INVESTIGATING THE IMPLICATIONS OF MANAGING DEPLETED NATURAL AND LOW ENRICHED URANIUM THROUGH GEOLOGICAL DISPOSAL

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An Integrated Project Team (IPT) comprising Radioactive Waste Management Limited (RWM) [a] *staff and members of the contractor base has been formed to work collaboratively on the topic of depleted, natural and low enriched uranium (DNLEU). This is referred to as the "uranium IPT". The project focuses on the disposability and associated full lifecycle implications of managing the UK inventory of DNLEU through geological disposal. The work of the uranium IPT is being carried out in two phases: Phase 1 (August 2012 to March 2013) involved planning and prioritising a programme of work to address knowledge gaps and advance understanding. This led to development of a strategic framework and project plan for Phase 2 (Ref. 1). Phase 2 (January 2013 to March 2016) involves implementing the planned activities, including review, integration and communication of the work. This paper summarises the answers provided by the project at the half-way point of the work programme and the forward programme.*

I. INTRODUCTION

DNLEU forms part of the UK inventory of higher activity materials set out in the Government's 2008 White Paper on Managing Radioactive Waste Safely[2] and the subsequent 2014 White Paper on Implementing Geological Disposal.[3] The great majority of UK DNLEU comprises uranic materials produced in the UK thermal reactor fuel cycle: these materials are currently in the form of depleted uranium (DU) hexafluoride (UF_6) tails – from uranium enrichment operations – and uranium trioxide (UO_3) – from reprocessing of spent nuclear fuels. The DU tails are mainly owned by URENCO UK Limited (UUK) and the reprocessing uranium is mainly owned by the Nuclear Decommissioning Authority (NDA) and EDF Energy. The Ministry of Defence owns relatively small quantities of DNLEU, and there are relatively small quantities of foreign-owned DNLEU in the UK. The most recent publicly available estimate of the total quantity (stocks plus arisings) of UK-owned DNLEU from civil nuclear operations is that reported in the 2013 UK Radioactive Waste Inventory (UKRWI), amounting to ~170,000 tonnes uranium (tU).[4]

The current UK Government strategy is for DNLEU to be safely and securely stored indefinitely in existing (or replacement as required) facilities. At present, the UK Government considers DNLEU to be a zero-value asset radioactive material, but Government strategy for long-term management of uranics materials is under review and a change could potentially cause this material to be reclassified as a higher activity radioactive waste.[5] If this were to happen, then DNLEU could need to be disposed of in a geological disposal facility (GDF). Important considerations regarding the potential disposal of DNLEU derive from the following observations:

1. DNLEU represents a significant fraction (~17%) of the total volume of packaged UK higher activity materials considered as a reference case in the NDA's 2010 generic disposal system safety case (DSSC) [6] (based on data in the 2007 UKRWI). However, DNLEU represents much less than 1% of the total radioactivity in this mass of materials at the assumed disposal date of 2040. The primary constituent of DNLEU is the uranium isotope U-238, which has an extremely long half-life (~4.5 billion years). Over the first 10,000-100,000 years, DNLEU has a relatively low radioactivity and radiotoxicity compared to other waste streams considered in the generic DSSC. However the radioactivity and radiotoxicity of DNLEU both increase by about an order of magnitude over a period of one million years because of the ingrowth of uranium daughter isotopes such as Ra-226 that were removed by enrichment and reprocessing. In contrast, the radioactivity of other components of the inventory decrease by three to four orders of magnitude over this timescale.

[a] RWM was established on 1 April 2014 as a wholly-owned subsidiary of the Nuclear Decommissioning Authority (NDA). The role of RWM is to implement the UK Government's policy of geological disposal of higher activity radioactive wastes by delivering a geological disposal facility (GDF) and by providing independent assessment of packaging of higher activity waste such that it is suitable for interim storage and eventual disposal in a GDF. Previously, RWM had been a part of the NDA – the Radioactive Waste Management Directorate (RWMD).

2. Conceptual understanding of the nuclear fuel cycle and the change in radioactivity and radiotoxicity with time of different waste streams lead to the conclusion that DNLEU (U-238 and daughters) becomes the most radiologically significant component of the UKRWI in the very far future. This is borne out by calculations based on the reference illustrative disposal concept presented in the 2010 generic DSSC; these calculations indicate that, in comparison to other components of the UKRWI, DNLEU becomes the largest contributor to calculated risk at timescales beyond 100,000 years. To provide context, the calculated peak mean risks are of the order of 10^{-6} per year, which is just consistent with the UK regulatory risk guidance level (RGL) for the post-closure period. The RGL is equivalent to about 1% of the risk from exposure to the average annual background radiation dose in the UK.
3. RWM's current planning assumptions for packaging, transport and disposal of DNLEU – as set out in the 2010 generic DSSC – are based on conversion of DNLEU to the form triuranium octoxide (U_3O_8), grout encapsulation into 500-litre drums, transport to a GDF in Type B containers, and disposal as remote-handled intermediate-level waste (ILW). The 2010 generic DSSC was the first attempt at an integrated consideration of disposal of DNLEU in the UK, and elements of this approach are non-optimal, as acknowledged in the generic DSSC itself.

The issues involved in improving understanding of the management of DNLEU via geological disposal cut across most RWM functions (e.g. disposal system specification, safety assessment, design, research, advice on waste conditioning and packaging, regulatory dialogue, stakeholder engagement). The main objective of the uranium IPT is for RWM staff and contractors to identify and address the issues associated with the potential need to dispose of DNLEU in a GDF, in order to inform wider decision-making on the long-term management of UK DNLEU. The strategic framework and project plan for Phase 2 of the uranium IPT work programme is illustrated in Fig. 1.

Key technical issues being considered by the uranium IPT relate to the following set of points:

1. The need for an improved **understanding of the UK DNLEU inventory**:
   - What is the inventory potentially requiring geological disposal?

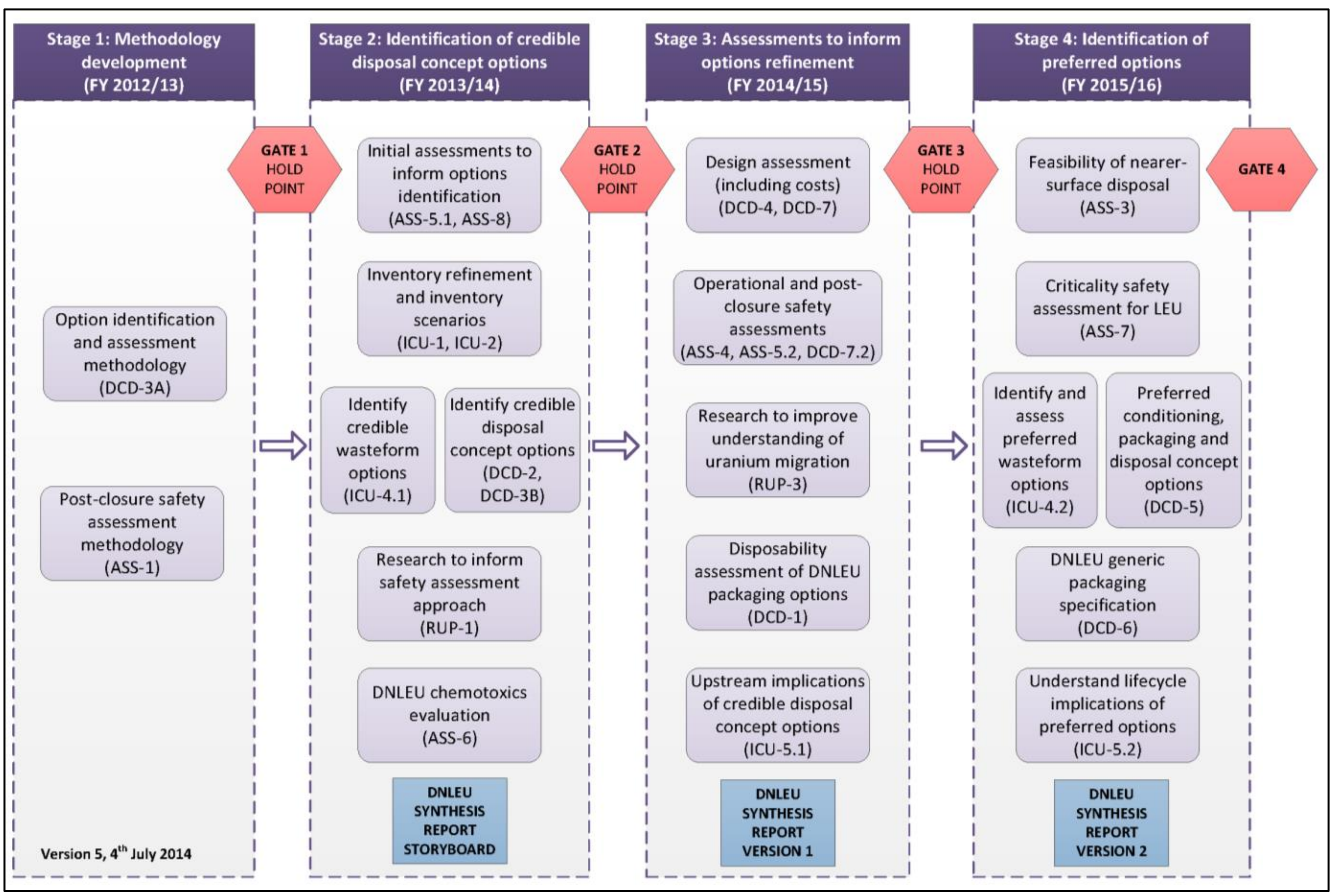

Fig. 1. Strategic framework and project plan for Phase 2 uranium IPT activities. Task reference numbers are provided in parentheses. Phase 2 also includes a number of integration activities not shown in the figure (see Ref. 1).

- What is the extent of contamination of this inventory by actinides, fission products and organic materials, and what are the safety implications?
- How much DNLEU could be used to realise benefits in a GDF (e.g. in place of backfill materials in other parts of a GDF)?

2. The identification of **preferred chemical and physical form(s), and preferred packaging and disposal concepts** for DNLEU in a GDF:
   - Can the conditioning and packaging assumptions made in the 2010 generic DSSC be improved from the standpoint of transport and operational safety? In particular, would it be feasible to transport and dispose of DNLEU in the form (powdered U_3O_8 and UO_3) and packaging (DV-70 containers,[b] 200-litre and 50-litre steel drums) considered acceptable for long-term storage?
   - What are the preferred conditioning, packaging and disposal concepts?
   - What are the upstream (conditioning and packaging) lifecycle implications of the preferred DNLEU disposal concepts?
3. The approach to **safety assessment of DNLEU** disposal:
   - Given that the ingrowth of daughter radionuclides leads to increasing radioactivity over a period of about one million years, what is the most appropriate approach to assessing long-term safety, given the very long half-life of U-238 (and some daughter radionuclides), which occurs naturally?
   - Is there a better approach to the treatment of uncertainty in the generic DSSC, particularly with regard to assumptions that underpin operational safety assessments and radionuclide migration parameters in the post-closure period?
   - Would disposal in a nearer-surface facility be feasible for any part of the inventory, that is, disposal of DNLEU at depths below which major human intrusion (i.e. wholesale excavation of waste) can be ruled out?
   - Are assessments that focus on radiological safety of DNLEU sufficiently bounding in terms of the chemotoxicity of uranium?
   - Can criticality safety of the LEU component be demonstrated for possible wasteforms, waste packages and disposal concepts?

International safeguards will apply to some of the inventory (i.e. that arising from the civil use of uranium), and inspection requirements could be a challenge to implement once material is disposed of in a GDF. However, this issue is not specific to the geological disposal of DNLEU alone, and is not being considered within this project.

This paper summarises a review of DNLEU management practices worldwide,[7] and key findings of the work conducted by the uranium IPT at the halfway point of the project.[8] The international review was conducted to provide context for the work in the UK.

II. INTERNATIONAL CONTEXT

Worldwide DNLEU stocks arise mainly from uranium enrichment operations, with significant quantities also arising from reprocessing activities, particularly in France and the UK. France, Russia and the US hold by far the largest stocks of DU. DNLEU stocks held by these countries significantly exceed UK stocks. Germany also holds significant stocks of DU, with other countries having relatively small inventories.

In other countries – with the notable exception of the US – DNLEU is considered to be a current or potential resource, and it is held in storage with this in mind. Some countries reuse uranium derived from reprocessing within fuel for nuclear power plants, either as conventional uranium-oxide fuel or within mixed-oxide fuel. This is the case in Belgium, France, Germany, India, the Netherlands, Sweden and Switzerland. The lower the burn-up of fuel in a reactor, the easier it is to reuse the reprocessed uranium. The trend for many years has been towards higher enriched fuel and higher burn ups; hence, it is historic stocks that have most potential for reuse in the nuclear fuel cycle.

Contaminant levels in overseas stocks of uranium derived from reprocessing do not appear to preclude their reuse in new nuclear fuel, nor do they present insurmountable issues from an operational safety perspective. An alternative use of DNLEU could be as a component of fuel for fast breeder reactors; however, this would require development of an industrial-scale fast breeder fuel cycle, for which there is currently little appetite.[c] Note that re-enrichment of DU tails or reprocessed uranium and recycling into a thermal reactor fuel cycle still leaves almost all of the uranium as secondary tails or enrichment tails. In contrast, recycling DU into a fast reactor fuel cycle has the potential to consume part of the uranium over the very long term, which could change the political view, i.e. DU could be seen as a valuable asset rather than as a potential waste. Nonetheless, given the large existing stockpiles and planned production of DNLEU, and the limited ability of

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[b] A DV-70 container is a painted mild steel box, approximately 3 m^3 in volume.

[c] For example, the UK stopped research into fast breeder reactors at Dounreay in the 1990s.

even a fast reactor fuel cycle to consume DU, some of this material may eventually become waste even in those countries that still consider it to be a potential resource.

The US is the only country in which some DNLEU is currently managed as a waste. The US Nuclear Regulatory Commission classifies DU as low-level waste (LLW), and regulatory policy providing for disposal of DU in near-surface facilities is under development.[d] However, waste classification schemes, regulatory requirements, and the available physical environments for near-surface disposal differ significantly between the UK and the US – all of which affect the value of knowledge transfer between the two programmes.

DU would not be classified as LLW in the UK, because it exceeds the UK LLW limit for long-lived alpha activity of 4 giga-Becquerels (10^9 Becquerels) per tonne by about a factor of three. It is therefore classified in the UK as a higher activity material. This does not, however, mean near-surface disposal of DNLEU would be precluded in the UK. The Phase 2 work programme includes a task to assess the feasibility of nearer-surface disposal of DNLEU and to evaluate the minimum depth of overburden that could be needed to safely dispose of such wastes, considering UK regulatory guidance. This option is worth investigating because disposal in a nearer-surface facility opens the option for a purpose-built facility at shallower depth with the potential for significant cost and time savings compared to disposal in a deeper GDF.

There is relatively little published experience from overseas that is directly transferable to geological disposal concept development for DNLEU in the UK:

- In Germany an assessment has been conducted of the safety of geological disposal of significant quantities of DU (in a salt dome at Gorleben).[9] The work shows that volumes on the order of 100,000 m^3 could be disposed of safely in a low-permeability evaporite host rock; however, there is limited published information available on the details of the disposal concept considered or the assessment approach. Conduct of this assessment suggests that the German government is considering the implications of classifying some DU as waste.
- Consideration has been given in older US research projects to alternative (beneficial) uses of DU in a GDF for spent nuclear fuel.[10] Although these studies indicated some potential benefits to the use of DU in geological disposal, as noted above DU is now classified as LLW in the US and geological disposal of this material is no longer being seriously considered.

[d] So far only relatively limited quantities of DU have been disposed of to near-surface disposal facilities in the US.

U_3O_8 is generally regarded as the preferred chemical form for long-term storage of UF_6 tails, where no immediate use has been identified, and reprocessed uranium is generally held in the form UO_3. DNLEU can be disposed of as a range of chemical oxide forms, e.g. U_3O_8 or UO_3 as in the US. UF_6 is not generally considered to be a suitable chemical form for disposal because the fluorine can be extracted and has value as a resource, and because its chemical form and toxicity may give rise to safety concerns.

Approaches to packaging DNLEU for storage are similar across different countries. UF_6 is typically stored in steel cylinders. Deconverted U_3O_8 and UO_3 from reprocessing are typically stored in steel containers. DV-70 containers are used to store U_3O_8 in France, Germany, the Netherlands, and the UK (planned).

Transport of DNLEU stocks in a variety of chemical forms (including UF_6, liquid uranyl nitrate, and U_3O_8 and UO_3 powders) is routinely undertaken in many countries. An outer transport package is generally not required to meet transport regulations, except where older packages may have become non-compliant (e.g. owing to corrosion). DNLEU is usually classified as Low Specific Activity and Industrial Packages are used for its transport.

III. URANIUM IPT KEY FINDINGS TO DATE

III.A. The UK Inventory of DNLEU

As noted above, the projected inventory of DNLEU in the 2013 UKRWI is ~170,000 tU.[4] Nearly 40% of this is projected future arisings. About 35% of the DNLEU inventory is owned by the NDA (mainly from nuclear fuel reprocessing operations), and almost all of the rest is owned by UUK (from uranium enrichment operations).

The actual amount of DNLEU requiring disposal could range from being significantly less than this (if another use is found for the material) to many times this. In particular, the inventory could increase significantly if UUK carried on uranium enrichment activities beyond the end date of 2023 assumed in calculating DU arisings in the 2013 UKRWI. UUK expects its future nuclear fuel production operations at Capenhurst to generate approximately 5,000 tU per year of depleted UF_6 tails. Because URENCO supplies a worldwide market, its decision making on continued enrichment activities in the UK is partly independent of the future UK demands for enriched uranium.

It is likely that whatever happens to the inventory, at least some DNLEU will need to be disposed of to a GDF. The credible non-disposal nuclear-related options for long-term DNLEU management are unlikely to use more than a small part of the inventory on any reasonable timescale.

III.B. Conditioning and Packaging for Long-Term Storage and Transport

Previous UK work had suggested that operational safety (during fire or impact accidents) could be a concern if DNLEU were disposed of as an unconditioned powder[11] – leading to the concept in the 2010 generic DSSC of cement conditioning. However, work conducted by the uranium IPT demonstrates that it would be possible, in principle, to make an Operational Safety Case (OSC) for disposal of DNLEU in a powdered form, based on comparison of conservatisms in the NDA's 2010 generic OSC with OSCs for long-term storage of DU (in particular, that for URENCO's Tails Management Facility and Uranium Oxide Store). This means that disposal of DNLEU in a powdered form can be considered a credible option.

An outer transport package would almost certainly be required to ship DNLEU to a GDF after 50-100 years of storage. There is a range of widely used containers available for transport of DNLEU to a GDF that would offer significant cost savings compared to RWM's current reference transport container for DNLEU.[12] The transport packaging could be reusable or could be disposed of directly. This provides a range of transport packaging to consider in developing disposal concept options.

III.C. Credible Geological Disposal Concept Options

In identifying and evaluating geological disposal concept options, the uranium IPT is considering the implications of the options for both RWM (at a GDF) and for DNLEU owners (upstream, at the storage sites). Consideration of the implications at a GDF is based on the assumption of a single GDF designed for disposal of all UK higher activity waste types (consistent with UK Government policy[3]). The upstream implications include such issues as chemical conversion, conditioning and packaging costs, and safety and environmental impacts.

A representative set of credible vault-based geological disposal concept options for DNLEU has been identified for further evaluation:

- The set includes four variants of disposal of DNLEU in its existing or planned form and packages for long-term storage; these concept options avoid the expense and hazard associated with repackaging of the existing oxide powders. In three of these concepts, the storage containers are packaged in an outer container for both transport and disposal, while the fourth concept dispenses with the outer container for disposal and makes use of a reusable transport package. In two of the three concepts that include an outer container for both transport and disposal, relatively simple outer packages (steel or soft-sided packages) are considered to have the potential to offer some operational benefits in terms of reduced handling, improved accident performance, duration of integrity of the waste package, and operability from standardisation of waste packages. The third such concept uses a more robust outer container with the aim of providing future retrievability in a GDF.
- A fifth concept option includes a briquetted wasteform (UO_2) and long-lived packaging (copper outer container). This option has been identified in order to explore the opportunities for volume reduction and the advantages and disadvantages of a disposal option that provides for a significantly longer containment period in a GDF.
- A sixth option is the one considered in the 2010 generic DSSC (see Section I), which is retained for further consideration and comparison.

In addition to DNLEU-specific vault-based disposal concepts, there are several ways in which it would be theoretically possible to beneficially "reuse" the inventory of DNLEU many times over in other parts of a GDF, e.g. as mass backfill or as part of structural components. The most promising re-use option is considered to be disposal of containerised DNLEU in the service and transport tunnels for other waste types (e.g. in place of some mass backfill in the disposal module for high-level waste and spent fuel), and this is the only re-use option being assessed further.

III.D. PCSA Methodology for Long Timescales

RWM's post-closure safety assessment (PCSA) methodology has continued to be refined, to highlight the central place of a narrative of disposal system evolution. This narrative is supported by a range of safety arguments based on reasoned arguments, deterministic calculations, and probabilistic calculations restricted to timescales when it is meaningful to treat uncertainty using probability density functions (PDFs). As shown in Fig. 2, three periods in GDF evolution, derived from an analysis of system uncertainties, could be used to define an appropriate timescale for the probabilistic calculations important in evaluating compliance with the environmental regulators' risk guidance level for the post-closure period. The duration of these periods could differ between waste types, disposal concepts, and geological environments. Different types of assessment calculations and argument are required to appropriately consider these periods. Fig. 2 shows indicative durations for the three periods based on a generic disposal concept for DNLEU in higher strength rock and a generic understanding of site characteristics in the UK.

The kinds of reasoned arguments and comparisons with natural systems that could be used to help build a safety case for geological disposal have also been summarised, focusing particularly on the very long timescales relevant to DNLEU. The complementary safety arguments fall into three categories, the first two of which roughly correspond to timeframes based on stability and extent of uncertainty associated with site-specific geosphere properties:

1. *Arguments for the geological barrier*. This category applies for the period after about 100,000 years up to about 1 Ma, when the uncertainty about the evolution of the engineered barrier system (EBS) for DNLEU disposal concepts may have increased to the extent that claiming benefit for its continuing barrier function may be difficult to justify. At this time, depending on the site and disposal concept, the geosphere will become the most important barrier ensuring continued isolation and containment of the wastes. In this timeframe, the properties of the geosphere are largely stable or change in a reasonably quantifiable manner in response to climatic change that results in periods of glacial or peri-glacial conditions.
2. *Arguments for continuing safety*. This category applies to the period after more than about 1 Ma, when large-scale geological processes such as uplift, erosion and tectonics could have significantly affected geosphere properties. Quantitative arguments are more difficult to justify owing to increasing uncertainty although, in very stable geological environments, robust quantitative arguments might be possible for several Ma.
3. *Comparisons to 'acceptable practices'*. This category comprises arguments based on comparison of disposal to acceptable practices in other industries dealing with radioactive materials (e.g. naturally occurring radioactive materials, or radioactive by-products produced during power generation).

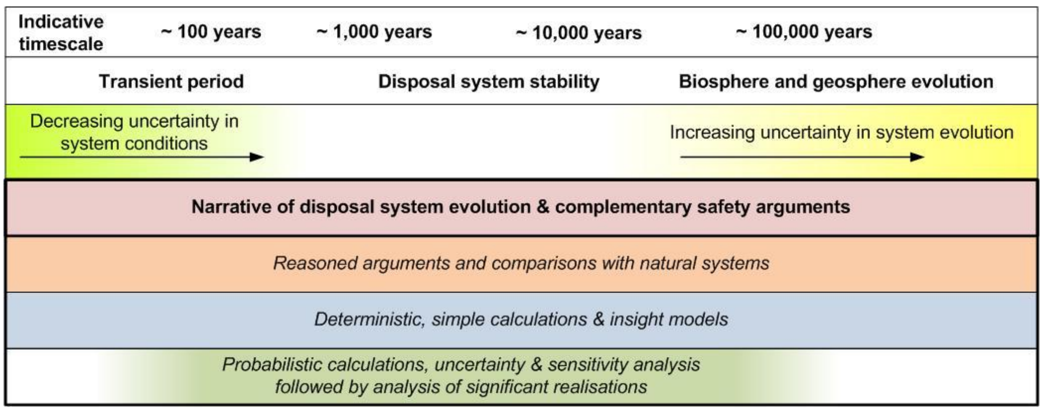

Fig. 2. Illustration of the PCSA methodology proposed to deal with the issue of communicating safety over long timescales.

III.E. Key PCSA Uncertainties

The uranium IPT has been reviewing the geochemical uncertainties that most affect calculations of post-closure safety.[13,14,15] Selected conclusions are as follows:

- Analysis of the 2010 generic PCSA illustrative risk calculations has shown that a relatively small number of simulations that sample from the tails of the PDFs for geochemical parameters (near-field solubility, far-field sorption) significantly affect the outcome of the calculations.[e] These PDFs were consistent with understanding of geochemical uncertainties at the time. If improved geochemical understanding could reduce uncertainties and, in particular, the extent of the tails of the PDFs, the calculated peak risks from disposal of DNLEU might also reduce.
- Consideration of specific near-field conditions illustrates the potential implications that knowledge of a specific site may have on post-closure performance assessment calculations. Natural and industrial analogues imply low solubility for uranium and its daughters under a range of relevant near-field conditions.
- Uranium series radionuclides are already present in EBS materials and in the geological environment (host rock and groundwaters), and the natural uranium concentrations in some groundwaters already exceed the solubility of amorphous uranium dioxide (UO_2). Depending on site-specific groundwater chemistry, any uranium series radionuclides derived from a GDF could be incremental to those in the EBS and natural system, and might be unlikely to migrate far before re-precipitating owing to solubility limitation. This means that there may be a need in PCSA models of the geosphere to consider geochemical processes other than reversible sorption (e.g. solubility limitation). In practice, the use of simplified PCSA representations based on the use of equilibrium

[e] The volume flux of groundwater through the disposal vaults was also a key control on calculated doses reported in the 2010 generic PCSA.

distribution coefficients (K_d concept) may be under-conservative or over-conservative, depending on assessment timeframes and site-specific groundwater chemistry.

The conceptual model for uranium migration in a geological disposal system consists of a series of transitions, as illustrated in Fig. 3. There is a transition from near-field conditions (on the left) through to adjacent host rock, and then further through the far field comprising, as an example, more permeable rock ('aquifer'), and a final transition to the biosphere ('shallow bedrock & soil'). There are four alternative chemical states for the near field: high pH if cementitious backfill is used, otherwise neutral pH, and reducing or oxidising redox depending on the influence of oxidised uranium in the wasteform.

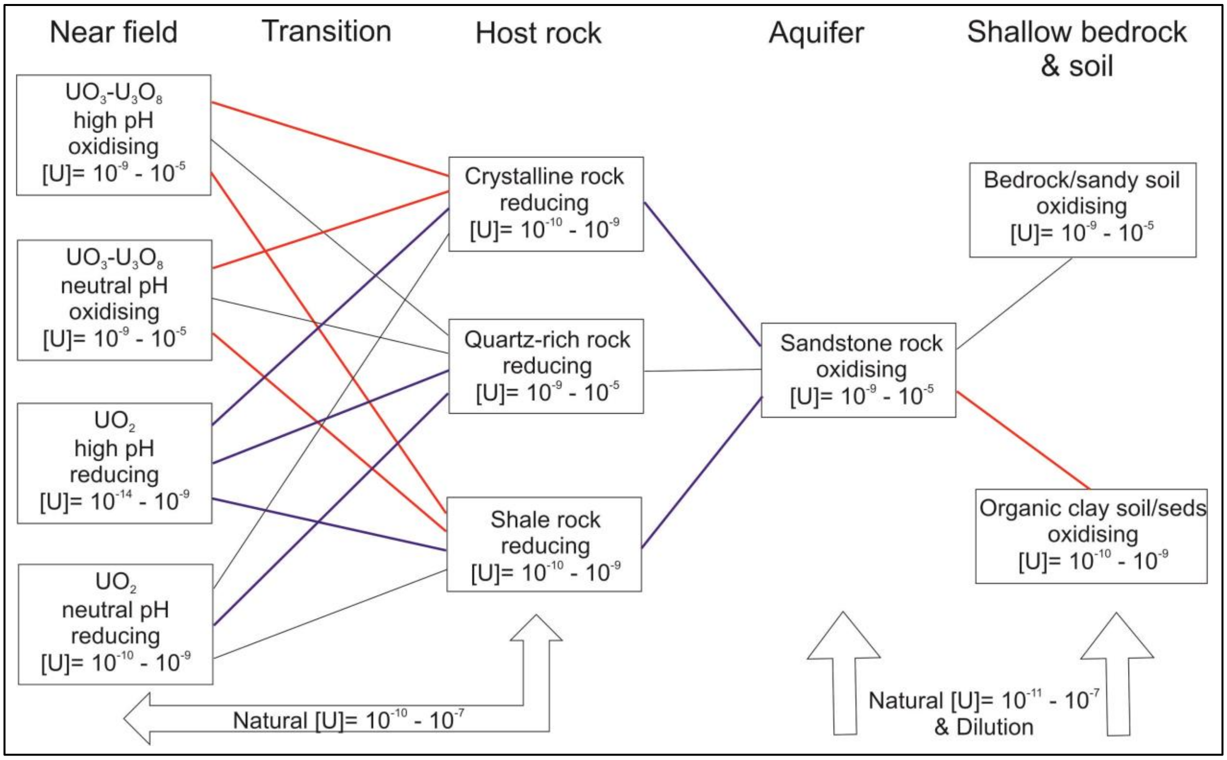

Fig. 3. Schematic illustration of alternative evolution of uranium solubility through a series of transitions in a geological disposal system. Lines connecting alternative conditions for each of the schematic compartments indicate the potential changes of uranium solubility (indicative order of magnitude values in mol.dm^{-3}): red = precipitation, blue = dissolution, black = no change. Typical concentration ranges for naturally occurring uranium in groundwaters in deep and shallow parts of the far field are shown at the bottom of the diagram.

Bounding calculations for the post-closure period show that long-lived radioactive contaminants in DNLEU (e.g. Tc-99, Np-237 and U-236 arising from reprocessing) do not contribute significantly to dose. This means that no further investigation of radioactive contaminants in DNLEU is needed from a post-closure performance perspective within this project. However, the U-232 content of some LEU from reprocessing will need to be considered in selecting suitably shielded transport and disposal containers, to ensure safety during transport and GDF operations.

III.F. Uranium Chemotoxicity

Biokinetic modelling of uranium chemotoxicity indicates that in the post-closure period, the radiological impact of uranium is likely to be more limiting in terms of safety.[16] However, during the operational period, separate assessment of chemotoxicity under accident conditions (acute exposures) would need to be considered, particularly for more rapidly adsorbing uranium compounds (e.g. U_3O_8), and acute chemotoxic effects on workers could be of greater concern than the radiological impacts.

In terms of synergistic effects, the radiological impact of uranium is of little significance in influencing its chemotoxicity but, as uranium is known to be a chemical mutagen, its chemical toxicity may influence its radiological impact in some contexts. These potential effects have not been considered in the project, but are not expected to be important at anticipated maximum exposure levels.

IV. CONCLUSIONS

The findings of work to date have been used to update the project plan for subsequent project stages, as shown in Fig. 1. The work of the uranium IPT is continuing to advance, and the findings summarised in this paper will be updated at the conclusion of Phase 2 activities. By working to better understand the implications of long-term management of DNLEU through geological disposal, the uranium IPT has provided value to RWM's wider activities, including the approach to disposal concept optioneering, the development of safety assessment approaches, and the understanding of uranium and daughter geochemistry and uranium chemotoxicity. Significant value is still to be provided, including:

- The development of more informed decisions on the preferred disposal concept(s) for DNLEU to take forward into formal change control and future updates of the DSSC.
- The development of a generic Packaging Specification for DNLEU, which will provide guidance to DNLEU owners on acceptable packaging approaches for disposal.
- Further development of safety assessment methods that will be of wider use across RWM's activities.

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