7. DISPOSAL OF HIGH LEVEL WASTE

Basic principles of disposal (Chapman 1987)

The two fundamental options available for disposal of any material are either to endeavour to keep it in the same place for as long as necessary, or to allow natural processes to mobilize and disperse it harmlessly. The first concept (F 1) is known as containment (or concentrate and confine) and is clearly very difficult to ensure for very long periods of time. The second is commonly referred to as dilute and disperse. There are obvious attractions to the latter concept for materials which remain toxic for extremely long times. Containment of such materials means that they always remain a potential hazard at one place since their concentration is only slowly reduced by decay. It would clearly be better to design a disposal system which allowed for their mobilization and transport by natural processes, resulting in their dilution in a widely dispersed form at concentration levels which presented no unacceptable hazard.

The attractions of dilute and disperse are naturally counter-balanced by some very difficult problems. If we are to allow the dispersal to take place, then we must be able to model it confidently to ensure that any eventual releases of radioactivity are acceptable. This involves the understanding of a very wide range of processes which can take place during mobilization and transport of the waste.

At present there is an increasingly widespread move towards adopting both concepts in any single waste disposal system. The way this is usually envisaged is that short-lived radionuclides are contained until a sufficient number of half-lives have passed that their concentration in the waste is extremely low. Since containment of very much longer-lived radionuclides for any equivalent number of half-lives is impossible to demonstrate, the system is also designed to allow for their eventual slow mobilization and dispersal. The definition of how long the initial containment period should last depends very much on the waste type and the predicted behaviour of the environment chosen for disposal.

For example, short-lived wastes might be earmarked for shallow-land burial where total containment may be required for around 300 years. After this the total concentration of both short- and long-lived radionuclides in these low-level wastes is very low and almost any degree of dispersal may be acceptable; hence the choice of a shallow rather than a deep disposal environment. In the case of HLW the initial containment period may vary from around 1000 years, up to a million years or more, depending on concept. Some groups suggest that since the dilution and dispersal process in the deep geological environment is so efficient, there are no technical reasons why any initial containment period is necessary, apart from during the operational, pre-closure life of a repository. The basic principles of disposal thus involve the selection of a disposal environment and a system of engineered emplacement of the wastes to provide the required containment and/or dispersal. A general requirement for disposal of any waste is to ensure that the disposal location is remote from man's environment and thus the two methods which have always been chosen historically are burial on land or dumping in the sea. Consequently these two methods have emerged as the principal contenders for disposal of radioactive wastes. It is worth bearing in mind that there have been no adequate controls on disposal of most industrial wastes until very recently (the early 1970s in most countries). This includes the, early disposals of radioactive wastes, so there is consequently already an established history of both burial and sea disposal prior to the present day rigorous scientific controls.

Moving back to a more technical summary of the functional requirements of any disposal system, the following list highlights the main points to be considered assurance of low release rates once the period of complete isolation is past, removal of the wastes from the effects of man's activities or catastrophic natural events, the technology to implement disposal should be available and the task should be achievable at reasonable cost and we must be able to model adequately the processes which control the long term performance of the chosen system, be they physical, chemical or even biological.

Geological disposal

Geological formations (F 4.1) are characterized by their great age, and in most environments by their considerable long-term stability. The lengths of time over which certain rock formations have retained their structure, integrity and properties is beyond the scope of human experience, and although the rocks are subject to constant change, the rates are generally extremely slow. This permanence is anecdotal and part of our own folklore: an Icelandic legend has it that there exists in the vastness of the cold northern oceans a huge, smooth dark rock which rises from the ice and the waves. Every year a great black bird goes to this rock to sharpen its beak on the hard surface of the stone. When the bird has eventually worn the rock away then one era of geological time will have passed. Although colourful, this highlights our difficulty in comprehending not only geological time, but by analogy, the isolation periods of long-lived wastes.

Burial of wastes in a suitable geological formation removes them from man's environment and puts them out of reach of major disruptive processes, both natural and man-made. It also provides more than adequate shielding from the radiation emitted by the waste. The wastes are put in an environment where all natural processes which can affect their behaviour are very slow. The deeper the burial, generally speaking, the slower and less pronounced are these processes, and we are effectively matching their rates to the required isolation period of the wastes. Thus it is clear that long-lived wastes will generally require deep disposal, but disposal at shallower depths may be acceptable for shorter-lived wastes. Political expediency also plays a role in choice of disposal depth. Thus some countries have chosen deep disposal for even low-activity wastes because they consider that the extra costs involved are more than matched by the political gain of demonstrable over-design of safety barriers.

The natural geological processes which will affect the behaviour of buried wastes are dominated by the behaviour of ground water in the host rock unit and in under- and overlying formations. All rocks below the water table are fully saturated with water; that is their pore spaces, which can vary from a few to several tens of % of the rock volume, are full of water. Even above the water table a considerable proportion of the pore space is water filled. This groundwater moves slowly through the rock, and in doing so it can leach and dissolve the waste and its containers and carry radionuclides away and potentially back to the biosphere. The behaviour of groundwater in the rock (hydrogeology) is thus a very important factor in selecting a host geological environment for disposal, and one is generally looking for very slow and predictable rates of water movement. Many geological environments fit this requirement and a variety of suitable rock types are available for construction of a disposal system, either deep or shallow. Again these are considered in much more detail in later sections. The disposal systems themselves fall into:

- deep burial in a purpose-built or modified existing mine, usually called a repository (F 6.13);
- deep burial in boreholes (F 6.11) drilled from the surface.

Alternative disposal options

Although geological disposal of radioactive wastes is generally favoured, a number of other options have been studied for long-lived wastes. Generally speaking the high potential costs of these other methods has meant that, with the exception of sea disposal, they have not been considered seriously for low-level wastes. The alternatives for long-lived wastes include space disposal, ice sheet disposal, disposal beneath the seabed, rock melting and nuclear 'incineration'.

Space

Space disposal (F 3.2) is one of the most commonly suggested solutions for nuclear waste offered by the 'man in the street'. It has the great apparent attraction of removing waste from the biosphere for all time. Studies have been carried out by NASA which show that the launch of nuclear payloads into a solar orbit halfway between the earth and Venus is feasible, based on space shuttle technology. The main problems involved are the extremely high cost of this option and the risk of launch failure. The disastrous loss of the US shuttle-craft 'Challenger' in January 1986 highlighted this risk, and reinforced the previously estimated rocket launch failure rate of 6 in 100 dramatically. The cost alone means that this option could only be considered for the very highest specific activity wastes, while the launch risks have all but eliminated this disposal route from further consideration. A launch failure could cause widespread, not to say politically embarrassing, pollution to almost any region of the world. The repercussions of the $8 million search and clean-up exercise, carried out in the Arctic winter of northern Canada in 1978, following the re-entry of a satellite nuclear power-pack, and its break up in the atmosphere, will long be remembered.

Ice sheet

Ice sheet disposal (F 3.3) is another option with great popular appeal. Apart from the remoteness of the polar ice-caps, the fact that heat-emitting wastes would be self-sinking in the ice has clear attractions. The problem areas are the very high transport and handling costs, uncertainties in the evolution of these areas over the geological time spans for which containment must be considered, and legal constraints. At present the controlling limitation is probably the latter, as waste disposal in the Antarctic is expressly forbidden by international law, while alternative sites in Greenland are controlled by Denmark.

Ocean

Disposal of high-level and some types of intermediate-level wastes on the bed of the ocean (F 1,3.4) is prohibited by international agreement (the London Dumping Convention), although it can be shown that the disposal of some wastes by this method presents very low individual risks. In recent years considerable research has been carried out on disposal beneath the ocean bed. Advantages of sub-seabed disposal in the deep oceans are physical remoteness, chemical and geological stability of the sediments and rocks, and the great dilution capacity of the sea in the event of releases occurring. In addition, disposal techniques involving self-burying containers ('penetrators') which are allowed to free-fall from the disposal ship could be relatively inexpensive. Technical problems mainly involve ensuring the performance of simple disposal techniques in the deep waters considered, or the high cost and engineering problems with more sophisticated alternatives involving, for example, emplacement in holes drilled in the ocean floor. Potentially more critical, however, are international concerns. In the foreseeable future, pressure from non-nuclear countries with marine seaboards is likely to cause further tightening up of agreements about the use of the oceans for waste disposal, rather than any relaxation. Research and development programmes into sea disposal of long-lived wastes are being reduced, not for technical reasons, but because political factors seem likely to rule out these disposal options.

Islands (Brookins 1984)

The island disposal concept would involve placing the radioactive wastes in deep, stable geologic formations on islands. The repository requirements are like those for mined repositories on continental land masses, only the wastes must be transported to the islands. The natural barriers at such a site would include the repository rock plus the oceans. The oceans surrounding the island would ensure rapid dilution of any escaped waste. The status of island disposal is not clear. Investigation of the island disposal concept is not being actively pursued by the U.S., although the United Kingdom Royal Commission on Environmental Pollution has pointed out the advantages of possible use as an international repository. Many islands contain very little groundwater, and exchange with the oceans is slight. Thus little water would be available for possible attack on stored waste, and if such unlikely reactions were to take place, the surrounding ocean would so dilute the products of the reactions that they would be rendered harmless. Basically two different below-ground sites are available for inspection. The near surface waters are fresh waters, which do not readily exchange with the deeper, saline waters, and whose shape is that of a lens sitting in heavier, saline water. Three types of islands available for repository considerations are continental, oceanic and arcs islands. The continental islands are found for the most part on the continental shelves and are made up of igneous, metamorphic, and sedimentary rocks. The oceanic islands are located in oceanic basins and are made up of volcanic rocks, usually basalt. The island arcs are those islands located at plate boundaries near continental masses. They have originated by tectonic processes and frequently are the sites for andesitic volcanism. There are serious disadvantages to this method. First, it is logistically impractical. In addition to the secure ocean vessels, there would have to be seaport and handling facilities both on the mainland and on the islands, and land transportation both on the mainland and the islands. This and the fact that handling time and transportation times are much greater than those for land repositories make this method economically less attractive than other alternatives. The U.S. Department of Energy has estimated in 1980 that an island repository would cost at least twice as much as a land repository. A possible attractive feature would be use of an island site for the storage of international waste. This may not seem necessary now, but, in view of the fact that 35 nations now rely to some degree on the nuclear option, such an international site may be necessary in the not-too-distant future. Another serious disadvantage for many of the islands of the earth is possible volcanism. Volcanism is a risk on too many islands and ocean islandic provinces to unequivocally say that it will not occur on a particular island. When only continental islands are considered, the volcanism risk is reduced, but these islands are perhaps less well situated geographically than the oceanic islands in order to be considered for the disposal of internationally generated wastes.

Rock melting

A more speculative alternative to any of the geological emplacement techniques for high-level waste is 'rock melting'. This involves emplacement of highly concentrated solid or liquid HLW into a deep borehole or cavity. The radiogenic heat produced by the waste melts both the waste itself and the surrounding rock and, upon eventual cooling, the wastes are incorporated into a natural rock matrix. As yet, the technical knowledge required to evaluate fully waste/rock interactions in this system does not exist and hence it has been considered with only very low priority.

Transmutation

The final technique considered-nuclear transmutation, or 'incineration'-is not a disposal option as such, but provides a method for destroying the long-lived radionuclides which present most of the problems in other disposal systems. Long lived radionuclides are chemically extracted from the waste and emplaced in the reactor core. This causes them to be fissioned or activated to form shorter lived products. This technique is effective, but it is expensive and can result in considerable radiation exposure to the work force involved.

Processes and events causing release from the repository (Chapman 1987)

Radionuclides in the waste might escape from a repository by natural causes or as a result of human intervention. The prime objective of disposal of waste is to remove it from the biosphere and man, so one of the aims of burial in a repository is to make it as difficult as possible for future human activities to result in inadvertent exposure to the still active waste. A backfilled and sealed repository will prevent unintentional access to the waste while some record remains of where a repository is, and what it contains. It is much more difficult, if not impossible, to prevent intentional exhumation of the waste. It must however be argued that we cannot be responsible for such action by an informed group at some time in the future. Since such an action would, of necessity, involve a major feat of mining engineering it must be assumed that it would occur only with the full realization of the consequences and risks involved. This type of 'release' of the waste is something which is not taken into account in safety and risk assessments.

Caused by man

The most important RI-releasing events caused by man are human intrusion and direct alterations in hydrogeology (e.g. through draining or storage), injection of liquid waste, drilling, construction of a dam and geothermal energy production. What must be taken into account first is accidental human intrusion into a repository at some time in the future when all records of its existence have been lost. Our experience of myth and legend would suggest that records may endure for thousands of years but this period is very short compared to the half-lives of some important radionuclides in wastes. If technology is still at the same level as at present, or more advanced, it would seem unlikely that any major intrusion by mining activity would occur without advance warning of the presence of such an anomaly in the ground being provided by the type of routine geophysical reconnaissance surveys that generally accompany such operations. The single most significant possibility of future human intrusion is thus considered to result from inadvertent drilling of a borehole through a contaminated zone in the repository, or through a waste container. Some repository designs attempt to prevent such a possibility by emplacing a thick metallic cone above each container to deflect a drill bit. Such features are clearly gestures only, to minimise the chance occurrence of what is in fact, for a deep repository, a very remote possibility. Careful site selection to avoid regions of exploitable natural resources (e.g. aquifers, ore bodies, geothermal zones), is the most important means of reducing the probability of this event occurring. This is a particularly problematic feature of disposal in evaporate formations. Many evaporate minerals are themselves natural resources, and have been mined in several countries. Such mining may be by remote means, such as solution extraction (pumping water down boreholes to dissolve the salt and return it to the surface in the form of brine) where direct access to the formation is unnecessary. In addition, such formations are increasingly being used for underground storage of oil, gas, and so on,owing to their extremely low permeabilities. This has led some countries to downgrade evaporates as potential host formations, owing to the 'resource utilization' factor, and the higher probability of future intrusion into a repository. As far as research into the effectiveness of the containment barriers is concerned, we are thus only interested in considering natural processes which might lead to release of the waste. These fall into isolated events which may either individually or in conjunction cause decreased efficiency or failure of the barriers, and the processes of slow evolution of the repository and its geological environment which will gradually but inevitably lead to the breakdown of each barrier and eventually result in release of the waste. This latter category forms the prime consideration in safety assessment, since evolutionary processes are bound to occur and inevitably lead to escape of the waste, and it is onto these processes that the effects of any isolated events from the first category can be superimposed. We have omitted mention of the more extreme results of penetration by nuclear weapons and intentional sabotage during times of war. With such natural or man-made events occurring, a release of radioactive waste would be the least of mankind's worries.

Caused by disposal of waste

The most important RI-releasing mecanisms caused by the disposal of waste are radiation damage of the matrix, radiolysis of groundwater, nuclear criticality, canister movement in backfill, mechanical canister damage, differing thermal expansion of waste matrix and canister, interactions between canister and backfill, backfill and host rock, decompressed zones from mining, host rock zones, thermal convection, thermally induced chemical changes, drying out and resaturation, chemical changes due to corrosion, geochemical changes in backfill and host rock, physico-chemical processes (e.g. colloid formation), microbiological processes, gas production and failure of shaft sealing. The near-field is the zone altered by the presence of the emplaced waste. It comprises all the engineered barriers and extends a small way into the host rock itself. The near-field zone is one of great chemical complexity, influenced by properties of both the waste package and the host rock. The main aim of analysis of the near-field is prediction of the mobilization rates of all significant radionuclides into the far-field, which acts as a 'source-term' for subsequent migration calculations. Parameter variation in such analysis also allows the relative performance of individual engineered barriers to be assessed, as a first step in system optimization. In this Chapter the processes occurring in the near-field will be reviewed in very general terms and then the performance of individual barriers will be considered in more detail. Finally, the construction of a source-term model will be explained with some indication of the simplifications required and the advantages and disadvantages of different approaches to this problem.

Caused by nature

Slow natural processes are climate changes, sea-level changes, erosion (fluvial and glacial), sedimentation, tectonic crustal movements, magma intrusion, volcanism, diapirism, diagenesis, metamorphism, weathering, mineralization and groundwater changes. The probability of such an occurrence can be estimated from our knowledge of such impacts in the geological record and of the origins and distributions of such extraterrestrial bodies, but readers should make up their own mind as to the scientific or social validity of considering such esoteric events in an objective safety assessment. Climatic events (rather than processes) can be accounted for in the prediction of evolutionary processes, but in general it is more satisfactory to superimpose the likely effects of, for example, future glaciations or changes in sea level caused by melting of polar ice, onto a normal case groundwater flow model. Thus a probabilistic assessment of events is superimposed on the deterministic model of 'normal case' processes. Increased rates of erosion must be allowed for in upland terrains; caused by glaciers, deepening of river valleys, tropical erosion and so on. However, it would appear reasonable to assume that no individual climatic event is in itself likely to cause release of waste from a deep repository, but will instead have a more or less predictable effect on regional groundwater flow patterns. If such factors are taken into account when designing and siting a repository, then future climatic changes, even those involving marine incursion over the site or submergence beneath an ice-cap, need present no problems of containment. Certain environments would naturally prove unsuitable in such circumstances, and after preliminary consideration would be discarded.

Rapid natural events are earthquakes, volcanic eruption, meteor impact, flooding with extreme erosion, hurricane, storms, movements at faults and formation of new faults. Tectonic processes are not 'sudden', but tend to occur over very long periods of time (1E+7 years) and do not in themselves constitute a hazard to the barriers in a repository. However, tectonically active regions of the world are often characterized by volcanic activity, earthquakes and major crustal flow, and accelerate the normal-case process of mobilization and migration. In the case of intrinsically dry host rocks such as salt domes, faulting could result in access of water to the waste from overlying water-bearing strata, hence initiating the slow process of leaching and migration. Since it is impossible to design completely against such events, the safety assessment of a disposal system must take them into account by examining the probability of their occurrence and the effect they would have on release. Information is thus required on the frequency and probability of seismic events of varying magnitudes occurring in specific areas, how these may change with geological evolution, and how they would affect the normal case release model. Reading through the list of events which might speed up the release of the waste one is struck by the cataclysmic nature of some of the natural processes involved, submergence beneath the oceans or beneath an ice-cap, penetration by volcanic conduits, rupture by massive earthquakes and exhumation by meteorite impact. It is tempting to ask whether, by seriously considering the effects of some of these hazards in the safety assessment, we are becoming obsessed with the containment of radioactive waste when, in the event, the physical damage to the environment would have orders of magnitude greater consequences for mankind.

Groundwater

An example of the two groups is given by the presence of groundwaters and their movement through a host rock. This will eventually cause the corrosion of the waste canister and the dissolution of the waste. Combined with the slow processes of erosion it will result in some of the waste radionuclides returning to the biosphere, probably at very dilute concentrations, and far into the future. However, a single natural event, such as a massive earthquake resulting in local fault development may 'superimpose' itself on this background pattern of processes, to the extent that a significant direction change or increase of groundwater flow occurs, resulting in earlier or more concentrated return of waste to the biosphere. In order to examine the significance of such sudden events, we must fully understand the background geological processes which they might affect. This has led to the definition of a 'nominal-case' release scenario, which can be modelled deterministically. In the normal case, release results from the natural processes of geological, climatic and chemical evolution which occur within the Earth's crust. Research focused on particular geological environments is used to examine whether the normal case leads to releases which are radiologically acceptable or not. The normal case deterministic models are used as a basis for designing research programmes. Natural events leading to release are related to climate, tectonic patterns and random catastrophes. The last group includes such unpredictable events as the impact of a massive meteorite or asteroid on a repository site which is of sufficient magnitude to lead to rapid or immediate release of the waste.

Normal-case release model

In the preceding section it has been proposed that in any of the major geological environments considered for the siting of a repository, there exist progressive geological processes that must inevitably cause the return of some of the waste to the biosphere. These can be considered using what has come to be called a 'normal-case evolution model (F 4.9). In fact one basic model can be used to describe almost all environments. The normal-case model can be constructed around the only credible natural mechanism by which components of the waste can be transported back to the surface, namely transport in groundwaters. Free water is present in all rocks, and it moves through the pores and fissures of the various lithological units in response to differences in hydraulic 'head' from point to point. Even where no flow occurs, the body of water contained in the pores in the rock can allow diffusion of radionuclides, although at very slow rates. The corroding and leaching action of water will eventually mobilize the waste, in solution or in suspended form, and it will migrate and diffuse with the local and regional movement of the groundwater until it eventually finds its way back to the biosphere. The route which is takes, how long it takes to travel along it, how far and in what form, are the fundamental questions which must be answered before realistic safety assessments can be made. An exception to this simple model of release of the waste is provided by repositories in dry salt deposits. Since there is little or no interconnected water movements. Regions such as the Mediterranean area, parts of the Middle East, central Africa and the Pacific Ocean margins display such symptoms of continuous tectonic activity, and in these areas difficulties may arise in the prediction of repository performance. Knowledge of the mechanisms and driving forces of these global tectonic processes has increased dramatically over the last twenty years, to the extent that we are able to predict with some confidence which regions of the earth's crust are likely to remain inactive in terms of vulcanicity and frequent major seismic events over the next few millions of years. Clearly, major earthquake damage or exposure to the many paroxysmic processes associated with volcanic activity can only be deleterious to the integrity of a disposal facility. This indicates that repositories should be sited away from tectonically active or marginal areas. The risk of seismic damage is, however, impossible to avoid completely, since all regions of the crust are seismically active to some extent. It is known that underground structures (such as a mined repository) suffer very little damage as a result of even quite major earthquakes, since the host rock and the components of the mine all undergo the same degree of acceleration when a seismic wave passes. The perceived risk lies in the possibility of faults being initiated in the repository rock, or of existing major fractures being reactivated. Once again, it is extremely unlikely that such events in themselves, would lead directly to release or exposure of the waste, but they could lead to modifications in ground-water filled porosity in pure evaporate formations, there is no possibility of water flow through these units. (Impurities such as silty or sandy layers can give rise to some component of permeability, and if an intergranular film of water is present, then slow diffusion may be possible, but at rates lower than in free pore waters).

Evolution of the near-field environment

During operation, a deep repository will be drained and ventilated, ensuring dry conditions while waste emplacement proceeds. After emplacement, the conditions in the near-field will change gradually with time and a number of distinct stages in the evolution of the normal-case model can be recognised.

Groundwater intrusion

Water re-invades zones of the host rock which have been drained during operation. The hydrogeological environment begins to stabilise and water starts to penetrate the backfill. For some backfill materials, such as bentonite, this causes them to swell. In confined conditions this builds up a 'swelling pressure' which causes plastic flow to seal any constructional gaps and, possibly, any disturbed rock zone around the emplacement tunnel. For high-level wastes, wetting of the backfill from the outside may be coupled to a drying from the inside as radiogenic heat warms the engineered barriers and some region of the surrounding rock. A thermal profile is relatively quickly established, within decades, which slowly decays to insignificance over some thousands of years. As groundwater penetrates the backfill (or possibly even during transit through surrounding warmed rock) its chemistry alters due to a complex series of reactions including dissolution and precipitation of particular minerals and ion-exchange. If the backfill is simply crushed host rock such alteration may be negligible, but if materials such as bentonite or concrete are used, water chemistry will be completely altered. For HLW, some additional chemical changes (radiolysis) may be caused by neutron and gamma radiation which can penetrate the intact canister. After water has penetrated the backfill it comes into contact with the canister, which begins to corrode. In many designs, the saturated backfill has much lower permeability than the host rock and hence negligible flow occurs in this region and all solute transport (of corrodants in groundwater inwards and corrosion products outwards) occurs by a diffusive process. Such diffusion may be very slow and thus limit canister corrosion rates.

Degradation of engineered barriers

At some point canister failure occurs due to corrosive penetration or mechanical effects (for example due to external hydrostatic pressure or backfill swelling pressure). Even after its failure as a complete barrier to water ingress, remnant canister material and corrosion products may act as a further diffusive barrier and alter the chemistry of water penetrating through the backfill. When water contacts the waste itself it will begin to degrade and radionuclides will be released, either into solution or as particulates or colloids. Prediction of the chemical environment in this region is very complex, being determined by all the reactions previously considered, plus reaction with the waste form itself (which may be somewhat heterogeneous). For HLW especially, there is the further complication of additional radiolysis from short range alpha and beta particles which can now contact the groundwater. In the low-flow or diffusive environments generally expected, waste degradation is unlikely to proceed by simple dissolution but is likely to involve formation of solid reaction products (secondary minerals) in situ. As the waste form breaks down, radionuclides are dissolved or mobilized as particulates and begin to migrate through the near-field.

Radionuclide migration

The rate at which radionuclides are released into solution is generally determined by the breakdown rate of the matrix (although it is possible for some radionuclides to be located preferentially on the waste surface as will be seen later) but may be further constrained by their low solubility. The rate of migration of radionuclides through the far field is determined by the hydrology of the different rock formations along the transport path and the extent of retardation processes, for example due to sorption onto secondary minerals from the waste, canister corrosion products and backfill. Further constraints may arise from precipitation (or co-precipitation) due to changes in pore-water chemistry along the flow path. Evaluation of all the stages above, taking into account radioactive decay (or ingrowth), allows prediction of the release of any radionuclide into the far-field to be assessed as a function of time. For some cases, the near-field/far-field boundary can conveniently be considered to lie within the mechanically disturbed rock zone extending at most a few metres from the tunnel or borehole wall. In others chemical perturbations (e.g. oxidizing conditions caused by radiolysis) may penetrate significant distances into the host rock. In the latter case, the development of the chemical perturbation front in terms of solute buffering by the mineralogy of the host rock and radionuclide transport within this region must also be evaluated.

Factors affecting the near-field environment

In this section we consider the guidelines used to define the requisite performance of the individual engineered barriers and the techniques used to quantify them. Before considering the barriers individually, some parameters dependent on waste type, overall repository design, and location need to be specified.

Temperature

The most important of these factors is probably temperature, especially for HLW. If a repository is constructed at, say, a depth of a kilometre or so, then the ambient temperature will be about 25-50 C, largely regardless of rock type. The decay heat of emplaced HLW will be additional to this, and the exact temperatures attained in any part of the near-field (F 5.1) can be predicted very reliably using heat transfer calculations which take into account the slowly declining heat output of the waste and the thermal conductivities and diffusivities of the materials involved. For more exact calculations it may be possible to include the effects of backfill resaturation to form a coupled heat/water transfer model. Such complex systems are much harder to quantify, and the differences between the results obtained and those from a simpler model are generally small. Similarly, complete backfilling of all tunnels and voids is generally assumed, and heat transfer is thus taken to occur by conduction only, although the small effects of remnant air-gaps can also be evaluated. Thermal convection on a larger scale was once considered to be of potential significance, but a model of thermal buoyancy in fissured rock has shown that for expected disposal environments, while it may result in large scale water movements, it does not contribute significantly to the transfer of heat.

The maximum temperatures attained in any part of the near-field depend principally on the thermal loading of the HLW containers and the geometry in which they are emplaced. In the rock types considered for disposal there is a predicted rapid increase in rock-edge temperature over the first few years after emplacement, with very steep thermal gradients away from the centre lines of individual waste packages, across the backfill and out into the rock. After between 50-100 years these gradients would start to flatten out such that temperatures within the repository itself would be fairly uniform. This is more obviously the case in the 'cubic' repository (F 5.2) than in the 'flat' type. Most countries engaged in repository design exercises have opted for an arbitrary value of 100 C or less for this maximum temperature, although research continues to optimize this figure for the various rock types and configurations involved. In the USA, maximum values of up to 250 C are suggested for some salt and basalt host rocks and 150 C is suggested for tuff, 80 - 100 C remains the design target for plastic clays. It is likely that any eventual design for fractured crystalline rocks such as granite could tolerate temperatures between 100 and 200 C. A more serious constraint on the maximum admissible temperature may well be the stability of engineered barriers. Both metal corrosion, and many backfill alteration reactions (e.g. montmorillonite to illite in a bentonite backfill) are much slower at low temperatures. Although, in some cases, the reactions encouraged by high temperatures may be favourable (e.g. formation of protective layers on metals), important properties may be lost; for example, the swelling capacity of bentonite. If long term pre-disposal storage is considered, then it may well be feasible to keep near-field temperatures below 100 C. Regardless of the maximum temperatures attained in any part of the repository, the near-field is characterized by steep thermal gradients over a period of several years to some decades depending on precise rock properties and repository design, reaching a maximum temperature after a few decades, and followed by fairly uniform near-field temperatures which decline very slowly over several hundreds of years until they approach what would be natural ambient values during the first millennia. Thereafter, while the actual amount of heat given out by the waste is still considerable (integrated over time for the ensuing 10000 -100000 years it dominates the total thermal energy output) its production rate is so small that it has little or no effect on processes in the near-field. However, as will be seen in the next chapter, this steady trickle of heat is a major driving force for groundwater movements in some rocks for very considerable periods.

Stress field

A second important factor comprises the hydrostatic and lithostatic stresses. In a hypothetical repository, it about 1000 m depth in a fractured or porous rock the water present (assuming complete interconnection of all fissures and pores) in the rock will exert a hydrostatic pressure on the near-field components approximately equivalent to a 1 km head of water. Naturally, pumping during repository operations, recovery of hydraulic pressure profiles after repository closure, and local hydraulic gradients will have some effect on this value, but it represents the minimum stress to which the waste packages would be subjected. In some mixed sedimentary environments deep groundwaters may be at substantially more than hydrostatic pressure, due to transfer of part of the lithostatic load, or long-range artesian effects. Over longer periods of time, as stresses in the rock-mass itself readjust to hole boring and tunnelling and backfilling, the near-field components will be expected to be subject to lithostatic load pressures caused by the one kilometre overburden of rock. These stresses are often anisotropic, particularly in some areas of ancient crystalline rock, and horizontal components of stress can be up to four or more times the vertical values, which are themselves two to three times greater than the maximum hydrostatic pressure, owing to the density of the overlying rock units. Stress readjustment will take place very quickly in some of the plastic clays and salt deposits being considered for disposal.

A third component of the stress field acting on the waste and its packaging is that caused by the heat output of the waste itself. This can cause highly anisotropic stress build-up in response to the steep thermal gradients and differential thermal expansion (or contraction) of rock and near-field materials. Such point stresses tend to congregate around any unconfined surfaces (although these will be uncommon in a backfilled repository) and can cause spalling and microfracturing in the more brittle crystalline rocks. The plastic rocks, such as salt, demonstrate quite rapid creep response to thermal stressing, which would tend to close any openings which may be present. The crystalline rocks will also display creep, but on a very much longer time scale, which may be significant in locally modifying the hydraulic properties of the rock. If a repository is designed such that local temperatures are allowed to build up to high values, then such thermomechanical responses must be considered when assessing the stability of mined openings. Where thermal stresses occur in a completely backfilled repository they are of considerably less importance.

Hydrogeology

Solute transport in the near-field is influenced by the regional hydrogeology of the host rock formation, the design and construction of the repository and the physical characteristics of the engineered barriers. Even though the regional hydrogeology constrains the maximum water flux through the entire repository, the rate and distribution of solute transport between the near and far-fields depends on the layout of the near-field. To illustrate this, we can consider the case of uniform flow through an homogeneous porous medium. If an emplacement tunnel is drilled through such a formation and backfilled with material of lower hydraulic conductivity than the host rock, water will tend to flow round the tunnel rather than through it (F 5.3), and hence the net water flux through the near-field would be less than that in the far-field. The near-field is generally more complex than this, however, and any tunnel will tend to be surrounded by a damaged rock zone of higher hydraulic conductivity caused by the construction process. In reality, of course, the situation is further complicated by heterogeneities/anisotropies in the properties of both the host-rock and near-field materials. Additionally, in very low-flow environments, molecular diffusion will be the dominant solute transport mechanism and this may need to be coupled to advective transport in more conductive regions. In order to assess barrier performance, a simpler model of solute transport is often used in which it is assumed that the entire water flux passing through the repository can exchange solute with the near-field, and is shared equally between individual waste packages. This is often termed the 'equivalent flux' and expressed in litres/container/year, as if all solute transport occurs by advection-even in a diffusion dominated environment. The extent of conservatism in this approach can be evaluated by more complex two- or three- dimensional advective/diffusive transport calculations.

Chemistry

The final major characteristic of the near-field is the water chemistry. The chemistry of the far-field groundwater, together with the hydrogeology, will define the input rate of solutes to the near-field and hence its extent of interaction with the various barrier materials and with the waste itself. A full specification of groundwater chemistry requires knowledge of at least pH, Eh (the extent to which the water is oxidizing or reducing) and the concentration of major ions ($Na+$, $K+$, Ca^2+, $Cl-$, SO_4-, etc). In addition further trace species might need to be defined to evaluate their reaction with particular barriers or their effect on radionuclide mobilization. As groundwater flows or diffuses from the far-field into the near-field, its chemistry will change due to reaction with its surroundings. Even before contact with the engineered barriers, hydrothermal reaction with damaged or heated host rock may alter the water chemistry, a process associated with the formation of secondary minerals. The changes in chemistry associated with such reactions, and indeed reactions with backfill, container or waste matrix, can be evaluated by chemical thermodynamic calculations. A large number of computer codes exist which can predict the result of reaction of water of known initial chemistry with a range of solid phases at any given temperature. If the timescales of the reactions are such that chemical equilibrium can be assumed, the main limitation to the use of such codes is the availability of appropriate thermodynamic data. For such calculations, basic thermodynamic data (free energies and entropies) are required for all reactions which may occur in the system. Even if only the most likely reactions are considered, extensive gaps in the thermodynamic database currently exist. This is especially problematic for some of the rather uncommon elements such as Se, Tc, Sn, Pd and the actinides. For such species, data either have very large associated uncertainties or are completely missing. Apart from bulk water chemistry, trace element solubility and speciation (i.e. the chemical form of the dissolved material) can be derived from such models. The solubility limit is obviously an important constraint on the release rate of radionuclides, while their speciation (as anions, cations, or unchanged molecules) greatly alters their retardation during transport. Rates of formation or reaction of solid phases can be derived by coupling such chemical models to models of solute transport. The chemical interaction of groundwater with various barriers and the application of thermodynamic models to predict speciation and solubility limits are discussed further in the following sections. Before this, however, some factors which may possibly affect the applicability of simple chemical models are briefly considered.

Colloids can be defined in this context as particles in the nanometre to micrometer size range which can form stable suspensions in a liquid phase. Metastable solid phases are unstable thermodynamically but exist due to the very slow kinetics of their alteration into more stable products. Neither of these can be adequately taken into account in present water chemistry models. Colloids are especially problematic, as they could be mobile in the aqueous phase and thus cause barrier degradation and radionuclide transport at rates greater than expected on the basis of solubility/speciation constraints.

Micro-organisms are well established as causes of extensive material degradation, even at considerable depths. Even the rather harsh environment of the near-field of a HLW repository cannot be guaranteed to be, or to remain, sterile. The effects of micro-organisms may either be direct-enhancing degradation of structural materials by enzymatic catalysis or increasing transport rates by acting as living colloids or indirect-perturbing the near-field with metabolic by-products such as gases or organic complexing agents.

A wide range of organic molecules may be naturally present in deep groundwaters, or may be produced in the near-field by microbial action. Such organics may more readily form stable complexes with key radionuclides than do many of the important inorganic complexants present in groundwaters. The very wide range of chemistry of such organics, together with the almost complete absence of any relevant thermodynamic data, makes the modelling of the behaviour of such species almost intractable at present. Given this background information on the physico-chemical environment of the near-field, we can now examine the role and performance of the individual engineered barriers.

Performance of the engineered barriers

Waste confinement by the disposal system should remain effective until the radionuclides have decayed to acceptable levels for entering the human environment. With a sufficient number of natural and man-made barriers, the release of radioactive material can be limited or delayed, its transport retarded or its concentration sufficiently diluted to assure that the impact on man will remain within acceptable levels.

Backfill (or Buffer)

The backfill may fulfil a number of roles, including minimization of groundwater access to the waste package, alteration of groundwater chemistry, and retardation of solute transport. It may display plastic properties-filling any gaps formed during or after emplacement and may contribute to the even spread of near-field mechanical stresses. It also acts as an important conductor of radiogenic heat. In fractured crystalline host rocks the most widely advocated backfill material is bentonite clay (which consists principally of sodium montmorillonite) compacted to a high density prior to emplacement, or a compacted mixture of bentonite and quartz sand which conducts heat more readily. Bentonite displays the ability to swell as it takes up water from the surrounding rock, and in doing so would squeeze into fissures around the borehole wall and fill any other cavities, while exerting a considerable swelling pressure on the waste package itself. Since it has an extremely low permeability to water (in the order of 1E-13 m/s hydraulic conductivity) it effectively prevents advective flow of mobile groundwaters to the package once it has taken up water and expanded. Diffusion of dissolved material through the extensive connected pore space in bentonite occurs, but may be greatly limited by sorption processes or the small size and surface charge of the pores. At temperatures below about 100 C, bentonite tends to buffer pH in the mildly alkaline range and the high surface area of the material ensures saturation of dissolved silica (SiO_2).

Ion exchange of cations in solution for sodium in the clay structure would also occur which may have further secondary effects. For example, sodium exchange with calcium in solution encourages calcite ($CaCO_3$) dissolution. This increases the concentration of carbonate in solution and may increase pH, which could alter near-field chemistry significantly. Bentonite may also directly affect the rate of canister corrosion-for example it may react with iron or steel to form layers or iron silicates which protect the canister from further corrosion. Apart from bentonite, several other buffer materials have been proposed, such as cement grouts or concretes which also display very low permeabilities but buffer pH to much higher values (more alkaline) than bentonite and have a rigidity and ease of emplacement that may be useful in many cases, particularly for ILW. For HLW disposal in basalt it has been proposed that finely powdered basalt would be an ideal buffer since the glassy matrix of the rock is highly reactive to the warm groundwaters which would initially be present in the near-field, and would rapidly crystallize as a very fine-grained and impermeable cement-like hydrothermal reaction product, which might then seal the waste package from further water ingress. In a dry formation such as salt, the requirements for backfill performance are much less extensive than for wet conditions, being mainly concerned with ensuring stress and heat transfer. Crushed salt is probably sufficient in itself. In very dry climates where a deep repository is above the water table (e.g. in tuffs at the Nevada Test Site), it has been proposed that an air gap may be the best 'backfill', as the capillary suction in the pores of the unsaturated rock ensures that the gap will not fill up with water and. Finally, very complex admixtures have been proposed to tailor a buffer to act as a 'getter' which would scavenge certain leached radionuclides from solution and fix them. Such buffers aim to control the oxygen activity of passing groundwaters so that radionuclides, principally the actinides, are reduced to valence states where they are most strongly sorbed on the buffer matrix. Apart from redox control components such as metals and sulphides, and sorbing media such as clays or zeolites, these backfills might also contain minerals which are known to react with specific elements and remove them from solution. These processes and their effects are considered in detail in the modelling section. At present the stability, longevity and reactivity of such tailored buffers in the warm hydrothermal environment of the near-field are not well understood. Backfill material will be selected on the basis of the general considerations above, and its performance under site and design specific conditions has to be evaluated.

Physical and engineering properties (e.g. thermal conductivity, strength, plasticity, swelling pressure) can be readily measured by standard techniques. Evaluation of chemical reactions and backfill stability is generally more problematic. In most cases the rock/groundwater/backfill/canister system will be thermodynamically unstable and undergo slow but continuous chemical reaction until backfill and canister are totally altered into stable products. The modelling of such reactions from first principles using rigorous thermodynamic calculations is beyond present capabilities and may be further complicated by kinetic factors at lower temperatures (100 C). At present, therefore, performance is mainly evaluated based on empirical laboratory experiments. In order to be applicable, such experiments must rigorously duplicate the expected environment (in terms of temperature, pressure, chemical components and so on) and even then extrapolation from laboratory to geological timescales is difficult to do with confidence. In the case of bentonite (perhaps the best studied backfill material), all relevant physical and engineering properties have been well measured, and predictions made on the basis of such measurements have been validated by laboratory and field experiments. In granitic groundwaters, the montmorillonite clay of which bentonite consists is expected to alter into another clay (illite). This alteration requires a supply of potassium ions from the groundwater which, given the low flow rates expected, ensures backfill lifetimes of millions of years, even without consideration of further limitations caused by slow kinetics. Although this conclusion is supported by observations of bentonite behaviour in natural geological systems, alternative alteration mechanisms have been proposed and a rigorous mechanistic model for this process does not currently exist. Similarly, although simple models of bentonite/water reaction exist, they do not consider the minor components of the bentonite (i.e. other than montmorillonite), do not take kinetics into account, and are based on rather poorly specified laboratory data. Radionuclide sorption onto bentonite has been extensively studied but in most cases experimental conditions were not relevant to those expected in repositories and the retardation predicted often agrees poorly with that observed in situ migration experiments.

Container

The function of the canister in most disposal concepts is to protect the waste from groundwater for a minimum period of time during which a certain proportion of the contained radioactivity will have decayed away and any thermal transient caused by radiogenic heat will have passed. The container may also be designed for ease of handling during emplacement operations or for optimization of packing geometry in the repository. The radiation shielding effect of the container limits possible exposure to operators and radiation damage to external engineered barriers after emplacement (including the radiolysis of groundwaters). The mechanical performance required to ensure that it will withstand emplacement operations and subsequent pressure re-equilibration generally leads to the choice of a metal for the container. More sophisticated designs (for example an inner metal canister surrounded by a ceramic overpack) are also being studied, but analysis of their behaviour is complex. Two basic conceptual choices of container design exist. The first aims at corrosion resistance, using materials which are thermodynamically stable in the repository environment, or form passivating layers of corrosion products. For example, native copper is found in some deep crystalline rock environments in Sweden, which leads to expectation that a copper canister would be stable over a period of millions of years. Although titanium would corrode quickly in this environment, it is known to form an oxide layer which limits further corrosion, which again provides a very long canister life. As very little (or no) corrosion is expected, resistant containers can be relatively thin and thus may be constructed out of expensive materials. The alternative concept is corrosion allowance in which container corrosion is expected, but is allowed for by designing the container to be thick enough to give the desired lifetime. In this case the container may be very massive, and made out of a cheap material such as cast iron or steel.

Much of the research on containers is involved with the practicalities of their construction and in particular, their sealing. Lid seals, welds and seams are all potential sources of weakness and likely sites for enhanced, localized corrosion. For most of the materials being considered seriously, techniques currently exist which seem capable of solving such problems. In the case of copper, for example, techniques such as electron beam welding and hot isostatic pressing have been demonstrated to provide reliable quality control. Nevertheless, most safety assessment models incorporate terms to allow for containers which have been inadequately sealed and escaped quality control checks prior to emplacement. Such analyses can be made probabilistically, and essentially give a proportion of early container failures in the near-field source term. After choice of container material, it is necessary to demonstrate that desired lifetimes can be obtained in disposal systems where this is considered an important factor. In cases where the groundwater supply rate to the near-field is very low, this in itself may constrain minimum container lifetime. Otherwise the rate of corrosion under expected chemical conditions must be extrapolated from laboratory measurements. Again the total chemical system is usually too complex to be rigorously modelled at a thermodynamic or mechanistically based kinetic level.

Corrosion rate measurements may be complex in practice. For some materials, the rates of corrosion are so slow that simple weight loss measurements are impracticable on a laboratory time-scale and sophisticated indirect electrochemical techniques are required. In addition, some form of surface analysis or inspection is required to determine the spatial variability in corrosion rate. In general, enhanced local corrosion (pitting) is less important for thick corrosion allowance canisters than thin corrosion resistant materials as the pitting factor (the ratio of the maximum to the average corrosion depth) is usually observed to decrease with increasing amount of corrosion. Nevertheless, particular conditions which enhance pitting (e.g. electrical cells set up at the junction between different materials) should be carefully avoided. Measurements must be performed under realistic conditions of temperature and pressure. Apart from absolute pressure, the stress field expected on the canister may be important as corrosion may be enhanced at positions of greatest stress, and mechanical failure may occur earlier than expected due to so-called stress/corrosion cracking. Finally, the measurements should take into account the effects of the backfill (both on water chemistry and direct reaction) and, where relevant, the radiation field. For HLW in particular, the latter has two main effects : direct damage to the container structure and radiolysis of water. The 'ageing' effect on the metal caused by the low radiation field acting over long periods of time can be simulated to some extent in the laboratory by short exposure to much higher radiation fields in a reactor. For most of the materials considered, such work shows that this is not likely to be a significant problem. Breakdown of surrounding water by radiation (radiolysis) can give rise to a mixture of oxidants (such as hydrogen peroxide), and reductants such as hydrogen gas. If the latter is lost, by rapid diffusion, then a net oxidising environment is produced, which generally results in enhanced container corrosion. Interaction of radiation with components dissolved in the water may result in the formation of acids (e.g. nitric acid from trapped air dissolved in the water) which again are generally corrosive. The effects of radiolysis can be quite significant for the thin corrosion resistant canisters as external radiation fields are higher and even small amounts of localised corrosion could cause premature failure. For this reason, some corrosion resistant overpacks include a radiation shield, such as lead, inside the resistant metal.

The final model for calculating canister lifetime may simply extrapolate from measured corrosion rates or also take into account the rate of supply of corrodants and radiolytic effects. A further factor which could be significant in some cases is microbial corrosion but scoping calculations indicate that its effect is small and can generally be ignored or assumed to be negligible. Apart from container lifetime, the chemical effects of its corrosion on other near-field processes may have to be considered. For example, corrosion of metal under anoxic conditions often results in the formation of hydrogen gas. For corrosion-allowance containers within a low permeability backfill such as compacted bentonite it has been calculated that this could build up extremely high pressures which could cause premature canister failure, disrupt the integrity of the backfill or even damage the surrounding rock. At present it is expected that such pressure build up will be self-limiting by decreasing the corrosion rate or causing gas flow (rather than diffusion) through the backfill, but the inclusion of a fine sand layer around the canister to decrease corrosion rates further as gas builds up is also considered. Even after water has penetrated the container it may continue to perform some barrier role. For the chemically-unreactive containers this may simply be as a physical constraint on solute transport resulting from remnant overpack and corrosion products. More reactive materials may, after failure, also act as chemical buffers and radionuclide sorbants. In particular iron oxides resulting from corrosion may buffer both pH and redox conditions (ensuring a chemically reducing environment) and strongly sorb many radionuclides. Quantitative evaluation of these chemical effects indicate that they could be very important, but the requisite laboratory background data are somewhat limited at present.

Waste matrix

The main function of the waste matrix is to limit the rate of release of radionuclides after failure of the container. In this section we will consider the behaviour of two HLW matrices (borosilicate glass and the unreprocessed uranium dioxide spent fuel itself). The first is unreprocessed spent fuel, and comprises the used, irradiated fuel rods, which are inserted in the disposal canister which may subsequently be evacuated or filled with gas or lead. The second is borosilicate glass, which incorporates the waste radionuclides intimately dispersed throughout its structure. It can be formed either by in-can melting or by pouring molten glass into a steel fabrication container. It is generally assumed that during cooling and subsequent handling and transport, the manufactured blocks of glass would fracture, and hence present relatively large surface areas to any water which penetrates the engineered barriers. This is a conservative assumption used in the design of leaching experiments and in calculating release rates. These two waste types are treated separately below.

Leach testing

Readers may come across many references to 'leach-testing' of all types of solid radioactive waste destined for geological disposal and it is worth pointing out that there are basically two types of experiment which should not be confused. The first is essentially a standard sorting technique, used to compare the overall quality of waste forms, for example batches of a vitrified waste with slightly different compositions. These are tests only, and give information on the bulk 'leachability' of a product. Various standard techniques are used, the IAEA recommended procedure being commonest. Some dynamic tests use continuously replenished fresh water (e.g. Soxhlet tests) as the leaching agent. The second type of leach testing is an experimental method which attempts to replicate realistic disposal conditions. The disposal environment will be characterized by virtually zero groundwater flow, so these experiments are generally closed-system, static leaching tests. Data are produced in the form of individual element concentrations in solution as a function of time, temperature, solid to fluid ratio, and so on. This second type of experiment is the only reliable means of providing data for release modelling and, as many authors have pointed out, data on bulk leach rates from flow-through tests should not be applied to realistic safety assessments. The leaching of various waste forms has been very intensively studied and is now quite well understood. This can be illustrated by considering borosilicate glass in some detail.

Borosilicate glass behaviour

While a variety of glass formulations have been studied as containment media, including phosphate and syenite glasses, borosilicate compositions have demonstrated greater stability or flexibility. Their formation temperatures are quite low, they are able to incorporate the bulk of the HLW components satisfactorily, and their leach behaviour is well understood. A typical waste glass composition (COGEMA specification 1982) is:

Table 44 Composition of high-level borosilicate waste glass.
Container weight = 480 kg; glass weight = 405 kg/container.
Added oxides (%) : SiO_2 45, B_2O_3 14, Al_2O_3 5, Na_2O 10, CaO 4, Fe_2O_3 3, NiO 0.4, Cr_2O_3 0.5, P_2O_5 0.3, ZrO_2 1, Li_2O 2, ZnO 2.5.
Fission product oxides : 11.1%, Actinide oxides : 0.9%, Metallic particles 0.7%
Actinides g/container: Am 423, Cm 33, Pu 80, Np 573, U 1920.

The major glass network forming complex is the SiO_2 molecule, with silica comprising up to about 50 % of the glass and dominating the dissolution behaviour of the glass in water. Borosilicate glasses react with water very slowly at low temperatures but can be highly reactive at elevated temperatures and pressures. The mechanism of waste dissolution changes with increasing temperature, and as a function of time. At temperatures approaching 200 C the glass begins to break down quickly under hydrothermal conditions. However, the breakdown of the glass matrix may not be the principal factor which controls the release rate of radionuclides into the near-field groundwaters. As the waste breaks down, a wide variety of secondary products are formed. These can be amorphous materials, colloids or stable mineral phases, depending on the temperature and the groundwater composition. The solubility of any radionuclide is thus dependent on the extent of its incorporation into these new materials and the relative stabilities of each in the groundwater environment. These in turn will vary with the rate of turnover of water, and with temperature and redox conditions. Thus radionuclide mobilization, either in solution or particulate/colloidal form, will be a rather complex function of the thermodynamics of a number of reactions in the near-field. This argument is true for any waste form where groundwater access is limited by the barrier system and holds for ILW types as well as HLW types. Tracking the behaviour of individual radionuclides up to their point of mobilization.

The change in concentration (F 5.4) in solution of a radionuclide released from any metastable wasteform, such as glass, is a function of time. Up to certain time tc, which is very dependent on the temperature of the system, the rate of water exchange and the surface area of the waste form being leached, a wide variety of 'leach rates' can be produced; leach rate being equivalent to the gradient of the tangent to the curve at any given time, tA, tB, etc. These are clearly only applicable to that time, and a general trend of decreasing leach rate can be seen, as the tangents flatten out. All of these leach rates will overestimate releases. When the concentration curve flattens out the radionuclide will be present in solution at its solubility limit, with respect to whatever phase is dominating its behaviour at that time. With increasing time the whole near-field system will progressively readjust to replace metastable reaction products with more stable phases, and the consequent solubility limits of individual radionuclides will decrease in a stepwise fashion. The initial 'solubility' limit portrayed thus represents a conservative figure for radionuclides in solution at any time after disposal. An important feature of this approach is that experiments must aim to characterize fully the solid phase chemistry and aqueous phase speciations in order to interpret and predict the overall reaction path.

Taking one step back from this approach, simple empirical calculations can be made of mobilization rates in a near-field where water availability is considerably limited by the barriers and the surrounding far-field. This was attempted, using very simple solubility criteria. A further example of the use of this approach is presented in the detailed near-field model considered later. Depending on the leaching mechanism chosen, and the degree of significance attached to saturation effects, this highly empirical approach predicted very protracted waste mobilization episodes, suggesting that the waste glass and the secondary waste products will have extremely long lifetimes, in the order of millions, or tens of millions, of years, with releases taking place at very low concentrations.

Spent fuel behaviour

Spent fuel is a very different waste form to borosilicate glass. First, its content of nuclear-reaction by-products is more dilute, since the fuel has not been reprocessed and these radionuclides extracted and concentrated. Second, the waste matrix is either uranium metal or oxide, rather than glass, and the dissolution behaviour of the waste is thus largely controlled by the solubility of uranium in water. The matrix is itself radioactive and, when integrated over long time periods, radiation effects such as alpha radiolysis may be more significant. Third, the thermal output of spent fuel shows different characteristics, being significant over substantially longer periods of time (up to

10 000 years), rather than exhibiting steady decay over about 1 000 years. In addition, the presence of very labile elements (such as Cs and I), which diffuse through the waste matrix and concentrate on the outer surface during the operational life of a fuel element, must be accounted for when studying the leaching of the waste. Leaching is controlled by the solubility of uranium which is very dependent on redox conditions and the concentration of complexing species (ligands) such as carbonate in solution. In reducing conditions this solubility is very low and, in addition, deep groundwaters are usually already saturated with naturally occurring uranium. In much the same way as glass dissolution, individual radionuclide mobilization rates are controlled by the nature of surface layers formed, and are highly dependent on groundwater pH and temperature. Models of radionuclide mobilization often avoid the complex thermodynamic consideration of secondary reaction products by making the generally conservative assumption that releases are congruent with the matrix dissolution. An exception is immediate releases, such as the 'instant' iodine, or caesium. These elements concentrate at grain boundaries or the inner surfaces of cladding during reactor operation.

Long-term leach tests on spent fuel in natural and synthetic groundwaters have been performed in Canada and Sweden, while in the USA work has concentrated on their hydrothermal stability in brines related to disposal in salt formations. The UO_2 matrix dissolves rapidly under oxidizing conditions, and these may be produced in the very more rapid near-field as a result of radiolysis of groundwaters. Leaching experiments on spent uranium dioxide fuel from CANDU reactors at temperatures of 25-30 C have been underway for 8 years, using distilled water and tap water. Matrix dissolution of the order of 1E-6 to 1E-9 per day, with uranium concentrations in solution well below theoretical saturation limits. Crystalline reaction products are formed on the fuel surface, and the mobilisation rates of more labile species such as Cs decrease very rapidly with time, to a presumably steady-state level, reminiscent of the process outlined earlier for glass dissolution. Long term tests in Sweden in synthetic granitic groundwaters show that Sr behaves similarly, although the more significant radionuclides (U, Pu, Cm, Ce and Eu) enter solution more slowly, reflecting solubility limitation controlled by their precipitation or adsorption on to the fuel surface, again parallel to the glass dissolution mechanism. Earlier Swedish work also demonstrated that both Sr and U were present in the aqueous phase as colloidal material; up to 50 % of the amount in the leachate in the case of Sr.

Modelling of radionuclide release from the near field (Chapman 1987)

We can now follow through the calculations used in repository safety assessment to evaluate releases from the near-field. These are used as input for the far-field migration calculations considered in the next chapter. First of all we look in detail at one specific near-field layout which was recently proposed for a Swiss deep HLW repository. The general principles can be applied to any deep repository nearfield for all types of long-lived wastes, but some specific issues emerge which are briefly examined. Finally the problems and limitations associated with present models are discussed.

The Swiss near-field model

The base case of the Swiss analysis (NAGRA, 1985) envisages disposal of vitrified HLW in a cast steel overpack (F 5.6). The containers are emplaced horizontally into tunnels which are backfilled with compacted bentonite. The repository is at a depth of 1200 m in granite. Deep groundwater flow is heterogeneous, occurring mainly in widely separated, subvertical disturbed zones in the rock. The calculated water flux through the entire repository is 4200 l each year. This water is slightly reducing chemically, and at near neutral pH. The ambient temperature at repository depth is 55 C and the radiogenic thermal transient gives a maximum temperature of 160 C inside the backfill and 100 C at the disposal tunnel wall. These high temperatures drop off within the first few decades, and are insignificant after about 500 years as far as near-field analysis is concerned. The quantities of materials involved are:

Table 45 Material inventory (per waste container) in the near-field of a reference Swiss high-level waste repository.

Material	Volume (m^3)	Mass (kg)
Glass	0.15	405
Steel-fabrication container	0.01	75
Fabrication void	0.03	-
Canister	0.9	6500
Backfill	52.8	-
(a) Bentonite (dry)	32.7	88000
(b) Pore space (water-filled)	20.1	20000

Calculations indicate that the bentonite will be mineralogically stable for >1E+6 years and its chemical and mechanical properties will be retained for such a period, after corrosion of a 5 cm thick 'allowance', the container will fail mechanically. This corrosion will take at least 1300 year (rounded down to 1000 year in the safety analysis). The low solubility of the corrosion products ensures that they exert a chemical buffering role for >1E+6 years. At the time of canister failure, the glass will be fractured to an extent which increases its area by a factor of approximately 12 times its original value. The rate at which the glass breaks down is 1E-7 g/cm^2 of surface area/day. Chemical buffering reactions in the bentonite and canister ensure that the pH of pore water is slightly alkaline (in the range 7-8.5) and chemically reducing. The log. of solubilities of particular elements of interest under these conditions are equal to : high :

Cs, C, I, Rn; -2 : Ca, Ac, Am, Cm; -4 : Be, Ni, Ag, Sr; -6 : Pb, Th, Pa, Pu; -8 : Ho, Nb, Pd, Ra, Sm, Sc, Np; -9 : Sn, Zr and U. Combining all this information, we can build our first simple model of release. As the canister lasts 1000 years, we can take decay during this period into account and the table shows that within this time the quantities of some short-lived nuclides have already become insignificant.

Over the long periods involved, it is considered that the waste dissolves congruently which, immediately after canister failure, would release in the order of 1E-5 of the inventory each year. If the bentonite/canister barriers are ignored at present and all the radionuclides released are assumed to dissolve in the water flux of 0.7 l/canister/year, their concentrations in solution would be as given. However, these releases may be further constrained by limiting solubilities, and the concentration which would saturate the reference water flux is shown in the final column (where more than one isotope of the same element is present, total solubility is divided between them in proportion to their total inventories). It can be seen that the concentration which would actually be released to the far-field is the lower of the values in the last two columns and this is shown underlined. Releases of 137-Cs, 90-Sr, 108m-Ag, 121m-Sn, and 151-Sm are all negligible.

Table 46 Fission activation product inventory 1000 years after disposal in the Swiss reference HLW repository.

Release rate limited by :			solubility		congruent dissolution	
RI	T - y	I -mol	Mol/y	Bq/y	Mol/y	Bq/y
10-Be	1.6E+6	2.6E-5	5.1E-10	4.2E+0	7.1E-5	5.9E+5
14-C	5.7E+3	1.9E-5	3.7E-10	8.5E+2	high	-
41-Ca	1.3E+5	8.7E-5	1.7E-9	1.7E+2	7.1E-3	7.2E+8
59-Ni	8.0E+4	1.1E-2	2.2E-7	3.6E+4	7.1E-5	1.2E+7
63-Ni	1.0E+2	1.8E-6	3.5E-11	4.6E+3	1.2E-8	1.6E+6
79-Se	6.5E+4	9.3E-2	1.8E-6	3.7E+5	7.1E-9	1.4E+3
90-Sr	29	6.1E-10	1.2E-14	5.5E+0	7.1E-5	3.2E+10
93-Zr	1.5E+6	1.1E+1	2.2E-4	1.9E+6	7.1E-10	6.3E+0
94-Nb	2.0E+4	3.9E-4	7.6E-9	5.0E+3	7.1E-9	4.7E+3
93-Mo	3.5E+3	6.6E-6	1.3E-10	4.9E+2	7.1E-3	2.7E+10
99-Tc	2.1E+5	1.1E+1	2.2E-4	1.4E+7	2.3E-8	1.4E+3
107-Pd	6.5E+6	2.5	4.9E-5	1.0E+5	7.1E-9	1.4E+1
108m-Ag	1.3E+2	2.8E-8	5.5E-13	5.6E+1	7.1E-5	7.2E+9
121m-Sn	50	4.2E-11	8.2E-16	2.2E-1	8.3E-20	2.2E-5
126-Sn	1.0E+5	3.6E-1	7.0E-6	9.3E+5	7.1E-10	9.4E+1
129-I	1.6E+7	1.8E-3	3.5E-8	2.9E+1	high	-
135-Cs	2.3E+6	3.2	6.2E-5	3.6E+5	high	-
137-Cs	30	2.0E-8	3.9E-13	1.7E+2	high	-
147-Sm	1E+11	1.5	2.9E-5	3.5	7.1E-9	8.5E-4
151-Sm	93	7.0E-5	1.4E-9	2.0E+5	3.3E-13	4.7E+1
166m-Ho	1.2E+3	5.4E-6	1.1E-10	1.2E+3	7.1E-9	7.8E+4

I = Inventory

Although extremely simplistic, this type of approach is widely used in calculating release functions. Computer models can readily calculate releases of all nuclides as a function of time taking into account changes in glass surface area during leaching, and complex decay chains. A typical example (F 5.7) indicates that even though the glass matrix has been totally destroyed within 150 000 years, the limited solubility of the radionuclides from the secondary reaction products in which they are by this time incorporated, may greatly extend release times. The model above is very conservative as it ignores the diffusive resistance of the bentonite. We could improve it by assuming radionuclides are released into a small volume of water inside the canister (corresponding to the original void space, say) and that diffusive transport to the tunnel wall then occurs. If we assume that a water volume of 0.7 l is distributed around the wall (as a 'mixing tank'), and this is changed each year, this could be considered a reasonably conservative boundary condition. For the diffusion calculation, interaction between radionuclides and the bentonite is modelled very simply by a partition coefficient which assumes a constant ratio of concentrations in the solid and liquid phases, i.e.:

$$Kd = CR/CW$$

where Kd is the partition coefficient or distribution coefficient (m^3/kg); CR is the radionuclide concentration on the rock (e.g. Bq/kg) and CW is the radionuclide concentration in the pore water (Bq/m^3).

This distribution coefficient (Kd = 5 for Ac, Am, Cm, Pu, and Zr; = 2.5 for Ho, Nb, Sm; = 1 for Pb, Np, Ni, Pa,Th and U; = 0.2 for Cs, Ca, Ra, Sr, Tc; = 0.05 for Sn, = 0.01 for Be; = 0.005 for C, I, Mo, Pd and Se) can be transformed into a retardation factor (R) by taking into account the porosity (P, 0.38) and density (D, 2760 kg/m^3) of the rock:

$$R = 1 + (1-P)*D*Kd/P = 1 + 4500*Kd$$

The R value corresponds to the ratio of the speed of transport of an ideal, unretarded ion to that of the radionuclide concerned. The R values vary from 24 (Se, Pd,.) to 23 000 (Ac, Am..). The final factor which has to be taken into account is radioactive decay during transport. The maximum release rates for a number of fission/activation products calculated by a simple one-dimensional diffusion model are a function of the 'nuclide characteristic' product of decay constant and retardation factor. Given natural background concentrations of radionuclides, releases below about 10 Bq/l can probably be considered negligible. Hence, out of the very large number of fission/activation products in the original waste, only 135-Cs, 79-Se, 107-Pd, 99-Tc, and 126-Sn are mobilized into the far-field in significant quantities.

Release models for other near-field designs

The numerical values in the previous section are obviously 'concept-specific' but the general approach can be applied to any near-field design. The essential parameters required are the functional lifetimes of individual barriers, release rate from the matrix, radionuclide solubility limits in regions of different chemistry, and retardation rates during migration within the near-field. Some factors not considered in this basic model which might, however, be important for other layouts are now considered. For concepts in which the engineered barriers do not act as chemical buffers, radiolysis may be very important. Radiolysis is particularly significant in the case of spent fuel, and Swedish calculations indicate that for disposal in a copper canister oxidizing conditions caused by radiolysis may penetrate up to about 50 m into the surrounding rock (KBS, 1983). For the far-flow path. The evolution of this oxidizing region with time is important, as the solubility of the waste matrix and many important radionuclides is higher under oxidizing conditions, and the extent of radionuclide retardation is less. In addition, the decrease in solubility of some radionuclides at the boundary between oxidizing and reducing conditions could give rise to colloid formation. Heterogeneous distribution of radionuclides can also require variations from the basic model. The 'prompt' release of labile elements such as I and Cs from spent fuel due to their accumulation at the waste surface has to be taken into account in calculations.

Finally, temporal changes in the properties of individual barriers may have to be taken into account. For example, the initially very high pore water pH in cements will decrease with time as interaction with groundwater occurs, and the matrix permeability also changes as its physical structure degrades. Although these processes occur gradually, models generally simplify this to a stepwise progression in which the simple calculation chain is repeatedly applied for different time periods during which all properties are taken to be constant.

Limitations and problems

The models considered above are all obviously great simplifications of reality and the limitations of their application must always be kept in mind. For example, all analyses available assume that releases from a single canister can be treated in isolation and multiplied up to give total repository releases. An alternative approach involves associating distribution of properties, e.g. variations in canister failure time which can average out releases over a long time period. Processes could also be envisaged in which releases from one canister could affect others. In the case of a distribution of canister failure times, radiolytic oxidants might cause accelerated failure of containers downstream of previously failed ones in a kind of domino effect. Such interactions could also be favourable when one considers processes which are limited by solute supply from groundwater or solubility-limited releases.

Another tacit assumption in the model chain considered is that all transport occurs in solution and is thus controlled by solubility limits. If significant transport of colloids or suspended particulates occurred, the results of the model would be completely invalid. Formation of colloids can result from breakdown or erosion of barriers, radiolysis reactions or precipitation at the boundary between zones of different chemistry (e.g. at radiolytic redox fronts). Colloids may also be supplied naturally by the groundwater. Few data are available, however, on the likely concentration, mobility or stability of such colloids and they are often disregarded on this basis-assuming any effects involved are masked by other large conservatisms in the analysis.

The only information available on colloid migration in the far-field applies to aquifers with large open pores, where transport of colloids, suspended particles and micro-organisms is well established. Such materials can even be used as groundwater 'tracers'. In more relevant formations few, if any, data are available. Although colloidal size particles (i.e. those less than 1 micron in size) can be detected and measured in water samples from deep boreholes, there are strong suggestions that these may be artefacts of the sampling process. Some tracer migration experiments have shown mobility of quite large organic molecules in crystalline and argillaceous rocks, but significant long-range colloid transport has yet to be demonstrated.

The pore structure of the engineered barriers may act like a molecular filter, preventing movement of colloids. Compacted bentonite, in particular, is often assumed to prevent colloid migration completely and, while this has not yet been experimentally proven, their mobility seems to be at or below the present limits of measurement. In this case, if all expected formation mechanisms occur within this barrier, near-field radionuclide releases are unaffected. They may even be decreased if the immobilized colloids contain significant quantities of key radionuclides. Apart from problems caused by colloids, the use of solubility limits also assumes that these values can be measured or predicted by using chemical thermodynamic models. For many radionuclides relevant data are extremely sparse and some values used are little better than educated guesses. The applicability of current thermodynamic models to low temperature groundwaters is also debatable at present. Finally, perturbations caused by micro-organisms have not been considered. Apart from biodegradation of engineered barriers, such organisms could also produce metabolic by-products which considerably enhance the solubility and mobility of some radionuclides. Relevant background data are very limited, but simple calculations indicate that such effects would not be significant for the Swiss HLW concept. Shallower repositories and ILW types are more likely to be perturbed, in particular those containing large amounts of organic wastes or bitumen, but no detailed analysis has been carried out so far. Given the limitations and problems above, it is important to evaluate the extent of 'conservatism' in near-field models. We also need to evaluate the relative performance of individual barriers by means of sensitivity analysis, and to compare different disposal concepts. This requires the near-field models to be as realistic as possible, even though their development is still at an early stage. For illustrative purposes, however, we can again consider the Swiss HLW case. Even the most realistic model yet used, which incorporates diffusion in the backfill, has failed to consider in detail exactly what the physical system would look like. Glass degradation would form extensive suites of secondary minerals which incorporate radionuclides and form a diffusion barrier. Any cracks through the canister which occurred during its mechanical failure would be infilled by corrosion products or swelling bentonite and form low permeability 'bottlenecks' to radionuclide transport. Even after corrosion of all the canister material, the corrosion products would probably have very low permeability and a high retardation capacity for many radionuclides. In the Swiss case most of the water flows in widely separated disturbed zones in the far-field and may not be uniformly available throughout the near-field. Hence the three-dimensional diffusion path through the backfill may be much larger than that in the simple one-dimensional calculation considered previously. The total water flux through the repository occurs mainly between disposal tunnels and hence the volume available for saturation may be very low and further limited by swelling of bentonite into any open porosity of the tunnel wall. Development of 'realistic' near-field models is now an important research area, and the limited results to date indicate that under expected conditions, applying a realistic cut-off time (say a million years), the releases from the near-field are negligible for many repository concepts. Such results very much strengthen the whole multibarrier concept.

Modelling of radionuclide migration in the far-field (Chapman 1987)

The 'far-field' volume of rock, the most important of the barriers to waste migration, extends from the 'near-field' disposal zone of the repository itself back to the Earth's surface. This chapter examines in more detail the function of the geological barrier in deep disposal concepts, the various rocks and geological environments which have been proposed as hosts for long-lived waste disposal, the factors which control the migration of radionuclides through these rock formations, and the assembly of a complete 'normal-case' release model for use in performance assessment. The quantification of parameters and understanding the interactions of geological phenomena in the far-field are the basic objectives of this, fundamentally geological, research area, in order that predictive performance models can be developed and tested.

The basic requirement of a waste repository is that the combination of its natural and engineered barriers should act to control the release of radionuclides to the biosphere. The far-field acts as a massive physical and chemical buffer to processes in the near-field, principally by controlling geochemical fluxes dominated by the rates of water movement. The near and far-fields are primarily coupled by exchange of solutes (e.g.

corrodants and complexing agents from the far-field, and corrosion products and leached radionuclides from the near-field). Once radionuclides are mobilized into the far-field, then the two main factors of significance in a safety assessment are the length of time for the radionuclides to reach the biosphere, and the concentrations in which they arrive, which are very dependent on the nature of the surface water body or aquifer into which they are released. These two 'far-field' factors are controlled by the path length and velocity of water-borne migration (by both advection and diffusion) through the host rock, overlying formations and superficial geology and the physical and chemical environment along this path, and in particular the ability of the rock to retard the rate of movement of individual radionuclides and dilute and disperse them in groundwaters. Concealed within these two factors are a host of influential processes and parameters which must be understood and quantified under specific circumstances for the purposes of a migration model. In attempting to define what might be suitable sites for a repository the aim is to find an environment whose properties give a good balance or combination of these important features. In all the subsequent discussion it is assumed that leaching by and transport in groundwaters is the 'normal' mechanism by which the wastes eventually reach the biosphere. It follows that we must consider not just the rock alone but the total geological environment. This includes the rock formations, groundwaters and surface cover, and their behaviour as affected by climate, terrain, geological evolution and the effects of the waste itself, all of whose properties vary with time. Some of these factors are completely site specific (i.e. related only to geographical position, regional geomorphology or local rock properties and structures) while others, particularly some physicochemical properties, are common to certain types of rock or certain environments, and are said to be generic.

The purpose of this section is to evaluate what have broadly been termed geological requirements and to see how their application has led to both the choice of generically suitable rock types and geological environments and eventually, in many countries, to the definition of potentially suitable areas of land where deep disposal might take place. The earliest comprehensive work relates to disposal of HLW. Three principal rock types (argillaceous or clay-rich units, hard crystalline igneous and metamorphic rocks; and evaporites-principally halite formations) most adequately fulfil the needs of a host unit for HLW disposal, and they have dominated international research programmes. This selection was made partly owing to their ability to accept heat emitting wastes, but chiefly on hydrogeological grounds. It was considered that since they are all of low permeability they would have very low rates of water through-flow, in terms of both volume and velocity. The task has now become one of quantifying and testing these largely intuitive notions, often at specific sites. In particular it has become clear that the properties of the large scale groundwater regime in which the host formation lies are as important as those of the host rock itself. This is especially the case for evaporate and clay formations lying in thick sequences of mixed sedimentary rocks.

Evaporite deposits, commonly referred to as salt in the literature, were thought to be completely impermeable, and also to have the advantage of being plastic enough to allow self-sealing of fractures by creep in response to heat or stress. Argillaceous rocks, although very diverse in physical properties, ranging from plastic clays through to hard, well-bedded and often fractured units, are generally characterized by low permeability and high sorption capacity (and hence radionuclide retardation potential). Massive bodies of hard, stable, crystalline rocks can have considerable vertical and lateral extent, are easy to construct underground facilities in, and are often very poorly permeable. More recently volcanic tuff deposits have been added to this group. In addition all the rock types, with some exceptions in the argillaceous and evaporate classes, are thermally stable under the heat load imposed by the wastes and can conduct and diffuse the heat away over very long periods of time.

The arrival of deep disposal concepts for other long-lived ILW types has largely been advanced on the back of the early HLW studies, and consequently very similar rock formations in similar environments are being studied for these non heat-emitting wastes. Without the added problem of finding a repository host rock with adequate thermal stability, the emphasis is leaning in some countries towards wider varieties of mixed sediments with good regional hydrogeological properties. These will be discussed later. The three groups of rock (generically known to the 'waste people' as crystalline, argillaceous and evaporate) are sufficiently broad categories to encompass almost all formations with physical properties which might prove suitable for deep disposal of long lived wastes. Consequently they will be retained throughout the subsequent discussion. Since the mechanism of groundwater movement is different in the three generic rock groups, owing to their different hydrogeological properties, the factors to be investigated vary in each case. The following sections provide brief introductions to these rock types.

Crystalline rocks

The term 'crystalline rocks' has crept into the waste disposal nomenclature as meaning hard, massive igneous or metamorphic rocks such as granite, gabbro and basalt. The common feature of all these rock types is that they have very low total porosity, and are generally jointed and fractured, on the scale of tens of metres down to centimetres (F 6.1). Here we use the terms fissure and fracture quite loosely to apply to any physical discontinuity in the rock extending from small cracks to large shear zones. Although the contribution of the fractures to the porosity of these rocks is small, water flow through the rock is dominated and controlled by these fissures, in terms of their apertures, orientations, numbers, degree of connectivity and so on. Major fractures (faults, crush zones and other features which can be traced over many hundreds of metres and sometimes many kilometres) occur at irregular intervals, are often nearly vertical or horizontal, and can be either more or less permeable than the surrounding rock. With the exception of basalts the crystalline rocks tend to occur as massive deep-seated plutons or extensive regions of intensely metamorphosed rocks which may be buried under younger sediments, or outcrop in seemingly unchanging monotony such as the shield areas of Canada, Scandinavia and north-west Scotland. Major fracture zones are usually present every few hundred metres. In fact none of these rocks is homogeneous either in terms of its mineralogical composition, its physical and chemical properties, or its fracture pattern. While some properties remain fairly constant from one area to another, this inhomogeneity must be taken into account in any detailed assessment of rock behaviour.

Basalts (solidified lava) typically occur in flows of variable thickness (up to several hundreds of metres) often layered one on top of another such that the total thickness of a 'pile' can reach several kilometres and extend over thousands of square kilometres. They display similar jointing and porosity to the other crystalline rocks but have an extra discontinuity to be considered, caused by the discrete nature of the flows. Tops and bottoms of flows are often rubbly or foamy. If long periods have separated basalt lava eruptions, soil layers can form which are subsequently buried under the next flow and persist as subhorizontal bands of higher porosity and permeability within the lava pile. However, since individual units can be some hundreds of metres thick, the potential for siting a repository in basalt clearly exists.

Since all of the crystalline rocks have formed at very high temperatures and either crystallized from a molten liquid or recrystallized as a refractory residuum during high temperature, solid-state metamorphism, it follows that they display considerable thermal stability and present few problems from the viewpoint of heat emitting HLW. Their hydrothermal behaviour, the interaction with warm groundwaters in the repository near-field, can, however, act as a control on nearfield chemistry. They are also reasonably good conductors of heat, have high compressive strengths, and at moderate depths (up to about 1500 m) can be easily and economically excavated to form self supporting caverns. At the time of writing, the various national research projects in crystalline rocks (Sweden, France, Finland, Canada, Switzerland and, to a lesser extent, the UK and the USA) have concentrated their efforts almost entirely on granitic or metamorphic rocks. The only work on basalts is the BWIPP (Basalt Waste Isolation Pilot Plant) project at Hanford (Washington State) in the USA. In addition the USA is giving equal consideration to a further 'crystalline' rock type; volcanic tuff, at the Nevada Test Site (NTS). This rock type is essentially compressed air-fall volcanic ash which may have been partly welded by its own heat immediately on deposition. Vast thicknesses occur in many parts of the world. At NTS it is compact and quite porous, with good engineering and hydrogeological properties. Owing to the presence of zeolite minerals in the pores it can also display a high radionuclide sorption capacity, although zeolites generally possess low thermal stability and repository temperatures would probably need to remain below 85 C.

Argillaceous rocks

While the crystalline rocks have fairly restricted physical properties, the clay-rich (argillaceous) rocks can provide a wide variety of geochemical, physical and hydrogeological environments which could be suitable for waste disposal. Unfortunately, the various properties which make this rock type suitable for disposal are often incompatible, and any clay unit is likely to represent a compromise of the potentially useful parameters. For example, the softer plastic clays have negligible permeability, respond to stress without fracturing, and act as good sorbing media for reached radionuclides. However, they occur only at relatively shallow depths, are often associated with more permeable rocks, have low thermal stability, and are difficult to excavate without complex mining and tunnel-support techniques. Conversely, the harder and more massive argillaceous rocks (generally considerably older and having undergone compaction, diagenesis and often low grade burial metamorphism) behave more as crystalline rocks in that they are fractured and less porous, with a lower clay mineral content and hence a lower sorption capacity.

Because argillaceous rocks are nearly always sedimentary in origin they frequently occur as stratified components of very large scale geological structures and individual beds of clay-rich rocks may extend for many tens of kilometres (F 6.4). Consequently, groundwater movements both within the clay formation and in over- and underlying rocks (sandstones, limestones, chalk, etc.) tend to be on a larger scale than in the case of crystalline rocks; in other words the potential path lengths of radionuclide migration may be much longer. A highly impermeable clay unit will only allow the passage of significant amounts of water, probably upwards or downwards into adjoining more permeable beds, if the hydraulic gradient across it is relatively high (e.g, artesian conditions). If the gradient is small the advective flow of water could in fact be very much lower than rates of diffusion. As a result, such a unit would not act as a flow conduit along its length. If a repository were situated in it, any leakage would tend to occur into the adjacent units, and it is these which would act as the migration pathway to the biosphere.

Clearly a wide spectrum of properties and flow environments exist, from plastic clays, through more consolidated but still plastic units; compacted mudstones with poorly developed fracturing; shales with strongly oriented fracture planes but remaining relatively soft and plastic; to brittle and highly fissured slates and phyllites which behave more as crystalline rocks. There is adequate scope within this category to tailor the disposal system to a particular environment, for example by reducing the thermal loading in the case of HLW. This style of approach is evident in the national research programmes. Belgium now has considerable experience in studying plastic clays for HLW disposal and has mastered most of the technical problems involved in working in these rocks while taking advantage of the undoubted benefits. Italy, the UK and the USA are putting effort into somewhat more consolidated units but, on the whole, clays are receiving much less attention at present that either crystalline rocks or salt, at least as far as deep disposal is concerned.

Evaporites

Evaporites are units of soluble salts of Na, Mg, K and Ca, formed in the geological past by evaporation of shallow lagoons and lakes in cyclic episodes. Considerable thicknesses of simple and complex chlorides, sulphates, carbonates, and so on may be laid down in layers with various degrees of mixing of salts, and subsequently buried as part of a stratified sedimentary sequence. As part of the process of burial they have often recrystallized to simpler and more stable salts. The commonest salt is halite, rock-salt (NaCl), although it is generally accompanied by many of the 80 or so other principal evaporitic minerals. Despite the frequent inhomogeneities, very thick units of almost pure halite (and several of the other salts) can be found (F 6.6). Because of its plastic nature halite is very susceptible to tectonic stresses deep in the earth and can be mobilized en masse due to its own density disequilibrium (being less dense than the surrounding rocks), rising in inverted tear-shaped blobs known as diapirs (F 6.7). Halokinesis, as this phenomenon is known, is not universal in regard to salt deposits; for example there are no diapiric (or salt-dome) structures below the UK mainland (although there are extensive simple bedded evaporate units) nor does it always result in the salt domes reaching the earth's surface. Several of the widespread continental European and US Gulf States salt domes show no evidence of contemporary movement, while others are rising very slowly, at the rate of a few millimetres a year. The time scales for such movement are so long that they present no particular problems from the waste-disposal viewpoint. Dome salts appear to be thermally more stable than their simple bedded counterparts.

Salt is an excellent conductor and dispenser of heat and is also, to all intents and purposes, impermeable to water, while containing only small amounts itself. The historic lead that salt has acquired in the HLW disposal field reflects the emphasis that was placed on an impermeable host rock in the earliest days of the research. Despite its obvious solubility in freshwater, salt is extremely stable in the geological environment, contains few if any fractures, is self-sealing owing to its simple plastic response to stress, and can be mined with ease. The consequent problems which surely accompany such an apparently ideal medium are: the frequent and often unpredictable occurrences of highly unstable mineral bands such as carnallite which can decompose at low temperatures to give out large quantities of water, the presence of fluid inclusions which can migrate towards a heat source carrying corrosive mixed brine with them, and the risk of flooding an operating repository, which is essentially a dry mine operating well below the water table present in overlying sedimentary units, some of which might be important aquifers. An experimental underground facility in a bedded salt was in operation as long ago as 1967 in the 'Salt Vault' project at Lyons in Kansas. Since then, and after a long pause in the work, the USA has committed much more effort to this rock type with the WIPP (Waste Isolation Pilot Plant) project in New Mexico, the Avery Island in situ heater experiments in the Gulf Coast (Louisiana), and characterization of a potential repository site in Deaf Smith County, Texas. At the same time Germany has put considerable emphasis on the use of dome salt for storage and possibly disposal of intermediate level wastes at the Asse mine, and is presently involved in the exploration and development of the salt dome beneath Gorleben for high- and intermediate-level waste disposal. With the phasing of the current commercial waste programme in the USA, the additional WIPP commitment, and the entry of both the Netherlands and Denmark into the international salt programme, it appears that this option might well be among the first to be pressed through to an operating conclusion.

Factors controlling migration

Radionuclides can leave the near-field environment by either advection (entrainment in moving water) or diffusion (dominant in very slow moving or static pore waters). They may be in solution in groundwater, in particulate form (as colloids and suspensions) or sorbed on to other suspended material. The speciation behaviour of many of the significant radionuclides, in particular the actinides, is extremely complex. The principal controlling factor is the nature of the geochemical environment, dominated by the groundwater chemistry. For the present however, we will ignore chemistry and concentrate on the physical properties of the groundwater environment.

Hydrogeology

The water table is the level in the ground below which water saturates the void space or porosity of a rock, which is made up of fissures, fractures and pores. Even above the water table the pores may be almost saturated with percolating water. In most temperate regions of the earth the water table is within a few metres or tens of metres of the surface. In the case of many argillaceous and crystalline rocks in the UK, where there is high rainfall and modest relief, the water table is to all intents and purposes coincident with the surface, particularly in flat lying areas. In all but exceptional cases in arid desert environments (such as the Nevada Test Site, NTS) we can say that at proposed repository depths the rock will be saturated with water. At NTS the water table is many hundreds of metres deep, and a repository might be situated in the unsaturated (but still wet) zone above it.

Hydraulic pressure

Groundwater moves in response to variations in 'head from one point to another. It can also be driven through the rock by the thermal effects of the waste which produce density differences (buoyancy) in the water. Under natural conditions head differences are most frequently produced by topographic effects-simple height difference (F 6.8), giving rise to hydraulic pressure gradients down which water moves. They can also be produced by the presence of zones or units of rock with different hydraulic conductivity (the ability to transmit water; broadly equivalent to electrical conductivity). The hydraulic head can vary considerably in different regions of a thick sequence of sediments, or within different fracture zones of a crystalline rock body, with little or no obvious relationship to the depth below the surface. This indicates that there may be very poor hydraulic connection between these different zones caused by intervening regions of low hydraulic conductivity (generally referred to as 'low permeability') rock. High pressures cannot easily be dissipated through such regions, as the water movement required to transfer energy is very slow. Zones of elevated pressure may result from long-distance connection via high-permeability rock formations to topographically higher areas. They can also be caused by differential compaction processes, or the presence of hydrocarbons in sediments. As a consequence, quite complex patterns of head can occur within a given volume of rock, resulting in flow patterns which might not easily be predicted from first principles. Artificial pressure differences can be caused by pumping an operating repository to keep it dry, thus locally reducing the ambient water pressure and creating a pressure gradient towards the pump, drawing water into the repository. Depending on the hydraulic properties of the rock, these pressure perturbations can persist for some time after pumping has stopped.

Hydraulic conductivity

The hydraulic conductivity of a rock is a function of the size and degree of interconnection of its pores and fractures. The smaller and more tortuous the structure of the void space, and the greater the surface effects in the pores, then the lower is the hydraulic conductivity. These parameters of hydraulic conductivity and head gradient are linked in the well known equation which defines the simplest form of Darcy's Law of flow through porous media (F 6.9):

$$Q = KIA$$

where Q = volumetric rate of flow (m^3/s), K = hydraulic conductivity (m/s), I = hydraulic head gradient (m/m), A = cross-sectional area through which flow occurs (m^2).

There is some controversy as to the degree of applicability of Darcy's Law to fractured rocks and other rocks with very low hydraulic conductivities, such as plastic clays. It is basically a means of representing flow through an isotropic porous medium like a sponge, but is widely used in the treatment of fractured rocks where there are clearly varying degrees of anisotropy in hydraulic properties caused by the very nature of the fracture patterns themselves. It is thought, however, that for fractured rocks Darcyan behaviour is a reasonable assumption if a large enough volume of rock is considered, and attempts are presently being made to test this approach against network flow models which more accurately represent anisotropic behaviour. The Darcy equation is very similar to the equation linking electrical current, resistance and voltage. In the form above it provides a volumetric flow rate, but no velocity of flow from one point to another down the hydraulic gradient. To obtain this simply for unit cross-sectional area (A), we divide Q by the 'effective' or 'kinematic' porosity of the rock, or that part of the total void space which takes part in flow. In a fractured rock this essentially means the fractures only, and not the intergranular pore space.

Early work on fractured rock hydrogeology tended to treat the rock as a whole, and either looked at homogenized hydraulic properties on a large scale or averaged them. It is now clear that the hydraulics of the very low permeability rock between major fractures (i.e. blocks of rock tens or hundreds of metres in size) is of importance only to near-field modelling. Far-field transport will occur in major fractures (those which occur perhaps every few hundred metres, may connect to the surface, and have high porosities and hydraulic conductivities), linked by smaller hydraulically active fractures to the bulk rock 'blocks'. Within any of these fractures preferential pathways exist for water movement (channelling), so they should not be considered as planar features (e.g. the parallel plate model of equal fissure aperture all across the plane), but rather as planes within which 'worm tubes' lie. This is because fissures have variable apertures owing to the rough nature of their surfaces and differences in the properties of their infill minerals. Models for fractured rock hydrogeology have moved from average porous medium equivalents, to single fracture dominated models connecting porous medium blocks, to probabilistic fracture network models, and now to random preferential paths. There is some way to go yet.

It is at this point that we can begin to see more clearly the difference between fractured rocks and unfractured 'porous' rocks such as clay-rich sediments. The latter often possess very high porosities, but if they lack fractures, may have no well-defined preferential flow routes. In this case all flow must be through the total porosity, although this may be anisotropic, owing to silty bands, for example. If we took a piece of unfractured sandstone, which has a very high porosity and relatively large and open pores, the hydraulic conductivity, or ability to transmit water, is quite high. However, a plastic clay, while also possessing a high porosity, has very small pores which are interconnected in an extremely tortuous manner. In many cases only a very small proportion (perhaps less than 10 %) of the water in these pores is actually free to move. The remainder is physically bound to the surface of the clay minerals. Thus while a plastic clay may have a total porosity of 30 %, its effective 'flow' porosity may be much less than 5 % (F 6.11). In addition the hydraulic conductivity is exceptionally low, and these types of rock are often considered to all intents and purposes as impermeable. While flow through an open-pored sandstone would be very close to simple Darcyan behaviour, the flow through plastic clays of very low permeability, as with low hydraulic conductivity fractured rocks, is rather more difficult to model. In the case of clay, flow under the hydrogeological circumstances suited to a repository site (low hydraulic gradients) is so slow that we begin to consider time scales and rates which are comparable to molecular diffusion processes. Thus, in clays, the initial part of the migration process takes place by diffusion, with possible subsequent advective transport in flowing groundwaters in overlying sediments. Table bellow lists some representative values of the parameters discussed above, and the extreme ranges of values can be seen immediately. It is clearly possible, within several of the rock types, to choose values of the various parameters which will give rise either to very slow or very rapid transit times of groundwater from one point to another.

Table 47 Likely maximum and minimum values for the hydraulic conductivity (HC), porosity (P), gradient (G), flux (F) and velocity (V) of various sediments and crystalline rocks, in typical environments which might be considered for disposal purposes.

Rock type	Depth - m	HC - nm/s	P	G	F - l/y/m^2	V - m/y
Clay	0-100	0.1,	0.3,0.5	0.05,0.2	1.6,640	0.0005,1.28
Clay	<100	1E-3,10	0.3,0.5	0.05,0.2	0.002,64	5E-6,0.1
Shale	0-100	1,1000	0.2,0.3	0.05,0.2	1.6,6400	0.008,21
Shale	<100	0.1,100	0.05,0.25	0.05,0.2	0.16,640	0.003,2.6
Crystallines	0-100	1,100	0.01,0.05	0.001,0.1	0.03,320	0.003,6.4
Crystallines	<100	0.01,10	0.001,0.01	0.001,0.1	0.0003,32	0.0003,3.2
Aquifer		10,1E+5	0.05,0.1	0.0005,0.01	1.6,32000	0.03,320

Transport models

We have thus defined a group of very simple hydraulic parameters which are needed for the radionuclide transport model. These can be used to quantify groundwater flow rate, and hence to a first approximation the maximum transport rate of the released radionuclides. They do not, however, define what is probably the most problematic feature, the length and direction of the potential flow area migration paths; the path and path length. In this respect there is a considerable difference between massive bodies of crystalline rock and stratified units of sedimentary rock. In both cases it is likely that large scale structural features, such as major faults, will act together with topographic features as boundaries to flow units or 'flow cells'. However, in the case of crystalline rocks current thinking centres upon the idea of very frequent flow boundaries giving rise to many small-scale localized flow cells within a body of rock, rather than the extensive regional flow patterns found in stratified formations. In all cases the shortest pathlength to the biosphere is directly upwards from the repository to the surface. This is an extremely improbable escape route for the waste as few hydraulic conditions can be envisaged which would give rise to unmodified vertical flow. The possibility exists for some quite short pathlengths in certain geological environments. The flowlines depicted (by two dimensional finite element modelling) are in response to typical predicted hydraulic gradients. In a formation which displays decreasing hydraulic conductivity with depth, the greater the depth to which a flowline penetrates, then the smaller is the volume of water moving along it and the slower the velocity. In other words, a near-surface flowline might represent large volume transport over perhaps a few hundred years, whereas a deep line would depict a very small volume of water taking perhaps tens or hundreds of thousands of years to re-emerge.

For stratified sequences, the pathlengths can be considerable, and time scales may be comparably long. In a stratified sequence, leakage of the contaminated groundwater may take place out of the repository host formation, and subsequent transport may be in an entirely different underlying or overlying rock type with different hydraulic properties. Because of the variability in hydraulic properties of sedimentary sequences, anomalous pressure differences and flow patterns can develop which may drive water up or down through the host-rock unit. In some environments, locations can be chosen where any groundwater flow must take place in a unit which will conduct contaminated water under the sea before discharge to the biosphere, hence considerably diluting the release. Such environments can be found in both sedimentary and crystalline rocks. The logical extension of this idea is to select a small island as a repository site, where all discharge must of necessity be to the sea. Some island environments have a unique hydrogeological feature known as the 'Herzberg lens', which is the phenomenon of freshwater in the rock pores 'floating' on top of denser saline groundwater at depth. The depth to the saline/freshwater interface is controlled by the thickness of the freshwater lens (reflecting the island's topography and infiltration rate/rock permeability) and mass exchange across it is limited. It is thought that groundwater movement just below the interface (in the saline waters) is virtually non-existent and would thus prove a potentially ideal site for a repository. Sites on the coast of the mainland possess some of the advantages of an island and are also well worth considering.

The fundamental hydrogeological properties of permeable rocks required for simple transport modelling are thus: hydraulic conductivity; kinematic (flow) porosity; regional and local hydraulic gradients; geological features controlling flowpaths.

These factors may vary quite considerably even within a given repository site. When performance modelling,is carried out to assess a potential deep-disposal site a much wider area of ground than the locality of the repository itself must be considered. This is especially true of stratified units where data on these properties must be available along what might be quite considerable potential pathways. Some idea of the variability of parameter values within any given body of rock was given in Table above. In crystalline rocks the hydraulic conductivity is thought to decrease markedly with depth. In regions which have been glaciated and subjected to permafrost to considerable depths, the effects of loading and unloading stresses caused by the presence of hundreds or thousands of metres of ice overburden, have caused fracture enhancement and consequent increased permeability in the upper few hundred metres. This short list only defines the Darcian parameters in their simplest form. To obtain a true understanding of anisotropic flow behaviour the equations become more complex and require further parameters to be determined. Some of these parameters are essential derivatives of field experimental techniques which endeavour to determine values of anisotropic hydraulic conductivity, storativity, dispersivity, transmissivity and other factors.

Physical dispersion

As water flows through a rock, any chemical component dissolved in it might be expected to travel at the same speed as the water. If we take a single tube-like pathway of 'clean' water and inject it at some point with a short pulse of a tracer chemical, then we would expect to see that pulse passing a downstream monitoring point as a spike on a concentration vs time graph. The width of the spike would correspond to the time taken to inject the pulse initially. If the water movement is very slow, however, diffusion of the component into the surrounding water behind and in front of the pulse will become noticeable, the width of the spike will increase, and its height (maximum concentration) will decrease a little. Different types of flow within the pathway (for example turbulent or laminar flow) and frictional edge effects will also give a similar result. These may be quite marked over the extensive pathlengths described in the previous section, at the slow rates anticipated. They will, however, be relatively small when compared with the effect of physical dispersion. This takes place during flow through a network of fissures, or a complex interconnected pore structure, which provide numerous alternative paths between two points, all of different degrees of tortuosity, and hence of length. From first principles, the arrival of the tracer should be at the same time as in the single channel case, as this still represents the shortest route, but now the spike will tail out over a long period of time due to the 'late arrivals' from the longer paths. This phenomenon, known as dispersion, is clearly very dependent on the fracture pattern or pore structure, and the degree of isotropy of hydraulic parameters within the volume of rock being considered. Its net effect is to retard the arrival of the peak of a 'pulse' of radionuclides released into groundwater, and to decrease considerably the concentration in which they arrive at the outflow or measuring point. Dispersion results from mechanical (caused by flow) and physicochemical (molecular diffusion) mechanisms, due to properties of both the fluid and the material in which the flow is occurring. In general the 'dispersion coefficient' is a symmetric tensor quantity having three independent parameters in the two-dimensional case and six parameters in the three-dimensional case. However, if the material is isotropic and the dispersion is assumed to be proportional to the flow velocity then the mechanical component of the dispersion coefficient can be written in terms of just two parameters, the longitudinal and transverse dispersivities. The components of the dispersion tensor can be written in terms of the dispersivities, the components of fluid velocity, and the molecular diffusion. The properties of some rocks, such as those in which flow occurs in a few major fractures, result in low values of dispersion so that tracer concentrations remain high over large distances. Others, such as poorly consolidated sandstones, have higher dispersivities and tracers become quickly diluted.

Attempts have been made to model combined flow and dispersion in fractured rocks using a statistical approach to the fracture network. This technique constructs a probabilistic model of fracture pattern and interconnections using as a basis real statistical field data, and then uses percolation theory to calculate water movement through the model network. This approach is likely to be the most feasible way forward since a full in situ determination of a fracture system is not likely to be possible (or tractable) in practice. In principle this is a significant step towards bridging the gap between purely porous medium flow models and simple-fracture flow models. At present, results indicate that the former type of model is valid if the volumetric scale is large compared to the fracture intervals. Hydrodynamic dispersion is complemented by chemical retardation and in fractured rocks by a further phenomenon called 'dead-end pore diffusion' or matrix diffusion.

Diffusive retardation

In the discussion of the hydraulic properties of rocks, two styles of porosity were described. The effective porosity is what we have been dealing with until now, that is the active porosity which takes part in advective flow. In a fractured rock however, the effective porosity might amount to only 0.1 % of the rock by volume, whereas the total porosity may be up to 2 % or more. In this case only 5 % of the total void space in the rock is 'flowing', the remaining 95 % is filled with water which to all intents is static (F 6.17). The water flowing along a fissure system is in contact with the large reservoir of dead water, and any components in the moving water (e.g. dissolved radionuclides, colloids) will be free to diffuse down concentration gradients into the main body of unfractured rock, subject only to constraints set by the pore dimensions. As they diffuse out into the dead pore space, the situation is analogous to a man stepping off a jungle track into the trees. He will easily get lost in the complex undergrowth and may remain there forever. In the case of a chemical diffusing into static water, there is no mechanism other than diffusion to return components to the flowing stream, and this can only take place when the chemical gradient is reversed and concentration in the rock pore-waters exceeds that in the fissure water (i.e. when the main pulse has passed).

There has been much effort in the study of this phenomenon, and initial calculations indicate that this could be an extremely effective mechanism of retardation by physical means alone. If we also give credit to chemical reaction of the nuclides with pore walls in the dead space, this retardation mechanism is potentially so efficient that most if not all, radionuclides decay to insignificant levels before they are released to the biosphere. As yet, the existence of long-distance networks of connected pores extending from fracture zones throughout the entire matrix has not been proven for relevant rock types. The potential importance of the mechanism is such, however, that it is under extensive investigation in laboratory, field and natural analogue studies.

In an unfractured rock this particular retardation mechanism may not occur unless similar preferential flow paths exist. In stead diffusion takes place throughout the pore structure of the rock. However, since an 'effective porosity' can also exist in clays, the situation is complicated if any noticeable flow does occur (under high hydraulic gradients for example). Since the non-mobile water is present in the same pore space as the mobile water, chemical exchange by diffusion can take place between them. Owing to the high surface charge on clays however, anions may be excluded from the non-mobile water. An additional, poorly understood, process, known as surface diffusion may also be operative, whereby cations migrate more quickly in the non-mobile water layers at highly charged mineral surfaces. This rather complicated system has not been fully assessed, and for most modelling purposes it is usually assumed that free diffusion can occur throughout the whole available porosity. Any components of advection (flow) are superimposed on this, although under the majority of circumstances the rate is much slower than for diffusion. The dispersion and diffusion effects can be experimentally quantified.

Chemical interaction of radionuclides with the rock

As radionuclides migrate from the immediate environment of the waste they are affected by interaction, not only with the corroded package and buffer material, but also with the massive body of rock through which they must subsequently pass, and which acts as the principal chemical buffer to all solid-fluid interactions (F 6.18). The rock buffers the composition of the groundwater and limits its rate of supply and thus controls the way in which the nearfield performs. Radionuclides diffusing out of the buffer into the host rock will either be faced with a continuous static body of pore water (e.g. in a clay), in which case diffusion continues out into the far-field, or they will be entrained in slowly moving groundwaters in a pore or fissure system (e.g. in a crystalline rock). In both cases a contamination 'front' will develop around individual packages and slowly increase in volume, being shaped by flow or diffusion anisotropy, and by interaction with similar fronts from adjacent leaking packages. Rather complex concentration patterns might occur over very long time periods within the volume of a repository. In some scenarios, we might consider the repository as a whole being a source for subsequent far-field migration, although there is little doubt that the majority of waste components will be fixed or will decay to negligible levels within the actual repository volume, or even in the immediate neighbourhood of their source package.

Precipitation

As radionuclides pass from the near-field engineered barriers to the far-field, the progressive change in water chemistry will cause the chemical form of some elements in solution (speciation) to alter, which may be reflected by changes in solubility or extent of retardation. If a very sharp boundary exists between different chemical conditions in these regions, extensive precipitation of radionuclides associated with secondary mineral formation may occur at this interface. Such precipitation could act as a source of colloidal material but it may also block pores and thus decrease the permeability of some low porosity rocks. The thermodynamic modelling approach used to evaluate solubility in the near-field is also applied to the far-field with the slight advantage in the latter case that the chemistry is usually better defined. As the migration path of the radionuclides traverses different rock environments, groundwater chemistry will slowly evolve with consequent changes in speciation. At present levels of sophistication, this could only be modelled as a stepwise transition and only the most important groundwater characteristics (redox conditions, pH, temperature, concentrations of major ions) could be taken into account.

Sorption

The most important processes of radionuclide/rock interaction in the far-field are generally grouped together under the loose heading of 'sorption', although few of them would be included in a physical chemist's understanding of this term. The mechanisms involved are purely physical processes which retard migration such as molecular filtration, ion exclusion and diffusion into dead-end pores, direct chemical reaction with rock surfaces involving physical adsorption, chemical adsorption or direct incorporation (mineralization) into the rock structure and indirect chemical reactions e.g. precipitation caused by enhanced concentrations at the rock surface. In principle, most of these processes are applicable to suspended or colloidal material in addition to that in true solution. The inclusion of the category of physical processes may seem incongruous here, especially as they have been considered in the previous non-chemical section. The magnitude of these processes can, however, be influenced very greatly by radionuclide speciation (especially if formation of very large or charged complexes is possible) and, in practice, they are often impossible to quantify experimentally in isolation from chemical sorption. Sorption is usually described by a simple partition constant (Kd) which is the ratio of rock phase radionuclide concentration to that in solution in the groundwater. Use of this simple approach in migration modelling requires the assumption that sorption is reversible, reaches equilibrium quickly, is independent of variations in water chemistry or mineralogy in particular regions of the flow path, and is not affected by changes in rock water ratio, or in the concentration of the species involved or other solution components which are being sorbed as they migrate. Such assumptions rarely hold in reality but, in some cases it can be shown that this approach overestimates the extent of migration (i.e. is conservative) with appropriate choice of Kd values. More sophisticated migration models separate out several different retardation mechanisms, and may also include the concentration dependence of chemical sorption by using empirically derived functions, often called isotherms. The complexities of the resulting calculations (and lack of data) have, as yet, limited the application of such models.

Present safety assessment models generally use a simple empirical Kd approach and, on the basis of the discussion above, this would seem to be justifiable. Problems arise, however, in choosing suitable Kd values and demonstrating that their use is actually conservative. Although measurements of a Kd value seems on the surface to be quite straightforward, the large number of mechanisms involved can lead to many practical difficulties, as will be seen later. As a result, data for relevant rock/water environments are often very limited or poorly defined and, for some important elements, non-existent.

Surface chemistry

Finally, it should be noted that the nuclear waste literature is full of examples of misuse of sorption data and terminology, particularly of the Kd concept. For example, it is evident that sorption can only occur at any point where rock and water come into contact; that is on the surfaces of fractures and on pore-walls. In many types of fractured rock, the mineralogy of such exposed surfaces is considerably different from that of the bulk of the rock itself. This is because the fissures have always been able to act as conduits for the passage of reacting fluids, and in the geological history of a particular body of rock such fluids may have been hot or concentrated mineralizing liquids. The surface chemistry encountered by the radionuclides may thus be totally different from the bulk rock mineralogy. For example the fissure coatings in granite are often comprised of chlorite, montmorillonite, iron oxides, sulphides and so on, none of which is present in quantity in the primary granite mineralogy. Despite this, many older migration models (and some relatively recent ones) use sorption data (Kd's) measured on crushed host rock, which are obviously inappropriate. The speciation, and hence sorption, of many important radionuclides is also very greatly altered by changing redox conditions. Nevertheless measurements under oxidizing conditions have been used to predict migration in reducing groundwaters.

For the moment, however, it is sufficient to appreciate that on a simple qualitative scale, some elements are sorbed in certain rock water systems much more strongly than others. Plutonium is usually strongly sorbed, and in most assessments of migration behaviour shows up as being of less radiological significance in eventual releases than radionuclides such as 99-Tc or 239-Np, which are less well sorbed by most minerals. While there may be some doubt about our present ability to measure fully realistic sorption values, a qualitative understanding of the behaviour of radionuclide-rock systems is emerging. In some cases, specific groundwater systems at potential repository sites are well characterized, and the sorption behaviour of radionuclides under realistic conditions is known. The present generation of computer codes can readily handle quite sophisticated treatments of radionuclide retardation during groundwater transport. The main reasons for the relative simplicity of models used in recent safety assessments are the lack of appropriate laboratory data, and the very limited validation of even the simple submodels by in situ experiments and natural analogues.

The complete 'normal-case' release model

After mobilization of the waste as a leachate in the near-field, we have followed its progress through the geological barrier in a flowing or diffusing groundwater system. Transport takes place either by diffusion or in response to flow down pressure gradients, and may subsequently lead to release at the surface. During the course of this migration the radionuclides in solution are subject to dispersion and diffusion, and are reversibly sorbed or 'irreversibly fixed' on to the exposed surfaces of the rock. The net effect of these processes is to 'smear out' the concentration-time plot of arrivals at the surface to such an extent that the consequent radiological impact is limited. None of these processes of fixing and retardation described is a truly permanent means of isolating the waste from the biosphere, although radioactive decay means that increasing retardation causes increased destruction of radionuclides within the geosphere, especially the shorter-lived species. All of these phenomena contribute to the eventual dilution and dispersal of the long-lived components of the waste. These longer lived radionuclides will remain in the repository as a concentrated potential hazard until either this natural dilution has taken place or they have decayed to relative inertness. The far-field barrier with which we have been concerned so far in this chapter has been treated as either a sequence of sediments obeying normal porous medium flow laws, or as a massive fractured body of fairly homogenous rock. In either case the last part of the escape route back to the biosphere may take place through relatively unconsolidated superficial deposits lying on top of the host rocks, and varying very markedly from them in their properties. Weathering products, soils, glacial debris, alluvium and the many other varieties of surface cover can often be several tens of metres thick and show relatively rapid groundwater transport compared with the underlying rocks. Water seeping upwards into a thick sequence of sands and gravels is unlikely to take more than a few tens or hundreds of years to move into surface water bodies, and will mix to some extent with meteoric waters (infiltrating rainfall). Nevertheless, the importance of this section of the migration route must not be underestimated. Superficial deposits are indeed already used as a host medium for shallow burial of lower activity, short-lived radioactive wastes. In the context of deep disposal however, the dilution, dispersion and radionuclide interactions with unconsolidated soils, etc., which occur within this zone, are normally taken into account within the biosphere models.

Migration from a repository in evaporites

Throughout this treatment of migration in the far-field we have concentrated on the way water or mobilized radionuclides move through permeable rocks which obey the Darcy Law of flow, or which allow diffusive movement in an extensive interconnected porosity. The exception to this treatment is the case of evaporate host formations, such as salt domes, which are both impermeable to water and contain very limited free water in pores. In a well chosen unit of thermally stable evaporate minerals with few hydrous phases the only free water would be in the form of an intergranular film, and predominantly as brine inclusions trapped in individual crystals and not interconnected. While these inclusions can move up a temperature gradient, they do not in themselves constitute a radionuclide transport mechanism. In some situations the accumulation of brine around a container of waste may contribute to corrosion, but no flow mechanism is available to transport released radionuclides. Intergranular diffusion in the salt will be exceptionally slow and negligible movement into the far-field is anticipated. During site selection areas of rapid uplift (diapirism) would be excluded and extensive dissolution of the salt formation can generally be discounted on relevant time scales. Consequently the only natural release scenario is by some form of catastrophic flooding of an operating repository caused by ingress of groundwaters through faults, solution pipes, or access shafts from surrounding rock strata. In such a case the integrity of the salt as a far-field barrier is partially lost and we must consider possible transport through the under or overlying rock, where the processes are identical to those already described. The emphasis here is on 'operating repository', since once the disposal galleries and shafts have been sealed there is no means of access available to waters from overlying aquifer units. Thus, as far as thick evaporate sequences are concerned, provided the seal system is adequate, no sensible migration pathways can be identified which could lead to releases into surrounding groundwaters (with the exception of possible human intrusion into the repository).

Geological evolution of the far-field barrier

To complete this discussion or the far-field geological barrier and radionuclide migration from a deep repository, consideration must be given to the likely effects of climatic and geological change on the rock barrier. It has always been considered important to try to locate sites for repositories such that changes in sea-level, or the geological traumas of an ice age have minimum effect on the performance of the far-field barrier. How significant are evolutionary processes in a safety analysis? Fundamentally only two scenarios appear to be of any importance to deep disposal. The first is the case of the repository being exhumed and exposed at surface. The second is a radical change in groundwater flow patterns such that throughflow rates are considerably increased and the waste returns to the biosphere more quickly, or with less dilution. Erosion is the only credible mechanism by which the repository could be exhumed on a relevant time scale, short of massive tectonic disturbance, which is itself predictable over very much longer periods (in the order of 1E+7 years). Such erosion might accompany very marked changes in climate, leading to very hot or very cold conditions. In either case, the second scenario, that of perturbation of the flow conditions, would occur long before the waste was exposed at surface. The other principal cause of a changed hydrogeological environment would be the occurrence of sustained seismic activity in the region. The reactivation of capable faults (those still potentially subject to movement) may perturb flow patterns but is an unlikely mechanism for causing massive and large scale volumetric flows. The prediction of the effects of various geological evolution scenarios is based on observations of the effects of similar processes in the past. In the case of glacial effects this is made more straightforward by the fact that we can observe terrain which is presently, or has recently (within the last 10-20,000 years), been glaciated. Such terrain can be in close proximity to, or in very similar geological environments to, areas proposed for disposal purposes. This is especially the case in northern Europe. In fact all of the geological processes likely to be disruptive to a deep repository are active to varying degrees somewhere in the world, and much can be gained from direct study of volcanism, changes in sea and surface levels, major climatic processes.

The maxim of geology has always been that the present is the key to the past, and that to understand the structure of ancient rock formations we must observe how sediments, volcanic debris, and so on, are deposited today. This can be pushed in the other, albeit unfamiliar, direction of predicting the effects of these processes; what in France are known as 'geoprospective' studies. Very little considered work has been carried out in this area, although there are clear overlaps with probabilistic models. Perhaps the main problem lies in the need to be site specific about such predictions, and the parallel need for a substantial database, not only for the site and its surrounding region but also for other occurrences of the phenomena under study. A study in France is endeavouring to assemble a database for such exercises, and to develop a methodology for applying predictive geology to specific sites. One of the most convincing attempts to predict future geological changes was that of Vandenberghe et al. (1980), who concentrated on the potential for erosion at the Mol nuclear research site in Belgium as a result of glacial processes which might occur over the next 200,000 years. This study used data on the known depths of erosion in similar sediments in the area as a result of past glacial episodes. This, combined with data on possible rates of subsidence caused by ice loading, isostatic rebound, sea level changes and so on, produced figures on maximum depths of erosion above the proposed repository site. A further interesting 'case history' concerns the potential uplift of diapiric evaporate structures such as salt domes. Since these structures are in density disequilibrium with their surrounding formations they have a tendency to rise, and as can be seen in parts of the Middle East, to puncture the earth's surface, giving rise to salt hills and salt 'flows'. Under some circumstances, and over very long periods of time, this could clearly be detrimental to any repository contained within them.

Biosphere modelling

In previous chapters we have looked at the mobilization of radionuclides and their transport from the near-field to the biosphere. The final link of the safety assessment chain is evaluation of the consequences of any releases which are predicted, in terms of doses to the human population. This involves evaluation of processes occurring in the surface environment, for which a fairly comprehensive methodology of biosphere modelling has been developed to allow a logical approach to the problem, which is applicable to all waste types and repository systems.

Radionuclides reach the earth's surface in springs and other groundwater discharges, and are then redistributed by surface-water movements. They may finally reach man either directly, through drinking-water, or indirectly, via the food chain. Biosphere models attempt to estimate radiation doses by evaluating dilution in surface and groundwater bodies, and possible radionuclide concentration in certain biosphere 'compartments'. The first stage of the preparation of a model involves evaluation of the area in which releases are predicted to occur, in terms of groundwater and surface water classifications: springs, streams, rivers, lakes, seas and subsurface flows. For individual geographical regions a set of compartments can be defined which shows the progression of contaminants between different water bodies, along with associated dilution by uncontaminated groundwaters and meteoric waters. Flow between different regions, evaporation, transpiration and loss of waters with insignificant radionuclide concentrations (largely via rivers or seas) allows an overall water balance to be calculated. The area of discharge of contaminated groundwater is derived from the far-field transport model, which also yields radionuclide output as a function of time.

It is generally assumed that radionuclides are instantaneously and homogeneously distributed through each compartment of the model. This means that compartment sizes have to be chosen so that this is true over the time periods involved. Partitioning of radionuclides between water and solid phases (e.g. soils) is also taken into account, usually by use of a simple partition coefficient. It may be noted here that while underestimation of Kd values is conservative for the near- and far-field transport calculations, overestimations are generally conservative in the biosphere model, as they provide increased reconcentration within a particular compartment. The radionuclide transport rate from a compartment is proportional to its concentration in that compartment and a factor called the transfer coefficient.

Concentration ratios

In a second modelling stage, a food chain is constructed for each compartment (F 10.2). For example, a particular water compartment serves as a source of drinking-water, or crops and vegetables are planted on a particular part of a soil compartment. When a full set of such chains has been constructed, the change in radionuclide concentration along them is calculated by the use of concentration ratios (CR) between various links (e.g. water to grass, grass to cow, cow to milk or beef, milk or beef to man). The calculation uses either average values for consumption of various food types, based on present lifestyles, or maximum consumption values by particular critical groups, depending on the objective of the analysis.

Experimental determination of CR values can be quite complicated. In the last example, transfer from water to grass may be dependent on the season, and radionuclide transfer from grass to cow may vary with the type of grass, and also on whether the grass is cut and stored (as hay or silage) before being eaten. Uptake by man via various meat and dairy proucts will vary with processing and cooking. In addition, such transfer factors, like Kd values, are very dependent on the chemical speciation of the radionuclide involved. The difficulties involved in evaluation of speciation can be even more pronounced in the near-surface realm, owing to enhanced biological activity, high concentrations of a wide variety of organic compounds and marked pH and redox gradients in soils and surface waters. For this reason, and because environmental conditions vary from site to site, it is not surprising that the reported range of variation of concentration ratios from soils to plants is large: from two to six orders of magnitude for some elements.

Finally, doses are calculated from the activity concentrations in water and in the individual foodstuffs, the food quantities consumed, and dose conversion factors. These factors are calculated using models of human metabolism, and take into account total body mass, and the mass of each organ, radionuclide partitioning between different body organs, radionuclide retention in the body ('biological half-life') and differing radiosensitivity of specific body organs.

In calculating these factors, account can be taken of variation of dose per unit intake with the age of the individual, and with the chemical form of the radionuclide. The doses calculated are those received over the lifetime of an individual, following one year's intake of a radionuclide, because radiological protection standards are based on the concept of limiting the risk to an individual over the rest of his or her lifetime from a year of exposure to radionuclides in the environment.

In addition to the ingestion pathway outlined above, in certain cases other exposure routes must be taken into account, such as inhalation or external irradiation. Calculations involving radioactive gases, aerosols or suspened particulate material use similar compartment specifications, and intakes derived from standard inhalation rates. Resultant dose is again calculated using specific dose conversion factors. External irradiation from contaminated soil or from swimming in contaminated water can be calculated by standard techniques.

Although the procedure involved in calculating doses from radionuclide release estimates seems complicated, computer codes exist which greatly simplify the procedure. From a database of soil partition coefficients, concentration factors and dose conversion factors, the net doses can be calculated readily for any release scenario. Apart from the 'base case', which evaluates present conditions, alternative scenarios are often assessed. Changes in patterns of water use can be of particular significance. The drilling of wells might short-circuit some of the near-surface transport pathway and decrease the extent of dilution, while construction of dams or reservoirs could greatly increase dilution. A range of such scenarios is generally considered in a comprehensive safety assessment, and this may even go so far as to consider the effects of longterm climatic variations, for example over glacial epochs, ranging through tundra-boreal-icecap conditions. This would alter both the groundwater system and the relevant food chains. Such calculations must obviously be very speculative. If one considers the difficulty which someone in the Middle Ages would have had predicting present populations and lifestyles, it is apparent that detailed extrapolations over periods up to a thousand times longer are, at best, exercises in educated guesswork, but nonetheless necessary.

Radiological safety assessments (Chapman 1987)

As a preliminary, we shall begin by defining exactly what is meant by the term radiological assessment. Within radiological protection and radioactive waste management circles 'assessment' has become a jargon word: it is used to mean both the estimation of the radiological risks associated with a particular practice (e.g. waste disposal) and the comparison of these risks with standards or criteria. Thus radiological assessment is a two stage process consisting of mathematical modelling studies which provide estimates of risks, and use of these estimates to draw conclusions about the acceptability of the practices from a radiological protection point of view. In this chapter 'assessment' will be used in this sense, while 'analysis' will be used to refer to risk estimation alone. Radiological assessments differ from the environmental impact assessments which are required by legislation in the US, in European Community countries and elsewhere. Assessments of the total impact of waste disposal on the environment needs to include considerations of aspects other than those related to radiation: for example, noise levels during operation of a repository; the presence and effects of non-radioactive pollutants in waste; the effects of repository construction on subsequent use of the site and the land surrounding it. In contrast, radiological assessments deal only with risks resulting from the radioactive nature of wastes, and are thus only one, albeit important, part of an environmental impact assessment.

Purposes, objectives and scope of assessments

A full radiological assessment of any disposal method for radioactive wastes needs to consider three phases of the existence of the waste repository: the operational phase', when wastes are being emplaced; a possible 'post-operational, institutional management phase', when the repository has been closed but the site is still monitored and maintained; and the 'post-operational, post-institutional management phase', when the site has been abandoned. Assessments for the first two of these phases need to consider potential radiation doses and risks, both to the workers at the site and to the general public (i.e. everyone else). For the final phase, only the public needs to be considered and the assessment needs to span much longer time scales. It is the risks during this phase, when there are no active control measures, which cause most concern and thus most of the assessments which have been performed so far have focused on the post-operational, post-institutional management period. A substantial progress has been made towards internationally agreed long-term radiological protection criteria for disposal of solid radioactive wastes. The consenus is that the objective of disposal should be to ensure that no individual, either now or in the future, should be subject to a risk from waste disposal which is greater than other risks which are widely accepted today. Furthermore, the risks to both present and future individuals and populations should be reduced to 'as low as is reasonably achievable (ALARA)', social and economic factors being taken into account.

Basic features

These forms of criteria, together with the worries which are often expressed about radioactive wastes, lead to the idea that radiological assessments of waste-disposal options should be as comprehensive as possible, in order to provide the maximum amount of information on which to base disposal decisions. Ideally, a radiological assessment of waste disposal should:

(a) consider all the events and processes which could lead to release of radionuclides from the repository, or which could influence the rates of release and the subsequent risks;

(b) produce estimates of the risks to ecosystems, individuals and populations, as a function of time and space, and information about the various components of these risks (e.g. whether they are due to high probability/low consequence events or low probability/high consequence events, whether many or only a few people could be affected);

(c) indicate the uncertainties in the risk estimates, and identify which of the parameters and assumptions have most influence on the results;

As will become apparent from the examples given later in this chapter, none of the assessments of geological disposal which have been published so far have been fully comprehensive. The reasons for this are twofold: first, research into disposal of high-level wastes is not sufficiently far advanced to allow full assessments to be attempted; second, many of the assessments which have been made have been preliminary and designed to indicate research priorities or to establish whether it is worthwhile to continue to study particular disposal methods, rather than to provide a basis on which to take final decisions about waste disposal. It must also be emphasized that there are situations in which it is not worthwhile devoting a great deal of time, effort and money assessing disposal risks (e.g. of very low level wastes). There are cases when it can be shown through simple calculations, using pessimistic assumptions, that the radiological impact of disposal will be totally negligible.

The starting point for any assessment of the long-term radiological impact of waste disposal is to list all the events and processes (natural, human-induced and waste-related) which could initiate release of radionuclides from the repository and transport them through the environment to humans, or could influence release and transport rates. These lists are generally established simply by asking people who are concerned about waste disposal to use their imagination. Once this list is available, the next step is to decide which of the events and processes are to be included in the risk analysis and how they are to be taken into account. These decisions depend on the purpose of the analysis, the resources of those carrying it out, and the amount of information available about the prospective disposal site and the behaviour of the wastes once they have been disposed of.

Categories of events considered

In preliminary radiological assessments of geological disposal, the procedure generally followed is to select a few events and processes, and to combine these into a small number of 'scenarios' for risk analysis. In some of these assessments, the probability of occurrence of these scenarios is estimated, while in others the consequences of each scenario are calculated but no attempt is made to estimate the probabilities that these consequences will occur. For a comprehensive assessment the procedure would be as follows. First, the events and processes are grouped into those which :

(a) are certain to occur (i.e. are 'normal');

(b) might occur and which, if they did, would perturb conditions in and around the repository but would not lead to a radionuclide release and transport scenario which is completely different from the 'normal' one;

(c) might occur but which, if they did, would lead to very different radionuclide release and transport scenarios, but would not lead to large, direct releases of radionuclides into the human environment;

(d) are catastrophic events which could give rise to large direct releases with high consequences but which generally have very low probabilities of occurrence.

The combination of events in group (a) is used to define the 'normal scenario'. In some geological formations (e.g. salt) the consequences of this scenario will be essentially zero, that is, the most probable outcome of disposal is that there will be no risks to humans or the environment. Events in group (b) can be dealt with in two ways. Either they can be included in the analysis of the uncertainties associated with prediction of the consequences of the normal scenario, or they can be combined into a series of what are sometimes called 'altered evolution' scenarios. In either case, their consequences are calculated using the same mathematical models as are used for the normal scenario but the probabilities of occurrence of these events and processes must be taken into account. Group (c) events are usually dealt with by a conventional probability-consequence approach. That is, appropriate mathematical models are developed to calculate their consequences, and then these are combined with their probabilities of occurrence to give an estimate of overall risks. Group (d) events can also be dealt with by the same means as group (c). However, group (d) events are often given separate consideration, partly because their probabilities of occurrence are so low and partly because their non-radiological consequences may well be greater than their radiological ones. (Examples of events in this class are the impact of very large meteorites and the occurrence of extensive tectonic activity, such as the formation of volcanoes.) Because such events are rarely taken into account when taking other decisions, for example whether to build cities, bridges or chemical plants, there is a growing feeling that they can legitimately be excluded from radiological assessments of geological disposal of radioactive wastes, although some of them will need to be considered when selecting prospective disposal sites.

The grouping of events and processes into categories leads to a qualitative definition of all the radionuclide release and transport scenarios which are to be included in a risk analysis. The second step in this analysis is to provide a quantitative description of each scenario, assigning numerical values to all the parameters to be used in the mathematical models and to the probabilities of occurrence of the various phenomena. Even when the available database is extensive, this parameter assignment exercise is not an automatic, completely objective procedure. In every case it is necessary to make judgements both about the 'best estimate' values of particular parameters and about their ranges or statistical distributions. For this reason it is important to identify the crucial parameters and assumptions in the analysis, so that the effects of differing judgements can be investigated.

Examples of the results of assessments

At present there have been no fully site-specific assessments carried out which endeavour to treat all mechanisms of mobilization, transport, release and uptake realistically. All available assessments either include over simplifications (intended to be on the conservative side) or have large generic components (such as assuming that all wastes, of all types, from a nuclear power programme are contained in one repository). In this section we examine in detail two examples of the most advanced assessments to date. These are mainly concerned with deep disposal of long-lived wastes, principally HLW. Before doing this, however, it is useful to look back at some of the earlier, and somewhat simpler, work which laid the foundations for these projects. The generic studies have contributed greatly to developing procedures for modelling repository behaviour. They contain a selection of data which is very conservative and, to some extent, arbitrary. Such generic analyses have been used primarily in the development of methodology and in defining criteria for the first stage of site selection and repository design. Results of these studies could thus be used to give some indication of the relative significance of various components of the model chain (for example, indicating the relative importance of the various near-field barriers), and to illustrate the dependence of performance on site location. The numerical values derived from such studies must be treated with great caution as they are not intended to predict specific doses. The simple models also took no account of several factors which were subsequently found to be highly significant, such as solubility restrictions and realistic redox conditions for the definition of appropriate sorption data. The models are extremely sensitive to the choice of parameters and their values (F 10.3). It was shown that with somewhat more realistic data for the flow path, flow rate and dilution, the doses calculated for I and Tc are decreased by a factor of 500 and, for a period of more than 1E+7 years, no significant dose would result from any radionuclide. All studies predict releases significantly below natural background levels, it may be noted that the analyses tend to fall into two groups: those with maximum doses much greater than 1E-6 mSv/year, and those with doses much less than this value. This can be interpreted, to a large extent, in terms of the degree of conservatism of the approach; very conservative, using highly pessimistic values for all uncertain parameters, or using realistic, though still conservative, values for parameters. The differences in these approaches are illustrated in the case studies below.

Sweden the KBS-3 assessment

In 1977 the Swedish government passed a law stipulating that no further nuclear reactors could be built or fuelled until it had been shown that high-level waste could be managed and disposed of safely. Since then, three major assessments of disposal have been performed by the KBS organization, which is financed by the Swedish utilities (1977; 1978; 1983). One of these deals with vitrified high level waste, and two with direct disposal of spent nuclear fuel. The first and third of these assessments have been subject to extensive international review, and have been used by the Swedish authorities to decide that a limited number of new reactors can be brought into operation. Since KBS-3 is the most recent of these studies, and the one which has the benefit of the most field and laboratory research, we will use this as an example (F 10.4).

Despite recent advances in assessment methods, the approach used in KBS-3 is very similar to that utilized in earlier studies, such as those of the NRPB referred to above. Although most of the mechanisms by which radionuclides might be released from a repository and transported through the environment are discussed in the report, calculations of potential doses to individuals and populations are carried out only for a small number of scenarios involving radionuclide movement with groundwater. No attempt is made to estimate the probabilities of occurrence of any of these scenarios or to utilize possible ranges or distributions of the parameters used in the models in order to quantify uncertainties in predicted doses. Also, in common with most other assessments, it is assumed that conditions in the biosphere remain unchanged throughout the time period considered. This assumption would be reasonable only if this period were very short, but in KBS-3 it is some 1E+10 years (twice the age of the earth!). Having said this, however, KBS-3 does provide what is widely accepted to be a convincing demonstration that spent nuclear fuel can be disposed of in a way which presents very low risks. The models and parameters used tend to overestimate rather than underestimate doses, and the level of detail included in the models is roughly commensurate with our present confidence and ability to quantify and understand processes, and the links between them.

The most detailed part of the analysis concerns the near-field mobilization of radionuclides from the spent fuel in copper containers, surrounded by a bentonite buffer. A sophisticated model is built up to describe the movement of corrodants in and out of the buffer, and the rate of dissolution of container and fuel is derived. The development of a redox front in the near-field is assessed, and the transfer of key radionuclides from the near-field, across the redox front and into the far-field is calculated. At this stage the analysis makes the very pessimistic assumption that the far-field comprises only a 100 m long single fracture, which connects the repository to a major vertical fracture system where water movement is so rapid that it can to all intents be assumed to be part of the biosphere. Although realistic values of groundwater flux along the connecting fracture are used, and matrix diffusion is attributed a major role, this is nevertheless a very conservative approach to the far-field, given the amount of available data which could have been used. The biosphere part of the analysis uses the well known BIOPATH model and assumes groundwater releases either to a well (intersecting the major fracture zone) or to a lake of about 3 million cubic metres volume. Although the lifetime of the copper container (i.e. the initial total containment period) is calculated to be in the order of 1E+7 years, a pessimistic assumption of failure after 1E+5 years is made. This shows up in the predicted releases of activity to the biosphere for the 'central scenario', where no release occurs (F 10.4) before 1E+5 years. Thereafter, up to 1E+7 years, 129-I dominates releases and consequent doses. In some respects 129-I dose levels are an artefact of the lack of a realistic far-field component in the migration model. The 'instant' releases observed highlight the lack of any near-field retardation mechanism for iodine, although the spreading of release due to the 'mixing tank' effect of backfill and spread of canister failure times is taken into account. Beyond 1E+7 years, 226-Ra dominates doses, but the time scale to which the analysis extends is clearly invalid, as geological and climatic conditions will not remain stable for such periods. The maximum individual dose levels for this central scenario are 1E-6 to 1E-7 Sv/a, up to four orders of magnitude lower than that from natural background radiation in Sweden.

Four other scenarios were analysed, including the assumption of initial damage of a single container, the prevalence of near-field oxidizing conditions (F 10.6) which increases the mobility of certain radionuclides, transport by colloids, and concentration of released radionuclides in a peat bog rather than release to a water body with turn-over and dilution. The release to the biosphere under near-field oxidizing conditions shows how 99-Tc now dominates releases, and how releases of all radionuclides are accelerated owing to more rapid breakdown of the fuel. Nevertheless, the maximum individual doses remain well below background, at only 9E-6 Sv/a.

Switzerland NAGRA study

As with the Swedish case, the drive behind the Swiss programme was derived from government regulations in 1978 which required that safe management and disposal of radioactive waste should be demonstrated before operating licences for further nuclear reactors were granted. An extensive safety assessment called Project Gewahr (Guarantee) was published at the beginning of 1985 by the Swiss national cooperative for the disposal of radioactive waste (NAGRA). This analysis is extensively based on the previous Swedish KBS studies but is considerably extended in content, especially in its additional treatment of LLW/ILW types. Two repository types were considered in detail-a HLW repository in granite bedrock overlain by thick sediments, and a deep LLW/ILW repository in a marl. The HLW safety assessment examines in detail only the disposal of vitrified waste, but the option of the direct disposal of spent fuel is not precluded. Some of the highest actinide content ILW would also be included in the HLW repository. However, due to its small effect on total repository activity inventories, this category was not explicitly included in the safety assessment. Processes of dilution, dispersion and decay during transit have greatly reduced radionuclide concentrations and spread their releases over longer times. Analysis of the consequences of such releases into the biosphere leads to dose predictions for each of these radionuclides arising from a number of different food chains:

Table 48 The distribution of radionuclide ingestion doses over various pathways for the Swiss Project 1985. The values are given as a %age of the total dose from each radionuclide, and apply to the biosphere transport reference case used in the assessment.

RI	DW	Milk	Meat	LV	Wheat	RV
Np-237	84	1	1	1	2	13
Th-229*	30	1	1	6	28	35
U-238	96	1	1	1	2	1
Ra-226*	64	1	1	1	25	8
Pa-231	25	1	27	4	7	37
Ac-227*	78	1	21	1	1	1
Ni-59	86	3	2	1	5	3
Se-79	30	3	66	1	1	1
Tc-99	21	54	1	1	15	8
Pd-107	86	3	2	1	5	3
Sn-126*	12	5	1	3	63	17
Cs-135	37	22	23	3	8	7

DW = Drinking water, LV = Leaf vegetables, RV = Root vegetables, * Short-lived daughters taken into account, 1= less than 0.5%.

It should be noted that final doses from the NAGRA study are extremely low, and are in fact much less than those from the KBS studies. Although there are some technical factors involved (relating to the greater depth of the NAGRA repository, potential site geology and so on), the main reason for the large differences is that the KBS studies are designed to be very conservative (particularly in their treatment of the far-field), while in the NAGRA study the base case is as realistic as possible, though still tending to conservatism. The Swiss study also involves a fairly extensive examination of the effects of parameter variations, and small changes in the conceptual models of the near-field, far-field and biosphere. The LLW/ILW repository study investigates deep disposal of all wastes arising from the current Swiss power programme, plus those from medicine, industry and research produced over a period of 70 years. Although sites in crystalline rock and anhydride are also under investigation, the study is based upon a repository in marl beneath a hill side with an overburden of 450 m.

Fewer data are available on the reference site than for the HLW case (as no field site investigation for the former had then been initiated) and hence the approach tends to be more generic and more conservatively based. Although a variety of waste matrices and canisters are used, the analysis takes into account only diffusive resistance in the concrete/cement overpack filling tunnel liner, without considering, in the base-case, elemental solubility limits in this region. Far-field transport is conservatively assumed to occur predominantly in fissures without matrix diffusion playing a significant role. Although base-case analysis indicated low release rates, extensive parameter variations showed that significant doses could result under some circumstances. Values comparable with (yet still usually below) the safety limits set were calculated for certain human intrusion or pessimistic erosion scenarios. The fact that higher doses were predicted from the LLW/ILW repository than from the HLW repository can again be attributed mainly to the extreme conservatism of the former. As with KBS-3, the NAGRA study does not consider some factors which could perturb the mechanistic basis of the calculation chain, such as the role of colloids, micro-organisms and organic radionuclide complexing agents.

Probabilistic assessments

All of the assessments described so far are essentially deterministic. That is, they use the normal case model and use the most likely parameter values, plus educated guesses as to the upper and lower limits feasible for those parameters. These are often brought together in some kind of simple sensitivity analysis. More sophisticated ways of dealing not only with processes whose parameter values may vary quite widely, but also with events which have certain likelihoods of occurrence, is to carry out a probabilistic assessment. In this case, single parameter values are not used, but instead a probability distribution is drawn up for that parameter, which includes a range of the potential values. The calculation then randomly selects from such distributions for each parameter required, and may even randomly select events which may perturb the normal case model. A large number of calculations can be carried out and the results can be presented very simply, for example as the probability of any given dose occurring, or the probability of it occurring at any given time in the future. Atomic Energy of Canada Ltd. (AECL) have developed a probabilistic modelling system based on their Systems Variability Analysis Code (SYVAC) in which the maximum doses which result from each simulation are plotted in the form of a histogram (F 10.10). A single simulation takes randomly selected values from a range for each parameter involved in the complete release and migration model used in the code. The histogram thus shows the most probable performance of the disposal system. SYVAC shows that for times up to 1E+5 years none of the simulations leads to doses greater than natural background (around 1 mSv),and at very long times into the future (1E+7 years) the most probable doses which might result are around 1E-4 mSv, about the same as those calculated in the KBS-3 study. Even at such very long periods into the future the probability of doses exceeding background are very small, but in devising a disposal system it is clearly worth considering the factors in the specific simulations which give rise to these doses very carefully. Considerable interest is currently being shown in such codes and the CEC now has its own probabilistic code under development at the Joint Research Centre at Ispra, in Italy. This is called LISA. While probabilistic models have the advantage of looking at many possible scenarios, the large number of calculations that must be performed means that individual calculations need to be as simple as possible. This is achieved by forming a complete system model from a group of subset models (e.g. of leaching mechanism, diffusion through buffer, speciation, etc.). Thus one disadvantage of probabilistic models is that we must sacrifice some of the sophistication which is possible for many of the processes considered, while the second is simply the lack of data on parameter value distribution functions, which is particularly critical at present. Clearly, future development must aim at producing the most simple but realistic submodels, backed-up by comprehensive data distribution sets.

Model validation and the use of natural analogues (Chapman 1987)

Whenever we have discussed the construction of performance models or the assessment of their results, we have frequently dealt with very large-scale and complex interrelated processes, or with extremely long time scales. This is an inevitable consequence of trying to predict behaviour in the geological environment. It raises the question of how confident we can be in the results of the calculations and, at an even more basic level, how confident we are that the processes we are assuming to occur will actually occur at all. Probabilistic models can cater for the last of these questions, but provide no help with the prediction of the effects and consequences of processes. As a result there has been increasing concern about how we can validate the models which we use in performance assessment. Validation should not be confused with verification. The latter is simply the process of ensuring that the calculations associated with any particular model actually give the correct numerical answer for the assumptions made, and that they are reproducible by other similar or related models. For some years now the international modelling community have been carrying out detailed intercomparison of models to this very end, and as an aid in developing more sophisticated models. Two major projects have now taken place; INTRACOIN and HYDROCOIN, the first dealing with some general radionuclide transport codes and the second specifically with groundwater flow models. There is also an international biosphere model comparison underway, called BIOMOVS. This was initiated by the Swedish National Institute of Radiation Protection in 1985. A fourth major programme called INTRAVAL started in 1986 (initiated by the Swedish Nuclear Power Inspectorate-SKI), which will aim not only at comparing coupled transport codes in more detail, but at actually validating them against both in situ experiments and what are known as natural analogues.

Natural analogues are seen by many as the only reliable means of model validation where processes are involved which concern time scales or experimental system sizes/complexities which cannot be replicated reliably in the laboratory. While some aspects of the processes considered can clearly be validated in the laboratory (e.g. simple diffusion coefficients or sorption parameters in simple closed systems), many others cannot. Clear examples of this are the slow alteration of a bentonite buffer material to other clay minerals, the rate of corrosion of metallic containers in the geological environment, the rate at which pore-waters in cement/waste matrices exchange with surrounding groundwaters, and the rate of migration of a radionuclide through complex, mixed rock-formation systems, with differing redox conditions en route. It became clear that the only realistic means of validating models which incorporated such processes was by direct comparison with similar processes, or groups of processes, in the natural environment. These are called natural analogues, although the term is now often used to cover many 'man-made' systems, such as archaeological analogues; the corrosion of ancient metal artifacts for example. Natural analogue studies began by concentrating on the study of radioactive ore deposits, when it was hoped that an analogue for a complete disposal system might be available (i.e a deep ore body might be directly comparable with a deep repository). However, it soon became clear that this was a gross oversimplification which left the validation argument full of holes, and a more appropriate approach was gradually developed, leading to the definition of five 'principles' of the use of natural analogues:

(1) The process involved should be clear cut. Other processes which may have been involved in the geochemical system should be identifiable and amenable to quantitative assessment as well, so that their effects can be 'subtracted'.

(2) The chemical analogy should be good. It is not always possible to study the behaviour of a mineral system, chemical element or isotope identical to that whose behaviour requires assessing. The limitations of this should be fully understood.
(3) The magnitude of the various physico-chemical parameters involved (P, T, pH, redox conditions, concentration, and so on) should be determinable, preferably by independent means and should not differ greatly from those envisaged in the disposal system.
(4) The boundaries of the system should be identifiable (whether it is open or closed, and consequently how much material has been involved in the process being studied).
(5) The time scale of the process must be measurable, since this factor is of the greatest significance for a natural analogue.

Although analogue studies have been taking place for many years (the earliest and most widely known being the work on the natural reactors in the uranium deposits of Oklo in Gabon), there has been a recent upsurge in interest in many countries and there are now major efforts in hand to apply natural analogues in the manner indicated above: that is, process-specific, with very clear and limited objectives. Such has been the interest that the Commission of the European Communities formed a Natural Analogue Working Group in 1985 to coordinate effort in the field. In its first report the NAWG endeavoured to rationalize the use of analogues by identifying four principal means of application.
(1) As natural experiments which replicate a process, or a group of processes, which are being considered in a model. This is probably the most quantitative application of analogues, which allows confident constraints to be placed on, for example, extrapolations of laboratory experiments to larger time or space scales.
(2) For determining the bounds of specific parameter values. This application would be most useful at the stage where a modeller needs limiting values on a parameter, but can obtain these from any or many geological systems. The origins of the data are not particularly important, and need not be linked to the process being modelled. Diverse sources may be used and a statistical approach adopted. An example of this is thermodynamic or kinetic data, which could be obtained from any system.
(3) As simple 'signposts' indicating which phenomena can occur in the system being modelled by reference to a parallel natural system. This is a purely quantitative application which gives 'yes-no' answers, or indicates the 'direction' of long-term processes. It would be the first means of application used when carrying out scoping exercises.
(4) In an empirical sense to integrate the results of many processes at one site, over long time periods. Not all of the processes involved may be evident, nor may the manner in which they have been linked. Only the end result is important, and in this sense this application is the most directly useful to a safety assessment (as distinguished from the individual models which comprise it). An example might be to determine whether there is any

surface radiological manifestation of a deeply buried uranium ore body.

Many excellent analogue studies have taken place and rather than trying to summarize them all, we have taken as an example one area where they have been applied to the most important modelling aspects of a particular waste-disposal system. This example is their application to HLW disposal in crystalline rocks. The first part of the study was an attempt to define which aspects of a complete system performance model were most in need of analogue validation. This could be done by a simple assessment of the sensitivity of the results of the model to the various mechanisms which were included in the performance assessments.

Behaviour of borosilicate glass (HLW matrix)

The parameters of interest in assessing vitrified waste are long-term stability (with respect to recrystallization), kinetics and mechanism of reaction with water, formation and composition of surface layers and leach rate of specific elements and their possible immobilization in secondary reaction products. Volcanic glasses, an obvious potential analogue for waste glasses, arise from very rapid cooling of magma flows or intrusions. Their composition is very variable, ranging from rhyolitic obsidians (with more than 80 % SiO_2) to basalt glasses with about 50 % SiO_2, which roughly corresponds to the SiO_2 content of waste glasses. However, the Fe and Al rich basalt glasses contain practically no boron or lithium and thus have properties different from those of the waste borosilicate glasses. Nevertheless, basalt glasses show that recrystallization at low temperatures requires around 1E+7 years, which is considerably longer than the assumed lifetime of vitrified waste (1E+5 years). Recrystallization is therefore not expected to affect the behaviour of the waste glass within its lifetime. The hydration rate of natural glasses is basically dependent on the availability of water. Hydrated glass (e.g. perlite) releases elements such as uranium to warm groundwater faster than fresh glass. Cooling and hydration cause stresses which lead to fracturing of larger glass bodies and thus to an increased surface area with correspondingly increased corrosion. Basalt glass surfaces alter rapidly on contact with water. The iron-rich clay layer (palagonite) produced appears to protect the glass for about 1E+6 years. On leaching of the glass, uranium and the rare earths (mainly La and Pr, but not Ce) are retained in this surface layer. The layer often contains strongly sorbing zeolites such as chabasite and analcime.

To summarize, natural analogues for the glass matrix indicate that recrystallization (devitrification) is insignificant on time scales relevant to the disposal concept considered, 'natural corrosion mechanisms basically agree with laboratory observations. If only a small amount of water is available, the kinetics of corrosion can be slowed down considerably, hydration layers are produced in low water-flow conditions. These act as a crust which can protect the glass from further attack by water for a long time, some secondary minerals in the hydration (or transformation) crust show a high sorption capacity for certain elements.

The HLW canister (cast steel)

The model assumption for the canister in the NAGRA safety analysis was a corrosion rate of about 3E-5 m/year and a minimum lifetime of 1E+3 years. The corrosion products then form an efficient redox buffer for more than 1E+6 years. With respect to natural analogues, the issues of interest are the kinetics and mechanism of iron corrosion under the expected geochemical conditions and the long-term stability of the iron corrosion products and their effectiveness as reducing agents. Investigations of iron meteorites as analogues have, to date, given only qualitative results. They are composed not of iron but of Fe/Ni alloys and their corrosion conditions cannot generally be quantified. Nevertheless, a series of corrosion rates has been estimated and some samples have survived at or near the earth's surface for up to 20 000 years. The corrosion rates of archaeological finds of iron show a notable consistency. For investigated samples, values lie between 1E-7 and 1E-5 m/year with a few values below 1E-8 m/year. The objects came from a wide range of environments: dry, wet, oxidizing and anoxic. Some Roman iron objects which probably corroded in contact with air in the wet rubble of an old spring showed a corrosion rate of only 1E-6 m/year. Natural and archaeological analogues therefore suggest that the corrosion rate of 3E-5 m/year assumed for the safety model is conservative, especially under the anticipated reducing conditions.

The bentonite backfill

The most important functions of the bentonite layer surrounding the waste canisters as derived from the near-field analysis are forming a barrier with very low permeability (meaning that near-field transport is primarily by diffusion), preventing transport of radionuclides as particles or colloids and forming a thermal and mechanical buffer. The most important properties of the bentonite to be investigated by natural analogues are its stability and its effect on the transport of radionuclides. With regard to stability, it is known that smectite-rich clays can alter to illites (at high temperatures), provided there is a sufficient supply of potassium. This process can also be observed in nature, for example in thick sequences of sediments (Gulf zone, USA) the age temperature history and pore water chemistry of which are known. Such natural analogues show that illitization is much slower than would be expected on the basis of kinetic models and a regular supply of K+. Instead of a 100 % alteration in 1E+5 years, only 0.3 % in 1E+6 years was observed. Temperatures over 100 C are expected in the bentonite backfill only for a short time. However, natural hydrothermal systems show that, even at 100-200 C and with a sufficient supply of K+, alteration occurs only slowly (in the order of 1E+6 years).

Concrete and Cement

Concrete and cement are not only important immobilization matrices for some types of LLW/ILW but are also widely used as structural and sealing materials. Analogue studies should give information on their mechanical long-term stability and leaching behaviour in relevant chemical environments. There are two types of historic and archaeological analogues: industrial constructions (ca. 100 years old) with concrete of relevant composition, and mortars and cements up to 3000 years old, with widely varying chemistry. In the first group there are many examples of dams, piers and canals which have remained intact for more than 50 years. The second group is more variable but also contains hydraulic installations which have been very stable under relatively aggressive conditions. There are, however, some cases where rapid destruction of concrete was observed, either in aggressive chemical environments or through intensive microbiological activity. A valid conclusion from these findings would be that concrete structures can survive for 1000 years in surface environments but it is not yet possible to draw quantitative conclusions for particular corrosion or leaching conditions.

Bitumen

Bitumen may be used in the LLW/ILW repository as a solidification matrix for certain waste types. Geological deposits of tar and pitch can serve as natural analogues. These show good stability under very unfavourable conditions (e.g. on the earth's surface) which allows an expected lifetime of 1E+5 to 1E+6 years to be deduced. However, this can be drastically shortened by microbiological activity. Archaeological samples containing tar or pitch have proved to be very stable over ca. 500 years. There are, however, many examples to the contrary where bituminous covering material was rapidly destroyed in damp soil, possibly again with involvement of micro-organisms. In the absence of microbiological attack, therefore, bitumen can be regarded as very stable.

Radionuclide release and migration

Natural analogues of radionuclide release and migration have been extensively studied in the last few years. Two of the best known 'global' analogues -Oklo and Morro de Ferro- will be briefly described as illustrations of the general approach. In order to validate models used in the safety assessment chain, more specific analogues are, however, required and these are considered for solubility limits and speciation, retardation during groundwater transport and redox conditions.

The best known example of a natural analogue is the Oklo uranium ore deposit in Gabon, Africa. About 2000 million years ago, spontaneous chain reactions occurred in several identified 'reactor zones' and these continued intermittently for around 1E+5 to 1E+6 years. About 1000-2000 tonnes uranium were present as 'fuel', of which around 6-12 tonnes of 235-U underwent fission, and about 4 tonnes of plutonium was produced. The natural reactor operated at temperatures of 400-600 C and at pressures of 800-1000 bar, which corresponds to a depth of about 3.5 km. The uranium isotope ratios used as a measure of the 'burn-up' of the nuclear fuel are normally constant within small rock specimens and ore grains but characteristically different for different reactor zones, which indicates that no (or little) uranium was transported between the reactors. However, there is some indication of migration of uranium with anomalous isotopic composition into the surrounding altered rock for a distance of a few metres. Radioactive fission products decayed within the geological time periods involved but have left characteristic fingerprints in their stable daughters. The isotopic composition of the latter can be determined by mass spectroscopy and gives information on the extent of migration from the reactor zones into the surrounding rock during or after the reaction period.

Table 49 Summary of element retention in the Oklo fossil reactor zones (RM 2,2,26)
Elements which mostly migrated: Kr, Sr, Mo, Ag, Cd, I, Xe, Cs;
Elements which were mostly retained : Y, Zr, Nb, Rh, Pd, In, Sn, Sb, light REE, Po, Th, U, Np, Pu;
Elements which were locally redistributed : Rb,Tc, Ru, heavy REE

In recent years, Morro de Ferro in Brazil has also been investigated. This body of iron ore with an estimated 30 000 tonnes of associated thorium and 50 000 tonnes of rare earths (mainly neodymium and lanthanum) lies near the surface on a hill of strongly weathered and hydrothermally altered rock which is exposed to erosion and leaching by rain-water. The Th content in the out-flowing groundwater is however only ca. 0.05 g/l and that of La 0.28 g/l. From simple calculations, mobilization rates of 1.4E-9/year for La result. Data recently measured for 226-Ra gave 1E-7/year. The total uranium content of the ore deposit is only 100 tonnes. It is postulated that this figure was very much higher at the time of origin of the ore body some 60 million years ago, and that the U was washed out under the prevailing oxidizing conditions. Transfer factors for Th, Ra and the rare earths through the food chains to man and animals were also determined around Morro de Ferro. Th and Ra concentrations in plants, vegetables and milk are, at most, 10 times higher in the vicinity of Morro de Ferro than in similar products from typical agricultural areas in the northern hemisphere.

Solubility limits and speciation

In the NAGRA Project safety analyses, actinide solubility and speciation are determined by use of thermodynamic equilibrium models. These are checked by comparison of the calculated concentrations of selected elements (e.g. U, Th) with those found in natural groundwaters. The natural release of these elements from the rock is an analogue for release from the waste matrix. Although there are many summaries of water analyses in relevant granite formations, these are generally not extensive enough to allow speciation to be examined in a dependable manner. Inadequacies in sampling and lack of equilibrium are further sources of errors. Despite all this, data can be obtained from such measurements and provide important checks on the models used. Use of analytical data as an analogue for speciation has only been attempted in a few cases as, for example, the Th and La content in water from Morro de Ferro considered above. The following conclusions can be drawn. Extensive analysis of groundwater from the host formation allows the rock itself to be used as a source of certain relevant elements (analogue of the HLW matrix). Assuming solution equilibria, such data can be very useful in checking thermodynamic models. Direct extrapolation of speciation data from near-surface soil conditions to those expected in deep groundwater is inapplicable for many radionuclides. Such data are, however, useful for checking speciation models, particularly in the case of elements with uncertain or non-existent thermodynamic data.

Interaction of radionuclides during migration

During groundwater transport, there is interaction between the dissolved radionuclides and the mineral surfaces present. Separation of the particular mechanisms involved is very difficult in natural systems but, in general, they result in retardation of the dissolved species in relation to groundwater (or in relation to an inert species in the case of diffusive transport). This retardation may, for example, be due to sorption of the solute onto mineral surfaces which, in turn, is dependent in a complex manner on many parameters such as speciation, surface structure, temperature and so on.

There are many analogues for radionuclide migration over a wide time range. Preconditions for use of such an analogue system are spatially and temporally defined source of the elements of interest, a well-defined hydrogeological situation during migration and a measurable concentration profile resulting from migration. Examples of such analogue systems are the Oklo natural reactor, ore deposits, aquifer redox fronts, discontinuities in sediments and fallout from nuclear weapons tests. Investigations of the Oklo reactor give only guiding qualitative data. High-temperature processes (e.g. in connection with magmatic intrusions) are not relevant for most HLW disposal concepts. Investigations of the hydrology in the vicinity of ore deposits give much qualitative data on site-specific radionuclide migration which in turn contribute to quantitative development of relevant models. Weathering profiles and aquifer redox fronts can be investigated by isotopes of the U and Th decay series, whereby the extent and the kinetics of radionuclide retardation can be derived. Sediment profiles which have dateable geochemical anomalies have potential as a means of quantifying retardation during diffusion of solutes through clay strata.

Sorption can be calculated for equilibrium groundwater systems from the exchangeable radionuclide concentration on the solid and in the liquid phase. The greater the sorption, the greater the retardation. Analogue studies often give data for both sorption and total retardation. Such data on natural sorption (or retardation) were taken into account when specifying the database for the NAGRA Project.

Matrix diffusion

Rock profiles from a disturbed zone through which water with a sufficiently high uranium content has flowed can be used for matrix diffusion analogue studies. Interpretation is made difficult by a usually blurred transition from the strongly permeable alteration layer to the micro-fissures in the unaltered rock. For example, during a hydrothermal phase (100-800 C), the penetration depth of uranium enrichment may reach about 3 cm and the subsequent alterations at lower temperatures (<100 C) may remain within this distance. Unfortunately, these and other results are not sufficient to allow unambiguous assessment of the extent of matrix diffusion in relevant situations.

Redox conditions

A potentially important factor in near-field chemistry is the radiolysis of water. Because of the high radiation levels required over long time periods, the selection of natural analogue systems for the effects of radiolysis has been limited to the Oklo natural reactor. The radiolytic effects were investigated in 1983. The radiolytically formed H_2 did not behave inertly but diffused into the structure of the clay minerals and reduced Fe^{3+} to Fe^{2+}. This shows that a model with an exclusively oxidizing radiolysis effect can be considered as conservative. Although the Fe^{2+}/Fe^{3+} ratio in the reactor zones points to reducing conditions, the actual redox conditions are not known and apparently vary locally. Considering the complex nature of the redox conditions in the near-field, the simple assumption of an oxidation front moving through the rock does not appear to be justified in the case of Oklo. In the NAGRA concept, a possible oxidation front would be hindered, by canister corrosion products, from spreading beyond the bentonite barrier for at least 1E+6 years. Some uncertainties could be clarified by natural analogues, for example the mechanism by which natural redox fronts are produced and the behaviour of redox-sensitive species at such a front, the extent to which Fe^{2+} in minerals (primarily in intact granite) is available as a redox buffer for aqueous systems and the kinetics of such buffering and the effectiveness of corrosion products as a redox buffer.