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Use of natural analogues to support radionuclide transport models for deep geological repositories for long lived radioactive wastes

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USE OF NATURAL ANALOGUES TO
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FOREWORD

Plans to dispose high level and long lived radioactive wastes in deep geological repositories have raised a number of unique problems, mainly due to the very long time-scales which have to be considered. An important way to help to evaluate performance and provide confidence in the assessment of safety in the long term is to carry out natural analogue studies. Natural analogues can be regarded as long term natural experiments the results or outcome of which can be observed, but which, by definition, are uncontrolled by humans. Studies of natural analogues have been carried out for more than two decades, although the application of information from them is only relatively recently becoming scientifically well ordered.

This report is part of a the IAEA's programme on radioactive waste management dealing with disposal system technology for high level and long lived radioactive waste. It presents the current status of natural analogue information in evaluating models for radionuclide transport by groundwater. In particular, emphasis is given to the most useful aspects of quantitative applications for model development and testing (geochemistry and coupled transport models). The report provides an overview of various natural analogues as reference for those planning to develop a research programme in this field. Recommendations are given on the use of natural analogues to engender confidence in the safety of disposal systems. This report is a follow up of Technical Reports Series No. 304 on Natural Analogues in Performance Assessments for the Disposal of Long Lived Radioactive Waste (1989).

The IAEA wishes to acknowledge the contributions made by the participants of the two consultants meetings and an Advisory Group meeting noted at the end of the report. It is particularly grateful to J.-C. Petit of DESA/CE-FAR, France, and N. Chapman of QuantiSci Ltd, United Kingdom, for their substantial contributions. A. Bonne and J. Heinonen of the Division of Nuclear Fuel Cycle and Waste Technology were the responsible officers at the IAEA.

EDITORIAL NOTE

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1. INTRODUCTION

The principal objective underlying the concept of deep geological disposal of long lived radioactive wastes is to achieve long term isolation of the radioactivity from humans and the environment. In most disposal concepts, the main natural mechanism that may bring radionuclides from the repository to the surface environment is transport in groundwater[1]. Radionuclide transport models are thus the most important tool used to assess the long term behaviour of a disposal system with respect to confinement.

Such assessments must consider long periods of time, large spatial scales and complex, evolving conditions. Consequently, it is important to have confidence that the tools (models, computer codes and databases) and their application in performance and safety assessments are appropriate, reliable and sufficiently realistic to provide an adequate and credible representation of the system. The conceptual models and databases used in assessments are products of laboratory and field research programmes which are, of necessity, carried out over relatively short periods of time. Therefore, additional information and data are required to build confidence in the estimated safety and performance of a disposal system, as derived from assessment models, specifically because these are extrapolating to much larger spatial and temporal scales.

An important way to help evaluate and provide confidence in the models and data used in safety assessments is to carry out so-called 'natural analogue' studies [1–4]. These use a wide range of information drawn from natural systems and from natural processes that are similar to those that will occur in the repository. Natural analogues are defined more by the methodology used to study and assess them than by any intrinsic properties they may possess [5, 6]. They can be regarded as long term natural experiments whose 'results' can be observed, but which, by definition, are uncontrolled by man.

Natural analogues can include both natural and human-made materials provided the processes that affect them are natural. Thus, studies of archaeological and historical artefacts, ancient buildings, anthropogenic sources of radionuclides such as nuclear weapons fallout, and examples of pathways in plants and animals can be regarded as natural analogue studies. Natural analogue studies can shed light on both near-field processes and radionuclide migration in the far field and in the biosphere. In natural analogue studies, the initial and boundary conditions and other important parameters are often not known precisely, which can limit the degree of accuracy with which they can be interpreted. In general, the majority of analogue data currently available are related to chemical processes in the geosphere, since this has long been seen as the area of greatest need, but other issues of interest could be tackled by analogue studies such as thermal effects of the wastes on the rock, or mechanical deformation (e.g. of bentonite, over long periods of time).

The idea of studying natural systems to obtain information on processes relevant to radioactive waste disposal dates back to the late 1970s. Since then, such studies have made substantial progress, resulting in both a large information base and international recognition of the role of natural analogues in safety assessment. There have been several recent reviews of natural analogue studies [7, 8]. In addition, the application of natural analogues to safety assessment has been the focus of discussion over the last decade [3, 7, 9, 10].

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[1] In some disposal concepts, in particular those involving large masses of metals in saturated, anaerobic conditions, or high heat producing wastes in unsaturated conditions, radionuclide transport associated with a gaseous phase may also need to be considered. However, natural analogues for these processes are not considered in the present report.

This report presents the current status of the use of natural analogue information in evaluating models for radionuclide transport by groundwater. It also provides an introduction to, and overview of natural analogues, for those planning to develop a research programme in this field. Thus, the report has been written in a generic format to cover the wide range of deep geological disposal concepts being considered in individual countries or disposal programmes.

The introduction discusses the use of, and relationships between, natural analogue studies and the various components of the safety assessment of a radioactive waste repository. It then provides brief, basic information on different disposal concepts and on the host rocks being considered in various countries. The bulk of the report focuses on radionuclide transport and retention processes, as deduced from natural analogues, and the modelling of the data produced by such studies for the purpose of safety assessment. Both the usefulness and the limitations of natural analogues in supporting radionuclide transport modelling are highlighted. At the end of the report, conclusions and recommendations are provided regarding the status of natural analogue research.

2. NATURAL ANALOGUES IN RADIONUCLIDE TRANSPORT STUDIES

In essence, a natural analogue study examines the occurrence of materials or processes similar to those found in, or caused by, a repository. Such occurrences should be investigated with the predetermined aim of developing or testing concepts and models for repository safety, or with the aim of providing data, notably when they are not accessible by direct laboratory experimentation. This approach implies an appropriate degree of chemical, physical or morphological similarity between the radioactive waste repository, or some of its components, and the natural system or object under investigation. A natural analogue study provides information on repository behaviour that is often derived from one site, but is applicable to another: natural analogue derived information should, therefore, be transferable.

The development of a disposal concept and the research leading to assessing the safety of the concept are based on knowledge and understanding of the material properties and processes involved. This provides both the qualitative (conceptual models) and quantitative (databases) input to the assessment of the disposal concept. The assessment models, being the mathematical representations of the knowledge and understanding of the disposal system, are needed to calculate the long term predictions of the performance of the disposal system and, thus, to assess future safety and potential environmental impact.

Natural analogue information has proved to be essential to three aspects of safety assessment [4]:

- Conceptual model development
- Data input
- Model testing.

Each of these aspects has different requirements for natural analogue input, as discussed below. In addition, it is important to recognize that natural analogues can provide both qualitative and quantitative information, which are of equal importance. The recognition of these two potential applications, and the consequent nature of the information, is gaining ground [9, 11]. The distinction between the two is a result of the methodology employed or, rather, the objective of the study.

An example of qualitative information would be the establishment of the presence of colloids in natural groundwater systems and their ability to take up radionuclides. This may be contrasted with studies which try to use the same system to quantify colloid transport parameters. The qualitative form of investigation must precede the quantitative form at any site: a process cannot be quantified until it has been identified and described. However, both qualitative and quantitative information is needed when determining the role of colloids in radionuclide transport. This distinction must be understood in order to appreciate the role of natural analogue input in developing safety assessment models. The current use of natural analogue information in safety assessment is illustrated in Figure l.

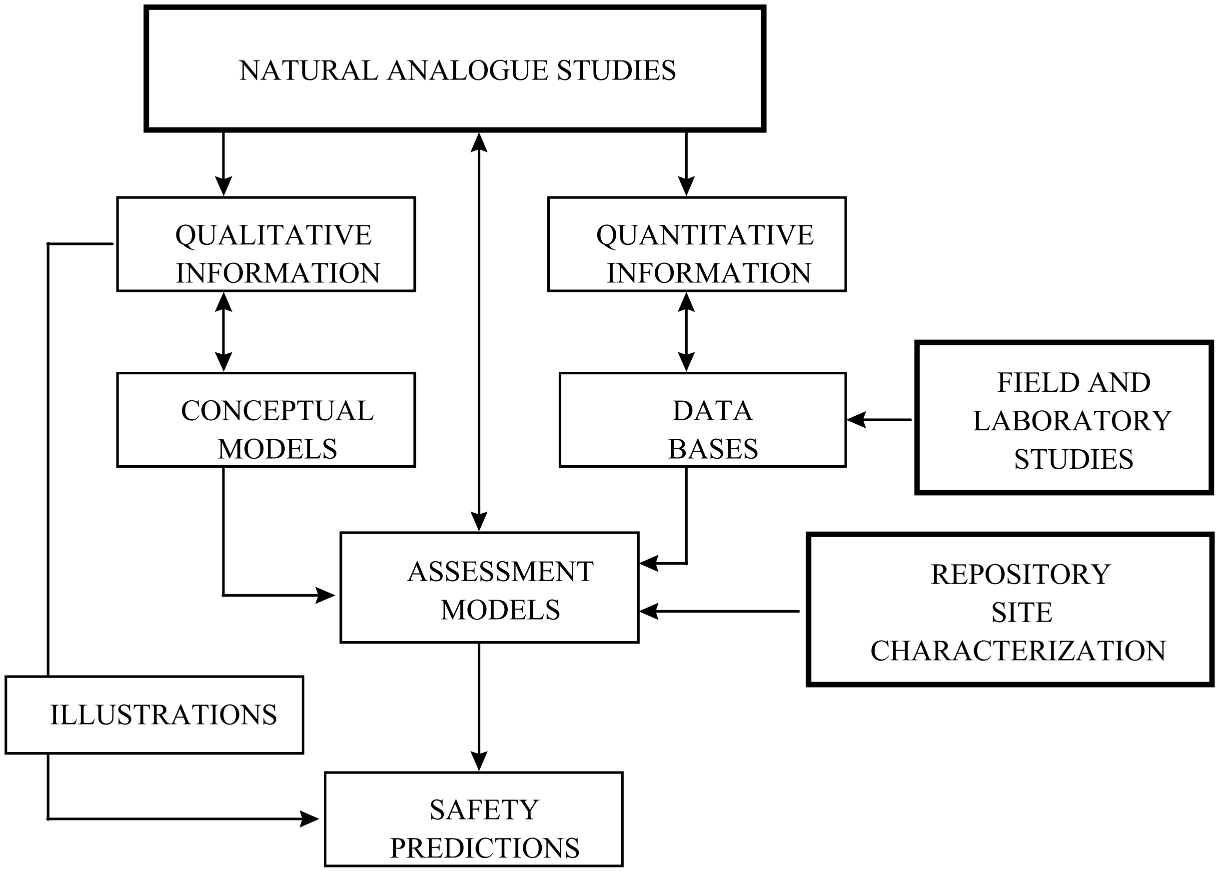

FIG. 1. Schematic diagram illustrating the relationships between natural analogue studies and the various components in the safety assessment of a disposal concept for nuclear fuel wastes.

Analogue information is primarily qualitative or semi-quantitative, because it is impossible, in most cases, to quantify all relevant parameters in natural systems. This is an inherent limitation in the study of all complex natural systems where long term processes have been active. For this reason, an analogue cannot be used, in general, in a quantitative sense for direct validation of a mathematical code such as a radionuclide transport code. In particular instances, quantitative information can be derived from a natural analogue (e.g. speciation of ions in groundwaters compared with the predictions of a geochemical code where the validity of the thermodynamic database can be tested). Both qualitative and quantitative analogue information can be used to test the robustness of the model and to enhance confidence in its predictions.

A recent appraisal of natural analogues and their limitations in performance assessment [10] has emphasised the need to carry out parallel and integrated laboratory and in situ studies at any analogue site under investigation. This is considered necessary in order to define further the initial boundary conditions for the geochemical processes and thus increase the quantitative applicability of analogues in performance assessment.

2.1. CONCEPTUAL MODEL DEVELOPMENT

It is clear that the development of many conceptual models of repository processes may proceed only on the basis of a good understanding of how the natural environment operates. For example, systematic assessment studies often break the complete repository system (near-field, far-field and biosphere) into a set of connected features, events and processes, or FEPs (e.g. in the form of a 'process system' [12]). The development of such systematic models should, ideally, involve natural analogue studies, as it is only after close observation of natural systems that it is possible to make the necessary decisions regarding:

- Which processes to include;
- Which processes are likely to be dominant and which of secondary importance;
- Which process interactions to model;
- The scales (both spatial and temporal) with which the model should cope; and
- Whether the basic premises of a model are appropriate.

Completion of a model of some part of a repository system may thus be the anticipated end-point for a qualitative natural analogue investigation. This is an important procedure contributing to the foundations upon which the entire safety assessment will be built. However, examination of published safety assessments indicates that this aspect of natural analogue studies, although carried out, usually goes unacknowledged.

Examples of natural analogue information used in developing conceptual models include the confirmation of important processes (e.g. complexation by organic compounds) and determining boundary values for critical parameters (e.g. the range of natural groundwater pH values in specific host rocks).

2.2. DATA INPUT

Obtaining well controlled parameter ranges may be more difficult compared with laboratory data derivation because natural systems are inherently complex. In fact, there are only a few examples of natural analogues providing quantitative data, e.g. the extent of solute diffusion into the bulk rock matrix and radionuclide solubility and speciation data. However, analogue studies prove invaluable for providing bounding values to processes, for which laboratory techniques prove inadequate or inappropriate, e.g. for processes too slow to be observed in the laboratory.

2.3. MODEL TESTING

This aspect has grown considerably in importance, as the models and databases used in safety assessment need to be tested against realistic conditions. There are two principal ways in which analogue data may be used to test mathematical models or computer codes. The first are 'simulation' tests, in which a code is used to simulate or reconstruct measurements from a natural system, such as a radionuclide concentration profile, but are not used to make predictions. The modeller may adjust the input parameters until the model provides a good fit with the measurements. In this case the test is whether the code can simulate the observations.

The second form of test involves 'predictive' testing, in which a code is conditioned on one set of data from a natural system and then used to predict the values of a separate set of parameters in the same system, which can (perhaps subsequently) be measured as confirmation of the validity of the predictions. This is similar to the way codes are used in actual safety assessment modelling. In these tests, the modeller might only be provided with a set of general system parameters (e.g. rock and groundwater major element chemistry and master chemical parameters), and then goes on to make so-called ‘blind’ (or ‘forward’) predictions (e.g. of groundwater trace element concentrations). In this case the test is not just whether the code and its supporting databases can produce realistic results, but also whether these results can be constructed from first principles, according to the methodology used in safety assessments. So far, only steady-state geochemical codes have been tested using blind predictive modelling in natural analogue studies, e.g. at Poços de Caldas, Oman, Jordan (Maqarin) and El Berrocal.

2.4. ILLUSTRATIVE ROLE OF NATURAL ANALOGUES

Natural analogues have an important role as providers of illustrative information to a broad range of audiences, both technical and non-technical. This aspect of their use is indicated in Figure 1. In some cases, natural analogues provide information to a wide, public audience as simple comparisons between nature and a repository. This type of wide dissemination is of great value in illustrating complex concepts in a straightforward and informative manner.

However, the illustrative value of natural analogues extends well beyond this general usage. More specific audiences can be identified, with particular interests and backgrounds. For example, well-informed but non-expert technical peers may find the concepts and conclusions from a detailed safety assessment credible only if they are provided with natural parallels for comparison. Other groups may include interested academic researchers, familiar with the broad scientific subjects (e.g. geology or materials science) but not with the intricacies of safety assessment methodology, or politicians charged with the final decision-making but lacking a technical training, who might need some form of everyday illustration of repository safety concepts.

The need to supply easily digestible evidence which indicates that geological disposal is a sound and credible technology should not be underestimated. A complex performance assessment cannot overcome the credibility problem concerning repository safety if no effort is made to explain the concepts in a language that is intelligible to all. Even the assessment modellers require illustrations from natural analogues that indicate that the underlying theory is correct. The illustrative uses of natural analogue information often derive from the ‘soft’, qualitative form of study. It is the identification of processes, the broad evaluation of material stability, and the indications of process rates and interactions that the qualitative study provides which are used in all forms of illustrative literature.

3. THE MAIN CONCEPTS FOR DEEP GEOLOGICAL DISPOSAL OF LONG LIVED RADIOACTIVE WASTE

The geological disposal of long lived radioactive waste involves a system of natural and engineered barriers whose function is to isolate the waste from people and the surface environment for thousands or hundreds of thousands of years. To achieve this goal, different disposal strategies have been chosen, based on the amount and nature of the waste, the available host rocks and the associated disposal concepts (e.g. whether the waste emits a substantial amount of heat, as with high level waste or spent fuel, or whether the host-rock is saturated or unsaturated).

In most disposal concepts, mobilization and transport in groundwater is the most probable natural mechanism to bring radionuclides from the waste package to the biosphere. However, other phenomena such as uplift/erosion, volcanism, human intrusion, etc., might also need to be considered in safety assessments. In all disposal concepts currently being considered, repository performance is based on multiple barriers, including natural and engineered materials. The natural barriers include the host rock and surrounding geological formations. The engineered barriers may include the waste form, waste package, buffer and backfill materials, and seals placed in the underground openings, boreholes and shafts. Together, these barriers are aimed at limiting groundwater contact with the waste and the subsequent transport of radionuclides to the biosphere.

Geological disposal concepts often place the repository in the saturated zone, below the water table, although the Yucca Mountain project in the USA has the proposed repository for spent fuel situated in a thick (hundreds of metres) unsaturated zone. The saturated zone is the region of the rock in which all the fractures and pores are filled with water, under hydrostatic and/or lithostatic pressure broadly equivalent to the depth of measurement. The unsaturated zone is the region above the saturated zone in which at least some pore space contains air or water vapour, rather than liquid water (with the exception of 'perched water'). The saturated and unsaturated zones are separated by the 'water table'. Hydrogeological flow and transport models make a distinction between the two zones, with separate mathematical codes for evaluating water and solute movement in saturated and unsaturated rocks. However, the above does not apply to salt formations.

Different countries are considering a variety of geological environments for deep disposal of long lived radioactive wastes. The following sections briefly describe the concepts, classified as follows:

Repository in the saturated zone	*Repository in the unsaturated zone*
* Hard, fractured (crystalline) rock formations	* Tuffaceous formation
* Argillaceous formations	
* Evaporite formations	

3.1. HARD, FRACTURED (CRYSTALLINE) ROCKS

Hard, fractured (crystalline) rocks form at high temperature and pressure either by cooling from a molten state or by deformation and recrystallization of pre-existing rocks. They occur as individual igneous intrusions (plutons or massifs) or as extensive igneous or metamorphic terrains whose formation and emplacement occurred during various phases of orogeny. These rocks are strong but brittle, and metamorphic rocks commonly show evidence of at least one period of deformation. Although the rock matrix has low porosity and low permeability, fractures and faults cutting through the rock can have much higher porosities and permeabilities. Fractures may separate large volumes of rock with significantly different groundwater flow and solute transport characteristics.

Several countries have considered locating deep repositories in crystalline rock, including Argentina, Canada, Czech Republic, Finland, France, India, Japan, the Republic of Korea, the Russian Federation, Spain, Sweden, Switzerland, Taiwan, Ukraine and the UK. Repository depths between about 500–1000 m are usually considered. A generic design might involve a mined repository, accessed by vertical shafts and containing parallel tunnels. Vertical deposition holes in the tunnel floors are considered in some countries (e.g. Canada and Sweden) whereas other envisage in-tunnel emplacement (e.g. Finland, Japan and Switzerland). Variations on this approach may involve a multilevel repository of a similar design.

3.2. ARGILLACEOUS ROCKS

Argillaceous formations (clays, mudstone, marls, shales) considered as host rocks may occur in extensive sedimentary basins. These basins normally consist of sequences of rocks which may have large contrasts in permeability between adjacent formations (e.g. sandstones and shales). Individual argillaceous formations may be laterally extensive and relatively homogeneous, allowing some confidence in the prediction of characteristics away from the point of measurement.

As a sedimentary basin develops, the formations become buried to great depths and their porosity and water content is usually reduced. Consequently, the formations will gradually lose most of their plasticity, which is derived from their content of water-saturated clay minerals. Conceptually, plastic clays can be considered as essentially continuous porous media, which simplifies hydrogeological modelling, while other clays may need to be treated as dual porosity (fractured) media, as crystalline rocks. Indeed, the majority of mudrocks and shales are fractured.

Several countries have considered locating deep repositories in argillaceous formations, including Belgium, France, Italy, Japan, the Russian Federation, Spain, Switzerland and the USA.

3.3. EVAPORITES

Evaporitic rocks are most commonly produced by the prolonged, in situ, evaporation of frequently recharged surface bodies of saline waters. They include halite and anhydrite deposits, with halite being the most commonly selected repository host rock. Evaporite sequences which provide an adequate thickness for hosting a repository are generally located in large sedimentary basins. Bedded evaporites may comprise complex sequences of interstratified evaporites, of various thicknesses and mineralogical compositions, and sedimentary formations, including clays, shales and sands. The evaporites may remain in this original sub-horizontal disposition (bedded salt formations) or, on increasing depth of burial, they may become unstable in terms of density and plasticity and become mobile, rising upwards through the overlying sediments as massive domes and diapirs (halokinesis).

Salt formations are potentially suitable as host rocks because they are often characterized by very low hydraulic conductivities, the absence of freely mobile groundwater, a relatively high thermal conductivity and a low porosity. They may also display plastic creep, which tends to close any openings. These properties favour exceptionally limited transport of radionuclides from a repository and effective dissipation of the heat generated by radioactive decay.

Halite was among the first geological formations identified as having good potential for hosting a repository [13]. Several countries have considered the possibility of using salt formations (bedded salt and salt domes) for the disposal of radioactive waste, including Denmark, France, Germany, the Netherlands, the Russian Federation, Spain, Ukraine and the USA.

3.4. TUFFACEOUS ROCKS

Tuffaceous rocks (tuffs) are the product of silicic and andesitic volcanism. Individual tuff formations may be deposited as ash-flows or ash-falls. Contraction of the ash during cooling produces a joint system that may be partially or completely filled by minerals that have crystallized from the vapour phase. Ash flow tuffs can become 'welded' into massive rock formations after deposition, by their considerable internal heat. Ash fall tuffs are deposited as relatively cool material, and are therefore non-welded, or only partially welded. Non-welded or partially welded tuffs are more porous and generally have larger primary permeabilities than moderately to densely welded tuffs, unless later alteration by zeolitic or clay minerals clogs the original pore structure. Cooling fractures are sparse or absent in non-welded to partially welded units. Tectonically-induced fractures and faults may be superimposed upon this generally stratified system. Additionally, secondary alteration of the primary rock matrix is promoted by the circulation of hot, mineralized fluids associated with volcanism.

A national repository in tuffs is proposed for the Yucca Mountain site, which lies in a semi-arid region of southern Nevada, USA. This site consists of a thick sequence of volcanic deposits, underlain by carbonates. The repository would be excavated in the unsaturated zone in the upper portion of this sequence, 200–400 m above the present-day water table. The design aims at limiting access of surface water to the repository. Japan also considers (saturated) tuffs as a possible host rock.

4. MODELLING RADIONUCLIDE TRANSPORT AND OTHER PROCESSES IN A DEEP REPOSITORY

This section discusses approaches to modelling radionuclide transport processes in performance assessments of these concepts (see Figures 2–4), in particular, approaches to conceptual model development, data input and testing of mathematical models and how these might utilize information from natural analogue studies.

Most natural analogue studies have focused on crystalline and argillaceous formations and, to a much lesser extent, volcanic rocks. Because of absence of flowing waters, evaporites have not been extensively investigated from a natural analogue point of view, with only a small amount of work reported [14, 15]. They are thus not discussed further here. This review reflects the status of analogue studies performed in the remaining, broad geological environments (crystalline, argillaceous and tuffaceous rocks). It should not be taken as an indication of the suitability of different host rock options.

4.1. THE TRANSPORT PROCESSES

Solute transport in groundwaters can occur by advection or diffusion. Advection is considered the dominant process in highly permeable features, such as fractures in crystalline and argillaceous formations, and sandy horizons within clay beds and densely welded tuffs. For tight plastic clays, diffusion is usually considered the dominant process of transport. This critical distinction is considered in some of the conceptual models discussed later in this section.

In the near field, fluid flow and mass transport may occur under elevated temperatures and variably water-saturated conditions.

Following canister failure, far field radionuclide is assumed to occur under ambient temperature and water saturated conditions, in most disposal concepts. A notable exception are the volcanic rocks in the vadose zone of the Yucca Mountain site, USA.

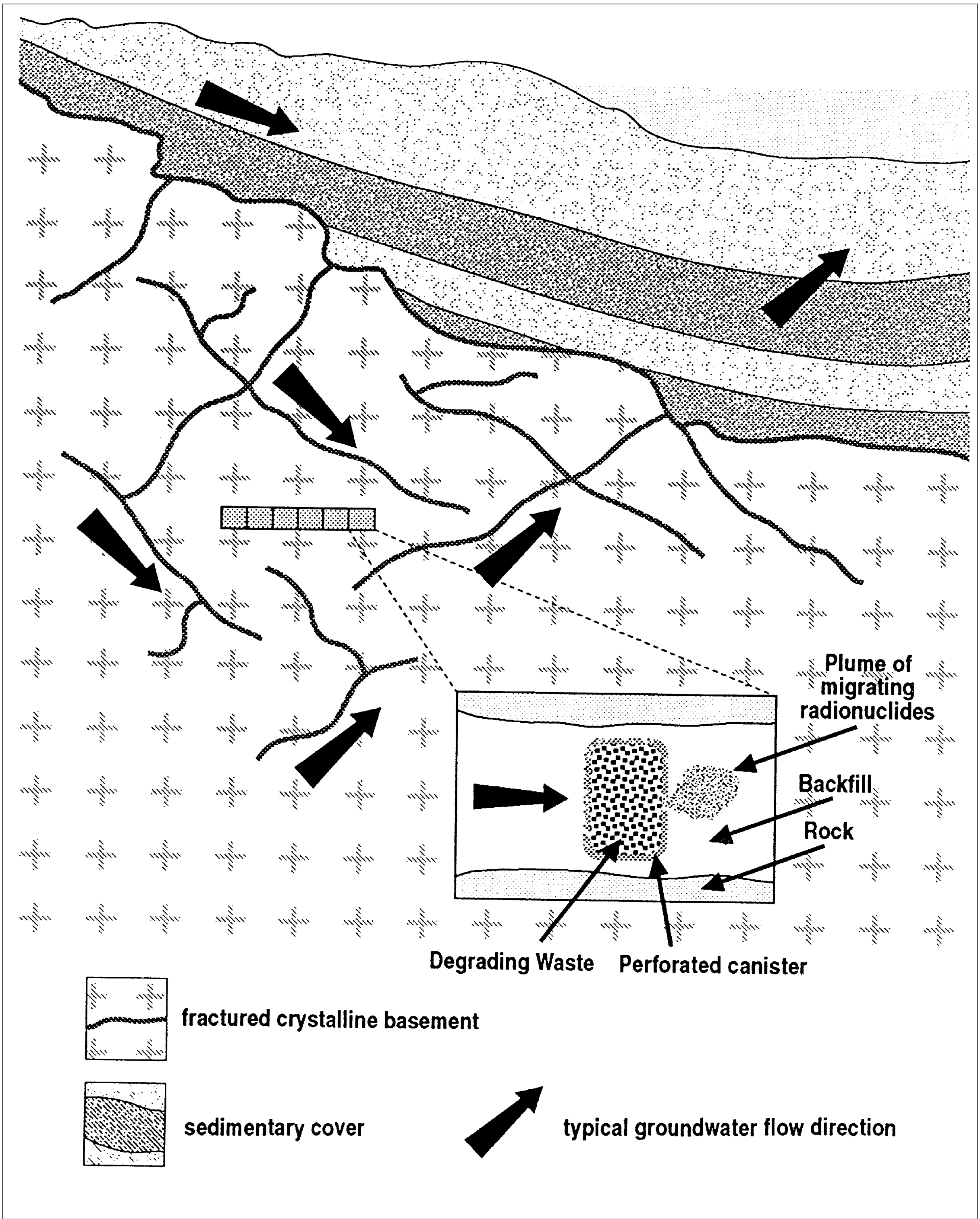

FIG. 2. Schematic release and transport pathways in hard, fractured crystalline rocks. Over a long time period the engineered components of the near field will degrade. Eventually, the waste will be exposed to groundwater and begin to dissolve. Radionuclides will be advected through the fracture network as solutes, or possibly, in particulate form. Their migration may be retarded by sorption onto the far-field rock or, in the near-field, onto the buffer material (e.g. bentonite) or the degradation products of the wasteform and canister. Diffusion into dead-end pores in the rock may also retard transport [7].

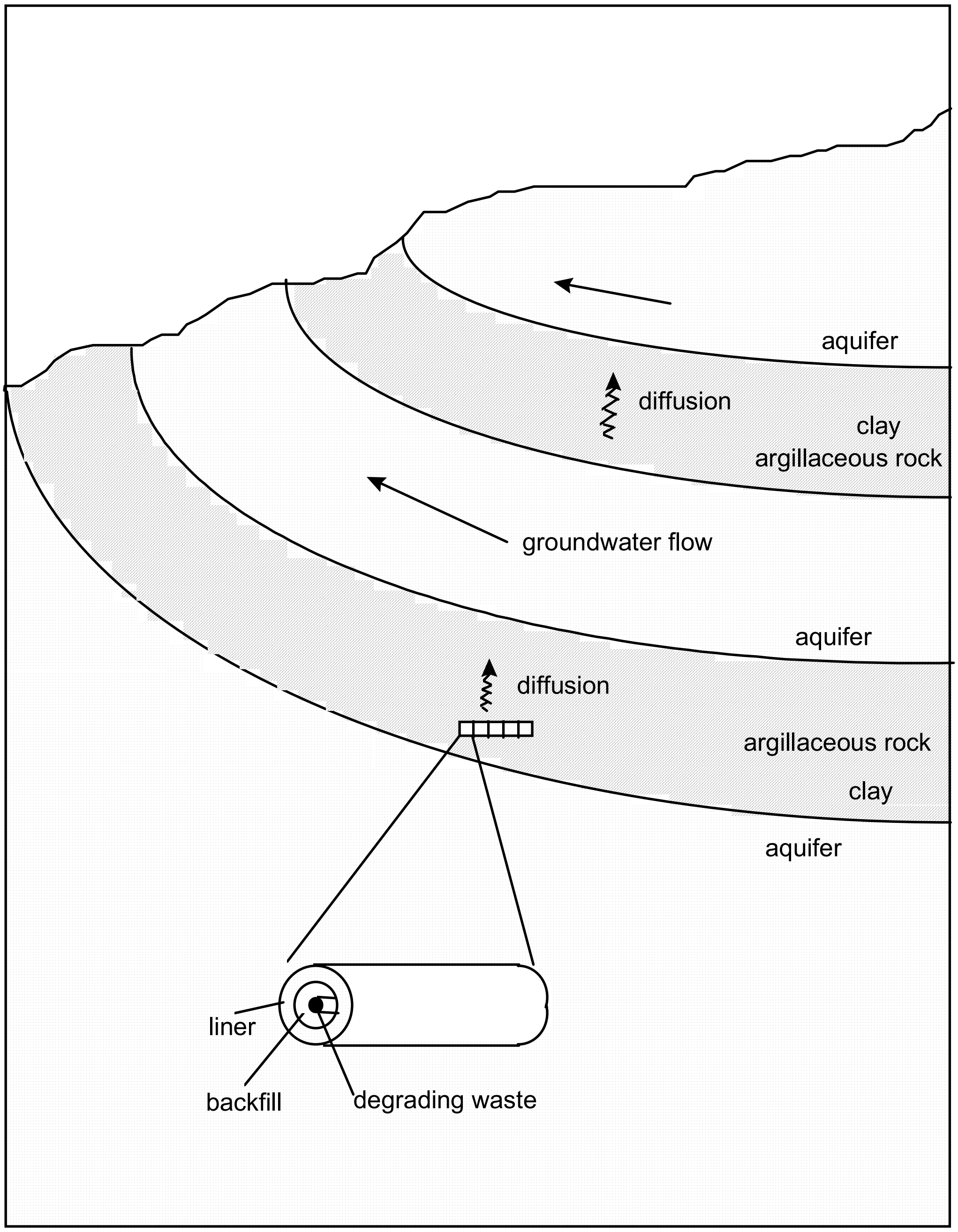

FIG. 3. Schematic release and transport pathways in argillaceous rocks. The release processes in the near-field are similar to those in Figure 2. In plastic (unfractured) clays, transport away from the engineered barriers may be dominated by diffusion rather than advection. The migration of radionuclides would be retarded by the high sorption capacity of the argillaceous rock. A fraction of radionuclides potentially reaching the aquifers will be taken up in the regional groundwater advection system.

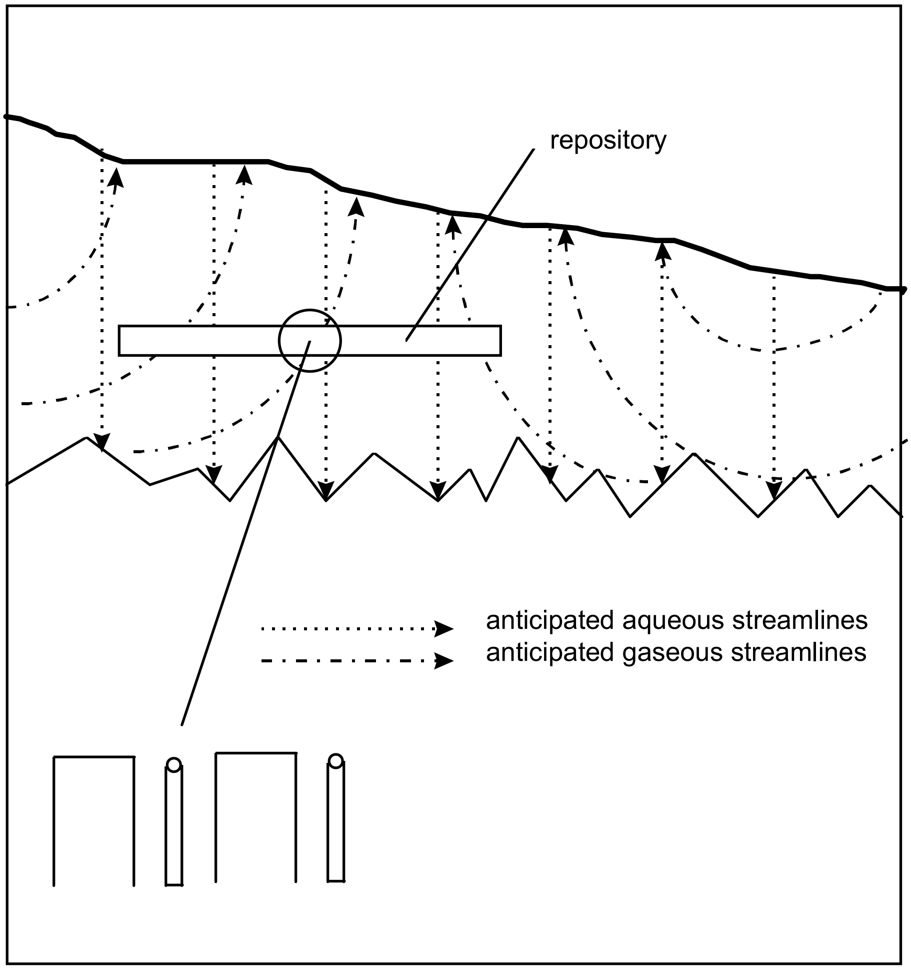

FIG. 4. Schematic release and transport pathways for a repository in densely welded tuffs, in the unsaturated zone. The temperature of the near field will increase over 10^2–10^4 years after disposal, affecting the fluid distribution in the near-field host rock. Dissolution of the waste under oxidizing conditions will potentially result in transport of radionuclides to the biosphere via advective water transport, gaseous transport or disruptive scenarios. Migration of radionuclides may be retarded by sorption on canister degradation products, fracture surfaces or mineral coatings in accessible pores within the tuff [16].

Natural analogue studies have identified a number of important processes influencing radionuclide mobility. For safety assessment purposes, these may be characterized as processes affecting radionuclide mobilization and transport and processes contributing to their retardation. Some processes, such as radionuclide uptake by colloids, can favour either transport or retardation, according to the geochemical and hydrodynamic conditions involved. The most significant processes are shown below, together with working definitions (see also Table I). While these definitions may not be widely accepted in terms of their precision, they do represent current general usage by the natural analogue community and, more broadly, by geoscientists. Other processes, not included in the following list (e.g. osmosis) have been postulated or suggested by laboratory experiments, but their importance has not been demonstrated by natural analogue studies.

Mobilization/transport

Advection	Bulk movement of fluid (and contained solute) induced by a pressure gradient
Diffusion	Movement of solute induced by a concentration gradient
Capillary action	Movement of water induced by negative pore pressure within the rock matrix or microfractures
Dissolution	Transfer from solid to liquid induced by chemical undersaturation in groundwater/porewater
Recoil	Expulsion of daughter radionuclide from a solid phase into solution in adjacent groundwater due to alpha decay of the parent radionuclide
Colloid uptake	Radionuclide sorption onto, or inclusion into, small organic or inorganic particles suspended in groundwater/porewater
Desorption	Removal of radionuclides from mineral surfaces by molecular scale interaction with the solution, thereby increasing liquid phase concentrations

Retardation

Sorption	Removal of radionuclides from solution by molecular scale interaction with mineral surfaces, thereby decreasing liquid phase concentrations
Precipitation	Transfer from liquid to solid phase, induced by chemical oversaturation
Matrix diffusion	Transfer of radionuclides from advective movement (e.g. in fracture-controlled groundwater pathways) to slower, diffusion-controlled movement within dead-end pores in the rock matrix
Particle filtration	Mechanically restriction of particle advection in porewaters by constricted pore connections

TABLE I. SIGNIFICANT RADIONUCLIDE TRANSPORT AND RETARDATION PROCESSES WHICH HAVE BEEN ADDRESSED BY ANALOGUE STUDIES IN THE THREE BROAD GROUPS OF DEEP REPOSITORY HOST ROCKS CONSIDERED IN THIS REPORT

	ROCK TYPE					
PROCESS	Crystalline		Argillaceous		Tuffaceous	
	Fractures	Bulk matrix	Fractures	Bulk matrix	Fractures	Bulk matrix
Mobilization						
Advection	x		x		x	
Diffusion		x		(x)	x	x
Capillary action		x				x
Dissolution	x	x			x	x
Recoil	x					
Colloid uptake	x					x
Complexation	x			x	x	
Retardation						
Sorption	x	x		x	x	x
Precipitation	x	x		x	x	
Pore/matrix diffusion	x	x		x		x
Filtration	x	x		x		

4.2. MODELLING THE TRANSPORT PROCESSES

The processes affecting or controlling the transport of radionuclides from the waste matrix through the geological barrier to the biosphere may be highly complex. It is, therefore, necessary for transport codes to simplify the real system and consider only the most important processes, whilst accounting for the uncertainties involved in such simplification.

Consequently, computer codes have been written with the objective of taking into account different combinations of the following processes:

- Advective transport of dissolved radionuclides by groundwater flow;
- Diffusive transport in low permeability rock formations;
- Retardation due to diffusion and dispersion through interconnected pores;
- Retardation due to diffusion and sorption into a low permeability matrix;
- Retardation due to (ad)sorption of solutes onto solids;
- Retardation due to precipitation/co-precipitation;
- Radioactive decay;
- Recoil effects;
- Complexation;
- Colloid transport.

For the purposes of this report, codes can conveniently be subdivided into those that describe:

- Fluid flow (e.g. VTOUGH, MOTIF);
- Geochemical processes occurring in a static system, including those affecting radionuclide solubility and speciation (e.g. MINEQL, EQ3/6, PHREEQE, SPECIATE);
- Solute transport, coupling flow with chemistry (e.g. CTOUGH, STEELE, TRANSIN).

The latter codes are still in the development stage and have different degrees of sophistication.

4.2.1. Flow models

The primary driving force for advective flow in a saturated medium is a hydraulic gradient (topographic control) and/or a thermal gradient. Capillary action or matrix potential (a negative pore pressure) acts as an additional driving force for advective flow of water and water vapour through unsaturated rock formations. Flow models are generally configured in either two or three dimensions. Multi-phase (gas and water) flow models are necessary in certain circumstances.

In addition to a description of system geometry, the input data that are generally required by flow models include: hydraulic head, hydraulic conductivity or permeability, diffusion coefficients, fluid density and viscosity, rock porosity, degree of saturation, temperature and the equations of state for water.

4.2.2. Geochemical models

The stability of the waste matrix, and the formation of water-soluble and mobile forms of radionuclides are dependent on the hydrochemical characteristics of the groundwater (chemical composition, redox state, pH, etc.). The evaluation of the heterogeneous chemical systems within a repository and the surrounding rock formations is complex, and the combined effects of these parameters are usually assessed by means of geochemical modelling tools.

Geochemical modelling can be based either on the assumption of thermodynamic equilibrium or can include the kinetic limitations of slow water–rock interactions. To date, equilibrium models have been preferred in repository performance assessments, owing to lack of key kinetic data. Geochemical thermodynamic codes calculate the equilibrium composition of solutions by simultaneously solving mass balance and stoichiometric relationships (e.g. PHREEQE, EQ3, Solgaswater). Some codes will also calculate the mass transfer changes between different phases as the system evolves towards equilibrium (PHREEQE). The results of these calculations may include:

- The equilibrium composition of the aqueous phase and the distribution of the soluble species in equilibrium;
- The degree of saturation for given solid and gas phases with respect to an equilibrated solution; and
- The resulting mass transfer across phase boundaries (dissolution-precipitation) as the system evolves towards equilibrium.

A basic limitation of geochemical thermodynamic codes is the notion of equilibrium among the different components and phases of the system. This is certainly not the case for slow processes such as some redox reactions, and water–rock interactions that do not reach equilibrium, even in the long times considered in geological disposal. Therefore, there is a need to incorporate the kinetics of these slow processes, if shown to be relevant in each particular case, into the geochemical modelling codes. This has been done for codes such as EQ3/6 and STEADYQL which will, in addition, calculate the time-dependent evolution of the chemical systems under consideration as they approach equilibrium.

4.2.3. Coupled models

Coupled (groundwater flow, plus solute transport) codes commonly model the water flow element using the continuum porous medium approach. Data on hydraulic conductivity and hydraulic head distribution within the modelled block are required. In continuum modelling, a set of equations representing conservation of mass of water and solute are solved. Numerical solutions are based on finite element or finite difference algorithms. In addition to the advection of the solute, the transport equation may also include:

- A zeroth order reaction (e.g. radioactive decay, heat dissipation);
- Diffusion and hydrodynamic dispersion;
- Some (often simplified) sorption parameters; and
- Colloid transport.

Fracture network and channelling models represent the rock as a system of interconnected planar or tube-like water-conducting features. Their spatial and temporal distribution may be statistically derived. The concepts modelled are basically similar to those dealt with by continuum modelling.

Both continuum porous medium and fracture network models can be modified to take into account the process of aqueous diffusion, under both thermal and chemical gradients. The solute flux between fast groundwater flow-paths (i.e. in macroscopic pores or fractures) and stagnant porewaters (i.e. held in microscopic pores of the rock matrix) is potentially a very significant process. This can markedly modify the transport of trace solutes relative to the advective mass flow of groundwater. It is incorporated into mass transport models by modifying the equation of mass conservation for solutes with an additional flux term between groundwater in a fracture and in the adjacent rock matrix.

More sophisticated versions of coupled codes, which directly couple the transport of solutes in groundwater with solid phase reactions, are now under development and testing. These codes do not form a direct component of safety assessments but can be used at a more detailed level to interpret or to evaluate the impact of changes in the chemical controls on performance. They impose further constraints on the transport of reactive solutes and on the development of system parameters (e.g. major cations, pH, redox state). The codes directly, or iteratively, couple equations formulating flow and mass transport, chemical equilibrium for major species and sorption.

Chemical reaction kinetics, including 'kinetic sorption', have also been incorporated by some modelling tools. Some recent developments follow the approach based on the original so-called quasi-stationary approximation [17]. In such approaches the mineral reactions coupled to the description of flow are described by kinetic rate laws of both precipitation and dissolution under variably saturated and thermal conditions. A further coupling takes into consideration heat transfer, as well as flow and chemical mass transfer in porous formations (e.g. [18]).

The transport of metastable particles, or colloids that carry sorbed or included radionuclides, requires special treatment. This is a transient process which could be important as it would allow radionuclides to be mobilized or retarded in groundwater outside the constraints imposed by equilibrium solubility with mineral phases. Most colloidal transport modelling takes into account both the physical (electrostatic) and chemical factors which control their metastability (e.g. [19]). Colloid stability and transport models are still largely under development at the conceptual level, for example, within the EU-MIRAGE programme. Most scientists involved in this field consider that colloidal stability and transport are time-dependent phenomena and should be treated accordingly. To date, only one code which models the transport of radionuclides by colloids has been applied directly in a performance assessment exercise (Kristallin 1 [20]).

Coupled models can be used to evaluate different scenarios for the evolution of a system over long periods of time. This can be accomplished by evolving a steady-state solute transport model through a series of time steps, either maintaining constant boundary conditions or varying these in relation to historical trends (e.g. glaciation, sea-level changes, rainfall changes, neotectonics). These models assume that the environmental factors and their interdependencies can be described adequately by reference to a present-day 'snapshot' in time. Another form of time-dependent model couples the environmental factors more strongly in order to predict phenomena which might arise from such interactions and from rates of environmental change.

The retardation of radionuclides along the flow path is considered in a very simplified way in the present generation of radionuclide transport codes used in performance or safety assessment. The term 'retardation' includes a number of theoretical mechanisms (diffusion into the matrix, molecular filtration, ion exclusion, physical sorption, ion-exchange, mineralization and precipitation, etc.) but, in practice, they tend not to be well-differentiated, and the net effect of the individual mechanisms is often (imprecisely) represented by a distribution coefficient, K_d.

There is an entire literature on the interpretation of such information, but it is sufficient to note that this coefficient only refers, sensu stricto, to an empirical representation of kinetically fast reversible sorption/desorption process. However, sorption is often found to be a function of various parameters and not fully reversible. In conservative calculations this irreversibility is commonly neglected. When sorption is represented by a K_d, the migration velocity of the reactive solute is reduced by a constant factor relative to the water velocity and may be termed a retardation factor, R_d. The precise meaning of retardation data should therefore be discussed on a case-by-case basis. The K_d approach is considered useful by experts involved in safety assessment exercises because it considerably simplifies calculations. More sophisticated transport codes which take into account individual retardation mechanisms are under development.

5. NATURAL ANALOGUE CONTRIBUTIONS TO TRANSPORT MODELLING

This section describes the contribution of natural analogues to radionuclide transport studies in crystalline, argillaceous and tuffaceous rocks. The distinction between different rock types does not imply significant contrasts in processes across these geological formations, but is used as a common and convenient classification to help the reader to refer to studies which are of most concern to them. The structure used follows the main headings of conceptual model development, data input and model testing.

5.1. HARD, FRACTURED (CRYSTALLINE) ROCKS

The migration of radionuclides with water through crystalline rocks can occur either by advective transport of groundwater along permeable pathways, such as fractures, or by diffusion through the interconnected water-filled pores in the low-permeability matrix of the intact rock. Depending on the degree of excavation damage in the rock surrounding the repository, the groundwater flowpath through cracks and fractures may include sections where fresh, unaltered rock is exposed to the groundwater. This is in contrast to existing fractures, unaffected by excavation damage, where alteration of the surfaces and matrix of the rock surrounding the fracture has established an assemblage of alteration minerals that are generally in chemical equilibrium with the groundwater. In intact rock, unaffected by stress release from repository excavation, the water in the interconnected pores will also be ‘equilibrated’ with the mineral surfaces surrounding the pores.

These criteria are important in determining the characteristics of potential transport paths for radionuclides through crystalline rock surrounding a repository.

5.1.1. Conceptual model development

Studies have been made of natural systems with characteristics analogous to those of interest for radionuclide transport modelling in both fractured and intact crystalline rock. These studies were aimed at identifying the physical and chemical processes that are significant with respect to either enhancing or retarding radionuclide migration (see Table I) and using them to develop the conceptual models for safety assessments of crystalline rock repository concepts. The following sections describe studies relevant to some of the key processes listed in Table I.

5.1.1.1. Dissolution and precipitation

Dissolution and precipitation occur in response to changes in phase compositions during water–rock interaction. Such changes may occur when water compositions change along a flowpath as a result of, for instance, mixing at flowpath (fracture) intersections. Similarly, migrating groundwater may encounter varying rock compositions along flowpaths that intersect freshly exposed rock resulting from excavation damage (i.e. in the so-called 'disturbed zone'). Dissolution and precipitation may also be significant in diffusion-controlled transport through unfractured rock, for example, when considering the diffusion of oxidants or reductants. For instance, a redox front, developed through diffusive ingress of oxygen, may precipitate ferrihydrites which can affect the migration of a diffusing radionuclide across this redox front.

The entrapment of colloids in tight fracture flowpaths, and filtration by the small apertures of pores in the rock matrix, can, in turn, retard the transport of radionuclides in colloidal form.

Examples of relevant analogue studies include those conducted on the characterization of fracture flowpaths in granites (e.g. [21–25]), on transport in, and alteration of, granitic rocks under various low-temperature conditions (e.g. [26–29]), and on diffusion-controlled migration of radionuclides in crystalline rock matrices (e.g. [23, 30, 31]). Examples of colloid formation in natural systems in crystalline rock are described in the following section.

5.1.1.2. Sorption

Studies on sorption have formed part of most natural analogue programmes, but they remain limited in number in crystalline rock system. However, results from these analogue studies have provided important input to the development of conceptual models for transport codes used in safety assessments. These results include information on the role of colloids, dissolved organics and alteration minerals in the transport of mobile radionuclides.

Examples of natural analogues of sorption in crystalline rock formations include studies on sorption properties of rocks and minerals (e.g. [32, 33]) and organic complexation (e.g. [34–36]). Studies on the 'sorption' of radionuclides by colloids have also been carried out (e.g. [35, 37–41]) and work is in progress in Sweden at Äspö and in Finland at Palmottu.

5.1.1.3. Recoil

The alpha-recoil mechanism may potentially enhance the mobility of those radionuclides located on mineral surfaces in contact with water. This process has primarily been observed in uranium-containing systems where measurements of uranium-series isotopes show the loss of ^{234}U from the solid to the aqueous phase under reducing, no-flow, conditions (e.g. [25, 42]). Alpha recoil of radionuclides may therefore be of significance in both fractured and intact crystalline rock for the transport of uranium. However, it is unlikely to be of significance for the transport of several 'safety-relevant' radionuclides in high level radioactive waste (e.g., ^{129}I, ^{99}Tc, ^{14}C, etc.) because these are not subject to alpha-decay. This mechanism may be important for transuranium elements.

5.1.1.4. Matrix diffusion

Matrix diffusion can act as a retarding and diluting mechanism by removing radionuclides from the groundwater flowing in the fractures and by enlarging the rock surface accessible to sorption of radionuclides. Results from laboratory experiments confirm the existence of connected pores in rock and fracture filling material (cm-scale) through which diffusion of solutes occurs. However, laboratory experiments are limited by the potential change in rock porosity of the de-stressed samples as well as the small space and time scale over which the diffusion mechanism can be studied.

Many potential natural systems exist which may be used to show that the system of interconnected pores in the rock matrix adjacent to fractures is accessible to mobile species in the groundwater. Studies have been made of the diffusion of natural decay series radionuclides (e.g. [23, 25, 30, 43–47]), of oxidants (e.g. [31]), and of elements in response to changes in groundwater composition (e.g. [48]).

The results of these analogue studies confirm the presence of a connected porosity, possibly extending up to several tens of cm from a fracture surface into the crystalline rock matrix, and that radionuclide diffusion into this matrix must be considered as a potential retardation mechanism in transport modelling.

5.1.2. Data input and model testing

5.1.2.1. Data input

Information obtained from natural analogue studies can provide a range of inputs to performance assessment databases, from simple quantitative parameter values to bounding ranges for selected parameters and processes. The following paragraphs provide examples of analogue studies which provided information that has been used to help develop databases for safety assessments or to give more confidence in data obtained from laboratory experiments carried out over short periods of time.

An attempt to estimate diffusivities from measured concentration profiles has been made by Shea [29], by matching the observed concentration profiles for chlorite grains to theoretical curves calculated by a diffusion model. Values of the apparent diffusivity, D_a (an empirical parameter which combines terms for the effective diffusivity of a radionuclide in water and for the physical properties of the porespace), in the range 10^{-21} to 10^{-18} m^2/s were obtained. For concentration curves observed in the solid rock matrix, estimated D_a values ranged from 10^{-19} to 10^{-16} m^2/s. However, the model applied in this study is very simple, in that it assumes constant conditions over the time of interest (of the order of 10^4 to 10^7 years), which may not be realistic.

D_a values for uranium in the range of 10^{-10} m^2/s can be obtained, for bulk rock close to fractures and pegmatitic veins [30, 43, 44]. It should be noted that the apparent diffusivity is dependent on the magnitude of sorption. Therefore, an estimate of diffusivity should be combined with an analysis of the geochemical conditions and an evaluation of the sorption parameters.

The chloride concentration profiles observed in granitic rock at the island of Hästholmen in Finland were compared with theoretical curves calculated using a diffusion model [48]. The best fit was obtained with an apparent diffusion coefficient of 2×10^{-9} m^2/s, which corresponds to an effective diffusion coefficient of 1×10^{-12} m^2/s. This value is about an order of magnitude higher than the effective diffusivity measured in the laboratory.

In the Swiss 'Kristallin 1' performance assessment, natural analogue data are explicitly cited as proof of the existence of matrix diffusion as well as providing an estimate of the depth of rock affected by this mechanism [43, 44].

5.1.2.2. Geochemical codes and databases

Some of the most valuable applications of natural analogues in crystalline rock to repository safety assessment have been in testing the geochemical models (i.e. codes and databases) used to calculate radionuclide solubility and speciation in groundwaters, and their sorption behaviour in contact with fracture mineral assemblages. Chemical thermodynamic models provide two main types of data for repository assessment purposes:

- Solubilities of particular elements or radionuclides;
- Speciation of such elements in solution (which allows their transport properties to be estimated).

These models were tested by carrying out a series of ‘blind’ predictions at several natural analogue sites (e.g. Poços de Caldas [49]). Using only the major element content and master chemical variables of groundwaters as input, the solubility-limiting phases, the saturation concentrations and the aqueous speciation at saturation for trace elements considered important in various safety assessments (e.g. U, Th, Pb, V, Ni, Sn, Se, Cr, Mo, Cs and Ra), were predicted. The predictions were then compared to observed aqueous concentrations, speciation and mineralogy. The redox behaviour of uranium has also been studied at Palmottu, and the results compared with theoretical calculations using different databases [50]. From all sites, there was a reasonable agreement between the predicted and observed trace element concentrations. However, discrepancies occurred in the predicted aqueous speciation, in particular for uranium. This underlines some of the large uncertainties which still exist in the solute speciation databases.

Recently, the application of steady-state kinetics to describe slow water–rock interactions, and the use of co-precipitation/co-dissolution approaches to describe the link between major and trace element geochemistry [51] have improved our capabilities to predict radionuclide solubilities in geological systems. The approaches have been successfully tested in El Berrocal [52] and by revisiting the Poços de Caldas trace element data [53].

There appears to be scope for extending this approach to the testing of other types of models. A limited attempt has been made on models of redox front movement, for example at Poços de Caldas [54]. The role of chemical equilibrium for some trace solutes, including radionuclides, is not yet well understood whereas others, such as uranium, have been studied extensively in different geochemical environments. However, the redox behaviour of uranium in natural systems still needs additional work. Perhaps the most obvious target would be solute transport models, where validation is currently based on short term tracer migration tests. Recently, the coupled transport code STEELE has been satisfactorily tested within the Oklo natural analogue project. The model results are supported by the observed pattern of the hydrogeochemical evolution at the site [55]. This is an example of the quantitative information which can be obtained from a natural analogue study.

In conclusion, whilst these predictive tests add considerably to confidence in the geochemical codes and databases employed in certain transport models which underpin safety assessments, their use is dependent on the quality of the input data. In addition, from a strict performance assessment point of view, refinement of codes to account for details may not be necessary, as long as simpler codes can be shown to be conservative. However, a complete understanding of the phenomena, processes and mechanisms involved and an evaluation of the quality of data are necessary to decide, with reasonable confidence, whether or not a simplified model remains conservative.

5.1.2.3. Coupled transport models

The validity of coupled models has been tested at Poços de Caldas [56] where the CHEQMATE and CHEMTARD codes were used to simulate the processes involved in uranium transport across a redox front system, moving into the far field environment. There was general agreement with the field observations; for example, the calculations showed that the amount of uranium associated with iron oxyhydroxides in the oxidized bedrock is significant compared to the amounts precipitated at the reduced side of the redox front. Furthermore, the rate of movement of the redox front was considered reasonable when compared to other estimates derived from geomorphologic observations and U-series disequilibrium studies. In contrast, the formation of secondary pyrite, normally associated with pitchblende nodules at the reducing side of the redox front, was not simulated. Generally, deficiencies in the modelling approach could be explained by inadequacies in the thermodynamic database.

Neither of the two approaches to modelling time dependency, i.e. a steady-state model ‘stepped through’ time by changing the boundary conditions, and a fully coupled time-dependent model which emphasizes transient phenomena, have been tested using natural analogue studies. The latter model approach may be relevant to some analogue sites which have been subject to known periodic changes in climatic conditions. In this respect, sites which have undergone repeated glaciations in Finland and Sweden have been studied so far.

For example, at Palmottu, fracture mineral studies have revealed the existence of young fracture-infilling calcites. Some of the fractures contain two or three generations of calcites, for which the U-Th dates coincide with the warm interglacial periods between succeeding glaciations [57]. The results indicate the cyclicity of the past climate-driven evolution of the site and give support to the predictability of the nature and possible impacts of forthcoming climate-change. Sulphate-rich groundwaters, probably generated at the end of the previous glaciation at the ground surface, are presently detected at a depth of 100 to 150 m [58]. The system offers good possibilities for time-dependent flow modelling which could account for such information.

5.2. ARGILLACEOUS ROCK

Section 2 described the disposal options being considered for argillaceous rocks. In addition to being used as a host rock, argillaceous materials (bentonite clays) may be used in the engineered barrier system as backfill/buffer and seals.

Geochemical transport processes in argillaceous rock are not as well understood as those in fractured crystalline rocks, owing largely to the complexity of the multiple coupled processes of solute–clay interactions, which not only affect radionuclide transport, but also control the movement of water and the development of the hydraulic properties of clay formations.

In homogeneous clays, whether as host rock or engineered barrier material, transport occurs dominantly by diffusion. Therefore, analogue studies of diffusional transport in clays may have relevance to both the near-field and far-field components of performance assessments.

Radionuclides transported in groundwaters through water-saturated clays may be sorbed or adsorbed onto clay mineral surfaces. In addition, precipitation and dissolution reactions can occur, depending on the degree of chemical-saturation of the groundwater. Movement of large particles (colloids and complexes) may be inhibited due to the small pore spaces between clay minerals.

Some argillaceous formations (mudrocks, shales, etc.) may be fractured, and/or may contain a high proportion of coarser grained material. In such cases, advective transport may be dominant over diffusion.

5.2.1. Conceptual model development

Where geochemical discontinuities occur as a result of the heterogeneity of a clay-rich sedimentary sequence, or from the intrusion of material into an argillaceous formation, they offer the opportunity to study migration and other processes at a small scale. Typical studies concern:

- The relative roles of diffusion and advection and the significance of small-scale physical heterogeneities;
- The estimation of elemental diffusion coefficients;
- Evaluation of mobilization and retardation processes (e.g. sorption, dissolution and filtration).

Several natural analogue studies have provided information to support the development of conceptual models involving these processes and some examples are described below.

One group of studies concerns the ability of saturated clays to limit geochemical fluxes around buried materials. Of particular interest is the study of fossil trees at Dunarobba in Italy [59, 60]. These fossil trees are still in their original vertical positions and, unlike most other examples of buried forests, are still composed of wood. In normal circumstances, wood alters to lignite and is subsequently lithified when buried.

At Dunarobba the remains of the 1.5-million-year-old trees were found embedded in a lacustrine clay, which is overlain by sand deposits with freely circulating, oxidizing water. The low-permeability clay envelope has protected the wood from degradation processes by preventing access of groundwater and/or air to the wood. Another example that indicates that clays can effectively prevent groundwater infiltration is found in a 2100 year old Chinese burial tomb [61]. Upon excavation in the 1970s, the body, and other buried objects (silk, wood, food and other artefacts) were found to be well preserved. The preservation resulted from the emplacement of a charcoal layer and a kaolin-clay layer around the wooden coffin structure, which prevented ingress of groundwater and air.

A second important characteristic of argillaceous formations is the ability to act as a colloid and macromolecule filter. This is demonstrated in the Cigar Lake uranium deposit, where the ore body is largely surrounded by a 10 to 50 m thick illite/kaolinite clay halo, locally isolating it from the overlying sandstone host rocks [62]. Studies of uranium and thorium distributions in colloids taken from the ore zone, the clay and the sandstones show higher concentrations in samples from the ore and the clay, suggesting that the latter zone has effectively filtered colloids and prevented them from migrating into the groundwaters in the surrounding sandstones [35].

Although much of the initial work on matrix diffusion was based on data from unfractured sedimentary rocks (e.g. [63, 64]), very little work has since been carried out on fractured sediments. The only known study on matrix diffusion in fractured argillaceous rocks [65], provides data from the Opalinus Clay (a potential HLLW repository host rock in Switzerland) which clearly show the presence of matrix diffusion to a depth of at least 8 cm in the fine-grained rocks.

Further work is ongoing at Nagra's potential L/ILLW repository site at Wellenberg in central Switzerland, where initial results indicate that the fractured and fine-grained marls of the Palfris Formation also display evidence of matrix diffusion to depths of 5–8 cm into the rock matrix [66]. Despite the fact that the rock matrix appears to be sealed by clays and/or calcite, matrix diffusion is evident in the U-series isotopic data in these profiles.

5.2.2. Data input and model testing

Available analogue studies on clays have tended not to distinguish between the provision of data and the testing of transport models. Both topics are thus discussed together in this section.

The studies at Loch Lomond and on the Maderia Abyssal Plain have both provided useful quantitative information on diffusion rates and have demonstrated that appropriate models can simulate observed diffusion concentration-profiles.

In the Loch Lomond study (e.g. [67, 68]), a range of concentration profiles was constructed for various elements, from analysis of sediment core samples which were particularly clay-rich, with up to 80% clay in some horizons. The deposition of a marine band was found to have occurred between 6900 and 5400 years ago, as determined by ^{14}C analysis, palaeomagnetic and palynological data. Fixation of iodine, bromine, uranium and ^{226}Ra was clearly identifiable within the marine layer, and could be correlated with the presence of organic carbon.

The porewater concentrations of bromine and, to a lesser extent, iodine decrease with distance from the marine sediments. The bromine concentration profile was modelled according to simple diffusion with reversible sorption, which neglects advective transport. When an initial bromine concentration of 60 ppm was assumed, the model produced an apparent diffusivity of 8×10^{-11} m^2s^{-1}. Batch sorption experiments were performed on the core material for iodine and bromine and it was discovered that, in both cases, the calculated apparent diffusivities were an order of magnitude less than those determined, possibly due to sample perturbation in the laboratory. This suggests that, although the laboratory studies are conservative, the calculated diffusivities are probably more realistic.

In addition, the laboratory experiments indicated that some sorption processes were apparently irreversible for several radionuclides. It follows that the assumption made in many models of migration, that sorption is instantaneous and reversible, is inaccurate and not realistic. The underlying kinetics of the process may thus need to be investigated further. From a strict performance assessment viewpoint, the major conclusion is that, whatever the simplifications, models must remain conservative.

Colley & Thomson, working on deep ocean sediments from the Maderia Abyssal Plain [69], examined the uranium enriched layers at inactive redox fronts and measured the longer lived parent–daughter pairs of the ^{238}U decay series; ^{238}U–^{234}U, ^{234}U–^{230}Th, ^{230}Th–^{226}Ra, and ^{226}Ra–^{210}Po. It was discovered that the only radionuclide to exhibit migration, since the front became inactive, was ^{226}Ra, whose symmetrical concentration peak around its parent ^{230}Th implies that transport occurred essentially by diffusion (although some advection has been detected in layers parallel to the surface). If significant advection of water had occurred in any particular direction, the resulting ^{226}Ra distribution should display asymmetry. The concentration profile for ^{226}Ra was used to calculate effective diffusion coefficients for this radionuclide of between 6×10^{-13} to 1×10^{-13} m^2s^{-1}.

Quantitative parameter values derived in natural analogue studies, such as those described here, could be used directly in mathematical models used in safety assessment. However, it is more likely that they will be used to support and provide bounding limits when extrapolating short term laboratory derived values.

5.3. TUFFACEOUS ROCKS

The migration of radionuclides through undisturbed saturated tuffaceous rocks may occur by advection through fractures and fracture zones or by diffusion into and through the rock matrix, depending on the degree of water saturation, the permeability and the temperature of the host rock. In an unsaturated host rock, such as that described in Section 3 at the Yucca Mountain site in the USA, where most recent interest has been focused, radionuclides may be transported in either a liquid or gaseous phase based on the fluid state. At the Yucca Mountain site, a further natural pathway for release has to be considered, via disruption of the disposal system by volcanic activity. Natural analogues are also useful in evaluating this scenario. Overall, there has been considerably less work carried out on natural analogues for transport processes in tuffaceous rocks than in argillaceous and crystalline rocks.

5.3.1. Conceptual model development

Several natural analogue studies have contributed to identifying processes of importance to the development of conceptual models for the performance assessment of a repository at Yucca Mountain. Studies of uranium ore deposits in a variety of saturated rocks, such as those at Poços de Caldas, Alligator Rivers and Marysvale, aided in initial development of conceptual models for uranium transport and sorption modelling. Due to the nature of the unsaturated, fractured tuff host rock at Yucca Mountain, dual continuum models of flow were developed to account for advective flux through fractures and matrix. Multi-phase (liquid and vapour) flow models were also developed to account for the variably saturated medium, and higher thermal flux due to waste emplacement.

The location of a repository in the unsaturated zone may expose the waste to an oxidizing environment, and a source term model applicable to such conditions has to be considered. Natural analogue studies have been an integral part of determining significant radionuclide migration and retardation processes under oxidizing conditions. These processes (Table I) have been used in developing the conceptual models for safety assessment exercises of the tuff-hosted repository in the USA. The following points address the contribution from analogue studies for each of the processes identified in Table I, and for additional transport processes due to potential volcanic disruption of a repository at Yucca Mountain.

5.3.1.1. Advective-diffusive flow and transport

The relative importance of advective and diffusive fluid flow in unsaturated fractured tuffs will affect the results of a safety assessment. In this respect, the existence of secondary fracture filling minerals (different from those in the bulk rock mineralogy) provides valuable information regarding the relative importance of fracture flow and flow through the rock matrix.

Examples of relevant analogue studies include those conducted on the characterization of fracture and matrix flow paths in tuffs (e.g. [70–74]). Natural analogue studies on transport of radionuclides include those conducted on the energetics of eruption and entrainment of host rock in a volcanic eruptive column (e.g. [75]) and those on the distribution of U in fractures and the host rock [74, 76, 77].

5.3.1.2. Capillary action

Conceptual models of flow and transport in unsaturated media must also consider capillary action as part of the flow processes. Capillary action can slow the movement of water and radionuclides through the host rock. This process has been studied by characterization of the fracture coatings and U content within the microfractures and rock matrix at the Peña Blanca site (e.g. [72, 73]).

5.3.1.3. Dissolution and precipitation

Dissolution and precipitation are processes occurring in response to mechanical and thermal disruption of the host rock during repository construction and after waste emplacement. Fresh rock surfaces may react when in contact with groundwater, causing dissolution and precipitation of minerals along fracture surfaces. The increased temperature of the repository environment due to waste emplacement will cause dissolution and precipitation of the host rock along fractures, thereby altering the flow path of groundwater and water vapour. These changes to the host rock may affect the transport of radionuclides. Degradation of the waste package will result in oxidizing fluids coming into contact with the waste, causing dissolution of the spent fuel, transport of radionuclides via fractures and/or matrix and, perhaps, precipitation or co-precipitation along the flow path.

Examples of relevant analogue studies include evaluations of dissolution, precipitation and co-precipitation along fractures and within the rock matrix (e.g. [70–74]). Secondary minerals, produced by oxidative alteration of primary phases, have also been studied (e.g. [78]), as has the oxidative alteration of natural uraninites [76, 79].

5.3.1.4. Sorption

Sorption studies have been included in most of the tuff-based natural analogue projects cited above. These include studies of uranium sorption on illite, iron oxides and iron hydroxides in fractures and within the rock matrix. The studies have provided useful supporting data and bounding values for the sorption databases used in safety assessments. Diffusion of U into the rock matrix has also been identified at the Peña Blanca site (e.g. [77]).

5.3.2. Data input and model testing

Geochemical modelling relies on the use of valid thermodynamic data for a specified mineral assemblage. Data from natural analogue studies have been used to identify the alteration sequence of uraninite, and hence the mineral phases (soddyite and uranophane) controlling the release of radionuclides in a chemically oxidizing, hydrologically unsaturated regime (e.g. [71]). Natural system studies confirm the long term stability of uranyl silicates under oxidizing conditions (e.g. [76, 79]) and have provided data on the reaction kinetics of ianthinite, shoepite and uranophane (e.g. [76]).

Uranium series data have been used to determine the transport of U along fractures and diffusion into the matrix at Peña Blanca, providing a bounding value for long term, episodic transport [78]. The relative importance of meso-fracture, micro-fracture and matrix transport were evaluated, providing a ranking of the long term effectiveness of different transport paths [80]. Observations of water seeping from fractures within the adit at Peña Blanca help bound the magnitude of the hydraulic conductivity in an unsaturated tuff. Observations of groundwater recharge in a well at Peña Blanca have provided bounding values for the recharge of a perched water zone within the unsaturated zone (e.g. [81]).

Only a limited amount of testing has been reported for groundwater flow and radionuclide transport models in tuffs. Hydrothermal models have also been tested against natural systems (e.g. [82]). Tests at natural analogue sites have identified gaps in thermodynamic data, and the EQ3/6 codes and data bases have been tested at both the Peña Blanca site [79] and at Alligator Rivers [83]. A blind test of the code CTOUGH was performed to compare the predicted transport of metals in an unsaturated tuff with the observed distribution beneath metallic artefacts at the Akrotiri archaeological site, Greece [84].

6. SAFETY ASSESSMENT AND NATURAL ANALOGUES

The long term environmental impacts of radioactive waste repositories must be quantified in order to be confident that they remain within acceptable limits, as defined by regulatory criteria. Impacts are evaluated using well established safety assessment methodologies which evaluate the performance of each part of the disposal system, as well as its integrated behaviour.

The methodology adopted must be capable of identifying and evaluating alternative evolution scenarios for the natural system and must be able to address the uncertainties inherent in the assessment process. Radionuclide transport models are a central part of all methodologies, regardless of the specific disposal concept and system design. This section illustrates the effective use of natural analogue information in safety assessment exercises carried out by various national programmes over the last 10 to 15 years.

The modelling approaches used in safety assessments and in natural analogue studies may differ. In safety assessments, simplified, conservative assumptions are generally used, whereas in natural analogue studies, the objective is usually more detailed and realistic modelling of processes. However, it is also claimed that analogue studies in many cases are limited to parameter fitting in the absence of site-specific data.

The mathematical codes used in safety assessment transport modelling require information that describes the site itself and the processes that will affect the repository and may lead to the release and transport of radionuclides. The site-specific information required by the transport codes should describe, in broad terms:

- Groundwater movement;
- Groundwater chemistry; and
- Bulk rock and fracture mineralogy.

Further information required by the transport codes describes in more detail the mobilization and retardation processes, including:

- Degradation of the containment system;
- Mobilization of the radionuclides from the waste into the groundwaters;
- Retardation mechanisms as the radionuclides move through the engineered barriers and surrounding formations to the biosphere.

However, many safety assessment exercises simply assume an instantaneous or steady release from the waste packages and do not include the progressive degradation of the containment system. Some assessments conservatively assume that all radionuclides are available for mobilization from the waste into groundwaters on the basis of elemental solubilities rather than the steady dissolution of the waste form.

In the near-field, close to the repository, the behaviour of radionuclides will be governed by the interactions between local pore fluids, inflowing groundwaters, the waste form and other repository components. At greater distance from the repository, the radionuclides may become minor components of a large, unperturbed, natural far-field geochemical system, due to dilution and retardation. However, in the case of a repository where large amounts of cement-based materials would be used (as geotechnical structures and/or engineered barriers), a high-pH plume may be generated by the interaction of groundwaters with such alkaline materials. This plume could progressively extend to great distances and could markedly affect the rock in the far field. Antropogenic analogues can provide a potential tool to study this situation.

6.1. EXPERIENCE FROM SAFETY ASSESSMENT EXERCISES

About a dozen comprehensive safety assessments of the deep geological disposal of radioactive wastes have been published. Table II identifies seven important studies which have made use of natural analogue information, and it is notable that six of these concern repositories in hard, fractured crystalline rocks, the seventh being for the Yucca Mountain tuff site. The crystalline rock studies apply to generic or reference repositories, and are designed to demonstrate the safety of the overall concept of deep geological disposal, rather than the safety of any particular design at a specific site. This section summarizes how information generated in natural analogue studies has been used in these assessments. The summary is based on previous work published [7, 85], with the addition of information from the more recent safety assessment exercises. Table II classifies the contributions with respect to their relevance to conceptual model development, data input and model testing.

Qualitative information derived from natural analogues is generally included in three main areas:

- In developing the basic justification for the underlying concept and choice of disposal environment and engineered barrier materials;
- In the derivation and discussion of natural evolution scenarios for inclusion in the safety assessments;
- In the justification for the content and structure of conceptual models.

These qualitative uses comprise a rather understated application of natural analogue information. Nevertheless, the conceptual derivations and justifications underlying both disposal system concepts and assessments are rarely based on scientific first principles, but almost always on a reasonable understanding of how the relevant natural environments have evolved and how they behave over long time periods.

Quantitative data from natural analogues often assume a higher profile in the reporting of assessment studies, and usually consist of either:

- Bounding values for certain key parameters (such as corrosion rates, sorption and dissolution values), which are used to demonstrate the conservatism of assumptions used in the assessment;
- Specific data actually used in the model calculations, in the form of parameter values or ranges (e.g. colloid concentrations, relevant mineral assemblages) or as contributions to the generic databases (e.g. thermodynamic) used in assessment sub-models.

This latter contribution is one which natural analogues are almost uniquely equipped to make, and is normally achieved by database and model testing.

The information in Table II shows that the use of analogue information in recent safety assessments has increased compared with the earlier exercises (i.e. KBS-3, Project Gewähr), and the use of both qualitative and quantitative analogue information is now being more explicitly mentioned in assessment reports (e.g. Kristallin I, TVO 92, AECL EIS).

TABLE II. SUMMARY SHOWING THE ACTUAL USE OF NATURAL ANALOGUE INFORMATION IN PUBLISHED SAFETY ASSESSMENTS ON DIFFERENT CONCEPTS FOR THE DISPOSAL OF NUCLEAR WASTE

Safety	*Development*	*Data*	*Model*
KBS-3 (Sweden, 1983)	* Radiolytic oxidation of spent fuel against observations from Oklo	* Max. pitting corrosion factor for Cu * Bentonite stability at T<100°C	
Projekt Gewähr (Switzerland, 1985)	* Stability of borosilicate glasses * Stability and instability of concretes and mortars * Stability of bitumen * Radionuclide release concepts against Oklo observations	* Long term steel corrosion rates * Constrain illitization of bentonite	
SKB-91 (Sweden, 1991)	* Support of bentonite stability from observations at Gotland * Redox front model supported by Polos de Callas observations * Inclusion of matrix diffusion	* Limit relevance of colloid transport by using data from Polos de Callas * Demonstrate conservatism in estimating radiolytic oxidation by using information from Cigar Lake	* Radionuclide solubility model testing and comparison with observed solubilities at Polos de Callas and Cigar Lake
TVO (Finland, 1992)	* Use of palaeohydro-geological data in development of Ice age scenarios * Observations from Cu deposits and Kronan cannon to support corrosion estimates * Use of colloid and microbial information from Polos de Callas and Palmottu to develop models	* Matrix diffusion profiles surveyed from various natural analogues	* Testing of UO_2 spent fuel dissolution models using Cigar Lake information
AECL EIS (Canada, 1994)	* Support development of conceptual models for: − Fuel dissolution; − Cu corrosion; − Clay buffer; and − Radionuclide retardation, particularly the role of colloids and organics	* Geochemical processes and parameter values for: − Redox control on UO_2 stability (incl. radiolysis bounding values); − Cu corrosion; − Bentonite-to-illite conversion; and − Radionuclide retardation (incl. matrix diffusion bounding values)	* Testing of models and databases for: − Radionuclide solubility; − Colloid formation and organic complexation; and − Cu-corrosion, using observations form Cigar Lake, the Canadian Shield and the Kronan cannon
Kristallin-I (Switzerland, 1995)	* Back-up in scenario development	* Bounding calculations on redox front development using information from Polos de Callas * Depths of matrix diffusion penetration	* Radionuclide solubility model testing and comparison with observed solubilities at Polos de Callas, Oman and Maqarin * Testing models for redox front development
NRC IPA (USA, 1995)	* Disruptive scenario development (volcanism) * Back-up source term conceptual model from Peña Blanca * Relative importance of meso-micro-fracture and matrix transport at Peña Blanca * Back-up for vapour phase transport from Valles Caldera * Back-up conceptual model for transport in fractures	* Identification of secondary phases for long term release at Peña Blanca	* Model testing for elemental transport in unsaturated media at Akrotiri

A number of broad areas can be identified from these studies, and from current developments, which have benefited considerably from conditioning using observations from natural analogue systems:

- The identification of relevant geological and climatic evolution scenarios to be included in safety assessments, of the processes that these involve and of the impacts that these might have;
- Definition of the long term evolution of engineered barrier materials under both 'stable' and thermally or geochemically perturbed conditions, including the corrosion and degradation behaviour of waste forms (borosilicate glass, uranium oxides), cements, concretes and bitumens and bentonite clays;
- Development of redox front models, particularly appropriate to spent fuel repositories, including models for the formation and movement of redox fronts and for the behaviour of radionuclides in their vicinity;
- The field identification of matrix diffusion, which had started as a largely theoretical model but which can now be observed and its effects quantified in a variety of rock types;
- The quantification of constraints on the significance of colloid transport in many geological environments, which have been difficult to determine from theoretical models alone;
- Contribution to the development of a much clearer understanding of the mineralogical and hydrochemical controls on radionuclide solution chemistry and sorption/migration behaviour, based on detailed characterization of natural migration systems and the effective testing of thermodynamic databases.

Nevertheless, there is still much development work to be done in terms of presenting assessments in a 'natural' framework. It is widely accepted that the credibility of results based simply on modelling, and aimed solely at quantitative risk and dose criteria, can be found lacking when the work is exposed to a wide readership. This is true, even within scientific audiences. It is thus important to consider the potential use of the natural analogue information in assessment studies, at the stage of deciding how the assessments are to be structured and the sources from which the data are to be derived.

7. CONCLUSIONS AND RECOMMENDATIONS

- The examples of natural analogue studies discussed in this report show that they have a potential to provide means of building confidence in transport models and in many other aspects of safety assessments.
- The use of natural analogue studies in confidence building can be considered in three interrelated areas: conceptual model development, data input and the testing of codes (mathematical models).
- Scenario and conceptual model development is improved by considering processes observed in natural analogue studies. This is particularly important for slow processes that operate over large spatial and temporal scales, and that may not be observable or simulated in the laboratory.
- Natural analogue studies may be used to test mathematical transport codes in two complementary ways. Simulation testing involves using codes to reproduce observations

made in natural systems. This type of testing is often employed in analogue studies as part of the system interpretation process. So-called 'blind' (predictive, or forward) modelling involves using codes conditioned on a restricted amount of data to predict independent observations of other parameters. This has proved to be one of the most useful aspects of quantitative natural analogue application in development and testing the models which, to date, has been mainly applied to steady-state geochemical codes. The approach is also currently being developed for other types of codes, such as coupled flow-geochemical codes.

- Since natural analogues can be described as complex, long term 'experiments' that are uncontrolled by people, great care must be taken when using them to test models. Successful testing of a model in a natural analogue study does not imply the correctness of its predictions under all conditions covered in a safety assessment. Rather, the analogue provides a useful means of identifying and scoping the principal uncertainties underlying the application of the model to a complex natural system.

- Natural analogue studies provide unparalleled sources for illustrative material. Illustrations are useful both for those experts directly involved in safety assessments and for decision-makers and the public.

- Confidence can only be built gradually, with the progressive increase in knowledge and understanding. The choice of future natural analogue studies should aim at improving our information base, for example, by guiding the closer integration of laboratory and in situ studies at both analogue and potential repository sites. This is essential for further refinement of models and for the detailed optimization of disposal systems.

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