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An overview of radionuclide behaviour research for the UK geological disposal programme

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ABSTRACT

In the UK, radioactive wastes currently planned for disposal in a geological disposal facility (GDF) are intermediate-level waste, some low-level waste and high-level waste. Disposal of other materials, including spent fuel, separated uranium and separated plutonium are also included in the planning of a GDF, if such materials are classified as wastes in the future. This paper gives an overview of the radionuclide behaviour research studies of the Nuclear Decommissioning Authority Radioactive Waste Management Directorate (NDA RWMD). The NDA RWMD's current understanding of the processes that control radionuclide behaviour in groundwater and how the engineered and natural barriers in a GDF would contain radionuclides is presented. Areas requiring further work are also identified.

Keywords: solubility, sorption, complexation, research and development.

Introduction

The Nuclear Decommissioning Authority (NDA) has established the Radioactive Waste Management Directorate (RWMD) to manage the delivery of geological disposal for higher activity radioactive wastes, as required under UK Government policy published in the *Managing Radioactive Waste Safely* (MRWS) White Paper (Department for Environment Fisheries and Rural Affairs *et al.*, 2008).

The multi-barrier approach to implementing geological disposal following International Atomic Energy Agency (IAEA) requirements (International Atomic Energy Agency, 2006) is the approach used by the NDA and other waste management organizations (e.g. Nagra, Posiva and SKB). This will involve designing engineered barriers that will work together with the natural environment to isolate and contain radionuclides and prevent their release to the surface environment in amounts that could cause harm to life and the environment. In order to demonstrate post-closure safety of a GDF, the implementer will need to show that the quantities of radionuclides which may eventually reach the biosphere will be sufficiently low that they will not pose an undue risk to living organisms.

The evolution of various engineered and natural barriers are discussed in a suite of research status reports (Nuclear Decommissioning Authority, 2012). The topic of radionuclide behaviour is covered within one of these status reports and describes NDA RWMD's understanding of how radionuclides may be released from a GDF and return to the biosphere through movement in groundwater. The focus is on the post-closure period (after the GDF has been sealed and closed) and application of this understanding to build confidence in long-term safety. Processes are considered that demonstrate how the engineered and natural barriers will provide sufficient containment or retardation of radionuclides. This builds understanding of the residual amounts reaching the biosphere to ensure they are sufficiently small that any possible radiological impacts are acceptably low.

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Currently there is no proposed site for a GDF within the UK. As a result of this, current research conducted by or for NDA RWMD is generic (i.e. not tailored to any specific geology or concept). At the point when a potential GDF site is identified, NDA RWMD's research and development (R&D) programme will become site-specific and will be tailored to the specific geological environment. Throughout this paper reference to the 'current preparatory studies phase' indicates the generic phase where no site has been identified.

This short paper describes aspects of the radionuclide behaviour understanding and research needs for NDA RWMD in the current preparatory studies phase and is based on information (Nuclear Decommissioning Authority, 2010*a*, 2011), which has been updated to reflect recent developments. The NDA RWMD recognize that there are significant studies of relevance elsewhere, but in this short paper the focus will be on work conducted by or for NDA RWMD.

Key processes

The key processes influencing radionuclide behaviour are illustrated in Fig. 1. The wastes that will be disposed in the GDF will be in solid form. They will be packaged in containers which, after closure of the GDF, will act to delay the access of water to the waste or wasteform.

Once the GDF has been closed and sealed, it will gradually resaturate with groundwater. Once the containers have been breached and the GDF is resaturated, water will enter the waste containers and contact the wasteforms. It will then be possible for radionuclides to be slowly released into the water in the engineered barrier system (EBS) and subsequently through the EBS and surrounding rocks. The important processes that affect radionuclide behaviour in the waste package, near field and geosphere are described in more detail in the *Geological Disposal: Radionuclide Behaviour Status Report* (Nuclear Decommissioning Authority, 2010*a*).

Scientific basis and current understanding

The technical approach adopted in the UK and internationally to achieve a robust evaluation of the processes influencing radionuclide behaviour is to use evidence available from a variety of sources, including laboratory-based experimentation, modelling studies, monitoring and practical experience in the nuclear and other relevant industries (e.g. construction), and long-term evidence from natural and man-made analogues.

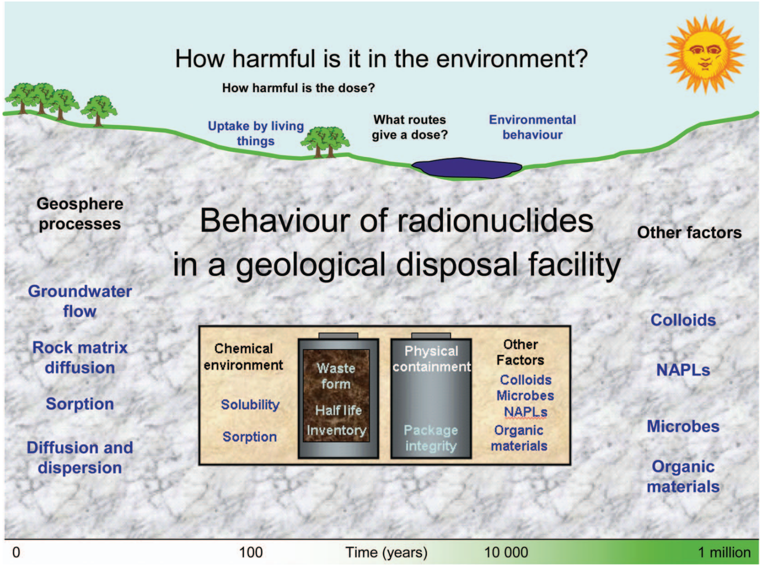

FIG. 1. Behaviour of radionuclides in a geological disposal facility; key features, events and processes. Figure published with the permission of the NDA.

Our understanding of radionuclide behaviour draws on a number of sources. These include: (1) R&D carried out by or for Nirex over a period of 25 years; (2) R&D carried out by or for the NDA since it was established; and (3) R&D carried out on radionuclide chemistry in academic institutions in the UK and overseas, which include collaborative studies, often partly funded through the European Commission (EC) or coordinated by the IAEA or Nuclear Energy Agency (NEA).

Understanding of the radionuclide inventory and the processes that determine radionuclide behaviour enable the evaluation of the containment provided by the multiple barrier system. Waste management organizations can build confidence in the safety of a GDF for the following reasons:

(1) The wastes that will be disposed in the GDF have been characterized and therefore there is a good understanding of the radionuclide inventory and the chemical form in which they will be disposed (wasteforms and waste containers[1] for different types of wastes may be considerably different).

(2) All radionuclides decay over time. The radiotoxicity of radionuclides is reduced through radioactive decay and, after a sufficient period of time has elapsed, the hazard posed by any radionuclide in the waste will have reduced substantially. The timescale for this to occur is dependent on the original inventory of the radionuclide in the waste and the radionuclide half life.

(3) After closure of the facility, the waste containers act to delay the access of water to the wasteform. Radionuclides will not be released from the wasteform during the period when it is isolated from water, except through gas production, which is discussed in the *Gas Status Report* (Nuclear Decommissioning Authority, 2010*b*).

(4) When contacted by water, the wasteforms will dissolve slowly, thereby slowing the release of the radionuclides that they contain.

(5) Processes such as solubility limitation, sorption and precipitation/co-precipitation retard or immobilize radionuclides and result in many radionuclides being contained in the engineered and natural barriers of the GDF.

(6) There is a good understanding of the mechanisms of uptake and potential radiological impacts of the very small levels of long-lived radionuclides that will reach the biosphere (Nuclear Decommissioning Authority, 2010*c*).

Radionuclide behaviour is represented in the disposal system safety case (Nuclear Decommissioning Authority, 2010*d*) and specifically in the generic post-closure safety assessment, which is an assessment of the safety and environmental performance of a GDF (Nuclear Decommissioning Authority, 2010*e*). The approach to assessing safety is to understand and illustrate the range of possible behaviours of the disposal system and the ways it may evolve, and to build confidence in that understanding.

Although it is possible to demonstrate an understanding of the basic processes at the current preparatory studies stage in the GDF project, this understanding will need to be refined once site(s) have been identified. Site-specific information, such as the chemical composition of the groundwater, will have an impact on the speciation of the radionuclides. The type of host rock and the nature of the minerals it contains will strongly influence radionuclide retardation and immobilization along with other influences on radionuclide behaviour which will be strongly site-specific.

Research needs for the current phase

The RWMD's forward programme for research on radionuclide behaviour is described in the *Status Report* (Nuclear Decommissioning Authority, 2010*a*) and in a programme overview document (Nuclear Decommissioning Authority, 2011). Specific objectives for radionuclide behaviour R&D during the current preparatory studies phase of the GDF programme are to: (1) provide understanding of the processes which control radionuclide behaviour in the various barriers of a GDF; and (2) provide UK-specific data for use in safety assessment calculations.

As previously mentioned, RWMD recognize that substantial amounts of work on geological

[1] A *waste package* is the combination of a *wasteform* (the waste as it arises or appropriately conditioned) and a *waste container*. The wasteform is inherently capable or purposely designed to *immobilize* the waste and its radioactive content as a solid with suitable mechanical properties and high durability. In some cases, the wasteform is manufactured with the use of a conditioning medium termed *encapsulant*. The waste container is designed to *physically confine* the radioactive content of the wasteform in a durable envelope with the required mechanical and chemical properties. Together the wasteform and the waste container contribute to achieve important safety functions of the waste package.

disposal have been conducted internationally and RWMD aim to fully utilize this work. At this stage in the MRWS process, RWMD are developing the understanding of key processes affecting radionuclide behaviour. The development of models to interpret experimental data is important to providing this understanding and the majority of RWMD's projects take the form of combined experimental and modelling studies. Figure 2 shows the structure of the planned future work programme on the topic of the radionuclide behaviour, which is described below.

Radionuclide release from the wasteform

This work area provides understanding of how radionuclides may be released into groundwater from the different higher activity wasteforms that are being used or considered for UK wastes. The dissolution behaviour of these wasteforms and associated release of radionuclides is influenced by the nature of the wasteform and by the chemistry of the groundwater that contacts it. This water is expected to have been chemically conditioned by the materials in the near field, such as cement, bentonite or metal. Where radionuclides are distributed throughout the wasteform, such as those in high-level waste (HLW) and spent fuel (SF), the rate at which the radionuclides are released is largely controlled by the dissolution rate of the wasteform. Where radionuclides are present at the surface of waste items or in a finely divided form, they will probably be released more quickly. However, such wastes items are generally encapsulated which slows the radionuclide release. Solubility limitation provides an upper limit to the dissolved concentrations of radionuclides in water within the waste packages. Slow wasteform dissolution or radionuclide sorption on surfaces in the waste package may reduce concentrations below this level. Once mobilized in solution, the radionuclides may move by diffusion or advection, from the waste package into the backfill or buffer.

Areas for further research

Reviews have been undertaken to understand glass waste forms, dissolution rates and durability (e.g. Utton *et al*., 2011; Harrison, 2011). Recently experimental studies have started to expand RWMD's knowledge in this area. An experimental programme on the durability of HLW and intermediate-level waste (ILW) glasses will measure the dissolution rate(s) of a selection of samples that encompass the range of compositions of HLW glasses under UK disposal conditions and obtain leaching data for a range of typical ILW glasses or slags (including the behaviour of inhomogeneous products) under UK disposal conditions. The work will also use existing mechanistic understanding from international programmes to interpret and understand the data measured in this programme of work for UK glass compositions under UK disposal conditions.

Measurements of instant release fraction (IRF) and matrix dissolution rates on different burn-up UK advanced gas-cooled reactor (AGR) fuel are being investigated. The programme will conduct one year leaching tests but may be extended to measure the dissolution rate over longer timescales (up to two years). The tests will be carried out in a representative groundwater at 20ºC in oxidizing conditions. This will allow a good comparison with data already published for LWR fuels under similar experimental conditions and work that will run concurrently with the EU 7th Framework Programme FIRST-Nuclides (see www.firstnuclides.eu) which is designed to measure the IRF from higher burn-up LWR fuels.

Intermediate-level wastes have been studied in the UK for more than 30 years. There is a good body of data and associated process understanding. A long term project to demonstrate the principle of chemical containment of radionuclides by a cement conditioned near-field environment is ongoing. This should provide evidence to demonstrate retardation of radionuclides within a high pH, low Eh cement conditioned environment representative of a cementitious near-field.

Radionuclide transport, retardation and immobilization

This topic covers the transport, retardation and immobilization processes occurring as a result of both the near-field, EBS and the far-field, geosphere. Physical barriers in the near field reduce the flux of radioactivity to the geosphere by reducing the flow of groundwater, in which radionuclides dissolve, in the vicinity of the waste packages. Chemical barriers achieve the same affect by causing the solubility of key radionuclides in the near-field porewater to be reduced.

Many radionuclides are retarded by cementitious and clay barriers. Sorption is the key mechanism by which this occurs.

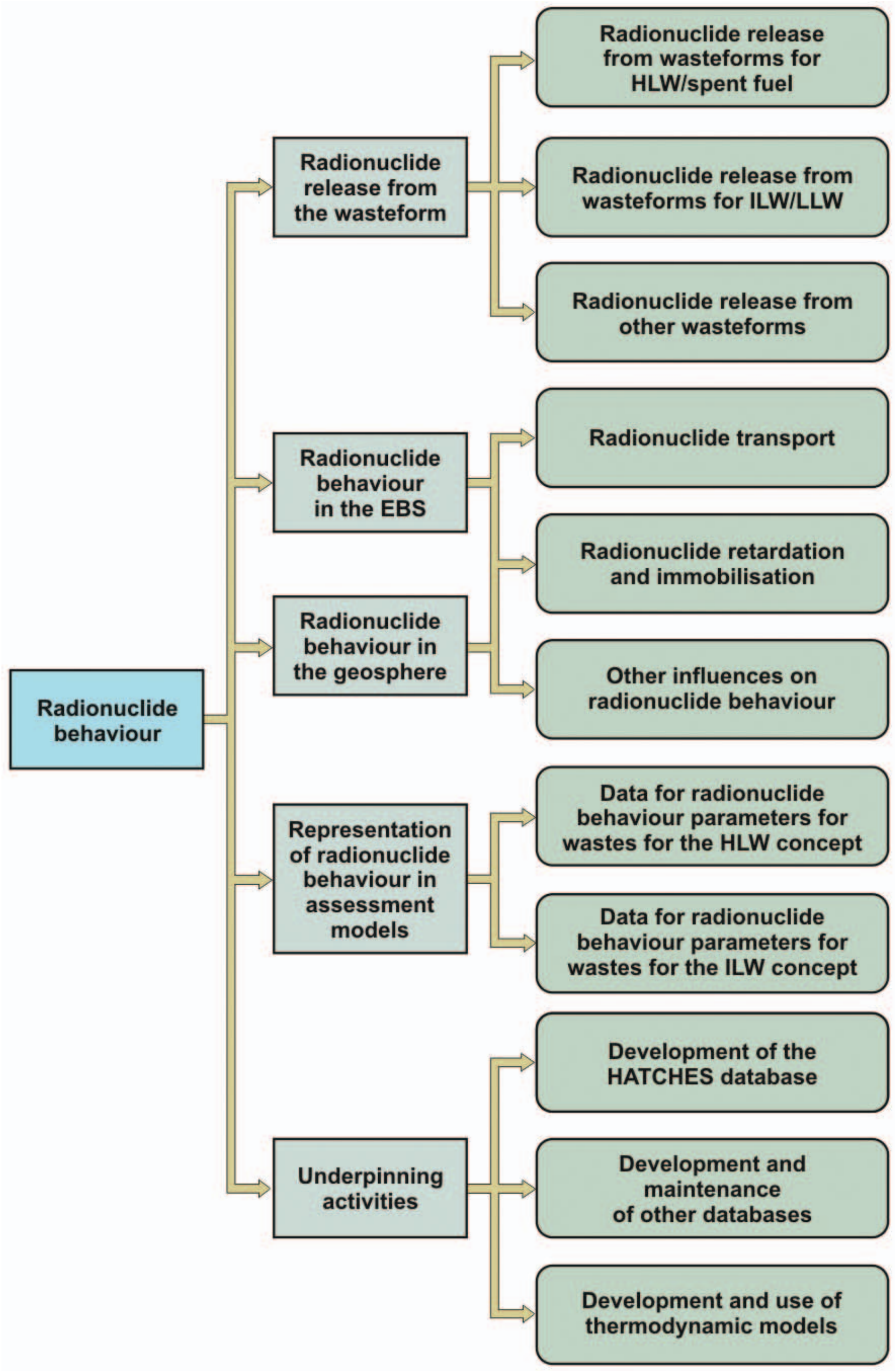

FIG. 2. Structure of the radionuclide behaviour research programme. Figure published with the permission of the NDA.

Immobilization by co-precipitation may be an additional containment mechanism, particularly in the alkaline disturbed zone (ADZ) around a cementitious GDF. In the geosphere, the key chemical processes contributing to containment are sorption onto the rock surfaces and precipitation or co-precipitation. Building understanding of the way in which natural groundwater tracers have migrated in the past helps the understanding of how radionuclides would behave in the future.

Areas for further research

Although radionuclide behaviour in bentonite-based buffers has not formed a significant part of the UK programme historically, it has been studied widely as part of overseas radioactive waste programmes (SKB, 2006; White *et al*., 2008).

Radionuclide sorption in cement-based materials used for grouts and/or backfills has been studied in the UK and overseas and there is an extensive dataset for radionuclide sorption onto cement-based materials (Heath *et al*., 1998; Baston *et al*., 1997). However, the majority of the measurements have been made using freshly prepared cements. Studies are planned to measure more sorption data on aged and leached cements.

The current status of R&D on radionuclide retardation and immobilization in the geosphere will be reviewed towards the end of the current preparatory studies phase of the programme. This will enable detailed plans to be prepared for the future site-specific phase of the R&D programme.

Other influences on radionuclide behaviour

There are a range of additional processes which may affect radionuclide behaviour within a GDF that are considered in the safety case.

Organic complexants

Organic material present in ILW (particularly cellulose) may degrade over time to produce a range of degradation products, some of which may form aqueous complexes with radionuclides in the waste (e.g. Humphries *et al*., 2010). These complexes could stabilize the radionuclides in solution, causing an increase in solubility and a reduction in sorption. Other considerations include cement additives, such as organic superplasticizers (Clacher and Cowper, 2011) and naturally occurring organics, such as hydrocellulose, lignin and humic acids (Heath and Williams, 2005).

Colloids

If there is a significant population of colloids, radionuclides could bind to the colloids and they could provide an additional transport pathway. Colloid stability, generation mechanisms and the ability of colloidal material to transport radionuclides all need to be understood in order to appropriately consider colloids in the safety case (Swanton *et al*., 2010).

Non aqueous phase liquids (NAPLs)

If there were sufficient NAPLs to form a separate NAPL phase in the GDF, radionuclides could partition into the NAPL phase and this could provide an additional transport pathway (Wealthall, 2002).

Microbes

All potentially suitable geological settings will have an indigenous microbial ecosystem. Microbial activity in the geosphere, as in any environment is generally located on chemical or physical interfaces, usually within biofilms. The impacts can be both physical (e.g. altering porosity) and/or chemical (e.g. changing sorption behaviour).

Areas for further research

There are a number of ongoing or planned NDA RWMD research projects to improve understanding and gather additional data, to inform future decisions about how to represent these processes in the safety case. These projects include:

(1) characterizing the degradation products from a range of cellulosic materials and measuring the impact on solubility and sorption in the near field and the geosphere (Randall *et al*., 2012);

(2) understanding the impact of commercial superplasticizers on radionuclide behaviour (e.g. Hayes *et al*., 2012);

(3) studying actinide chemistry in high pH colloidal ternary systems (e.g. BELBaR, bentonite erosion: effects on the long term performance of the engineered barrier and radionuclide transport: www.skb.se/lagerbladet____33716.aspx);

(4) studying the potential for plastics, such as PVC, to degrade to form NAPLs (e.g. Dawson, 2012; Baston and Dawson, 2012);

(5) developing the capability to model multi-phase flow, including modelling any potential NAPL migration (Watson *et al*., 2012).

The impact of microbes, naturally occurring colloids and naturally occurring organic materials is site specific and therefore these aspects may require significant experimental programmes in the site-specific phase of the programme.

Representation of radionuclide behaviour in assessment models

This area considers how radionuclide behaviour is represented in safety assessments. The rate at which radionuclides enter the groundwater; radionuclide accessibility to the pore space in certain barrier materials; solubility limitation of radionuclides in the GDF; perturbations to sorption and solubility from organic degradation products; radionuclide retardation provided by the barrier materials of the GDF; and an assessment of chemo-toxic materials need to be included.

Areas for further research

An important component of RWMD's R&D programme on radionuclide behaviour is the provision of data for safety assessment calculations. The overall approach to treatment of uncertainty and data elicitation will be reviewed. This will improve consistency in the treatment of uncertainty across different materials and concepts and improved documentation of the justification for parameter selection. The focus will be on key radionuclides which are known to be particularly important for post-closure safety. For example, report on data ranges for carbon was published recently (Small *et al*., 2011). Once site-specific information is available, datasets will be developed for the specific concept and geochemical conditions of the candidate site(s).

Development and maintenance of databases and thermodynamic models

Many assessment tasks and research modelling activities require the use of thermodynamic data which is comprehensive, internally consistent, internationally recognized and quality-assured. This work is to provide a fully referenced source of thermodynamic data, with a transparent audit trail of updates to the database(s), for use by NDA RWMD. Currently this data is held within the *HATCHES* database (www.sercoassurance.com/hatches/main.htm) which was developed by Nirex and contains information on the aqueous chemistry of key radionuclides. As part of this development NDA RWMD contributes to a multinational effort under the auspices of the Nuclear Energy Agency (NEA) to produce high-quality, peer-reviewed, internally consistent datasets for elements of interest in the geological disposal of radioactive waste (www.oecd-nea.org/dbtdb/info/publications).

Thermodynamic models are important for the interpretation of observations of radionuclide behaviour. Models of radionuclide solubility and sorption are used extensively to build confidence in understanding of radionuclide behaviour.

Areas for further research

The RWMD is reviewing its approach to the management of the quality and auditability of the data for use in assessment studies. This includes reviewing the approach to thermodynamic databases (Nuclear Decommissioning Authority, 2009). An alternative approach to that of developing the *HATCHES* database could involve joint development of a thermodynamic database with another waste management organization.

To ensure that NDA RWMD's approach is in line with current thinking, RWMD part-funded the NEA sorption project (www.nea.fr/jointproj/sorption.html), which studies the potential of chemical thermodynamic models for improving representation of sorption phenomena in the long-term safety analysis of radioactive waste repositories. Following on from this project; NDA RWMD will undertake further studies to improve its models.

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