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An introduction to package evolution and criticality research studies relevant to the UK disposal programme

C. PADOVANI*, S. J. WILLIAMS AND P. WOOD

Nuclear Decommissioning Authority, Radioactive Waste Management Directorate, NDA Harwell Office, Building 587, Curie Avenue, Harwell, Didcot, Oxfordshire OX11 0RH, UK

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ABSTRACT

In the UK, radioactive wastes currently planned for disposal in a geological disposal facility (GDF) include intermediate level waste, some low level waste and high level waste. Disposal of other materials, including spent fuel, uranium and plutonium is also being evaluated to inform the safety case for a GDF, if such materials were to be classified as wastes in the future. This paper describes the generic safety functions through which waste packages can contribute to the safety case of a GDF in the UK. It describes the engineering approach used or envisaged, in the UK and internationally, to ensure that waste packages retain their safety functions for the required periods of time and summarizes the scientific basis underpinning the current understanding of relevant evolution processes. Where gaps in the knowledge exist, the Nuclear Decommissioning Authority Radioactive Waste Management Directorate has identified specific research activities needed to close out such gaps to a level of maturity sufficient for this stage of the disposal programme (generic). This paper describes the latest results from their R&D programme and presents a summary of the research activities planned to meet the current needs of the disposal programme with specific reference to the topics of package evolution and criticality safety.

KEYWORDS: package evolution, wasteform, waste container, criticality.

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).

There are a variety of materials that may require disposal in a geological disposal facility (GDF), including intermediate level waste (ILW), high level waste (HLW) and potentially spent fuel (SF) uranium and plutonium. Wasteforms and waste containers[1] for different types of wastes may be considerably different. Some packages contain fissile material in relatively limited amounts (ILW). If destined for disposal, packages

[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 good 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 (as described below) the wasteform and the waste container contribute to achieve important safety functions of the waste package.

* E-mail: cristiano.padovani@nda.gov.uk
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containing materials such as spent fuel, uranium and plutonium would contain significant amount of fissile materials. Packages containing fissile materials need to be specifically designed and manufactured to take into account the potential for a *nuclear criticality*.

This paper describes aspects of the safety cases for geological disposal and the relevant underpinning engineering and scientific basis for two separate topics considered in the UK radioactive waste disposal programme: package evolution and criticality safety. This short paper is based on the information published in (Nuclear Decommissioning Authority, 2010*a*,*b*, 2011), updated to reflect recent developments.

The characteristics of waste packages are important during all periods of waste management, from interim storage to final disposal in a GDF (Nuclear Decommissioning Authority, 2010*c*,*d*,*e*). Waste packages need to provide specific *waste package safety functions* during different periods (storage, transport, operations and post-closure) dependent on the type of waste and disposal concept. In the current stage of the disposal programme, the NDA RWMD identified a set of *generic safety functions* for waste packages (Nuclear Decommissioning Authority, 2010*a*), as illustrated in Fig. 1. A specific safety function of waste packages is to ensure *criticality safety* preceding and following geological disposal (Nuclear Decommissioning Authority, 2010*b*).

Given current estimates of when a GDF in the UK may become available and considering the long nature of processes of radioactive decay, the length of time for which the safety functions of waste packages have to be retained represents a particular technical challenge to the disposal

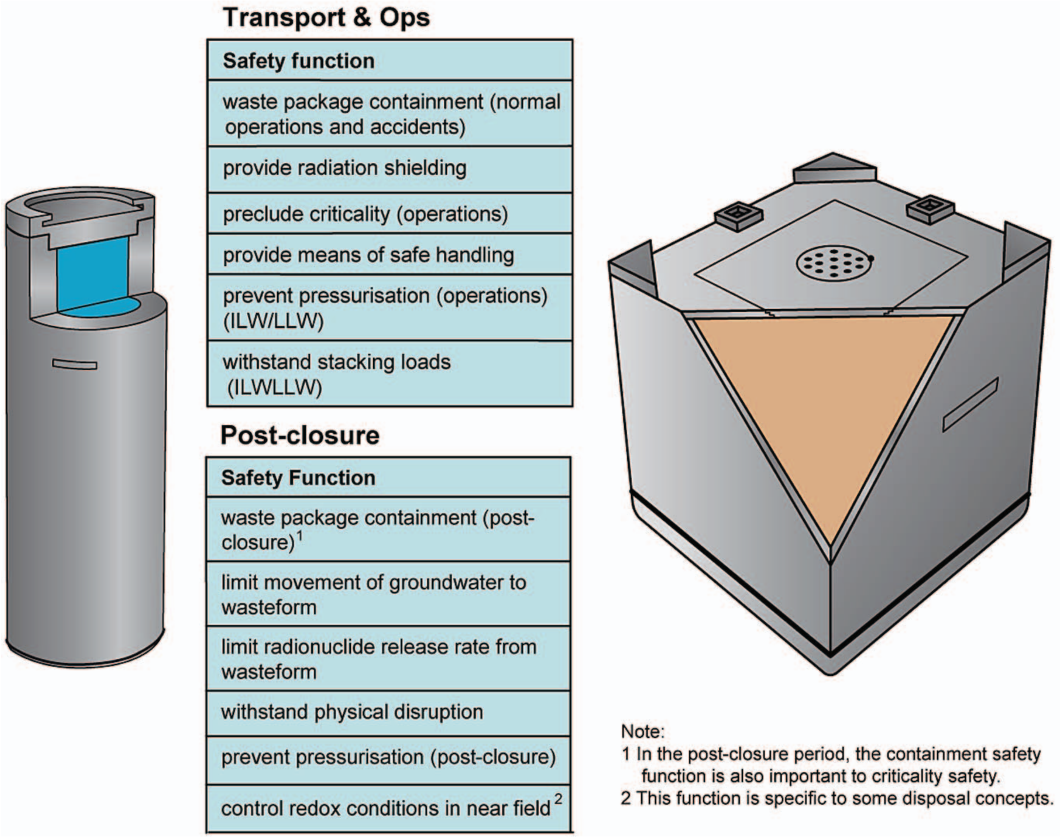

FIG. 1. Schematic representation of the generic waste package safety functions during the transport/operational and post-closure period identified in the generic disposal system safety case. Published with the permission of the NDA.

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programme. For some wastes (mainly ILW), periods of interim storage and in unbackfilled disposal vaults may be of the order of a hundred years or longer. For other potential wastes (e.g. spent fuel), the period of interim storage inside waste containers designed for final disposal may be significantly shorter but post-closure periods in which waste packages will have to maintain their safety functions can be extremely long, from thousands to hundreds of thousands of years. In order to retain their safety functions for the required periods, waste packages need to be designed to provide sufficient *durability*.

Engineering approach to meet the safety functions and underpinning scientific basis

Engineering approach to meet the safety functions

A sound evaluation of the nature of probable evolution processes and of their significance with respect to the safety functions forms the basis of the approach adopted in the UK and internationally to ensure that relevant waste package components are manufactured to achieve and retain their safety functions. A design principle underpinning the manufacture of waste packages is to design waste package components in a way that they are either: (1) able to tolerate change (for changes relatively slow in nature or mechanistically limited in amount); or (2) designed in a way that ensures that changes are very unlikely to occur (for changes difficult to account for in the design, or not limited in rate and extent) (Nuclear Decommissioning Authority, 2010*a*).

This may include, for example, designing waste containers with a corrosion allowance (so that any change is tolerable) (Nuclear Decommissioning Authority, 2010*a*), or setting specific limits for the amount of fissile material in a waste package, so that criticality cannot occur (Nuclear Decommissioning Authority, 2010*b*).

Designs of suitable engineering systems to ensure a benign chemical environment surrounding waste packages is also important, as the nature, rate and amount of change likely to occur in waste packages depends on their storage and disposal environments. For this reason, a design principle for modern storage facilities and for a GDF is to consider suitable means of ensuring adequate environmental conditions in contact with waste packages. Guidance on interim storage (Nuclear Decommissioning Authority, 2012) has recently become available through the work of an integrated project team (IPT) set up by the NDA. Such guidance may be also applicable to the case of a GDF but may need to be tailored to site-specific conditions.

Monitoring and inspection also play an important role in providing confidence in the durability of waste packages and in identifying any unexpected changes in time for corrective action to be taken. Techniques suitable to monitor package evolution in interim stores are being developed (Stanley, 2010).

Scientific basis underpinning the engineering approach: wasteform and container evolution

Given the long timescales over which the safety case for disposal needs to consider evolution processes, understanding and demonstrating the behaviour of packaging materials is a significant technical challenge. The technical approach adopted in the UK and internationally to achieve a robust evaluation of relevant evolution processes 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. Two examples are shown in Fig. 2. Significant amount of such work has been gathered in peer-reviewed scientific publications, including:

(1) For wasteform evolution: the *International Union of Laboratories and Experts in Construction Materials, Systems and Structures* (RILEM) workshop series (e.g. L'Hostis *et al*., 2009), the *Scientific Basis for Waste Management Proceedings* (Van Iseghem, 2006), *Proceedings of the Migration Conference* (e.g. Qaim *et al*., 2008).

(2) For container evolution: proceedings of the workshops on the prediction of corrosion damage in nuclear waste systems (e.g. Mansur *et al*., 2008; Kursten and Druyts, 2011) and the *Scientific Basis for Waste Management Proceedings* (e.g. Van Iseghem, 2006).

The understanding developed so far indicates that (Nuclear Decommissioning Authority, 2010*a*)[2]:

(1) The evolution of most ILW and relevant encapsulation materials within a waste package is unlikely to compromise the key characteristics of the wasteform over relevant timescales (a hundred years or longer). Most wastes can be immobilized effectively in cement, which has been used

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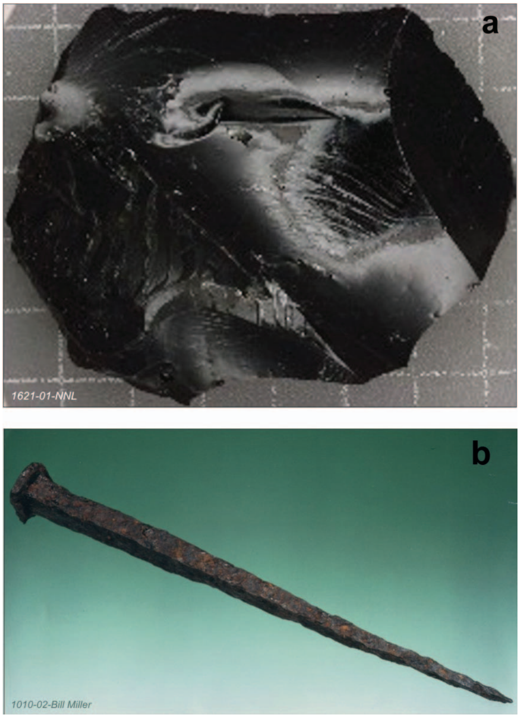

FIG. 2. Examples of materials that have been used in laboratory-based experiments and analogues studies to increase confidence in the understanding of long-term evolution processes; (*a*) sample of HLW glass simulant, courtesy of National Nuclear Laboratory; (*b*) Roman iron nail (almost 2000 years old), courtesy of Bill Miller.

successfully for more than two decades. Specific waste streams in which package evolution may negatively affect their durability are being identified. Such waste streams may not be suitable for encapsulation in cement and alternative options are being evaluated, including encapsulation in polymer or glass.

(2) Materials employed for ILW waste containers are very durable in appropriate conditions. Stainless steel has been extensively used in relevant applications for many years and experience with both the use of this material and relevant research and development show that, in appropriate environmental conditions (an interim store or underground vaults/tunnels with good environmental control), it is likely to show little degradation over relevant timescales (a hundred years or longer).

(3) Wasteforms for HLW and spent fuel are very durable in relevant environments (in contact with anoxic groundwater) and are likely to significantly limit the release of radionuclides in the near field once the waste container is breached.

(4) For waste containers that may be designed to contain HLW, and potentially SF, the required durability (from a thousand to hundreds of thousands of year) can be achieved with a variety of materials and designs. Durable containers can be manufactured either with very corrosion resistant materials or with materials which corrode in a slow and predictable fashion. In the latter case, thick-walled containers with a suitable corrosion allowance are required. With some materials (e.g. copper, titanium, nickel alloys) very long durability can be achieved.

Scientific basis underpinning the engineering approach: criticality safety

The approach adopted in the UK to ensure criticality safety of waste packages is to use well established methods with appropriate conservatisms to ensure sub-criticality during transport and operations. In some cases, methods used by waste producers to produce waste packages already stored under existing, approved, site safety cases can be used. Criticality safety is also assessed by the NDA RWMD as part of the assessment of post-closure performance of a GDF and associated radiological risk. The length of time over which evolution processes need to be considered represents a challenge in ensuring that criticality safety is appropriately considered in the safety case for disposal. Current understanding indicates that a criticality post-closure is unlikely, i.e. it is a low probability event. However with large numbers of packages and very long timescales it is difficult to guarantee that a criticality cannot occur. Therefore, research to understand how a criticality could begin, progress and end has been carried out. An example illustrating the type of scenario considered in criticality safety studies is shown in Fig. 3.

[2] The text below is extracted directly from (Nuclear Decommissioning Authority, 2010*a*). The text in brackets has been added for the sake of this work to give an indication of the environmental conditions in which waste packages are likely to operate and the length of times over which durability their key components (the wasteform and the waste container) is likely to be required. Such aspects are described in greater detail in (Nuclear Decommissioning Authority, 2010*a*).

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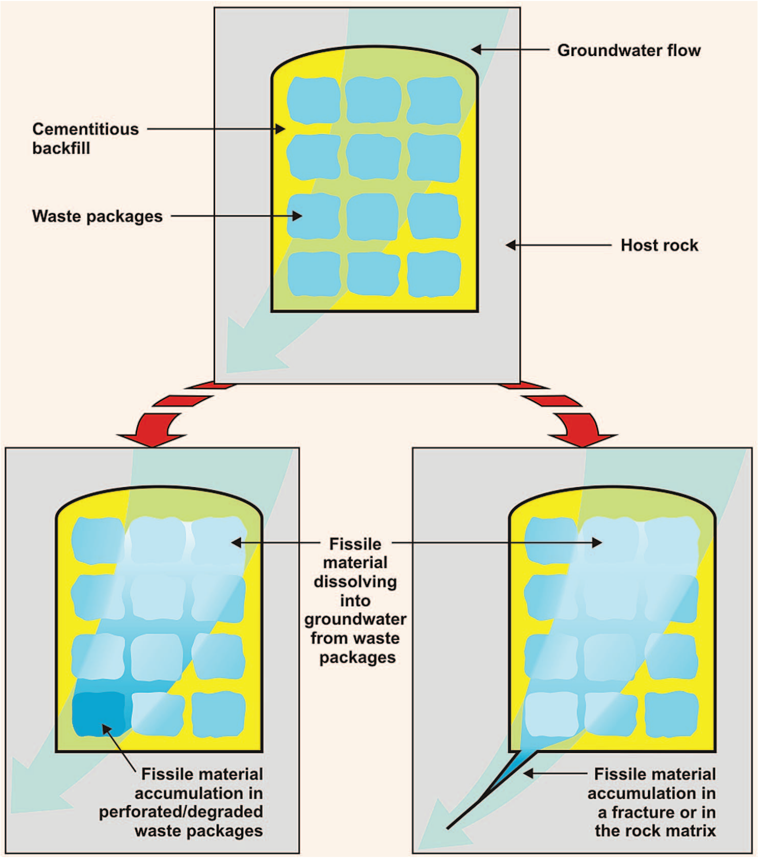

FIG. 3. Schematic illustration of potential evolution mechanism leading to the potential for a criticality in a GDF. Published with the permission of the NDA.

Relevant sources of information include the International Conference on Nuclear Criticality, held most recently in 2011 (Anonymous, 2011) and 2007 (Anonymous, 2007). The understanding developed so far indicates that (Nuclear Decommissioning Authority, 2010*b*):

(1) The likelihood of criticality inside a waste package is low because the fissile material will remain in a subcritical condition, either due to limits on package content or due to suitable design of the wasteform. Once in a GDF, most evolution processes are likely to make the system further subcritical. The potential effects of processes that may lead to increase in the likelihood of a criticality are mitigated and retarded through the use of multiple barriers.

(2) If it were to occur, the consequence of criticality once waste packages are in a GDF would be low because of direct shielding by the host rock and because the buffer/backfill would still isolate the radioactive waste from the surface environment. Criticality events would only affect a limited part of the GDF volume. Criticality events on a larger scale, if they were to occur, would only occur a long time after closure.

Relevant work planned in the NDA RWMD research and development programme

Wasteform evolution

Important work in this topic includes evaluating the characteristics (e.g. the dissolution rate) of UK spent fuels and HLW glasses, refining the understanding of the evolution of potentially challenging cement-based wasteforms, progressing understanding of suitable alternative encapsulants for ILW (e.g. polymers, glasses) and evaluating the characteristics of other potential

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wasteforms (including wasteforms containing uranium and plutonium). The research planned or being carried out by the NDA RWMD in this topic is subdivided as described in Fig. 4.

(1) The majority of HLW in the UK is based on either Magnox reprocessing wastes (Mg- and Al-rich) or blended waste (a mixture of Magnox and oxide fuel reprocessing by-products); these are unique to the UK (Harrison, 2010). The UK stockpile of spent fuel also includes fuels that are unique to the UK, such as those discharged from the UK fleet of advanced gas-cooled reactors (AGRs). In the research programme, studies aimed at evaluating the durability of the whole envelope of HLW compositions under a range of geochemical conditions relevant to the UK (including the effect of an alkaline plume in a co-located GDF) are in progress. For most of our spent oxide fuels, their performance in a future GDF can be bound using data from LWR (light water reactor) fuels measured by overseas waste management organizations (Karney and Thetford, 2011). However, the behaviour of AGR fuel is potentially different (Mignanelli and Rossiter, 2011) and experimental work is planned to measure the matrix dissolution and radionuclide leaching rates under a range of geochemical conditions relevant to the UK. These data are used to build understanding of the factors influencing radionuclides dissolution and to better underpin the parameter ranges used in post-closure safety assessment modelling.

(2) Experimental and modelling studies of issues associated with the packaging of specific types of wastes in cement are being carried out. Recent studies indicate that, if encapsulated in cement, the evolution of wastes containing reactive metals, supercompacted wastes, and some sludges and ion-exchange materials may affect the durability of waste packages and hence warrant further consideration in the research and development programme (Cronin and Collier, 2011; Utton and Godfrey, 2010). Additional experimental studies, modelling or more detailed review on these types of wasteforms are planned. Plans also exist to continue to undertake experiments aimed at improving current understanding of the long-term stability of alternative encapsulants in representative conditions expected in a GDF. A review study on vitrified ILW has recently being concluded (Utton *et al.*, 2011). Work on polymeric encapsulants has also

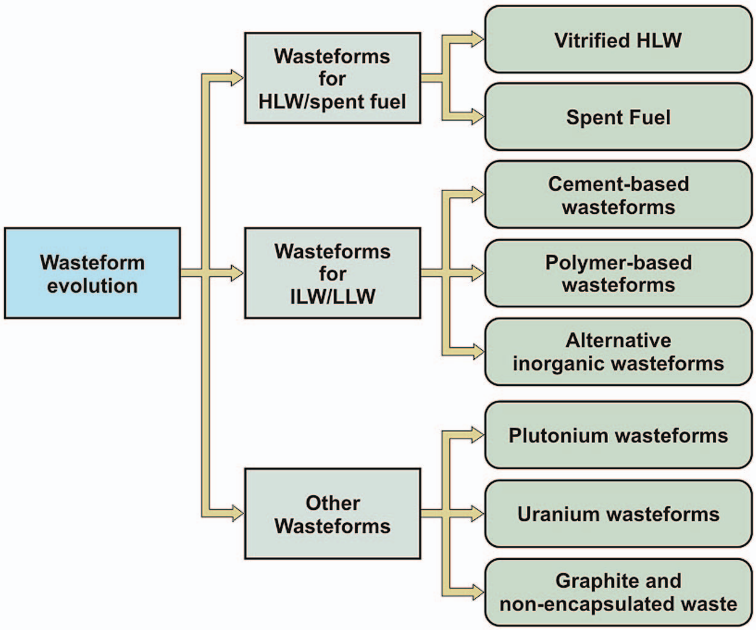

FIG. 4. Structure of the wasteform evolution topic in the NDA RWMD research and development programme. Published with the permission of the NDA.

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been carried out (Dawson *et al.*, 2010). Further work will be aimed at understanding the behaviour of vitrified products considering relevant wastes and at evaluating whether the formation of degradation products from candidate polymeric encapsulants has any potential impact on transport, operational or post-closure safety. Such work needs to be coordinated with work on potential for internal corrosion due to degradation products released by the polymers upon exposure to temperature and irradiation. Work may also need to be undertaken to examine alternative disposal options for this type of wasteforms.

(3) A study of the durability of potential plutonium wasteforms has recently been completed (Deissmann *et al.*, 2011). Work on uranium is ongoing. Particular issues for these materials are related to criticality safety (for plutonium) and volume reduction (for uranium). Work on criticality safety of plutonium is considered in the criticality topic of the research programme. Regarding uranium, work to understand the inventory and potential disposal routes for the various chemical forms present is being commissioned; this includes a series of scoping studies to measure the durability of candidate wasteform materials under a range of geochemical conditions relevant to the UK. Work on graphite is currently focussed on the potential release of ^{14}C during the post-closure period (see 'Gas' topic of the R&D programme). The NDA RWMD is continuing to participate in the CARBOWASTE European project to maintain awareness of any important developments in this area.

Container evolution

The work in this topic is focussed on progressing the evaluation of the options for different HLW/SF container designs in the absence of site specific information and on progressing understanding of the durability of ILW container materials, in particular in prolonged conditions of interim storage and GDF operations. For specific waste streams and encapsulation materials, it is also important to confirm that there is little potential for internal corrosion. The research planned or being carried out by the NDA RWMD in this topic is subdivided as described in Fig. 5.

(1) Two extensive reviews of the international experience and of the technical literature to evaluate the potential for corrosion of packages designed to contain HLW and spent fuel during the operational (King *et al.*, 2011) and post-closure (King and Watson, 2010) periods of a GDF have been completed. Further work is planned in this area to strengthen the current knowledge base by evaluating any information which may not have been previously considered, by supporting a more robust evaluation of container failure in the safety case and by considering in greater detail other important characteristics of container materials. Additionally, work is planned to evaluate the potential for internal corrosion and pressurization of UK spent fuel packages which may contain residual water due to previous storage operations and to identify suitable drying limits. An initial evaluation on AGR fuel is being completed and

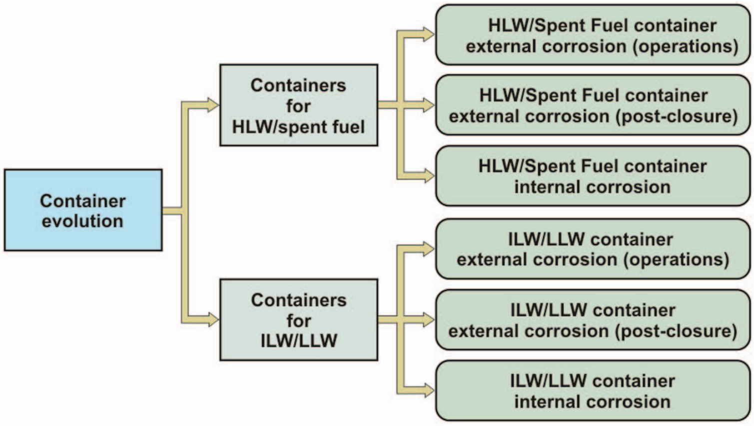

FIG. 5. Structure of the container evolution topic in the NDA RWMD research and development programme. Published with the permission of the NDA.

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further work is planned to expand and refine the previous analysis.

(2) A 10-year monitoring programme on a prototype waste container has recently been completed (Winsley *et al.*, 2011*a*). Some results of two experimental PhD studies supporting the development of a robust, mechanistic understanding of the evolution of stainless steel in relevant conditions have also been published (Albores-Silva *et al.*, 2011; Ghahari *et al.*, 2011). Both studies have been completed (Albores-Silva, 2011) and (Ghahari, 2012). Follow-up experimental studies aimed at identifying suitable environmental conditions for waste packages during surface storage and any operational period of a GDF are beginning. In particular, plans exist to evaluate the potential for localized corrosion and stress corrosion cracking in the presence of mixtures of salts that are likely to be representative of compositions in waste stores. Such studies include considerations on the likely evolution of alternative container materials (e.g. cast iron) (Morris and Winpenny, 2012). Models for the evaluation of corrosion in the long term are also being developed to evaluate the likely damage that can be produced over timescales of interim storage and operations of a GDF. Regarding the potential for internal corrosion, an experimental programme aimed at evaluating the potential for internal corrosion in the presence of polymeric encapsulants and their degradation products has been completed (Winsley *et al.*, 2011*b*, 2012). Further work is also planned to refine our understanding for the potential for internal corrosion (e.g. due to acidification, carbonation, release of chloride) in specific waste types through review of existing knowledge and, if required, experimental observation in specific conditions for which gaps in the data may exist.

Criticality safety

The work in this topic is focussed on extending the understanding of the likelihood and consequences of a criticality to bring it to a level suitable, in the current stage, to provide reassurance that criticality safety will be achieved in a GDF. This is an important area according to the views of some stakeholders. The research planned

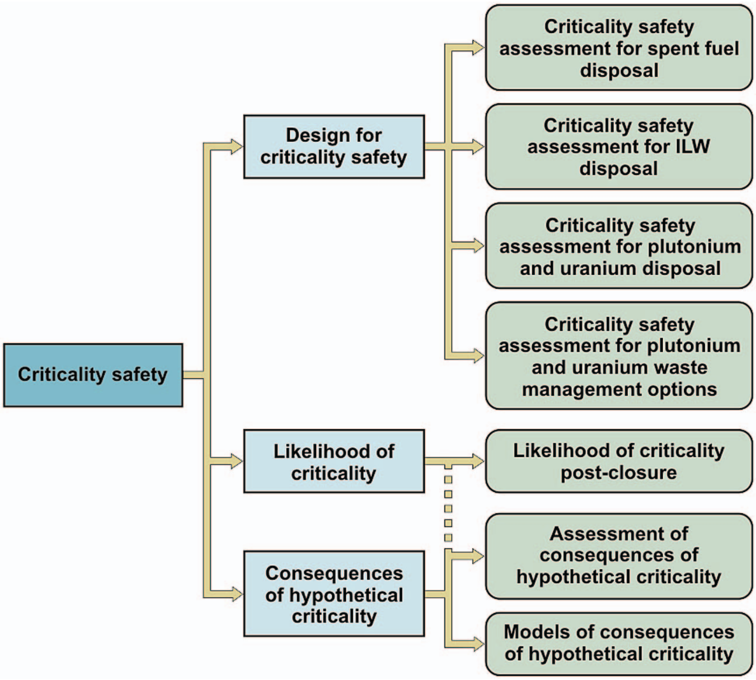

FIG. 6. Structure of the criticality safety topic in the NDA RWMD research and development programme. Published with the permission of the NDA.

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or being carried out by the NDA RWMD in this topic is subdivided as described in Fig. 6.

(1) Work is being completed to attempt to estimate the likelihood of a post-closure criticality occurring in ILW. This is based on the role of the backfill in limiting transfer of materials to possible accumulation sites. Scenarios will be constructed and analysed for a wider range of processes and for a wider range of system designs, wastes and other materials. The objective of this work is to develop, document and communicate the qualitative and quantitative arguments required to analyse the probability of criticality post-closure. The potential for accumulation of fissile material arising from disposed spent fuel, plutonium and uranium will be assessed in detail. Additional work may be needed to establish to what extent the likelihood of criticality can be quantified; alternatively, a set of qualitative arguments will be assembled relating to the various features, events and processes that hinder the accumulation of sufficient fissile material.

(2) In order to develop an understanding of how a criticality event might affect the ability of a GDF to contain its radioactive inventory, a programme of research was initiated entitled Understanding Criticality under Repository Conditions (UCuRC). This work (Nuclear Decommissioning Authority, 2010*b*), has been designed to explore the processes that would control the nature and magnitude of a hypothetical criticality under the particular post-closure conditions arising in a GDF containing ILW. Models of transient criticality have been developed (Mason *et al.*, 2009); as part of their verification and validation, one model has been compared with some of the behaviour of the Oklo natural reactors (Mason *et al.*, 2012). Further work is underway to apply models and approaches to scope the consequences of criticality transients in other materials and concepts and to recommend how the associated models and understanding should be developed and applied in future, beyond the preparatory studies phase.

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