Chapter 5
Waste Management and Performance Assessment

Jacob Philip and Steve Williams

Abstract Cement barriers play an important role in many radioactive waste management and disposal systems where they may provide physical and/or chemical barriers to limit the release of many radionuclides. Therefore, an understanding of how cement barriers are likely to evolve over the very long timescales that may be considered in safety cases and performance assessments is important. This can build confidence in the basis of a performance assessment or may reduce the need to make simple overly-conservative assumptions. This section begins with an overview of the role of cementitious materials in radioactive waste management and disposal, performance assessment and the treatment of uncertainty. It then summarises relevant papers presented at the NUCPERF 2009, NUCPERF 2012 and AMP 2010 workshops on the topics of the role and durability of cement barriers in waste storage and disposal and on the impact of gas generation and carbonation of cement systems. Some recommendations for future R&D are then made after drawing conclusions.

Keywords Disposal systems · Chemical and physical barriers · Radionuclides · Treatment of uncertainties

5.1 Introduction

Cementitious engineered materials have been used or proposed in a variety of waste management systems because these materials can be formulated with desirable performance characteristics (e.g. hydraulic isolation, chemical isolation and structural stability). Cementitious barriers are commonly engineered with a goal of achieving specific performance (e.g. minimisation of hydraulic conductivity,

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provision of required porosity or diffusivity or the maintenance of suitable chemical conditions). However a simple performance goal may not be optimum when practical considerations of design and performance characteristics are considered simultaneously. Laboratory-scale optimised designs may also have full-scale characteristics that are less than ideal (Esh et al. 2011).

Typical generic safety functions of cements employed in multi-barrier disposal systems may include providing a stable low solubility matrix that limits the release of many radionuclides by dissolving slowly (the wasteform), acting as a partial barrier limiting the access of water to the wasteform (the container), or conditioning the chemical characteristics of groundwater and porewater to reduce container corrosion or limit the dissolution of radionuclide-containing phases (the backfill) (Nuclear Decommissioning Authority 2010). Safety function indicators (a measurable or calculable property of a component that indicates the extent to which the safety function is fulfilled) and safety function indicator criteria (a quantitative limit such that if the safety function indicator to which it relates fulfils the criterion, the corresponding safety function is maintained) can be defined for specific safety functions.

The multi-barrier system approach has been applied to surface/near-surface and geological disposal concepts and designs. The wasteform itself often provides some degree of radionuclide retention. However, conservatively, no credit is usually taken for the retention of radionuclides by most wasteforms in deep disposal with the exception of vitrified high-level waste (HLW) and spent fuel (SF). For intermediate-level waste (ILW), depending on the disposal concept and host geology, their contribution may often be assumed to be negligible in comparison to components of the engineered barrier system (EBS) and to the geological host formation (which may be the main barrier to radionuclide migration). The main processes considered for IL-wasteforms are those related to their compatibility with EBS components and the host rock formation and any disturbances that might eventually be induced from these interactions. Typical examples include the release of complexing agents, gas generation and the effect of alkaline pore water migrating from wasteforms and the EBS into the surrounding rock (this is not an exhaustive list and depends on the disposal concept).

A similar conservative assumption about the contribution of the wasteform to radionuclide retention may also be made in surface disposal. In Belgium, for example, up to now the contribution of the wasteforms is not taken into account in the performance assessment, even if they would provide some degree of radionuclide retention in comparison to the other EBS components in this case (Wacquier et al. 2013). The reason is the great variety of wasteforms and the related complexity of the processes involved. Because of the remaining uncertainties, it would be difficult to change the conceptual model from something that is conservative and defensible (such as instant release of all the radionuclides from the wasteform) to more representative conceptual models that account for known physical and chemical processes. Adding model complexity adds to data requirements which have to be underpinned by extensive research and development (R&D) programmes. These added costs must be balanced against the benefits obtained for the entire safety assessment. Nevertheless in the future, for some particular cases, the contribution of a wasteform to radionuclide retention could possibly be considered if justified by a potential significant contribution to limiting the release of radionuclides and by a predictable behaviour. This could apply particularly to activated metals such as stainless and carbon steels where the ranges of the uniform corrosion rates of these alloys in cementitious environments have been determined with a good level of confidence.

Even if the contribution of the wasteform is not taken into account in performance assessments, their behaviour is the subject of R&D programmes. A minimum knowledge of their performance and expected evolution builds confidence (developing the scientific assessment basis), even if significant interactions with components of the EBS and with the host rock are not expected, and can quantify the safety reserve associated with them.

5.1.1 Performance Assessment

A performance assessment (PA) or safety assessment (a term used internationally) quantitatively estimates the potential post-closure impacts to human health associated with a radioactive waste facility. PAs are a means of helping decision makers to evaluate siting, design, operation and decommissioning of the waste disposal facility. PAs also identify a baseline point of compliance, require a sensitivity/uncertainty analysis and address requirements related to the protection of water resources. In some cases additional analyses are performed to identify doses to the public, not only from the disposal facility under consideration but from any other co-located sources that could contribute to a composite dose to a member of the public. These composite analyses are used to ensure that the total dose associated with the facility and any other source remains within levels allowed for exposure to the public. PAs of disposal systems are iterative processes involving site-specific modelling evaluations. The primary goal of a PA is to determine with reasonable assurance that the facility complies with regulatory criteria for the protection of human health and safety. The other goal of PAs is to identify critical data needed to support facility design and information needs. These are needed to make defensible and cost effective licensing decisions, and to maintain operating limits (e.g. waste acceptance criteria). The modelling conducted for PAs includes assessments of contaminant migration through environmental pathways (e.g. air, groundwater and surface water) and potential human exposures to the contaminants in various exposure media (e.g. soil, drinking water, crops and livestock). The potential for inadvertent human intrusion into the waste as an accidental pathway can also be modelled.

Figure 5.1 is a graphical representation of a PA process where the different components of the PA are represented as a function of time, infiltration, location and the chemical environment (adapted from US Nuclear Regulatory Commission 2000).

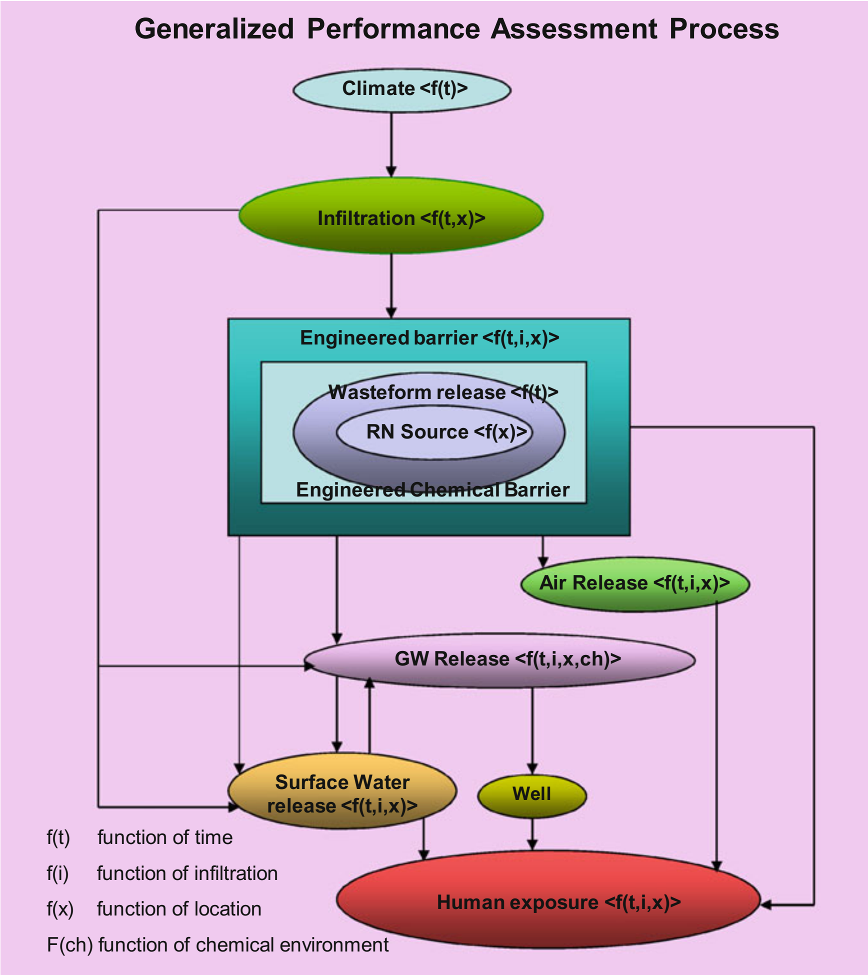

Fig. 5.1 Performance Assessment (PA) process

The central attribute of the process is that it is conducted iteratively, starting with a combination of generic and limited site-specific information in support of relatively simple conservative models and analyses, and progressing to more realistic, site-specific and detailed analyses, as necessary, to reduce uncertainty in assessing the performance of a disposal facility.

In current PA practice, in addition to the concept of an iterative process, PA is performed on a regular basis as information and data are acquired and as the performance assessment of a facility evolves. In the broader sense, PA has become a management tool in addition to a demonstration of compliance with regulatory criteria. This has led to increasingly sophisticated methods capable of representing physicochemical processes that can be used to guide improved designs for waste-forms, containments and facilities. As discussed later, there has been an increase in the use of stochastic modelling approaches to better capture the uncertainty in modelling results. Benefits of these improvements have been recognised in increased defensibility with stakeholders and improved efficiency of waste management practices.

As an example, in ONDRAF/NIRAS' proposed safety assessment methodology (ONDRAF/NIRAS 2013), safety assessments are carried out in two main phases at every programme stage (see Fig. 5.2):

- a phase of preparatory assessments
- a phase of formal assessments

Preparatory safety assessments are conducted continuously and repeatedly, at every programme stage, on the basis of phenomenological evidence from the

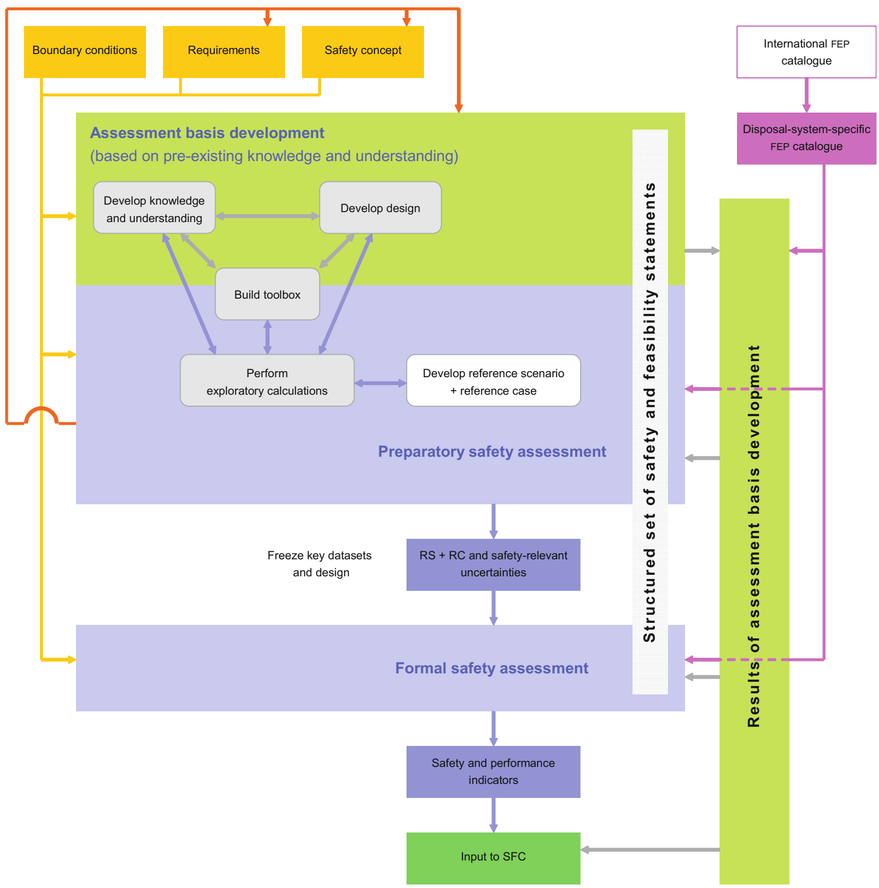

Fig. 5.2 ONDRAF/NIRAS proposed assessment methodology

assessment basis, and in parallel with system development. They take the form of sensitivity analyses and involve qualitative arguments. They thus entail sustained, structured interactions between safety assessors and experts in phenomenology and technology. Exploratory calculations are used, for example, to identify those contaminants that are potentially safety relevant, and therefore need to be considered in formal safety assessments, or to evaluate the impact of a particular process or uncertainty on system evolution and performance and hence determine its relevance to safety. Preparatory assessments also aim to identify any significant deficiencies in current knowledge and understanding and in the plans to address these in a R&D programme, thus providing feedback to assessment basis development. Modifications can then be made to the programme and, should this appear necessary, in the strategic choices (e.g. the choice of concept).

Through preparatory assessments, the safety analysts gain stepwise confirmation that the disposal system will evolve as defined in the safety concept and will fulfil each safety function as required (the reference scenario). They are then able to derive a reference case (i.e. a specific realisation of the reference scenario associated with conservative or a more realistic, but well-justified set of parameter values) as well as gaining insight into the impact of uncertainty on the expected evolution.

A methodological tool that can be used to examine the impact of perturbing phenomena and associated uncertainties on the safety and performance of the system in a systematic way is through a set of 'safety and feasibility statements'. Safety and feasibility statements are assertions regarding the safety or the feasibility of a proposed disposal system. They are derived from requirements and organised hierarchically in a top-down manner, starting with the most general (high-level) statements (e.g. 'The geological system is known') and progressing to increasingly specific (lower-level) statements (e.g. 'The hydraulic conductivity is sufficiently characterised'). The assessment of the impact of the uncertainties and perturbations on the validity of a statement, relative to the understanding needed, provides a useful tool to steer R&D.

Once the level of support available for the statements and the knowledge of the impact of uncertainties are judged sufficient (given the programme stage at hand) for proceeding with formal safety assessment, the key datasets within the assessment basis and the design are formally frozen. A formal safety assessment aims to show in a formal, quantitative way why the disposal system under consideration, despite the existence of remaining uncertainties, can be judged to be safe, satisfying all relevant regulatory and stakeholder requirements of the programme objective at hand. Formal safety assessments are quantitative, and as comprehensive as necessary, and are conducted prior to, and as part of, the preparation of a safety case (or safety and feasibility case). They aim to illustrate through a selection of calculation cases and indicators, the robustness of the system towards low probability scenarios affecting one or several safety functions (altered scenarios), alternative evolutions which are equally probable but do not affect the safety concept of the system (alternative cases within the reference scenario) and also, in particular, quantification of the safety margin with respect to the reference case. Formal safety assessments demonstrate how the system fulfils the regulatory and stakeholder requirements and highlight and prioritise the main open issues left to be treated in the next programme stage. Regulatory requirements may be, and are, different in different countries.

As part of the quality assurance of safety assessments, completeness checks are conducted during both preparatory assessments and formal assessments using a disposal-system-specific catalogue of features, events and processes (FEPs) (e.g. ONDRAF/NIRAS 2013) based on international FEP lists (NEA/OECD 2000).

PAs that include analyses of cementitious barriers have traditionally been associated with waste disposal. Current challenging environmental remediation and decommissioning activities, and disposal of low level, intermediate and high-level waste require PA-like analyses that may benefit from understanding the performance of cementitious materials. These involve numerous applications in many different locations (e.g. concrete vault-based designs for low-level waste disposal, in situ decommissioning of nuclear reactors and remediation of contaminated sites and old burial grounds). Typically, the performance of engineered barriers, including cementitious barriers, can be divided into those: (1) based on hydrological effectiveness or physical containment to reduce water-to-waste or waste-to-water contact; (2) chemical effectiveness to limit radionuclide transport; or (3) a combination of the two. Figure 5.3 depicts the time frames for apportioning approximate credit years for reliance on engineered/chemical barriers and site characteristics for Low Level Radioactive Waste (NRC Regulations 2016).

Concrete degradation mechanisms (e.g. sulphate attack, rebar chloride corrosion, cracking, alkali aggregate reaction, carbonation) can cause degradation and/or failure of the barrier and corresponding contaminant release. Obtaining data and

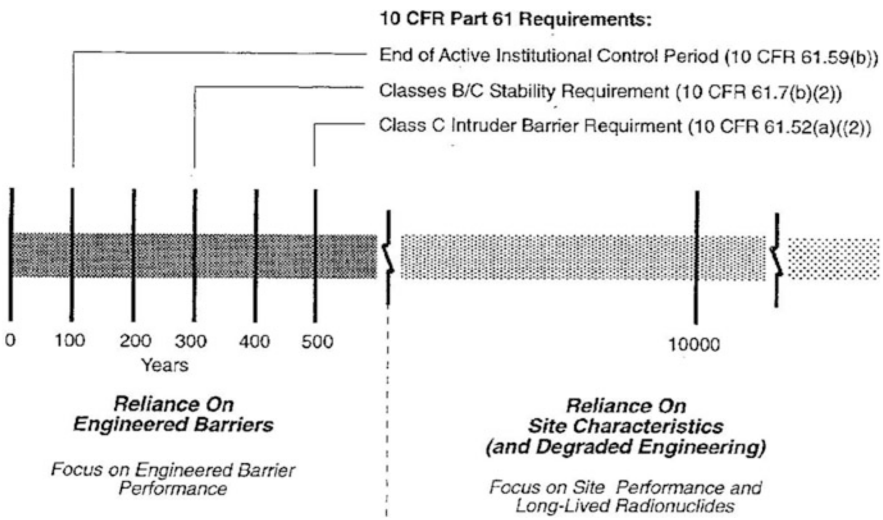

Fig. 5.3 Time frames for credits in PA for engineered barriers/site characteristics (taken from NRC Regulations 2016)

information from the cementitious barrier system facility is vital in the iterative PA process. This helps assure that the uncertainties in barrier performance are known and that any simplifying assumptions made for barrier performance in earlier PAs are still appropriate and, if not, revised. The revised PAs account for structural degradation for estimating whether end-of-life predictions in the PA are reasonable. Ideally, risk-based criteria are needed to evaluate how ageing and degradation affects structural capacity (structural margins) and whether PA accounts for it. For chemical containment, where the effectiveness of cementitious materials strongly depends on the source term, and source release characteristics, performance is very difficult to predict and is strongly related to both hydraulic properties and quantity of cement-based materials present. A cement-based barrier may also limit inadvertent intruder contact with waste, probably for at least a few hundred years, if it remains unexposed to aggressive environmental conditions. Because the performance of cementitious barriers may have to be assessed over hundreds if not thousands of years, the uncertainties for cementitious barriers are likely to be important to the PA.

5.1.2 Treatment of Uncertainties

The importance of sensitivity and uncertainty analyses has long been recognised as an integral part of PA for waste facility disposal. Figure 5.4 depicts an analysis process approach that incorporates parameter and model uncertainty in the PA calculations (US Nuclear Regulatory Commission 2000).

However there has not been general agreement regarding the specific approaches used to implement such sensitivity and uncertainty analyses. The views on sensitivity and uncertainty analyses can be different depending upon the regulatory environment, analyst preference and other reasons. For example PA for wasteforms from waste processing may have different goals than soil and groundwater assessment for remediation and may also be somewhat different from decommissioning assessments. Approaches to uncertainty analyses are an important consideration in the assessment of cementitious barrier performance in PA (Cementitious Barriers Partnership 2009).

Throughout the PA iterative process, sensitivity analyses are used to identify parameters with the greatest influence on the decision to be made and provide a means of focusing attention on those parameters for both the operations of the facility and compliance with regulations. For many years it was common to use deterministic approaches, which involved a base case and multiple sensitivity analyses targeted to explaining or better illustrating the effects of changes in different parameters on the overall result of the assessments over time. Lately, there has been increased use of probabilistic approaches to replace or supplement the deterministic calculation.

Two typical ways of classifying uncertainties are apparent in PA for health risk assessments. One method classifies uncertainties according to where in the risk

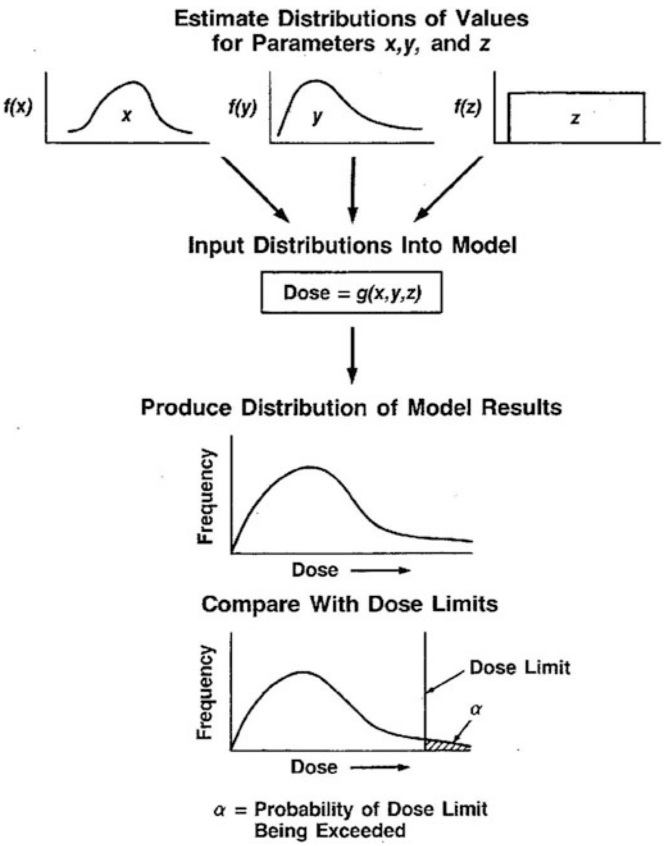

Fig. 5.4 Incorporation of model and parameter uncertainty in PA (taken from US Nuclear Regulatory Commission 2000)

assessment process they occur. Other approaches characterise uncertainties into more abstract, general categories, e.g. bias, randomness and variability. A more useful category is one based on parameter, model and scenario uncertainty. Parametric uncertainty occurs due to lack of knowledge of the 'true' value of an input parameter to a model. In model uncertainty, lack of knowledge about the structure and accuracy of the model is the major concern and includes simplifying assumptions and mathematical representations e.g. the use of 1-D and 2-D models. Scenario uncertainty arises from lack of information regarding missing or incomplete information needed to adequately define the model. This is an important aspect since PA of waste facilities involves predictions of performance, sometimes for hundreds to thousands of years, where the analysis of FEPs may play an important role. Overall, one element that runs through PAs and risk assessments is the need for expert judgment to be used to determine the appropriate parameters, values, distributions, models and scenarios. Expert judgment is valuable in that experts often have the greatest experience with these types of problems; however their judgment may suffer from the same biases as lay people. Use of a structured elicitation methodology is necessary in order to limit human bias (see, for example, United Kingdom Nirex Limited 2006).

It is important to consider the challenges specifically associated with the development of input distributions to support a probabilistic assessment. Multiple methods are available for characterising uncertainties in risk assessments. Two popular methods are Monte Carlo simulation and sensitivity analyses. For risk assessments Monte Carlo analyses involve characterising the uncertainty and variability in risk estimates by repeatedly sampling probability distributions representing risk equation inputs and using the results to estimate the range of risks. On the other hand sensitivity refers to variation in model inputs with respect to changes in model inputs that can provide a rank ordering of model inputs based on their relative contributions to model output variability and uncertainty. This can provide meaningful insight if the number of uncertain parameters is fairly small.

Characterising the properties and reducing uncertainties in understanding and predicting the fundamental behaviour of cementitious barriers is needed to evaluate and improve system design for near-surface engineered waste disposal systems (e.g., wasteforms, containment structures and entombments), environmental remediation and decommissioning. Uncertainty reduction can be informed by models coupling multi-scale and multi-physics processes, including physical chemical evolution and transport phenomena, and applying these to heterogeneous cementitious materials with changing boundary conditions. Ultimately, these can be integrated into a set of tools to predict the structural, hydraulic and chemical performance of cement based barriers over extended time frames [e.g. >100 years for operating facilities and >1000 years for nuclear waste management facilities (Cementitious Barriers Partnership 2009)].

5.2 Role and Durability of Cement Barriers in Waste Storage and Disposal

Zuloaga (2011) has discussed the ageing management program for the Spanish spent fuel and high level waste centralised storage facility. The planned operational life is 60 years, while the design service life is 100 years. Durability studies and surveillance of the behaviour have been considered from the initial design steps, taking into account the accessibility limitations and temperatures involved. The preliminary definition of the Ageing Management Plan, addressing the behaviour of spent fuel, its retrievability, the confinement system and the reinforced concrete structures, including test plans and surveillance design considerations, based on El Cabril LILW disposal facility developments, were presented.

Dry cask storage systems are used in the US for interim storage of spent fuel prior to the availability of a repository for these wastes in the future (Philip and

Graves 2013). Spent fuel is packaged within steel casks that are housed with an overpack, typically made from reinforced concrete, which acts as a storage module. These units are then placed vertically or horizontally on a concrete pad. These systems are currently licensed for 20 years with a potential for extension beyond this time. The concrete elements of these are subject to possible degradation through the effects of temperature and thermal gradients, ionising radiation, corrosive agents, water in air and soil, sulphate and chloride attack, alkali-silica reaction, carbonation and freeze-thaw cycling. A programme of modelling, inspection, monitoring, maintenance and repair of this type of container is required for successful ageing management. Snyder et al. (2006) discuss the application of Kalman filtering to combine computer model predictions with periodic measurements. An example scenario was presented based on the possible degradation of a concrete structure resulting from a concentration of a particular mobile species.

The impact of heat on the durability of concrete structures with regard to the long-term interim storage has been investigated by CEA (Lagrave et al. 2006). The technical approach is based on three areas of research: material characterisation; modelling to identify weaknesses in the structure; and validation by experimental tests on heavily instrumented structures subjected to representative loads. Models to estimate the effect of drying on the mechanical and the thermo-hydro-mechanical behaviour of concrete had been validated at the centimetre scale and were underway at full-scale (e.g. the GALATEE programme).

The role and performance of engineered barriers in present and future facilities for waste management and disposal in Romania have been described (Fako et al. 2013). Short-lived low and intermediate level wastes are conditioned by grouting with ordinary Portland cement in carbon steel drums and have been placed in disposal galleries in the low-level radioactive waste repository (DNDR) at Băita Bihor since 1985. Filled galleries are then isolated by the construction of a masonry wall. Since 1996 the galleries have also been backfilled with bentonite powder. The facility is located in an old mine and the geology and hydrogeology are highly disturbed and complex, leading to challenges in guaranteeing the long-term performance and safety of the facility. Current work has the objectives of increasing radiological safety by improving the technology for filling the galleries, developing a closure plan that is compliant with national and international requirements, and evaluating the performance of the closure systems. The concrete elements of the disposal system are the waste encapsulation matrix, which is an important physical and chemical barrier to the migration of contaminants and where improved formulations will be developed, and the concrete floors and walls, which provide a lesser benefit for the long-term safety of the repository. Spent fuel is stored at the Cernavodă interim dry spent fuel storage facility that was commissioned in 2003. Three of a total of 27 storage modules have been constructed so far and the condition of these is monitored. A near-surface facility (DFDSMA-Saligny) for LILW-SL (short-lived low-level and intermediate-level wastes) generated during the operation, refurbishment and decommissioning of the Cernavodă NPP is planned. The concrete barriers of the multiple engineered barrier conceptual design would play a more significant role in the long-term safety of the facility compared to those in DNDR and it is intended to model the system evolution. The waste encapsulation grout will be finalised during implementation of the conceptual project.

Kari and Puttonen (2009) discuss finite element modelling of the durability of concrete engineered barriers in the context of the disposal concept for low- and intermediate-level waste in Finland where these barriers are required to achieve a service life of at least 500 years after the facilities are sealed. The modelling considered interactions of atmospheric carbonation, chloride penetration, concrete degradation due to sulphate and magnesium, and leaching. Preliminary modelling results were compared with results from experiments and empirical models. It was concluded that the interaction of deterioration mechanisms is important and that the long-term deterioration of reinforced concrete may not be assessed with sufficient accuracy if only a single phenomenon is considered. Further discussion of this modelling approach was presented at NUCPERF 2012 (Kari and Puttonen 2013). It is important to identify the most critical model parameters in probabilistic calculations to assess the durability of concrete structures. Gulikers (2006) discusses this in relation to chloride ingress and shows that the age factor and critical chloride content have a pronounced influence on service life predictions. He concluded that the choice of mean values for model parameters has to be objective and care should be exercised when using values from limited data sets; this may lead to the necessity of making conservative choices. Reliable field data from real structures with information on measured chloride profiles, exposure conditions and concrete quality are required to derive more reliable statistical quantification of model parameters.

5.3 Impact of Gas Generation and Carbonation of Cement Systems

Simulations of atmospheric carbonation of concrete intermediate-low level waste (ILLW) packages during an operating period of up to 100 years with ventilation have been undertaken (Thouvenot et al. 2013). These considered the ILLW disposal zone within a facility based in the deep Callovo-Oxfordian formation in France according to the concept developed by ANDRA. Ventilation will lead to the desaturation of cement and atmospheric carbonation, which could potentially trigger corrosion of steel reinforcement. Atmospheric carbonation of concrete in unsaturated conditions is a complex process involving coupling of transport of water and CO_2 in the gas and liquid phases, capillary flow during drying and chemical reactions involving cement hydrates and CO_2 dissolved in the liquid water phase. Initial modelling of the drying and carbonation of concrete using the ToughReact code calculated a carbonation depth of between 1 and 4 cm in 100 years, depending on the assumed properties of the concrete. The degree of carbonation was greater than that found in experiments and this was attributed to decreasing reactivity as the degree of saturation of concrete fell from around 0.7 to

0.3. The model was refined to include this decreasing reactivity, which resulted in less intense but deeper carbonation of the concrete. Thouvenot et al. consider that the model could be further refined by taking into account the protective effect of secondary minerals, which would reduce carbonation intensity. In addition, they suggest that a more realistic simulation of the alteration phases may be possible in the future if kinetic data for the alteration of CSH at low Ca:Si ratios become available.

Carbonation was also the degradation mechanism considered in a risk analysis of possible concrete degradation with respect to safety of a proposed near-surface disposal facility for short-lived low and intermediate-level waste in Belgium (Capra et al. 2013). Carbonation, which could induce corrosion of rebar in the waste package monoliths or the module walls and slabs, was considered over a period of 2000 years from the commencement of operation of the facility until the time at which chemical containment of radionuclides was no longer claimed in the safety case. The risk analysis focused on the safety function "limitation of water through the system (thanks to the engineered barrier)". The time evolution of the carbonation front in the concrete elements was assessed probabilistically for several combinations of temperature, humidity and CO_2 content. The probability of the carbonation front being deeper than the thickness of the concrete cover of the rebars represents the end of the initiation period and the possible start of corrosion along the rebars. The impacts on the safety function were then ranked according to their probability, and their effect on the limitation of water flow for the component considered, in terms of localised, extended or global impact for the facility. It was found that there was no critical risk to limitation of water flow over a 2000 year period for any individual component. Minor risks were identified that could be managed by quality control during construction and the use of the specific concrete formulation proposed. Lower risks, after closure of the facility and the end of the nuclear regulatory control period after 350 years, related to the effects of inadvertent removal of the earth embankments or cover and could be managed by appropriate surveys of the disposal facility.

The Cementitious Barriers Partnership (Kosson 2013) has the aim of developing a set of tools to predict the structural, hydraulic and chemical performance of cement barriers used in nuclear applications over extended time frames (e.g. up to and greater than 100 years for operating facilities and greater than 1000 years for waste management) on behalf of the Office of Environmental Management in the US Department of Energy. A range of reference cases (e.g. cementitious wasteforms in a concrete disposal vault with a cap, grouted high-level waste tank closure and grouted vadose (unsaturated) zone contamination) have been identified for the comparison and demonstration of tools developed by the partnership. Codes such as STADIUM and LeachXS™/ORCHESTRA (http://cementbarriers.org/partner-codes) can be coupled to GoldSim (http://www.goldsim.com) in order to undertake probabilistic calculations and analyses of uncertainty and sensitivity. One important challenge that the Cementitious Barriers Partnership is currently addressing is the assessment of the integrity and closure of high-level waste tanks after they have been emptied and grouted internally (Brown et al. 2013). The HLW tanks may be empty for many years prior to closure and the performance of the closed tanks must be assessed over centuries, if not millennia. Carbonation-induced corrosion of the steel tanks (due to carbonation of the outer concrete shell) was identified as a primary degradation mechanism and possible failure mechanism prior to closure. After closure the impact of carbonation and concurrent oxidation of redox-sensitive elements may be to increase the release and short-range transport of some contaminants. LeachXS™/ORCHESTRA was used to model the penetration of CO_2(g) into the uncracked partially saturated wall of a representative HLW tank, the resulting carbonation reaction and leaching of constituents. Parameters including composition, soil-gas CO_2 concentration, degree of concrete saturation, porosity, CO_2 effective diffusivity, the mineral assemblage and thermodynamic parameters were varied to evaluate the sensitivity of the calculated results. The variations in these parameters tended to have less than one order of magnitude effect on the predicted extent of carbonation resulting in a pH value of less than 9, at which embedded carbon steel can be depassivated and become liable to corrosion. The influence of carbonation and chloride ingress in reinforced concrete structures is also of relevance to the corrosion of rebars in concrete secondary containment vessels for nuclear power plants (Petre-Lazar et al. 2006).

The importance of water content, cement porosity, iron content and gamma dose in determining the evolution of hydrogen from pore water radiolysis by βγ-emitting wastes encapsulated in cement binder was illustrated by the results of a study using the model DOREMI (Bouniol and Bjergbakke 2008) undertaken by Foct et al. (2013). This showed that the radiolytic production of molecular hydrogen depends strongly on water saturation with a maximum production rate occurring near 64 % saturation. Hydrogen production was calculated to decrease sharply at higher saturations due to the decrease in open porosity available for gaseous diffusion and a resultant increase in hydrogen recycling. The results also showed that increased cement binder porosity increased hydrogen production (because of the increased water content), as did increased dose rate. The presence of iron at greater than 64 % water saturation also increased radiolytic dihydrogen production because of the preferential reaction of Fe(II), rather than H_2, with the $O^{\cdot -}$ radical.

5.4 Conclusions

In summary, given that cementitious materials are engineered features, the key uncertainties in performance assessments tend to be related to the size of radioactive source term and near-field transport. The amount of credit taken in PAs for the role of cementitious barriers ranges from taking no credit at all, through to taking considerable credit for the physical and chemical properties (including the timing of any degradation). In some cases simplifying conservative assumptions may be made to take account of uncertainty when cementitious materials are considered in PAs. Such assumptions are generally made because of a lack of site- and facility-specific information for the cementitious materials or for expediency where conservative assumptions may be shown to be adequate. Alternatively, the uncertainty may be quantified and sampled in a probabilistic model.

In general, the role of cementitious materials as a physical barrier is for shorter periods than for their role as a chemical barrier. Cementitious barriers are designed to physically contain short-lived radionuclides, whereas their chemical properties limit radionuclide release rates for many longer-lived radionuclides. Because of the difficulties associated with quantifying the extent and initiation of cracking of cementitious materials when assessing the physical containment function, simple assumptions are often made (e.g. the cementitious barriers fail completely at the onset of through-wall cracking). For chemical barriers, the most common consideration has been the use of empirical sorption distribution coefficients (K_d values) that account for the radionuclide retardation properties of cementitious materials. The presence of reducing conditions in grouted wastes is an important consideration when considering redox-sensitive radionuclides (e.g. ^{99}Tc) in PA. This approach can lead to improved confidence related to taking credit for the long-term chemical performance as opposed to assessing the evolution of concrete cracking over time and its impact on physical containment.

5.5 RILEM TC-226-CNM Recommendations for Future R&D

There are a number of areas where improved understanding would be of benefit in underpinning the assessment of the performance of cementitious barriers in radioactive waste management.

Because of the difficulties associated with quantifying the extent and initiation of cracking of cementitious materials when assessing physical containment, simplifying assumptions are often made, for example that the cementitious barriers fail completely at the onset of through-wall cracking. A better understanding of how cracking evolves and how this leads to a developing loss of physical containment would allow credit to be taken for more gradual changes as cracking progresses.

The aqueous concentration of many radionuclides may be limited by the formation of low-solubility solid phases and through sorption to cement surfaces under the aqueous conditions of high pH imposed by the cementitious materials and low redox potential developed through components of the wasteform or engineered barrier system (e.g. metal corrosion, grouts containing BFS). The possible spatial and temporal evolution of these conditions is important when considering the range or uncertainty of the radionuclide parameters to be used in performance assessments. Research is ongoing to give increased understanding of how such conditions evolve (e.g. through improved thermodynamic and physical data and modelling of new phases) and is to be encouraged.

Carbonation affects the ability of cement materials to condition pore water to high pH. This can lead to increased rates of corrosion of embedded steels (e.g. reinforcement and closed underground tanks) and increased radionuclide migration.

Much modelling of carbonation has been carried out under partially-saturated conditions and further refinement to such models can be expected if data on secondary minerals and kinetics of carbonation of low Ca:Si gels are obtained. The rate and effect of carbonation of cements under fully-saturated conditions has been studied less extensively and such studies would be relevant to deep disposal facilities.

Evaluations of the performance of cement structures should include an appropriate sensitivity analysis to take account of variability in operations and materials. There is a general need for the development of probabilistic approaches to account for uncertainty in materials characteristics and performance. Overall the evolution of cement barriers should be considered in the context of the safety functions they are required to provide in a particular waste management concept.

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