Radioactive high level waste insight modelling for geological disposal facilities

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1. Introduction

The Nuclear Decommissioning Authority (NDA) is responsible for implementing the UK Government’s policy for the long-term management of higher activity radioactive waste by planning, building and operating a geological disposal facility (GDF). A GDF is an engineered facility for radioactive waste disposal which is constructed deep underground within a suitable rock formation to provide long-term isolation of the wastes from people and the human environment. Geological disposal is internationally recognised as the preferred approach for the long-term management of higher activity radioactive waste. Higher activity waste includes high level waste, intermediate level waste and a small quantity of low level waste that is not suitable for disposal in other facilities. The licensing process for such a facility includes examination of a safety case covering all aspects of the operational and post-closure safety of the facility and requires detailed numerical modelling to substantiate the safety case. Post-closure modelling assesses safety of a GDF over very long timescales following closure (e.g. a million years). In developing a safety case for such a facility the NDA believes that there is value in modelling the system at a number of levels of complexity to develop understanding and build confidence. This paper describes the development of an analytical model (NDA, 2013) to provide ‘insight’ into the key performance measures of a disposal system for high level waste. Similar insight models have been developed for the near-surface disposal of low-level waste (Kelly, 2011) and for the deep geological disposal of intermediate level waste (UK Nirex, 1995; Baker et al., 1997) in the past.

2. Model

The insight model described in this paper assumes a disposal concept similar to the Swedish KBS-3V concept, developed by the Swedish Nuclear Fuel and Waste Management Company (SKB) for the disposal of high-level waste (HLW) and spent fuel (SF). In the KBS-3V concept, waste is incorporated into a stable, low solubility matrix which is packaged inside a copper canister with cast iron insert for mechanical strength. The copper canister would be up to 5 cm thick to provide an extremely high level of corrosion resistance. The rate of corrosion of copper depends on the prevailing chemical conditions in the near-field porewater. Nevertheless, it is expected that such a canister will not be breached due to copper corrosion within a 100,000-year time frame.

For each canister, a deposition hole would be drilled into the floor of a disposal tunnel and lined so that, once emplaced, the canister is surrounded on all sides by a bentonite clay buffer. The bentonite clay would be around 40 cm thick and would provide a low permeability layer to prevent advective transport, sorb migrating nuclides and protect the canister. The disposal tunnel itself, which lies at approximately 500 m depth, would be backfilled with crushed host rock or further bentonite prior to closure of the facility.
Fig. 1 shows the barriers to radionuclide release from the KBS-3V disposal concept together with the properties and processes that contribute to its safety functions. The insight model presented here captures the key aspects of the KBS-3V concept. However, the model has been developed to be representative of other more general disposal concepts for HLW and SF. The model is able to represent any disposal concept in which the barriers to radionuclide migration are physical containment, slow release from the waste-form, diffusion through the bentonite (or other) buffer, and transport through the geosphere. The following sections provide a more generalised discussion of the physical and chemical processes incorporated into the model.

2.1. Disposal system and release pathway

The UK Government is using an approach based on voluntarism and partnership to identify a site for a geological disposal facility within the UK. The first stage of the process involves local communities expressing an interest in entering discussions about the siting process. At this stage no site has been selected and the insight model is based around a generic disposal concept and surrounded by a hard fractured rock. This geological environment is selected to provide a bounding estimate of radionuclide travel times; other geologies, for example a lower strength sedimentary host rock, would be expected to provide an increased travel time.

Within our generic disposal concept, waste is assumed to be vitrified or incorporated into a stable, low solubility matrix which is surrounded by a corrosion resistant disposal container. Each such container is surrounded by a low permeability buffer and placed into a disposal hole drilled into the host rock.

The waste containers are expected to remain intact for many thousands of years, but may eventually breach, for example due to corrosion, allowing groundwater to enter. When this occurs, radionuclides are assumed to dissolve slowly from the waste matrix and migrate through the buffer and geosphere via advection, dispersion and diffusion before finally reaching the biosphere and giving a radiological dose to a receptor. Decay and ingrowth of radionuclides occur throughout the system so that each transport equation takes the general form

\[
\frac{\partial C_n}{\partial \tau} = -v_x \frac{\partial C_n}{\partial x} + D \frac{\partial^2 C_n}{\partial x^2} - \lambda_n \frac{C_n}{\phi} + \lambda_{n-1} \frac{C_{n-1}}{\phi},
\]

where \( \lambda_n \) is the capacity factor for the \( n \)th radionuclide in a decay chain, \( \phi \) is the porosity of the surrounding rock, \( C_n \) is the radionuclide concentration in porewater, \( v_x \) is the transport velocity of porewater, \( D \) is the hydrodynamic dispersion coefficient (which includes a contribution from diffusion and dispersion) and \( \lambda_n \) is the decay constant.
2.2. The waste package and buffer

The canister provides physical containment of the waste through the use of a physical barrier, which is expected to gradually corrode with time. Depending on the material and thickness chosen this containment may be greater than a million years. Within the insight model a single failure mode for the canister is considered – total failure – in which absolute containment is provided for a period $T$ after which the container provides no barrier to any further release.

The waste is assumed to be vitrified or incorporated into the atomic lattice of a ceramic material. Once the waste container is breached, groundwater is assumed to ingress rapidly into the container, fill any void space and begin the process of dissolution of bound radionuclides. At this point a fraction of the total inventory remaining (which has been decaying throughout the assumed period of containment $T$) is released instantaneously, with the remainder dissolving slowly at a rate proportional to the mass of radionuclides still bound at that time.

It is assumed that radionuclides enter into solution with the porewater that occupies the bulk void space within the canister. Radionuclide sorption to canister corrosion products and the wasteform are neglected. Radionuclides are lost from the bulk canister void space through radioactive decay and diffusion into the buffer. Transport through the buffer is assumed to be diffusion-dominated, because water flow velocities through low-permeability buffer materials such as bentonite will be very low.

2.3. Geosphere

Radionuclides diffusing from the buffer are assumed to enter a fracture in the host rock that intersects the buffer. After travelling through the fractured host rock, radionuclides are assumed to enter a non-fractured geosphere with a significant transport time (several thousand years or more). The porous geosphere is assumed to consist of two layers, namely a reducing layer and an oxidising layer.

In the reducing layer chemical conditions are assumed to be reducing, and consequently redox-sensitive elements (e.g. uranium, technetium) will be in low oxidation states. Similarly, chemical conditions in the oxidising layer are assumed to be oxidising, and redox-sensitive elements will be in higher oxidation states. The distinction between reducing and oxidising is important because the solubility and sorption characteristics of the redox-sensitive radionuclides may be significantly different in the reducing and oxidising layers.

In the current model, the travel time through the fractured host rock is neglected. The transport processes through the reducing and oxidising layers are taken to be advection, dispersion and diffusion. Radionuclide sorption to rock substrates is taken into account. The diffusion of radionuclides into stationary porewater is neglected. In addition, solubility limitation of radionuclides in the geosphere is neglected.

2.4. Biosphere and exposure

Eventually, radionuclides will discharge from the geosphere into the biosphere, giving rise to radiation doses and risks to potentially exposed groups that come into contact with discharged radionuclides. In assessments undertaken to date by the NDA, an equilibrium biosphere is assumed. In this approach, radiological risk is proportional to the radionuclide mass transfer rate from the geosphere. Therefore, to test the effectiveness of the insight model, it is sufficient to compare insight model predictions of radionuclide mass transfer rates from the geosphere with those obtained from more detailed models. The biosphere is not considered further in this paper.

2.5. Insight approximation

At the heart of the insight modelling approach is the notion that, for a single-peaked, positive-definite function of time, $f(t \geq 0)$, the peak value $P_f$ of the function can often be estimated from knowledge of the total area $A_t$ under the curve and the width of the peak, expressed in terms of a suitably defined quantity $\sigma_f$, viz. $P_f \approx A_t / \sigma_f$. This approximation may be converted into an equality following the introduction of a shape factor $S_f$ (of order unity) which takes account of the relationship between the shape of the curve and the width $\sigma_f$, so that $P_f = S_f A_t / \sigma_f$. The exact numerical value of $S_f$ is generally unknown and will depend on the specific function $f$, but may be shown to be a constant for certain classes of function. For a Gaussian function the shape factor may be evaluated as $S_f = (2\pi)^{-1/2} \approx 0.4$ (Baker et al., 1997); for curves which are close to Gaussian the inclusion of this approximate shape factor then improves the accuracy of predicted peaks. Within this article the standard deviation of the curve is selected for the width, hence $\sigma_f^2 = \mu_2 / \mu_0(0)$ where $\mu_n(a)$ is the $n$th moment of the function $f$ about a point $a$, and $\mu_2 = \mu_1(0) / \mu_0(0)$ is the mean of the function. An approximation to the time $T_f$ at which the function $f$ has its peak is also provided by the mean, $\mu_f \approx T_f$. Fig. 2 illustrates these approximations.

In the context of radioactive waste disposal, $f(t)$ might represent a quantity such as the radionuclide concentration in part of the biosphere or the radionuclide mass transfer rate between two regions of the disposal system. In the latter case, $A_t$ then represents the total amount of radionuclide that is transferred, $\mu_f$ provides a measure of the time at which the radionuclide mass transfer rate is greatest, and $\sigma_f$ provides a measure of the timescale over which the bulk of the radionuclide transfer occurs.

2.6. Modelling approach

The basic structure of the model is relatively simple: once a radionuclide leaves from the wasteform it enters the groundwater in the bulk void space of the container, a concentration gradient is then set up across the buffer which leads to migration through the buffer and into a fracture in the host rock. From here the radionuclide migrates into the bulk host rock (the geosphere), where it combines with the radionuclide from other failed containers in

\[ \mu_{\alpha}(a) = \int [t - a^{t}] f(t) dt. \]
the disposal vault. Each disposal area within the vault is assumed to be identical in size and initial inventory. After transport through the geosphere rock (in reducing and then oxidising conditions), it finally discharges into the biosphere.

The insight model constitutes a system of differential equations coupled via conservation of mass boundary conditions at each interface. The pathway through which radionuclides travel is shown in Fig. 3; specific equations for each region will be introduced in the following sub-sections. Initially within the discussion the non-linear effects of solubility limitation are neglected. For certain radionuclides, solubility limitation may be treated through an approximation that neglects certain aspects of the release and dissolution of radionuclides from the waste matrix.

2.6.1. Container and wasteform leaching

Each waste container $i = 1, \ldots, N_c$ is assumed to provide absolute confinement for a specified period of time $T_i$, after which it is assumed to undergo an immediate failure, after which it provides no further confinement. Once confinement is lost groundwater is assumed to ingress rapidly into the container, fill any void space and begin the process of dissolution of bound radionuclides. At this point a fraction $f$ of the total inventory remaining (which has been decaying throughout the period $T_i$) is released instantaneously, with the remainder dissolving slowly at a rate proportional to the mass of radionuclides still bound at that time, and with constant of proportionality $k$.

The bulk void space within the container is represented by a well-mixed compartment (i.e. the timescale for equilibration within the container is much faster than the rate at which radionuclides enter or leave the container) which receives radionuclide mass as it dissolves from the wasteform. Let the rate of radionuclide mass transfer from the bound state to this compartment be denoted $R_{ni}^b(t)$, then

$$R_{ni}^b(t) = fM_n(t - \epsilon)\delta(t - T_i) + kM_n(t)H(t - T_i),$$

where $M_n(t)$ is the total mass of a radionuclide $n$ which is still bound within the waste matrix at time $t$, $\delta(t)$ is the Dirac delta function and $H(t)$ the Heaviside function. The small quantity $\epsilon \to 0$ is used to indicate that the instantaneous release depends on the bound mass immediately before container failure ($M_n^b$ itself being discontinuous at $t = T_i$). The function $M_n(t)$ varies as a result of ingrowth and decay in addition to dissolution so that

$$\frac{dM_n(t)}{dt} = -\lambda_\alpha M_n(t) + \lambda_{\alpha-1}M_{n-1}(t) - fM_n(t - \epsilon)\delta(t - T_i)$$

$$- kM_n(t + \epsilon)H(t + \epsilon - T_i).$$

For the release model we have adopted, which at this stage neglects solubility limitation, the release rates $R_{ni}(t)$ may be determined through the solution of these differential equations without further coupling to the remainder of the disposal system. As an initial condition we assume that $M_n^b = M_n(t = 0)$ is known from the radionuclide inventory for each container at the point of disposal.

2.6.2. Buffer and fracture

The material from which the buffer is composed is assumed to be sufficiently low permeability that radionuclide transport through advection may be neglected and diffusion becomes the dominant transport process. To represent the buffer we again adopt a well-mixed compartment approach, where, for two compartments (GoldSim Technology Group, 2010),

**Diffusive Mass Transfer Rate = Diffusive Conductance**

**× Concentration Difference.**

The diffusive conductance $D_{CB}$ between a compartment labelled $C$ and a compartment labelled $B$, having an interface area $A_{CB}$ may be calculated using the relation

$$D_{CB} = \frac{A_{CB}}{L_j^2 + D_j},$$

where $L_j$ is the diffusive length in compartment $j$ and $D_j$ the intrinsic diffusion coefficient\(^1\) in compartment $j$. The diffusive length is

\(^1\) The intrinsic diffusion coefficient is related to the free water diffusion coefficient $D_{hw}$ by $D_j = \phi D_{hw}$.
typically the distance from the centre of the compartment to the interface over which the diffusion is occurring, while the diffusion coefficient depends on the properties of the material and solute and would be determined through experiment. In the present model a single well-mixed compartment is used to represent the buffer. In the spirit of insight modelling this is a simple first approximation which is certainly reasonable for radionuclides which diffuse rapidly.

Once radionuclide diffuses through the buffer it is assumed to enter a fracture (subscript \( F \)) in the host rock, through which groundwater is flowing with a volumetric flow rate \( Q \). Even though the radionuclide travel time through the fracture is neglected, it is necessary to include a fracture compartment explicitly in the model. This is in order to apply the appropriate diffusive resistance that arises from the theory of diffusion into narrow fractures (Neretnieks, 1986).

If volume of compartment \( j \) is now denoted \( V_j \) and having concentration \( C_{j,n,i} \), we may write transport equations for the compartments representing the bulk void space of the container, buffer and fracture:

\[
V_c \frac{dC_{C,n,i}}{dt} = -D_C (C_{C,n,i} - C_{CB,n,i}) - \frac{\lambda_n V_c C_{C,n,i} + \lambda_{n-1} V_c C_{C,n-1,i}}{R_n(t)},
\]

\[\text{(5)}\]

\[
\frac{\partial C_{F,n,i}}{\partial x} = D_B (C_{F,n,i} - C_{F,n,i+1}) - D_B (C_{F,n+1,i} - C_{F,n,i}) - \lambda_n V_c C_{F,n,i} + \lambda_{n-1} V_c C_{F,n-1,i},
\]

\[\text{(6)}\]

\[
\frac{\partial C_{G,n,i}}{\partial x} = D_b (C_{G,n,i} - C_{G,n,i+1}) - Q_B C_{G,n,i} + \lambda_n V_c C_{G,n,i} + \lambda_{n-1} V_c C_{G,n-1,i},
\]

\[\text{(7)}\]

respectively.

The radionuclide concentration in each compartment is taken to be zero at the time of disposal to provide an initial condition for each of these equations. The parameter \( D_{BF} \) is chosen to provide the correct diffusive resistance between the buffer and the fracture, as discussed in (Neretnieks, 1986). Neglect of the travel time through the fracture is implemented by choosing a small value for the compartment volume \( V_c \). In the current study, the volume is chosen to be numerically equal to the water flow rate \( Q \). This ensures that the concentration in the fracture compartment equilibrates rapidly, and the radionuclide transfer out of the fracture compartment then equals the radionuclide transfer into the compartment.

2.6.3. Geosphere

The geosphere is divided into two regions with reducing (subscript \( R \)) and oxidising (subscript \( O \)) conditions. Within the geosphere transport is modelled as being one dimensional and the groundwater flow velocity is assumed to be uniform and time independent. The effect of diffusion into stationary waters in the rock matrix is neglected. If the reducing region has length \( L_R \) and the oxidising region length \( L_O \), then the transport equations become:

\[
\frac{\partial C_{R,n,i}}{\partial t} = -v_{R,n} \frac{\partial C_{R,n,i}}{\partial x} + D_R \frac{\partial^2 C_{R,n,i}}{\partial x^2} - \frac{\lambda_n x_o R_n C_{R,n,i} \partial C_{R,n,i}}{\phi_R} + \lambda_{n-1} x_o R_{n-1} C_{R,n-1,i} \partial C_{R,n-1,i},
\]

\[\text{(8)}\]

and

\[
\frac{\partial C_{O,n,i}}{\partial t} = -v_{O,n} \frac{\partial C_{O,n,i}}{\partial x} + D_O \frac{\partial^2 C_{O,n,i}}{\partial x^2} - \frac{\lambda_n x_o R_n C_{O,n,i} \partial C_{O,n,i}}{\phi_O} + \lambda_{n-1} x_o R_{n-1} C_{O,n-1,i} \partial C_{O,n-1,i},
\]

\[\text{(9)}\]

where \( D_{RO} \) is the longitudinal dispersion coefficient.

The radionuclide concentration in each compartment is taken to be zero at the time of disposal to provide an initial condition for each of these equations. In the full treatment of radionuclide transport across the reducing/oxidising boundary we require: continuity of porewater concentration at the reducing and oxidising geosphere boundaries,

\[
C_{R,n} |_{x=L_R} = C_{O,n} |_{x=L_O},
\]

that the contaminant mass entering the reducing portion of the geosphere equals the total radionuclide mass leaving the disposal vaults

\[
A \left( v_{R,n} C_{R,n} - D_{R,n} \frac{\partial C_{R,n}}{\partial x} \right) |_{x=L_R} = \sum_{i=1}^{N} Q_i C_{F,n,i}(t),
\]

\[\text{(10)}\]

where \( A \) is the assumed cross sectional area of the contaminant plume within the geosphere (reducing and oxidising geosphere areas are assumed to be equal); and that all contaminant mass leaving the reducing geosphere is assumed to enter the oxidising geosphere, implying

\[
\left( v_{R,n} C_{R,n} - D_{R,n} \frac{\partial C_{R,n}}{\partial x} \right) |_{x=L_R} = \left( v_{O,n} C_{O,n} - D_{O,n} \frac{\partial C_{O,n}}{\partial x} \right) |_{x=L_O}.
\]

For physical solutions we also require

\[
v_{O,n} C_{O,n} - D_{O,n} \frac{\partial C_{O,n}}{\partial x} \to 0 \quad \text{as } x \to \infty \quad \text{(assuming radionuclides in the biosphere are dilute)}.
\]

These equations can be solved by transforming into Laplace-transform space and using the boundary conditions to determine the various constants of integration. The result will be solutions that correctly capture the coupling between the reducing and oxidising regions. In particular, the concentration in the reducing region will depend on the properties of the oxidising region. The coupling arises because of the representation of longitudinal dispersion through a diffusion-like term in Eqs. (8) and (9). The coupling takes account of the fact that dispersion modelled in this way can occur either in the positive or negative \( x \)-direction, depending on the concentration gradient in the system. The coupling is likely to be most important in highly dispersive systems.

For advection-dominated systems, the coupling is less important, except within the vicinity of the boundary between the reducing and oxidising layers. For this reason, and in order to apply a simplification that is in the spirit of the insight model, it will be assumed that the radionuclide concentrations in the reducing layer do not depend on the properties of the oxidising layer. This can be implemented by imposing the following boundary condition on the flux from the reducing layer:

\[
v_{R,n} C_{R,n} - D_{R,n} \frac{\partial C_{R,n}}{\partial x} \to 0 \quad \text{as } x \to \infty \quad \text{(this is of course an approximation, but it is consistent with the approach adopted within the GoldSim modelling tool (GoldSim Technology Group, 2010) and other studies. It also has the value that the radionuclide concentrations and mass transfer rates in the reducing layer are uniquely determined by the nature of the radionuclide input and the properties of the reducing layer. Under these conditions, the output from the oxidising layer is the convolution of the input from the reducing layer and its own response function).}
\]

2.7. Solution

The coupled system of differential equations and boundary conditions, (2)–(12), defines the model we wish to solve. Since the equations are linear, the method of Laplace transforms provides a means for solution in Laplace space. From the discussion in
Section 2.5, an estimate of peak dose (or radionuclide concentration or mass transfer rate) will be provided by the moments of the function. A key result of the insight modelling approach is that these moments may be readily evaluated in Laplace space and the numerical inverse transform is therefore not required.

The solution of the various equations, and the derivation of moments, is set out in Appendix A.

2.8. Radioactive ingrowth

In growth is neglected in the solution to these equations; this simplification allows the governing equations for each radionuclide to be solved independently of the others in the decay chain. Nevertheless, it does mean that the insight model cannot be used to investigate the behaviour of radionuclides that are significantly influenced by ingrowth from a radioactive parent.

In principle, the “correct” treatment of decay chains increases the number of equations that are to be solved. For a decay chain with n members and a system of m compartments, there will be a total of nm equations to solve. However, one of the aims of the insight model is to develop analytic expressions for radionuclide concentrations and mass transfer rates, and for a large system of equations, analytic solutions may become very complex.

In some cases, however, it may be possible to use an approximation. If the half-life of a parent radionuclide is much shorter than the half-life of the daughter, then the mass or activity increase of the daughter due to decay of the parent is likely to be small. This is the case, for example, for the parent radionuclide of uranium-238, and therefore for uranium-238 the neglect of ingrowth will be an acceptable approximation. In other cases, it may be possible to augment the initial mass of a long-lived daughter with the initial mass of a shorter-lived parent.

Where the daughter radionuclide is much shorter-lived than the parent (as occurs for example in the uranium-238 decay chain), then it may be possible to use secular equilibrium arguments to estimate daughter radionuclide concentrations and mass transfer rates from those of the parent. Future work will investigate the use of these approximations and other approaches for including radionuclide decay chains within the insight model formalism.

2.9. Solubility limitation in the near field

In the analyses discussed above and in Appendix A, solubility limitation of radionuclides is neglected. In Appendix B, an approach is presented that enables solubility limitation in the buffer to be taken into account for certain radionuclides. The approach is based on the assumption that, following the failure of a container, the time period for which solubility limitation is active is much greater than the timescales for the release of radionuclides from the wasteform and diffusion of the radionuclides into the buffer.

3. Comparison of results with numerical models

Results from the insight model have been compared with those from a more detailed numerical model. The NDA total system model, as described in the generic post-closure safety assessment (NDA, 2010), was selected for this comparison. The total system model is a probabilistic performance assessment model created using the GoldSim software in which a number of key parameters are specified as Probability Density Functions (PDFs). The model is run 200 times and in each run (termed a ‘realisation’) different parameter values are randomly sampled from the PDFs, thereby producing a set of 200 results. These are combined to produce statistics for the desired performance metrics (such as radionuclide concentrations and mass transfer rates and risk to the public). In this way parameter uncertainty is included in safety assessment calculations for the GDF and its effect quantified.

The parameter PDFs that were used in these calculations, and a more detailed discussion of the detailed calculations, can be found in the NDA generic post-closure safety assessment (NDA, 2010).

To demonstrate the level of agreement between the insight model and the GoldSim model, peak values for various quantities and nuclides have been plotted from each model, for each of the 200 realisations. Perfect agreement between the insight model and detailed model would then result in a graph with all points lying on a straight line passing through the origin and with unit gradient.

The generic post-closure safety assessment (NDA, 2010) for high level waste predicts that the radionuclides caesium-135, selenium-79 and iodine-129 will provide the top three contributions to mean radiological risk in the period up to around one million years after GDF closure. These three radionuclides have therefore been selected to test the insight model and Figs. 4–9 show the degree of agreement. For caesium-137 and iodine-129, the approach set out in Appendix A is used. For selenium-79, the approach used in Appendix B is used for the realisations in which selenium-79 is solubility limited in the buffer. Uranium-238, chlorine-36 and palladium-107 were also investigated but are not reported here. The degree of agreement between the detailed and insight model results for uranium-238 and chlorine-36 is very similar to that for caesium-135 and iodine-129. For palladium-107 (a radionuclide that is solubility limited in many model realisations) the results show a similar degree of agreement to those for selenium-79.

Two quantities have been evaluated for each: the contaminant mass leaving the near field (buffer compartment) and entering the reducing geosphere.

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4 The key parameters that vary among the 200 realisations in this comparison are bentonite and geosphere sorption coefficients, solubility limits, diffusion coefficients, waste dissolution rates, effective flow rates and container failure times.
Fig. 5. Radionuclide mass transfer rate (g yr\(^{-1}\)) of caesium-135 leaving the oxidising geosphere and entering the biosphere.

Fig. 6. Radionuclide mass transfer rate (g yr\(^{-1}\)) of selenium-79 leaving the near field and entering the reducing geosphere.

Fig. 7. Radionuclide mass transfer rate (g yr\(^{-1}\)) of selenium-79 leaving the oxidising geosphere and entering the biosphere.
radionuclides with a low diffusivity in the buffer could therefore potentially be overestimated. The final situation is more subtle and concerns the insight model approximation itself. Radionuclides whose concentration or mass transfer rate curves do not approximate a Gaussian shape will not have their peaks well estimated by the formula given in Section 2.5, or may require a different numerical value for the shape factor.

As shown in Figs. 4 and 5 the agreement between the insight model and detailed model is very good for caesium-135 (which is neither solubility limited nor within a decay chain), especially for the contaminant mass entering the geosphere. The agreement for iodine-129 is also generally good, as shown in Figs. 8 and 9, although this offset in log space suggests an alternative choice of shape factor would produce better agreement. The insight model appears to consistently overestimate the radionuclide mass transfer rates. This offset in log space suggests an alternative choice of shape factor would produce better agreement. The insight model approximation itself. Radionuclides with a low diffusivity in the buffer could therefore potentially be overestimated. The final situation is more subtle and concerns the insight model approximation itself. Radionuclides whose concentration or mass transfer rate curves do not approximate a Gaussian shape will not have their peaks well estimated by the formula given in Section 2.5, or may require a different numerical value for the shape factor.

4. Conclusions

Within this paper we have described an analytical insight model for the radiological risks arising from the disposal of high level radioactive waste in a geological disposal facility. The model includes the processes of waste matrix dissolution, container failure, a diffusion-limited buffer and advection and dispersion in the geosphere. When coupled to an equilibrium biosphere model the insight model may be used to approximate the peak risk to a receptor, together with other useful performance metrics. The current model neglects the effect of ingrowth, which is important for some radionuclides, and employs a relatively coarse compartment model for the buffer. Despite this, for all of the significant radionuclides in the HLW assessment in the NDA generic post-closure safety assessment (NDA, 2010), namely chlorine-36, selenium-79, palladium-107, iodine-129, caesium-135 and uranium-238, predicted peak values for geosphere radionuclide concentrations and mass transfer rates show good agreement with more detailed numerical modelling.

The results presented here show that the insight model can provide a valuable second line of argument to assist in confirming the results of numerical models. In addition the simplified conceptualisation and analytical form allows the sensitivity of performance metrics against important parameters to be quickly investigated, which provides greater insight and understanding of the performance of a potential GDF, which in turn aids the creation of a robust safety case.

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Appendix A. Mathematical solution

A.1. Wasteform leaching

Release of the radionuclide \( n \) from the wasteform within container \( i \) is determined through the release rate \( R(t) \) which is determined through (2) and (3). Taking the Laplace transform of (2), and noting the initial condition, produces

\[
\mathcal{R}_{n,i}(s) = e^{-ts} M_{n,i} - \kappa \mathcal{M}_{n,i} - k \int_0^t e^{-\alpha t} M_{n,i}(t) dt,
\]

while (3) produces

\[
s \mathcal{M}_{n,i} - \mathcal{M}_{n,i}^0 = -\lambda_{n-1} \mathcal{M}_{n,i} + \lambda_{n-1} \mathcal{M}_{n-1,i} - fe^{-xt} M_{n,i}(T_i - \epsilon)
\]

\[
- k \mathcal{M}_{n,i} + k \int_0^{T_i} e^{-\alpha t} M_{n,i}(t) dt.
\]

If ingrowth is neglected we may obtain an expression for \( M_{n,i}(t) \) during the period \( 0 \leq t < T_i \) by integrating (3) in this region to produce:

\[
M_{n,i}(t) = M_{n,i}^0 e^{-\lambda t} \quad 0 \leq t < T_i,
\]

which in turn may be used to remove \( M_{n,i} \) in terms of \( \mathcal{M}_{n,i} \) from the expressions above. After some algebra the following expression is produced for the release rate

\[
\mathcal{R}_{n,i} = M_{n,i} \left( f(s + \lambda_i) \right) s + \lambda_i + k \right) e^{-(s + \lambda_i) T_i},
\]

A.2. Container, buffer and fracture

Concentration of radionuclide \( n \) in the container, buffer and fracture associated with container \( i \) is determined through (5)–(7). Taking the Laplace transform of each of these equations and noting the initial condition produces

\[
s + a \mathcal{J}_c_{n,i} = \frac{\mathcal{R}_{n,i}}{V_i} + \beta_i \mathcal{J}_b_{n,i},
\]
\[(s + b)\mathcal{C}_\text{R,n)} = \beta_1 \mathcal{C}_\text{R,n)} + \beta_1 \mathcal{C}_\text{F,n)}\]

and

\[(s + c)\mathcal{C}_\text{F,n)} = \beta_1 \mathcal{C}_\text{R,n)} \]

respectively, where

\[\beta_1 = \frac{D_B}{V_c}, \quad \beta_2 = \frac{D_B}{2s_{\text{act},n} V_B}, \quad \beta_3 = \frac{D_B}{2s_{\text{act},n} V_B}, \quad \beta_4 = \frac{D_B}{V_c} \]

and

\[a = \lambda_0 + \frac{D_B}{V_c}, \quad b = \lambda_0 + \frac{D_B}{2s_{\text{act},n} V_B} + \frac{D_B}{2s_{\text{act},n} V_B}, \quad c = \lambda_0 + \frac{D_B}{V_c} + \frac{Q_e}{V_c}.\]

These equations may be solved simultaneously to give the following expressions for \(\mathcal{C}_\text{C,n)}, \mathcal{C}_\text{R,n)}\), and \(\mathcal{C}_\text{F,n)}\):

\[\mathcal{C}_\text{C,n)} = \left[\frac{(s + b)(s + c) - \beta_3 \beta_4}{(s + a)(s + b)(s + c) - \beta_1 \beta_2(s + c) - \beta_1 \beta_4(s + a)}\right] \cdot \mathcal{R}_n(s), \quad \mathcal{C}_\text{R,n)} = \left[\frac{\beta_2(s + c)}{(s + a)(s + b)(s + c) - \beta_1 \beta_2(s + c) - \beta_1 \beta_4(s + a)}\right] \cdot \mathcal{R}_n(s), \quad \mathcal{C}_\text{F,n)} = \left[\frac{\beta_1 \beta_4}{(s + a)(s + b)(s + c) - \beta_1 \beta_2(s + c) - \beta_1 \beta_4(s + a)}\right] \cdot \mathcal{R}_n(s).\]

These expressions may also be used to derive similar expressions in Laplace space for the rate of diffusive mass transfer between container compartment and buffer compartment,

\[\mathcal{F}_\text{CR,n)} = D_B(\mathcal{C}_\text{C,n)} - \mathcal{C}_\text{R,n)}),\]

between the buffer compartment and the fracture compartment,

\[\mathcal{F}_\text{BF,n)} = D_B(\mathcal{C}_\text{B,n)} - \mathcal{C}_\text{F,n)}),\]

and between the fracture compartment and the reducing geosphere,

\[\mathcal{F}_\text{RF,n)} = Q_e \mathcal{C}_\text{F,n)}.\]

### A.3. Geosphere

In a real GDF the number of waste containers for disposal will be large (typically several thousand) and the explicit summation in (10) may be approximated by an integral. Suppose that the probability of a container failing between \(t\) and \(t + \Delta t\) is \(p(t)\Delta t\). In addition, suppose that the containers are identical, including having the same initial radionuclide inventory \(M^0\), so that the number of containers expected to fail between \(t\) and \(t + \Delta t\) (the failure density) is \(\mathcal{N}(t)\Delta t = N(t)\Delta t\). Taking the Laplace transform of (10) and substituting for \(C_{\text{R,n}}(t)\) using (20) produces

\[A \left(\nu_{\text{R,n}} \mathcal{C}_{\text{R,n}} - D_B \frac{\partial \mathcal{C}_{\text{R,n}}}{\partial x}\right) \bigg|_{x=0} = Q_e \mathcal{C}_\text{F,n)}(s) \int_0^\infty \left[\sum_{n=1}^N \delta(t - T_n)\right] e^{-(s + \lambda_0)T} dt,\]

where we introduce

\[\mathcal{C}_\text{F,n)}(s) \equiv \frac{M^0}{V_c} \left[\frac{\beta_1 \beta_4}{(s + a)(s + b)(s + c) - \beta_1 \beta_2(s + c) - \beta_1 \beta_4(s + a)}\right] s + \lambda_0 + k\]

and

\[\int_0^\infty A \left(\nu_{\text{R,n}} \mathcal{C}_{\text{R,n}} - D_B \frac{\partial \mathcal{C}_{\text{R,n}}}{\partial x}\right) \bigg|_{x=0} = Q_e \mathcal{C}_\text{F,n)}(s) \mathcal{N}(s).\]

where \(\mathcal{N}(s)\) is the Laplace transform of \(\mathcal{N}(t)\).

Eqs. (8)–(11) can be solved subject to the requirement that radionuclide concentrations approach zero as distance approaches infinity (as discussed in the main text) to give the following mass transfers from the reducing to oxidising layers, and from oxidising layer into the biosphere (subscript \(E\)):

\[\mathcal{F}_{\text{R0,n}}(s) = Q_e \mathcal{N}(s + s) \mathcal{C}_\text{F,n)}(s)\exp(-K_{\text{R0,n}}(s)L_e)\]

and

\[\mathcal{F}_{\text{OE,n}}(s) = Q_e \mathcal{N}(s + s) \mathcal{C}_\text{F,n)}(s)\exp(-K_{\text{OE,n}}(s)L_e)\exp(-K_{\text{O,n}}(s)L_o).\]

where

\[K_{\text{R0,n}}(s) = \frac{-\nu_{\text{R0,n}} + \sqrt{\nu_{\text{R0,n}}^2 + 4D_{\text{R0,n}}(s)} s + \lambda_0 + s}{2D_{\text{R0,n}}}.\]

### A.4. Moment relations

The insight approximation described in Section 2.5 attempts to obtain an estimate of the peak \(P_t\) and time of peak \(T_t\) for suitable function \(f(t \geq 0)\). This requires knowledge of the first three moments of the function when \(P_t = S_t \mu_t(0)/\sqrt{\mu_t(0)/\mu_t(0)}\) and \(T_t = \mu_t\) where \(\mu_t = \mu_t(0)/\mu_t(0)\). If the Laplace transform of the function is denoted \(\mathcal{F}(s)\) then the moments may be written

\[\mu_t(0) = \mathcal{F}(s)_{s=0}, \quad \mu_t(0) = -\frac{\partial \mathcal{F}(s)}{\partial s} \bigg|_{s=0}, \quad \mu_t = \frac{\partial^2 \mathcal{F}(s)}{\partial s^2} \bigg|_{s=0} \right) \cdot \mathcal{F}(s)_{s=0} \bigg|_{s=0} = \left(\frac{\partial \mathcal{F}(s)}{\partial s} \right)_{s=0}.\]

### A.5. Moment evaluations

The various moments of the mass transfers described by (21) and (22) can be evaluated by noting that these are products of functions in Laplace Transform space. The zero moment is the product of the zero moments of the individual functions, and the first and second moments are the sums of the suitably normalised first and second moments of the individual functions (Baker et al., 1997).

We begin with the failure density function \(\mathcal{N}(t)\). In NDA assessments, canister failure is assumed to occur at a uniform rate between times \(t = T\) and \(t = 2T\). If this is the case, if the initial number of canisters is \(N_c\) we have

\[\mathcal{N}(s + \lambda_0) = \frac{N_c}{(s + \lambda_0)^T} \left[\exp(-(s + \lambda_0)T) - \exp(-2(s + \lambda_0)T)\right]\]

and the moments are given by

\[\mu_{\mathcal{N}_c}(0) = \frac{N_c}{s + \lambda_0 T} (\exp(-\lambda_0 T) - \exp(-2\lambda_0 T)),\]

and

\[\mu_{\mathcal{N}_c}(0) = T + \frac{1}{\lambda_0} \cdot \frac{1}{\exp(-\lambda_0 T) - \exp(-2\lambda_0 T)},\]

and

\[\mu_{\mathcal{N}_c}(0) = \frac{1}{\lambda_0} \left(1 - T^2 \left(\exp(-\lambda_0 T) - \exp(-2\lambda_0 T)\right)^2\right) \cdot \exp(-\lambda_0 T) - \exp(-2\lambda_0 T)^2\right)\]

From (14) and (20), the function \(\mathcal{C}_\text{F,n)}(s)\) is the product \(uv\) of two functions \(u\) and \(v\) as follows:
\( u(s) = \frac{M_0}{V_C} \left[ f(s + \lambda_n) + k \right] \)

and

\( v(s) = \frac{\beta_3 \beta_4}{(s + \alpha)(s + \beta)(s + \gamma) - \beta_3 \beta_4(s + \alpha) - \beta_3 \beta_4(s + \beta) - \beta_3 \beta_4(s + \gamma)} \).

The moments of these functions are as follows:

\[
\begin{align*}
\mu_{u,0}(0) &= f\lambda_n + k, \\
\mu_{u,1}(0) &= (1-f)k, \\
\mu_{u,2}(\mu) &= \frac{(1-f)(1+f)k^2 + 2fk\lambda_n}{(\lambda_n + k)^2(f\lambda_n + k)^2}, \\
\mu_{v,0}(0) &= \frac{r}{h}, \\
\mu_{v,1}(0) &= \frac{g}{h}, \\
\mu_{v,2}(\mu) &= \frac{g^2}{h^2} - \frac{2e}{h}.
\end{align*}
\]

The following variables have been defined in these final three expressions:

\[
\begin{align*}
e &\equiv a + b + c, \\
g &\equiv ab + ac + bc - \beta_3 \beta_4 - \beta_1 \beta_2, \\
h &\equiv abc - \beta_3 \beta_4 a - \beta_1 \beta_2 c, \\
r &\equiv Q_1 \beta_2 \beta_4.
\end{align*}
\]

The moments of the exponential \( -K_{R,n}(s)L_\lambda \) are as follows:

\[
\begin{align*}
\mu_{K,n}(0) &= \exp \left( \frac{\nu_k L_k}{2D_K} \left( 1 - \frac{1 + 4D_K \bar{\gamma}_{n,K} L_k}{\phi_k v_k} \right) \right), \\
\mu_{K,1}(0) &= \bar{\gamma}_{n,K} L_k \frac{\phi_k v_k}{\phi_k v_k} \left( 1 + \frac{4D_K \bar{\gamma}_{n,K} L_k}{\phi_k v_k} \right)^{-1/2}, \\
\mu_{K,2}(\mu) &= \frac{2D_K \bar{\gamma}_{n,K}^2 L_k}{\phi_k v_k} \left( 1 + \frac{4D_K \bar{\gamma}_{n,K} L_k}{\phi_k v_k} \right)^{-3/2}.
\end{align*}
\]

Similar expressions will apply for the moments of the exponential \( -K_{Q,n}(s)L_C \).

The total moments of the function \( F_{R,n} \) (see (21)) are then given by

\[
\begin{align*}
\mu_{F,R,n}(0) &= \mu_{K,n}(0) + \mu_{K,1}(0) + \mu_{K,2}(\mu), \\
\mu_{F,R,n}(1) &= \mu_{K,n}(1) + \mu_{K,1}(1) + \mu_{K,2}(\mu) + \mu_{K,2}(\mu), \\
\mu_{F,R,n}(2) &= \mu_{K,n}(2) + \mu_{K,1}(2) + \mu_{K,2}(\mu) + \mu_{K,2}(\mu).
\end{align*}
\]

The total moments of the function \( F_{D,n} \) (see (22)) are similarly obtained.

### Appendix B. Solubility limitation

In this appendix, an approach is presented that enables solubility limitation in the buffer to be taken into account. The approach is based on the assumption that, following the failure of a container, the time period for which solubility limitation is active is much greater than the timescales for the release of radionuclides from the wasteform and diffusion of the radionuclides into the buffer.

Under these assumptions, and following the failure of a container, all of the disposed radionuclides are instantaneously transferred into the buffer. The radionuclides will partition between the dissolved and sorbed phases. If the inventory of a given radionuclide is sufficiently large, then that radionuclide will become solubility limited in the buffer.

Suppose that the solubility limit of a given radionuclide (assumed to be the only isotope of a given element in the disposed inventory) is \( C_S \). Solubility limitation will apply if the initial disposed inventory \( M_0 \) satisfies the following condition:

\[
M_0 \geq V_K C_S (\phi_0 + \rho_b K_D K_B)
\]

where \( K_D K_B \) is the sorption coefficient of the buffer. The time period \( T_S \) for which solubility limitation exists can be estimated. If the condition above applies, then \( T_S \) is the time required for the amount of radionuclide in the buffer \( M_S \) to satisfy:

\[
M_S = V_K C_S (\phi_0 + \rho_b K_D K_B)
\]

That is, it is the time required for an amount \( M_0 - M_S \) of radionuclide to diffuse out of the buffer.

During the period of solubility limitation, the governing equations for the radionuclide masses in the buffer compartment \( M_B \) and the fast flushing compartment \( M_F \) are as follows:

\[
\frac{dM_B}{dt} = -\lambda M_B - D_{BF}(C_S - C_F)
\]

and

\[
\frac{dM_F}{dt} = -\lambda M_F + D_{BF}(C_S - C_F) - QC_F.
\]

The second of these equations is easy to solve. Noting that \( C_F = M_F/V_F \), assuming that the fast flushing compartment initially contains no radionuclides, and taking \( t = 0 \) to be the time of container failure, it is easy to show that

\[
M_F = \frac{\alpha}{\beta} (1 - \exp(-\beta t)),
\]

where

\[
\alpha = D_{BF} C_S
\]

and

\[
\beta = \lambda + \frac{D_{BF} + Q}{V_F}.
\]

Now, an important characteristic of this solution is that it contains an exponential term with the factor \( Q/V \) in it. In order to get the properties of the fast flushing compartment correct, this ratio is of order unity. The consequence of this is that the exponential term in (28) will be negligible for times \( t \) greater than about 10 years. That is, the radionuclide mass in the fast flushing compartment rapidly reaches its equilibrium value.

A little algebra then leads to

\[
T_S = \frac{1}{\beta} \ln \left( \frac{M_0 - A/\beta}{M_S - A/\beta} \right)
\]

The peak radionuclide concentration in the buffer is of course just equal to \( C_S \). The peak concentration in the fast flushing compartment is given by
\[
C_{p,\text{peak}} = \frac{\alpha_{\text{VF}}}{V_{\text{BF}}} = \frac{C_{\text{c}}D_{\text{BF}}}{Q + D_{\text{BF}} + \lambda V_{F}}.
\]

The peak mass transfer rate from the buffer into the fast flushing compartment, and hence (approximately) the fast flushing compartment into the reducing geosphere, is given by

\[
F_{\text{BF}} \approx F_{\text{FR}} = QC_{p,\text{peak}}D_{\text{BF}}(C_{s} - C_{p,\text{peak}}) = \frac{QC_{\text{c}}D_{\text{BF}}}{Q + D_{\text{BF}} + \lambda V_{F}}.
\]

In order to use these results in the moments approach, it can be assumed that the time period of solubility limitation will be long compared with the time period for removal of the remaining radionuclide mass from the buffer, once the period of solubility limitation has ceased. In this case, the various moments are given by

\[
\mu_{F,0}(0) = \frac{Q C_{\text{c}} D_{\text{BF}} T_{S}}{Q + D_{\text{BF}} + \lambda V_{F}},
\]

\[
\frac{\mu_{F,1}(0)}{\mu_{F,0}(0)} = \frac{T_{S}}{2}
\]

and

\[
\frac{\mu_{F,2}(0)}{\mu_{F,0}(0)} = \frac{T_{S}^{2}}{12}.
\]

References


