possible (see Sects. 2.2 and 2.3). If proven successful and economic, storage of CO_2 in basalts may be considered sometime in the future. RW can be disposed of in a larger range of rock types, including many types of hard, fractured (crystalline) rocks, sedimentary rocks of various types (but probably most likely mudstones and clays, as these have the necessary low permeabilities) and in evaporites (most likely halite) (see Sect. 3.2).

4.2 Emplacement Characteristics

Seven attributes, associated with the characteristics of the emplacement of CO_2 or RW, are considered below: the mode of disposal, the volume of the waste, the disposal depth, the physical state of the waste, the mode of containment, the timescale of interest and the containment period.

5. *Mode of disposal*: The disposal of CO_2 will take place via direct injection from wells. In contrast, RW will be emplaced in or from tunnels or in vaults, e.g. in tunnels (e.g. Fig. 1), in boreholes or holes (both horizontal and vertical) drilled from disposal tunnels (e.g. Fig. 7).
6. *Volume*: The volume of CO_2 that requires disposal is extremely large, in the order of gigatonnes (equivalent to billions of cubic meters) per year whereas, in comparison, the volumes of RW, especially HLW and SF, are very small—many orders of magnitude lower. As of 2003, the total volume of all long-lived RWs, both in storage and already disposed of was approximately 650,000 m^3. Table 2 lists some figures which indicate some of the volumes of RW that require disposal. The total volume of conditioned RWs produced per year in all the OECD countries is 81,000 m^3, and much of this waste is relatively short-lived and does not require geological disposal.
7. *Depth*: The disposal of CO_2 is likely to take place in the range 800–5,000 m, for the reasons given in Sect. 2.1. The disposal of RW will almost certainly take place over a smaller and shallower depth range (probably >200 m, and more likely >400 m and <1,000 m) with the exception of the deep borehole disposal concept, where disposal could take place at depths as great as approximately 5,000 m (see McEwen 2004).
8. *Physical state*: CO_2 will be disposed of as a fluid, and most likely a supercritical fluid, whereas RW will definitely be disposed of as a solid (but see item 5 above).
9. *Containment mode*: No engineered barriers are present in the disposal of CO_2 (at least not in the disposal zone itself) and, given the size and nature of the disposal operations, none can be constructed; although, of course, the necessary boreholes are sealed so as to prevent leakage, and there has to be at least one, but preferably several, natural barriers (i.e. confining strata) in the geological succession (see Sects. 2.2 and 2.3). An important axiom of RW disposal is the necessity of having multiple barriers (normally referred to as the multi-barrier concept), in which an EBS (see Figs. 1 and 7 and discussion in Sect. 3.2) acts in tandem with the geological barrier. The EBS is likely to be composed of several components (each of which is itself a barrier). The relative significance given to the EBS and the geological barrier in the safety concept will vary with the type of repository under consideration and, in particular, with the geological environment for disposal, but both types of barrier will always be present.
10. *Timescale of interest*: There are two timescales currently considered for the disposal of CO_2. The first of these is that associated with global warming, which is likely to be in the order of centuries to millennia, and the second is that associated with local risks related to the injection of the CO_2 and to possible leakages (Figs. 4 and 5), in the order of decades to centuries. The first timescale is associated with the stabilization and subsequent decrease of CO_2 concentration in the atmosphere, whilst the second timescale is associated with the immobilization of CO_2 in the ground and the disappearance of the potential for and risk of leakage.

The timescales for the disposal of RW are considerably longer and the distinction between the different timescales considerably more complicated. Figures 8 and 9 illustrate the decreasing risk or hazard of the waste with time, so that the events that are of most interest in the short term, i.e. during the operational phase of the repository and immediately post-closure (less than about 150 years), will be different from those in the far future, and the contribution of the different radionuclides will change with time. Detailed, quantitative calculations of doses or risk will be required for at least 10,000 years, and more likely as much as 100,000 years, with more qualitative calculations and reasoned arguments being presented for times up to, and possibly exceeding, one million years. Figure 8 shows that for SF the toxicity of the waste is similar to that of the uranium ore from which the fuel was fabricated after about 100,000 years; for HLW this period will be less, of the order of 10,000 years. The timescales of interest can be related to this changing toxicity of the waste, and therefore the risk associated with its disposal, so that several waste management organizations have developed timescales of interest related to the development and evolution of the repository, for example:

- Operation (approximately 100 years);
- Near future (post-closure monitoring phase, phase of global warming, no expected release from repository via groundwater pathway, etc.) (perhaps up to 1,000 years);
- Period where the integrity of the EBS is guaranteed, where climate change does not include glacial phases, where there is considerable confidence in the behaviour of the disposal system (up to 10,000 years approximately);
- Period during which the toxicity of the waste approaches and/or equals the toxicity of the uranium ore, when there is the possibility of major climate change and when the confidence in the behaviour of the disposal system may be considerably reduced (10,000–100,000 years approximately);
- Period when the processes and events associated with the disposal system are those illustrated in Fig. 9 and when major climate change may be even more significant (>100,000 years).

11. *Containment period*: For CO_2 disposal, containment will likely be required for several centuries up to millennia, although to what extent this needs to be absolute containment, or whether such absolute containment is indeed possible, is unclear. There are some organizations that, by analogy with RW disposal, are suggesting that containment (absence of leakage) should be demonstrated for 10,000 years, but so far this view has not gained acceptance. Recognizing that some CO_2 leakage may be unavoidable, several studies have suggested that, from the point of view of climate stabilization, small global leakage rates (0.01–1%/year) would still be acceptable (Pacala 2003; Hepple and Benson 2005; IPCC 2005). However, these are peak leakage rates, and the long-term average permissible leakage rates should be much lower. The extent of absolute containment for RW (i.e. no escape from the EBS) depends on the waste type and disposal concept and could vary from less than 10,000 years (perhaps as little as approximately 1,000 years) to as much

as 100,000 years—possibly as much as one million years in some circumstances. Some disposal concepts assume that the waste containers have no containment function immediately after closure of the repository—although this does not imply that release of radionuclides takes place immediately. There may be problems with the escape of radioactive gas relatively soon after repository closure, especially in the disposal of ILW-LL, although whether this gas ever enters the biosphere depends on several factors, many of which are site specific. Releases are treated in a probabilistic manner and are acceptable if the consequences of such releases are below a risk or dose target.

4.3 Effects of Emplacement and Potential Migration from the Disposal Site

The subjects discussed below concern the effects of the disposal of CO_2 or RW and mechanisms by which they could migrate from the disposal site. These include: the direct effects of disposal, the effects on the natural barrier, the transport mechanisms of CO_2 or radionuclides and the possible return to the biosphere, hydrosphere and atmosphere.

12. *Direct effects of disposal*: For CO_2 disposal there are a variety of effects resulting from the emplacement and presence of CO_2 in the subsurface, such as pressure increase, stress changes and deformation, and geochemical reactions, including mineral dissolution and/or precipitation in the presence of formation water in a weakly acidic environment, and also brine displacement. Another possible effect will likely be cooling of the disposal reservoir or aquifer due to the fact that the injected CO_2 will likely be at a lower temperature than the initial formation temperature. This thermal effect may in turn affect stresses in the disposal unit and overlying confining layer. For RW the direct effects are thermal, geochemical reactions and processes, in both the near and far fields, and hydrogeological and geomechanical effects due to repository construction and operation; although the extent of any such effects depends on many factors, such as the types and characteristics of the wastes (there may be several different waste types in a single repository), the rock type, the repository design, the depth of the repository and the length of the operational period.
13. *Effects on the natural barrier*: With regard to the disposal of CO_2, no significant structural changes to the geological environment are expected due to the drilling and sealing of boreholes, although the CO_2 itself may affect the integrity of the natural barrier and there will be physicochemical changes to the rock mass in the disposal zone, which have been discussed in Sects. 2.3 and 2.4. The physical (pressure and temperature) and geochemical changes induced by CO_2 disposal may locally affect the integrity of the natural barrier at the interface between the disposal unit and the overlying natural barrier, but they should not affect the barrier's confinement/containment ability, otherwise the site should not be selected for disposal.

With regard to the disposal of RW, the construction of the repository and the EBS will directly affect the natural barrier (although mainly only locally). These changes may be chemical, due, for example, in some but not all repositories, to the effects of the alkaline plume caused by the cementitious components of the waste, the EBS and the rock support. Also, heat emitting wastes will directly affect the natural barrier, although any thermal effects will be limited to a few thousand, years, at most, with the dominant thermal phase lasting perhaps 1,000 years.

14. *Transport mechanisms of CO_2 or radionuclides*: With regard to CO_2 disposal, the mechanisms that drive the flow of free-phase CO_2 are the excess pressure, due to the injection process (Fig. 5), and its own buoyancy (see Sects. 2.1 and 2.4). In the case of CO_2 dissolved in formation water, the transport mechanism is the hydrodynamic drive of formation water and/or free convection induced by density differences in the order of 1% between the CO_2-saturated brine and unsaturated brine, if unstable conditions develop. The dominant transport mechanism for radionuclides is either advection in the groundwater or, in some host rocks with very low hydraulic conductivities such as clays and halite, diffusional transport. There are likely to be elements of both advective and diffusional transport in many repositories, e.g. perhaps diffusion in the EBS and advection in the geosphere. Transport in a gas phase is also possible—some wastes, such as LILW-LL, may produce considerable volumes of radioactive gas, due to geochemical and biochemical reactions and radioactive decay, and the potential effects of pressurization of the repository system need to be considered; other wastes, such as HLW, can produce much smaller volumes of gas, mainly due to processes such as anaerobic corrosion of the steel components of the waste form. Transport of radionuclides can also take place in colloidal form.
15. Return to the biosphere, hydrosphere and atmosphere: With regard to CO_2 disposal, the potential for leakage exists only as long as CO_2 is in free, mobile phase. There are no engineered barriers against CO_2 leakage, and leaky wells, fractures and other local geological features may provide pathways for the return of free-phase, mobile CO_2 to shallow potable groundwater, the vadose zone, soil, the biosphere and/or atmosphere (see Sect. 2.4). Evaluating the potential impact of wells and well integrity may be one of the key issues of assessing the performance of the disposal system. Such wells include both those used for CO_2 injection and those that may already be present or that may be drilled in the future and penetrate the disposal horizon. The issue of long-term cement and casing integrity is central to CO_2 disposal. With regard to RW, some of the long-lived wastes are encapsulated within containers that will remain intact for considerable times, e.g. those contained in copper or iron canisters may last for periods in excess of 100,000 years (see Figs. 7 and 9). Other wastes are encapsulated in concrete, which is designed to limit the release of the radionuclides. Even after canister failure, the EBS will delay the release of radionuclides for further times (see Fig. 9 for the situation regarding HLW and SF). The safety case for a repository for long-lived wastes needs to consider the effect of the return of any radionuclides to the biosphere. A variety of scenarios are normally modelled, in which different evolutionary paths for the repository are considered, i.e. different future

climates, premature failure of waste packages, etc. The return of radionuclides to the biosphere needs to be determined for all of these scenarios and the doses to critical groups determined for different times in the future. The processes that can take pace at the geosphere–biosphere interface can be complex, as can the processes and pathways in the biosphere. Again, as in item 14 above, both waterborne and gaseous releases need to be considered.

4.4 Site Activities

Included here are the activities that take place on and around the disposal site (except for the mode of disposal, which is considered above in Sect. 4.2). The subjects considered below are: site characterization, monitoring and future access, and intrusion or penetration of the disposal site.

16. *Site characterization*: Adequate site characterization is essential in ensuring proper site selection and performance of a CO_2 disposal site (IPCC 2005). Sites will be characterized based on well and seismic data, rock and fluid samples, laboratory analyses, and computer modelling, but a degree of uncertainty will always remain. Site characterization will likely be continuously refined after the start of injection as monitoring data are acquired, interpreted and fed back into the system. Research programmes will most likely accompany site characterization in the case of CO_2 disposal operations deployed in the near term, as is currently the case with the few pilot, demonstration and commercial operations implemented to date. With regard to RW disposal, a very comprehensive and lengthy investigation programme is inevitable—almost certainly more comprehensive and more extensive than one for CO_2 disposal. Such a programme is likely to involve a number of phases, may last many years and be accompanied by an extensive R&D programme. Eventually, underground access will be required, which will allow a considerably greater level of detail to be obtained regarding the near field (a prerequisite for the development of a final safety case). Site investigations at sites that are likely to be developed into repositories are currently taking place (or one phase of such investigations has been completed, with the investigations planned to be resumed later) in Finland, Sweden, Canada and France, and other countries are planning to carry out similar investigations starting in the next few years. Investigations are also taking place at underground research laboratories. Information on these investigation programmes can be obtained from the respective websites (e.g. www.posiva.fi, www.skb.se and www.andra.fr), whilst www.radwaste.org provides a list of the majority of websites of waste management organizations, waste repositories, regulatory organizations, etc.
17. *Monitoring*: With regard to CO_2 disposal, monitoring is likely to be required for defining the baseline conditions (before any disposal takes place), during injection, and decreasingly as the risk decreases (see Fig. 5) after the cessation of injection for site closure and for ensuring the long-term safety of the system.

With regard to RW disposal, all disposal concepts have extensive barrier systems, so, although monitoring will be a requirement (due, in part to public concern regarding the possibility and consequences of any release of radionuclides), no releases are likely to be detected for a considerable time (i.e. several millennia) after closure. Monitoring will also be required as part of the site investigation programme in order to study medium-term changes in parameters such as hydraulic head and seismicity, but a monitoring system will also be required to be in place before any underground construction takes place so as to monitor the effect of such construction on groundwater heads, groundwater chemistry and rock stress, i.e. to define the baseline conditions (EC 2004). Monitoring systems for this purpose are currently in operation at Olkiluoto, Finland (e.g. Pitkänen et al. 2007), where construction of what will become the access ramp to the SF repository is currently taking place. Monitoring will also be required in association with any requirement for the reversibility of the waste emplacement process or the retrievability of the waste.

18. *Future access, intrusion or penetration*: Sites used for CO_2 disposal may be penetrated in the future by exploration and/or production wells due to hydrocarbon exploration in, or production from, deeper strata. In the case of CO_2 disposal in coal seams, if successful, consideration should be given to coal mining or underground coal gasification at some future date. The possibility of such future intrusion appears, however, not to be an insurmountable problem, although it has been suggested that such potential intrusion be included in any future performance assessments for CO_2 disposal (Maul et al. 2007; Maul 2011). Sites for an RW repository will only be selected where the future intrusion risk, i.e. after closure of the repository, is considered to be low. This is likely to exclude all areas where there are mineral resources (of all types), and there has been extensive work in this area. The retrievability of the waste and the reversibility of the disposal operations need also to be considered—and all these subjects have been extensively discussed over recent years (see discussion in Chapman and McCombie 2003). Opponents of deep disposal would prefer to leave the wastes indefinitely in monitored surface or underground stores. Proponents argue that this is not a sustainable solution and that it is a higher risk option and that one should proceed in a stepwise manner towards eventual disposal. It may be necessary, in order to obtain public support for the disposal of RW, at least in some countries, to evolve a strategy that includes the possibility of retrieval of the wastes at all stages of the repository development programme. Such a programme is likely to last at least 100 years before final closure of the repository takes place and a stepwise approach, in which waste is slowly emplaced in the repository, combined with extensive monitoring of the performance of the repository, may be sufficient to allow the programme to proceed. The subjects of the accidental penetration of a CO_2 storage or RW disposal site and of the retrievability of RW from a repository are large subjects by themselves and much of their discussion is not geological in nature, and, being outside the scope of this chapter, are discussed elsewhere (e.g. see West et al. 2011).

5 Insights and Implications

5.1 Main Insights from the Comparative Assessment

It can be seen from the above discussion that there are similarities, but more, and sometimes significant, differences between the disposal of CO_2 and the disposal of RW. In both cases tectonic stability is preferred, and both require at least one, and preferably several, natural barriers against migration. Monitoring after emplacement will be required in both cases (for CO_2 monitoring, see Brunskill and Wilson 2011), although, in the case of RW disposal, unless monitoring takes place very close to the waste, it is very unlikely that any releases of radioactivity will be detected (and even close to many of the waste forms no releases are likely for very considerable periods in the future (EC 2004)). Also, in both cases there will be local effects on the geological environment as a result of the emplacement, although not identical (e.g. thermal cooling versus heating, different geochemical and geomechanical effects, etc.).

Whilst the current thinking is that CO_2 can be stored only in certain types of sedimentary (soft) rocks, RWs can be disposed of in hard rock as well. Very large volumes of fluid CO_2 will ultimately be disposed of through wells at great depths in natural geological media, whilst considerably smaller volumes of RW are or will be disposed of in solid form in tunnels or vaults at relatively shallow depths, using a combination of engineered and natural barriers. The timescales and containment period are significantly shorter for CO_2, in the order of centuries to millennia, whilst for RW they are in the order of at least ten thousand to possibly as much as a million years.

Whilst the main mechanism for the possible migration of CO_2 out of the disposal unit is its own buoyancy, being lighter than water (i.e. it is self-propelled), the main mechanism for the transport of RW once outside the EBS is transport by groundwater (i.e. it needs a carrier).

Site characterization in the case of CO_2 disposal is inherently simpler, but the results of such characterization are likely to be less certain because of sparser data and information. In contrast, a site characterization programme at an RW disposal site is likely to be considerably more comprehensive, lengthier and also more expensive. The programme will also include eventual access to the disposal zone via shafts or inclined tunnels, which will provide considerably more data on the rock mass (see the example of the current construction of the ONKALO at Olkiluoto, Finland (Posiva 2009; Andersson et al. 2007)), and this forms part of the site characterization programme. The extent of the safety case that will be required for obtaining permission to dispose of RW appears likely to be more comprehensive than the equivalent for CO_2 disposal (and this applies also to the associated R&D programme). Regarding the costs of characterization, they are likely to be lower for CO_2 disposal than for RW disposal, except possibly in the case of offshore CO_2 disposal in the deep sea in a region with limited data coverage.

Whilst the intent is that disposal sites for RW will never be penetrated or accessed (with disposal taking place only in areas with no mineral reserves), it is likely that in some cases the injected CO_2 will encounter existing wells, and it is possible that CO_2 disposal sites will be penetrated in the future by wells drilled for other purposes, given the very large areal footprint of CO_2 disposal operations. The potential impact of such penetration is unclear, as it may lead to CO_2 leakage into other strata, potable groundwater and even to the surface, posing local risks and also reducing the efficacy of the CCS process and limiting its usefulness. For this reason operators and regulatory agencies must take care in properly drilling, completing and abandoning such wells, and possibly new regulations will have to be developed for such situations.

There is considerably more experience in the disposal of RW, as there have been research programmes for several decades in many countries and there are now some operating repositories, although only one for long-lived RW—at the WIPP in the USA—whilst the disposal of CO_2 in geological media has been considered as a climate change mitigation measure only in the last decade. Several additional repositories for long-lived RW should, however, be developed over the next 2 decades, whilst large-scale demonstration projects for the disposal of CO_2 are under way. The science of performance assessment for RW is also well developed and there is general international consensus as to the suitability and efficacy of geological disposal for such wastes, whilst the science and criteria for performance assessment in the case of CO_2 disposal are currently under development. Consequently, the nascent CO_2 disposal industry can learn from the mistakes and successes of the RW disposal industry. Also, each national and subnational jurisdiction should develop a proper legal and regulatory framework for the selection, characterization and acquisition of CO_2 disposal sites, and a framework for the management of this new natural resource that is the CO_2 disposal pore space.

5.2 Implications

There are two areas of a geological nature where the disposal of CO_2 in geological media, which is in an incipient phase, can benefit from the experience gained to date from the disposal of RW:

- Site selection;
- Performance assessment (which itself involves three main elements and is itself part of the safety case) including the development of a good site understanding.

The mistakes made in earlier attempts at selecting sites for RW repositories, and the public opposition that was generated, made the nuclear industry in many countries realize the importance of openness and transparency during not only site selection, but in all elements of their RW disposal programmes (e.g. McEwen 2007; see Reiner and Nuttall 2011). An open and transparent approach is likely to be beneficial, not only in selecting sites for the disposal of CO_2, but during all phases of any subsequent disposal programme.

It is probably in the area of performance assessment (and the associated more comprehensive safety case) that the greatest opportunity lies for the proponents of the disposal of CO_2 to make use of what has been learned regarding the disposal of RW. As emphasized in Maul et al. (2007) and Maul (2011), the development of methods for undertaking performance assessments for the disposal of CO_2 is at an early stage. Much can, therefore, be learned with regard to the disposal of CO_2 from the experience of RW disposal over 3 decades: the need to employ a systematic and transparent methodology; the advantage of using system-level models; and the need to make maximum use of information from natural systems (i.e. natural analogues) (see also Maul 2011), where the safety case is discussed. There are, for example, both natural and engineered analogues for CO_2 disposal (Pearce et al. 2004; IPCC 2005). Natural accumulations are being studied in Australia, Europe and the US (IPCC 2005), supplying information on trapping and migration mechanisms and the potential impacts of leakage, as well as providing field-based testing grounds for deep, shallow, surface and atmospheric monitoring tools. This subject is discussed in greater detail in Maul et al. (2007, 2011).

In RW disposal, in addition to the performance assessment itself, the requirement to demonstrate a good understanding of the geology, hydrogeology and hydrogeochemistry of the site, and the region in which it lies, has grown in importance over the years, and is now a major element in developing a safety case for a potential disposal site. As outlined in items 1, 2 and 16 in Sect. 4, this site understanding needs to include an understanding of the way in which the site has developed over at least the last million years and how it is likely to develop in the future over a similar time frame. It seems unlikely that this element will be a requirement in a safety case for CO_2 disposal, at least not to the same extent as that required in the disposal of RW, as the timescales of interest are so much shorter. In all other respects, however, a similar emphasis is likely to be placed on the requirement to develop a good site understanding, and in placing the site in its regional geological context, thereby requiring more geological information than would be required for the performance assessment itself. This has important implications for the design, areal extent and operation of a site investigation programme at a potential CO_2 disposal site and is likely also to require more work in areas such as model validation, together with associated R&D, than is normally the case in the hydrocarbons industry.

Broadly speaking, CO_2 and RW disposal operations follow, or should follow, similar processes of site selection and characterization, application and permission, design and construction, disposal, site closure and post-closure monitoring, with remediation being an activity to be considered in case of leakage and/or migration out of the disposal zone. Whilst there is a large body of experience in the RW disposal industry with regard to site selection and characterization, there is no such experience yet in the incipient CO_2 disposal industry. The characteristics the geological media should possess for CO_2 disposal and, where relevant, any analogies with the RW disposal industry, should serve as a basis for developing policy and regulatory procedures for the site selection, characterization and siting of CO_2 disposal operations.

Given the large volumes of CO_2 for disposal, and the corresponding large footprint of such operations, having the necessary rock volumes available that meet the basic requirements for CO_2 disposal becomes a matter of resources, i.e. the pore space suitable for CO_2 disposal becomes a natural resource to be appropriately managed within each jurisdiction. Thus, as a matter of policy at the national and subnational level, each jurisdiction (state, province, country) should proceed with an inventory of its CO_2 storage capacity in terms of its size and distribution. Such inventories have been completed in some countries (e.g. Australia and most countries in northern Europe), or are in the process of being completed in other countries, such as Canada, the USA and other European countries. A lack of resources and data makes such inventories a challenge in developing countries. On the other hand, the lack of suitable geological environments or sufficient capacity for CO_2 disposal in some countries, as is the case in Japan and South Korea, will affect their policies with regard to a reduction of greenhouse gas emissions because carbon capture and storage, as CO_2 disposal in geological media is known, is an option with limited or no potential. Preliminary studies also indicate that large CO_2 emitters such as China and India may not have sufficient CO_2 storage capacity compared with their current and projected CO_2 emissions. The same limitations regarding the availability of potentially suitable geological disposal environments do not apply to RW, as there is greater flexibility in the use of different geological environments and the volumes of rock required for disposal are so much less than those required for CO_2 disposal.

There are certain legal implications regarding this new natural resource because currently it is not covered by existing legislative acts that would provide for acquisition of the right to dispose of CO_2 and access to the disposal site (i.e. mineral, mining, oil and gas acts, and alike, provide for access to and production of mineral and energy resources, but not for the utilization of the pore space for CO_2 disposal on a large scale). Similarly, regulatory agencies that will be mandated with regulating the CO_2 disposal industry will have to develop clear criteria for site selection, for predicting and monitoring the fate of the injected CO_2 and its effects on the subsurface environment, and for permitting CO_2 disposal projects. (See Bachu (2008b) and Wilson and Bergan (2011) for a review of legal and regulatory aspects that need to be addressed in relation to the management of this new resource.)

Finally, regulatory agencies will have to keep track, both geographically and stratigraphically, of the location of the CO_2 disposal operations and of their large footprints because of the potential for their penetration in the future by well drilling activities. Similar requirements will, naturally, also apply to RW disposal sites, although for the majority of countries there is likely to be only one deep geological disposal site. As already discussed, the likelihood of any future intrusion of such a disposal site is considerably lower than that for a CO_2 disposal site as, in contrast to perhaps the majority of CO_2 disposal sites, such sites will only be located in areas with no mineral reserves.

The potential impacts of CO_2 disposal on RW disposal programmes, or vice versa, also need to be considered. In most countries, RW repository projects are a long way from completion. On the other hand, if it is to be an effective means of mitigating climate change, CO_2 disposal will probably need to be implemented on a large scale within the next 10–20 years. Therefore, the majority of countries with both CO_2 disposal and RW management programmes will probably implement CO_2 disposal first. Thus, in these cases there is no danger that the implementation of CO_2 disposal itself will cause human intrusion of an RW repository. However, although RWs will be disposed of at shallower depths than CO_2, when siting an RW repository in sedimentary rocks, it might be necessary to determine the spatial extent of rock volumes that are likely to be affected by CO_2 disposal in the future and/or the footprint of CO_2 disposal operations. Potentially, large-scale implementation of CO_2 disposal could effectively rule out large areas of a country from consideration as possible locations for an RW repository. This factor may have to be included as part of a site selection programme, although it was not specifically included in the list of initial subsurface screening criteria for the geological disposal of RW published recently in the UK (Defra 2008). On the other hand, in the future the presence of RW disposal sites may reduce the potential for CO_2 disposal in their vicinity.

6 Conclusions

Various geological media are suitable for the disposal of the many products of human activity, among them RW and anthropogenic CO_2 captured from large stationary sources. For both these particular types of waste, the objective of disposal is their isolation from the hydrosphere, atmosphere and biosphere for very long periods of time—in the order of centuries to thousands of years for CO_2, and in the order of tens of thousands to perhaps a million years for RW. However, there are some fundamental differences between the two products that, consequently, dictate the types of geological environments that are suitable and required for their disposal. RWs are in solid form and are relatively limited in volume, but are extremely hazardous. As a result, RWs are to be emplaced in subsurface engineered systems (often known as repositories) at depths of a few hundred metres, using both tunnels and shafts to provide access to the point of waste emplacement. CO_2 is a fluid that can be emplaced in the disposal unit only by its injection through wells, usually at depths greater than 800–1,000 m. The volumes of CO_2 that need to be injected to achieve significant reductions in atmospheric CO_2 emissions are huge, with consequently a very large subsurface footprint (tens to hundreds of square kilometers at a single disposal site). As a result, no barriers to CO_2 escape can be engineered, unlike RW, and containment has to rely entirely on the natural system, or natural barrier.

For both RW and CO_2 disposal a stable geological environment is preferable, with additional safety and remediation measures possibly being necessary if the disposal site is located in a less tectonically stable region. Currently, only sedimentary rocks with suitably high porosity and permeability are considered for CO_2 disposal, although laboratory and field experiments are being carried out to assess the possibility of disposing of CO_2 in other rocks, such as basalts. Hard crystalline rocks, but also sedimentary rocks and evaporites are considered potentially suitable for RW disposal, but it is important that all such rocks should have low to very low permeability. For both forms of waste, any site should be geomechanically sound and associated with low velocities of groundwater flow, although the requirements in this regard are more onerous for RW than they are for CO_2.

The main effects of the disposal of CO_2 are an increase in pressure at depth, with accompanying changes in the stress regime at the disposal site, thermal effects as a result of the temperature difference between the injected CO_2 and the disposal zone (CO_2 being cooler), and geochemical changes as a result of the dissolution of CO_2 in formation water, forming a weak carbonic acid. In the case of RW disposal, the main effects due to waste emplacement are thermal (at least for HLW and SF), as a result of the heat emitted by the waste, geochemical and biochemical, as well as geomechanical and hydrogeological, due to the construction and operation of the repository. The natural barrier is affected, in the case of CO_2 disposal, by the drilling of wells and may be affected by the CO_2 itself and, in the case of RW disposal, mainly by the construction of the repository, but also by the boreholes required to investigate the site. CO_2 may escape from the disposal unit, if a pathway is available, as a result of its own buoyancy and due to the pressure build-up caused by injection, whereas the main escape route for radioactivity is via groundwater, although the release of radioactive gas also needs to be considered. Consequently, extensive site characterization and monitoring is or will be required in both cases, although it seems likely that any such site characterization programme is likely to be more detailed and more prolonged in the case of RW disposal.

A significant difference between the two types of disposal is retrievability, which may be a requirement in the case of RW, but which is not, and may in any case be impossible, in the case of CO_2. There may be cases in the future where wells will be drilled into a plume of CO_2 to release CO_2 and reduce the pressure, but this scenario is envisaged only as a remediation measure in case of uncontrolled leakage. In any case not all of the CO_2 will be recovered because some will be dissolved in formation water and some will be immobilized in the pore space at irreducible saturation or through mineralization. The subject of the retrievability of RW is included, although not discussed in detail, in item 18 of Sect. 4.4. Although the subject has been extensively debated, the possibility of waste retrieval does not necessarily influence the type of disposal environment selected nor does it necessarily exclude specific host rock types from consideration.

There are two areas of a geological nature where the disposal of CO_2 in geological media, which is in an incipient phase, can benefit from the experience gained to date from the disposal of RW: firstly in site selection and, perhaps more importantly, in performance assessment. It is, therefore, probably in the area of performance assessment (and the associated more comprehensive safety case, which includes the development of a good site understanding) that the greatest opportunity lies for the proponents of the disposal of CO_2 to make use of what has been learned regarding the disposal of RW. There are many areas in the development of a comprehensive safety case for a disposal site where geological input is required.

The analysis presented in this chapter indicates that there are, therefore, similarities, but also many significant differences, between the geologically related issues and requirements for the long-term emplacement and isolation of CO_2 and RW, although, in carrying out such an analysis, useful comparisons can nevertheless be drawn between the disposal of these two types of waste.

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