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A PERFORMANCE ASSESSMENT MODEL FOR GENERIC REPOSITORY IN SALT FORMATION

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The U.S. is currently re-evaluating the policy on high-level nuclear waste (HLW) management and has been studying generic disposal system environment (GDSE) concepts to support the development of a long-term strategy for geologic disposal of high-level nuclear waste. The GDSE study focuses on the comparative analysis of different GDSE options, and a salt repository is one of the options currently under study. The immediate goal of the generic salt repository study is to develop the necessary modeling tools to evaluate and improve understanding on the repository system response and processes relevant to long-term HLW disposal in salt.

This paper presents an initial version of the salt GDSE performance assessment model and discusses the preliminary analysis results, emphasizing key attributes of a salt repository that are potentially important to the long-term safe disposal of HLW. It also discusses the preliminary results on the repository response to the effects of different waste types (commercial UNF, existing DOE HLW, and reprocessing HLW), and radionuclide release scenarios (undisturbed and human intrusion). In addition, the paper elaborates on the identified knowledge gaps and path forwards for future R&D efforts to advance understanding of salt repository system performance for HLW disposal.

I. INTRODUCTION

The U.S. is currently re-evaluating the policy on high-level nuclear waste (HLW) management. As part of the Fuel Cycle Research and Development (FCRD) program supported by the U.S. DOE Office of Nuclear Energy, the Used Fuel Disposition (UFD) campaign has been studying generic disposal system environment (GDSE) concepts to support the development of a long-term strategy for geologic disposal of high-level nuclear waste including commercial used nuclear reactor fuels (UNF). The GDSE study focuses on the comparative analysis of different GDSE options, and a salt repository is one of the options currently under study.

The immediate goal of the generic salt repository study is to develop the necessary modeling tools to evaluate and improve understanding on the repository system response and processes relevant to long-term HLW disposal in salt. This initial phase of study considered, where applicable, representative geologic settings and features adopted from the literature data for salt repository sites. The conceptual model and scenario for radionuclide release and transport from a salt repository was developed utilizing the literature data. The current version of the salt GDSE model consists of four major model components: source-term, near-field, far-field, and biosphere. The model was developed for a probabilistic analysis framework.

This paper presents an initial version of the salt GDSE performance assessment model and discusses the preliminary analysis results, emphasizing key attributes of a salt repository that are potentially important to the long-term safe disposal of HLW. It also discusses the preliminary results on the repository response to the effects of different waste types (commercial UNF, existing DOE HLW, and reprocessing HLW), and radionuclide release scenarios (undisturbed and human intrusion). In addition, the paper elaborates on the identified knowledge gaps and path forwards for future R&D efforts to advance understanding of salt repository system performance for HLW disposal.

II. SALT GDSE MODEL DESCRIPTION

II.A. Conceptual Model

The geologic settings and features of the generic salt repository in this study were adopted, where applicable, from the Waste Isolation Pilot Plant (WIPP) site.[1] The analysis assumes that repository is located in a bedded salt formation. A horizontal marker-bed with a significant thickness exists below the repository, which runs in parallel with the repository horizon to an extended distance, and a carbonate aquifer exists above the repository. Fig. 1 shows a schematic of the geologic setting and the conceptual model for radionuclide release and transport in a salt GDSE. Two scenarios are considered for repository radionuclide release and transport: the reference case, and the disturbed case. The reference case releases radionuclides by a sequence of typical processes that are expected to occur in a salt GDSE, and the disturbed case represents a non-typical process that provides a fast pathway for radionuclide to the far-field due to human intrusion.

In the post-closure repository, the waste decay heat would cause the near-field brines to boil during the peak thermal perturbation period, driving the water away from the waste disposal area leaving behind salt minerals in the pore space. This would create a dry-out zone around the waste disposal area, and its duration would depend mostly on the waste heat output characteristics and repository thermal loading. The thermal perturbation and its

associated moisture movement would also enhance creep closure of salt in the open space of the waste disposal area. As the disposal area temperature decreases following the peak perturbation, brines could start flowing into the waste disposal area driven by higher lithostatic pressures away from the disposal area. Brine flows into the disposal area would be enhanced by the deliquescence of rock salt minerals and precipitated salts in the dry-out zone. In general the deliquescence relative humidity (DRH) of these salt mixtures is lower than the individual pure salt and decreases with increasing temperature.

Corrosion of waste package and other engineered materials in the disposal area would greatly be enhanced in contact with hot concentrated brines, and gases generated as a result of the corrosion in chemically reducing condition. Subsequent to waste package corrosion failure, corrosion of waste form, its canister, and waste package internal structure materials would ensure, releasing RNs and generating additional corrosion gases. Combined actions of the corrosion gas generation and decreasing confined space in the disposal area by salt creep closure would pressurize the disposal area; this would result in brine flows and transport of dissolved RNs away from the disposal area to the surroundings.

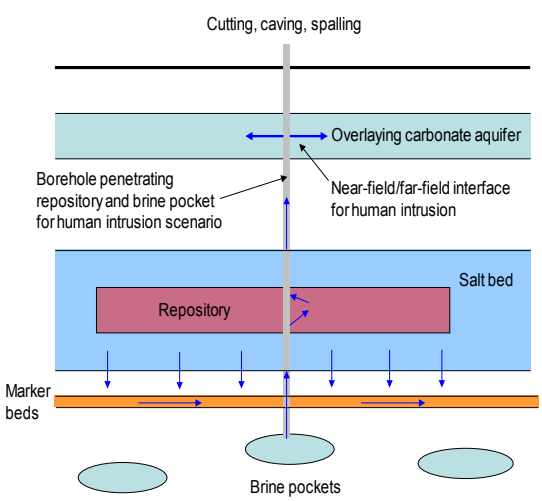

Fig. 1. A schematic showing the conceptual model for radionuclide release and transport from a salt generic repository.

Because information on thermal perturbation and other associated processes including brine flows, salt creeps, etc., for a representative generic salt repository is not available, the analysis assumes an ambient temperature for the generic repository and continuous brine flows from the disposal area from time zero. It also assumes no performance credit for waste package and waste form canisters and waste form degradation at the beginning of analysis. These are highly conservative assumptions.

II.B. Near-Field and Far-Field Models

The reference case assumes that a marker bed below the repository provides the major pathway for radionuclide release and transport from the repository. The marker bed is assumed to be composed of a mixture of evaporite minerals (such as anhydrite) and clay. As discussed above, no waste package performance is considered, and the waste form degradation starts at time zero, releasing radionuclides into the near-field. Dissolved radionuclide concentration in the near-field is determined by the amount of radionuclides released from the waste form (constrained by the waste form degradation rate), the amount of water available in the near-field, and the solubility if it is subject to its solubility limit. The solubility for the near-field concentrated brine is applied (Table 1).

Dissolved radionuclides in the near-field are released to a marker bed below the repository, which is assumed to run horizontally in parallel with the repository (Fig. 1). The model does not consider the distance between the bottom (or floor) of repository and the marker bed and assumes no resistance to the flow and radionuclide transport over this distance. The marker bed is assumed to be one-meter thick, with its width to be the same as that of repository; the marker-bed cross sectional area to water flow is the bed thickness times the width. As shown in Fig. 1, dissolved radionuclides are transported into the marker bed over its length below the repository; this portion of the marker-bed is referred to as the near-field marker bed in the analysis.

Radionuclides are transported advectively in the marker bed to a drinking well location 5 km down-gradient from the edge of the repository, where a hypothetical biosphere exists. This portion of the marker-bed is referred to as the far-field marker-bed. The brine flow rates in the marker bed away from the repository are based on the WIPP site analysis, for which the brine flows are driven by the pressurization in the disposal room by the gases generated from degradation of organic materials.[1] Table 3 lists key transport parameters and their values for the marker-bed.

Radiation exposure, or dose, is used as a performance metric for the salt GDSE analysis. The IAEA Example Reference Biosphere 1B (ERB 1B) dose model[2] was used to calculate the dose from the dissolved radionuclide concentration at the hypothetical drinking well location. It is acknowledged that it is unlikely that groundwater drawn from a bedded salt formation would be potable without significant treatment. It is assumed that recharge in the marker beds would sustain well withdrawal over a long period of time.

The disturbed scenario is represented by a “stylized” human intrusion scenario. In this scenario it is assumed that a single borehole penetrates a waste package and a brine pocket below the repository at 1,000 years after

repository closure. The number of waste packages affected (one penetrated plus, if any, neighboring packages affected) is randomly sampled between one and five (uniform distribution), and this represents the total amount of waste inventory that becomes available for the fast pathway release by the human intrusion. Dissolved radionuclides from the affected waste packages are carried upward through the borehole by pressurized brines from the brine pocket, and released to an overlying carbonate aquifer. The brine flow rate through the borehole is sampled between 0.1 and 5.0 m^3/yr (uniform distribution).[1] The overlying aquifer is assumed to comprise primarily dolomite matrix with clays dispersed in the matrix.

The dissolved radionuclide concentrations in the aquifer are evaluated against the solubility for the far-field water (Table 2) before they are transported in the aquifer to a drinking well location 5 km down-gradient from the repository boundary. Table 4 lists key transport parameters and their values for the overlying aquifer. As for the reference case, the same hypothetical biosphere is assumed to exist at that location, and the reference biosphere model (IAEA ERB 1B model) is applied to calculate the dose.

Two repository waste inventory scenarios were considered for each of the cases. For the reference case, the waste inventory for Scenario 1 comprises the commercial UNF and DOE HLW. The waste inventory for Scenario 2 comprises the DOE HLW and reprocessing HLW. Scenario 1 takes a square repository footprint with a side of 3,270 m for disposal of a total of 37,157 waste packages (32,154 commercial UNF waste packages plus 5,003 DOE HLW waste packages). Scenario 2 needs a smaller square repository footprint with a side of 1,615 m for a total of 9,058 waste packages (5,003 DOE HLW waste packages plus 4,055 reprocessing HLW waste packages). For simplification, for the disturbed case, Scenario 1 considers only the commercial UNF waste packages are affected, and Scenario 2 only the DOE HLW waste packages are affected. More detailed discussions of the source-term and near-field model are given elsewhere.[3,4] Additional discussions on the far-field model are given in Ref. 4.

The salt generic repository model was implemented in Goldsim.[5] The repository performance analysis was performed probabilistically, with 100 realizations for each case and for a time period of one million years.

III. MODEL RESULTS

The model results are presented in terms of the mean radionuclide (RN) mass release rate from the near-field and far-field as the intermediate result, and the mean dose (mrem/yr) by individual RN at the hypothetical accessible environment (AE). It is noted that this model is the initial effort of the salt GDSE analysis tool development and needs further improvement and refinements as the study progresses. Also note that using the mean dose is an arbitrary choice to present and discuss the analysis results in order to facilitate comparative studies among the GDSE options and does not indicate any realistic dose implications. Therefore, the results presented in this paper *should not* be construed as being indicative of the true performance of a salt GDSE or compared to any regulatory performance objectives regarding repository performance.

Fig. 2 shows the model results for the waste inventory scenario 1 (commercial UNF plus DOE HLW) of the reference case: (A) the mean RN mass release rate at the edge of the near-field marker-bed below the repository, (B) the mean RN mass release rate from the far-field marker bed, and (C) the mean dose by individual RN at the hypothetical AE. RNs released from the near-field marker-bed (Fig. 2(A)) are greatly retarded in the far-field marker-bed mainly by sorption on to the marker-bed filling medium (Table 3); non-sorbing or weakly sorbing RNs with a significant inventory (^{129}I, ^{79}Se, ^{126}Sn and ^{36}Cl) are released from the far-field marker-bed at noticeable rates (Fig. 2(B)). The dominant long-term dose contributor is ^{129}I (Fig. 2(C)), and this is expected based on the following characteristics of the radionuclide: 1) unlimited solubility, 2) weak sorption on marker-bed filling materials, 3) extremely long half-life (17 million years), and 4) a significant inventory in the waste (about 950 g per commercial UNF waste package and about 65 g per DOE HLW waste package). The mean dose for ^{79}Se is comparable to that of ^{129}I for up to about 530,000 years, and this is expected because of radionuclide's properties that are similar to those of ^{129}I listed above, except a shorter half-life (295,000 years). There are conflicting data on the ^{79}Se half-life in the literature, which has been variously reported as 6.5×10^4 years, 2.95×10^5 years (used in the model), 4.8×10^5 years, 6.5×10^5 years, and 1.13×10^6 years.[6] Also the selenium solubility in water is highly uncertain. The metal selenium is insoluble in water, but it can also be released as soluble selenate ion (SeO_4^{2-}), which is not readily sorbed onto geologic materials. More work is needed to better characterize and quantify dissolution and sorption behavior of selenium in a geologic repository environment.

The results for the waste inventory scenario 2 (DOE HLW plus reprocessing HLW) are shown in Fig. 3. Compared to the inventory scenario 1, this scenario requires a smaller number of waste packages (9,058 waste packages vs. 37,157 waste packages) and a one-fourth of the repository footprint area. This in turn takes a smaller volume of near-field, therefore a smaller amount of near-field water, resulting in higher concentrations of soluble radionuclides (such as ^{129}I and ^{79}Se) in the near-field water. In addition, because of the assumptions made for the reprocessing HLW, the fission products inventory on a per-waste package basis is higher than that for scenario

1. For example, each reprocessing HLW waste package contains about 7,500 g of ^{129}I and about 250 g of ^{79}Se, which is about eight times greater than the per-waste package inventory mass of the radionuclides of commercial UNF. Their impacts are shown in Fig. 3 as having about two orders of magnitude higher mean peak dose for ^{129}I than the inventory scenario 1. The mean peak dose for ^{79}Se is about 10 times higher than that for inventory scenario 1. Note that the ^{36}Cl dose is absent in the result because both the DOE HLW and reprocessing HLW do not have ^{36}Cl inventory.

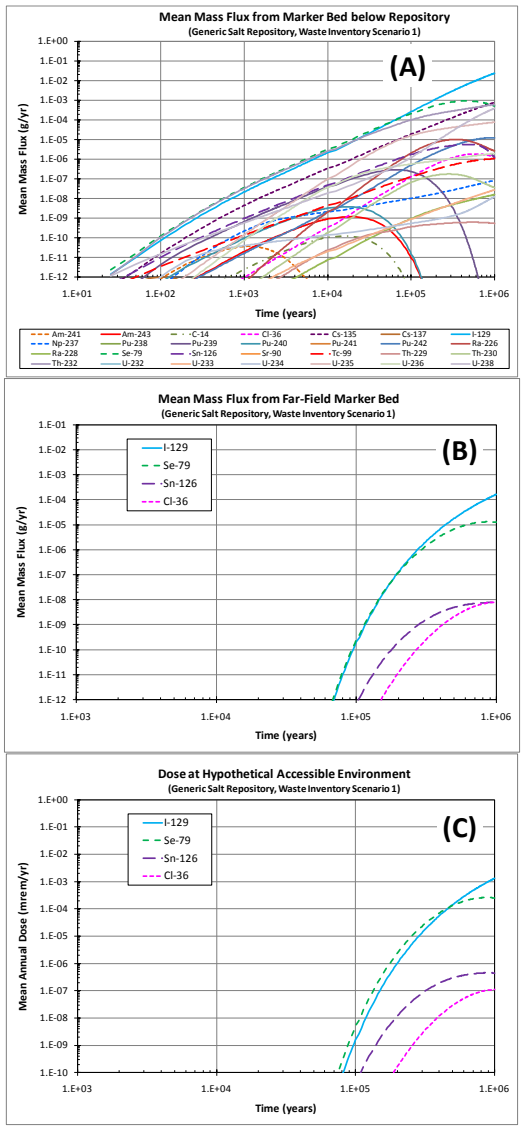

Fig. 2. Model results for the waste inventory scenario 1 (commercial UNF plus DOE HLW) of the reference case: (A) mean RN mass release rate from the marker bed below the repository; (B) mean RN mass release rate from the far-field marker bed; (C) mean dose by RN at the hypothetical AE.

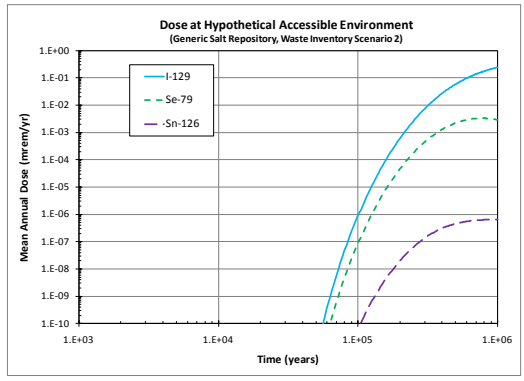

Fig. 3. Mean dose by RN at the hypothetical AE for the waste inventory scenario 2 (DOE HLW plus reprocessing HLW) of the reference case.

Fig. 4 shows the model results for inventory scenario 1 of the disturbed case: (A) the mean RN mass release rate from the far-field overlying aquifer, and (B) the mean dose by individual RN at the hypothetical AE. The mass release and dose histories are very different from those for the reference case. In this case, only commercial UNF waste packages are assumed to be affected by a single borehole penetration at 1,000 years, and the number of waste packages affected (i.e., the amount of waste inventory affected) are sampled between one and five. The dissolved radionuclides in the near-field water are transported upward by pressurized brine flow through the borehole and released directly to the overlying aquifer. The aquifer water flow rate is several orders of magnitude greater than the flow rate in the marker bed, and the radionuclides are transported advectively at much greater rates (Tables 3 and 4). ^{226}Ra is the dominant dose contributor, which is a daughter of the decay chain:

^{242}Pu (3.76×10^{5} yrs) → ^{238}U (4.46×10^{9} yrs) → ^{234}U (2.45×10^{5} yrs) → ^{230}Th (7.54×10^{4} yrs) → ^{226}Ra (1.6×10^{3} yrs)

The initial inventory of ^{226}Ra is very small (9.6×10^{-6} g per commercial UNF waste package), so most of the dose for ^{226}Ra is likely to originate from the daughter of the above decay chain. In addition, due to a lack of data, radium is modeled as having unlimited solubility and as non-sorbing. However, radium is known to readily sorb on soil, clays and other geologic materials.[7-9] The radium solubility in water is dependent on the type of radium-containing minerals; for example, the solubilities of radium sulfate and carbonate are low, but solubilities of radium nitrate, chloride and iodate are high.[10-12] In general radium is not mobile in groundwater. If more representative values for the radium solubility and sorption were implemented, the dose for ^{226}Ra would have been much lower. Additional studies are needed to better characterize and quantify the dissolution and sorption behavior of radium in geologic environments.

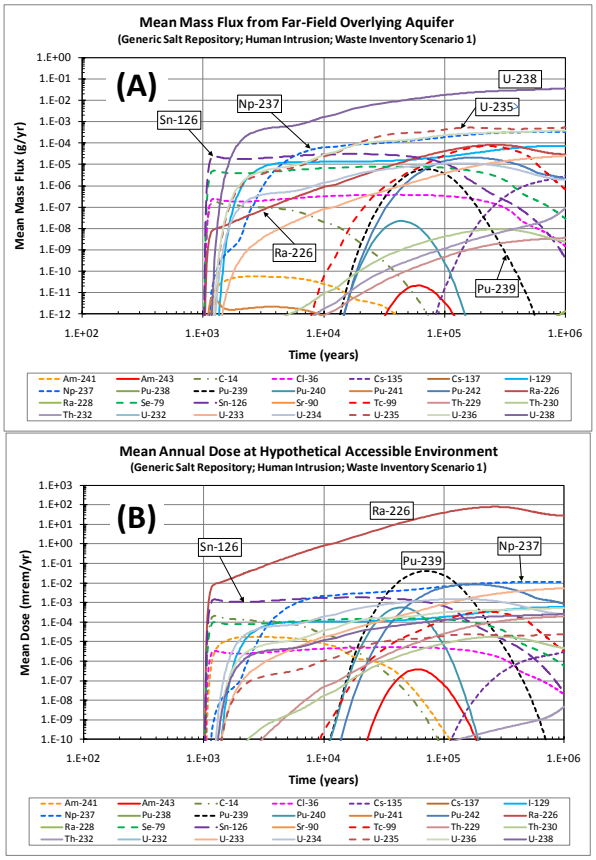

Fig. 4. Model results for the waste inventory scenario 1 (commercial UNF) of the human intrusion case: (A) mean RN mass release rate from the far-field overlying aquifer; (B) mean dose by RN at the hypothetical AE.

Compared to the reference case results, the doses for the soluble, non-sorbing fission products, particularly ^{129}I and ^{79}Se are much lower, while the doses for actinides including ^{239}Pu, ^{242}Pu and ^{237}Np are much higher. The lower doses for the fission products are due to their lower total inventory available for release (i.e., up to five affected waste packages), and the higher doses for the actinides are due to the direct release of the radionuclides in the aquifer with higher water flow rates, resulting in an early arrival of higher concentrations of the radionuclides at the biosphere drinking water well prior to their significant decay.

IV. SUMMARY AND CONCLUSIONS

This paper discusses an initial version of the salt GDSE model and the preliminary model results. The current model is an initial outcome of the long term effort to develop a salt GDSE analysis tool and it will be further improved and refined as the study progresses. The current model analysis helps to draw the following important considerations and suggestions for the near-term improvement of the on-going efforts on the salt GDSE model development and analysis.

Soluble, non-sorbing fission products, particularly ^{129}I and ^{79}Se, are the major dose contributors. In the current model, ^{79}Se is modeled as soluble and non-sorbing in generic salt repository environments. However, the solubility and sorption behavior of selenium in reducing geologic environments are uncertain, and improvement is needed to better characterize and quantify the chemical properties. In addition, the half-life of ^{79}Se has been reported variously ranging from 6.5×10^4 to 1.13×10^6 years. A more accurate half-life estimate of this radionuclide is needed.

Radionuclide release pathways and scenarios are important to the analysis of a generic salt repository, and this could be true to any generic repositories. Additional studies are needed to improve the conceptual models for the release pathways and scenarios that are representative of a salt GDSE.

The salt GDSE model analysis has also identified the following important knowledge gaps to improve and enhance the confidence of the future repository performance analysis.

- Repository thermal loading by high-level radioactive waste, and the effect on the engineered barrier and near-field performance.
- Creep closure and consolidation of salt rocks under the influence of thermal perturbation, and the effect on the engineered barrier and near-field performance.
- Brine flows and radionuclide transport under the influence of thermal perturbation in generic salt repository environment, and the effect on the engineered barrier and near-field performance and far-field performance.
- Near-field chemistry and radionuclide mobility in generic salt repository environment (high ionic strength brines, elevated temperatures and reducing condition).
- Degradation of engineer barrier components (waste package, waste canister, waste forms, etc.) in a generic salt repository environment (high ionic strength brines, elevated temperatures and reducing condition).
- Waste stream types and inventory estimates, particularly for reprocessing high-level waste.

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TABLE 1. Elemental Solubility of Select Radionuclides in Near-Field Concentrated Brine

Element	Distribution Type	Solubility (molal)
U	Triangular	4.89E-08 (min); 1.12E-07 (mode); 2.57E-07 (max)
Pu	Triangular	1.40E-06 (min); 4.62E-06 (mode); 1.53E-05 (max)
Am	Triangular	1.85E-07 (min); 5.85E-07 (mode); 1.85E-06 (max)
Np	Triangular	4.79E-10 (min); 1.51E-09 (mode); 4.79E-09 (max)
Th	Triangular	2.00E-03 (min); 4.00E-03 (mode); 7.97E-03 (max)
Tc	Log-Triangular	4.56E-10 (min); 1.33E-08 (mode); 3.91E-07 (max)
Sn	Triangular	9.87E-09 (min); 2.66E-08 (mode); 7.15E-08 (max)
C, Cl, Cs, I, Se, Sr	n/a	Unlimited solubility

Note: Elements Ac, Cm, Nb, Pa, Pd, Ra, Sb, Zr are known to be solubility-limited, but are implemented as unlimited solubility in the near- and far-field model because their solubility calculations have not been completed.

TABLE 2. Elemental Solubility of Select Radionuclides for Far-Field Dilute Brine

Element	Distribution Type	Solubility (molal)
U	Triangular	9.16E-05 (min); 2.64E-04 (mode); 7.62E-04 (max)
Pu	Triangular	7.80E-07 (min); 2.58E-06 (mode); 8.55E-06 (max)
Am	Triangular	3.34E-07 (min); 1.06E-06 (mode); 3.34E-06 (max0
Np	Log-triangular	1.11E-06 (min); 1.11E-05 (mode); 1.11E-04 (max)

Element	Distribution Type	Solubility (molal)
Th	Triangular	8.84E-06 (min); 1.76E-05 (mode); 3.52E-05 (max)
Sn	Triangular	1.78E-08 (min); 4.80E-08 (mode); 1.29E-07 (max)
C, Cl, Cs, I, Se, Sr, Tc	n/a	Unlimited solubility

Note: Elements Ac, Cm, Nb, Pa, Pd, Ra, Sb, Zr are known to be solubility-limited, but are implemented as unlimited solubility in the near- and far-field model because their solubility calculations have not been completed.

TABLE 3. Far-Field Model Parameters for Marker Beds for the Reference Case

Parameter	Distribution Type	Parameter Value	Source
Thickness	Constant	1 m	Ref. 13
Porosity	Constant	0.01	
Density	Constant	2500 kg/m^3	
Brine flow rate below repository (m/yr)	Log-uniform	1.0E-08 (min); 3.0E-02 (max)	Ref. 2 (Fig. 7.6.2)
Brine flow rate away from repository (m/yr)	Log-uniform	1.0E-08 (min); 2.0E-02 (max)	Ref. 2 (Fig 7.6.5)
Longitudinal Dispersivity	Constant	10% of flow conduit length	
Kd for Radioelements (ml/g) :			
Uranium	Uniform	0.2 (min); 1 (max)	Refs. 14-17
Plutonium	Uniform	70 (min); 100 (max)	
Neptunium	Uniform	1 (min); 10 (max)	
Americium	Uniform	25 (min); 100 (max)	
Thorium	Uniform	100 (min); 1000 (max)	
Technetium	Uniform	0 (min); 2 (max)	
Cesium	Uniform	1 (min); 20 (max)	
Strontium	Uniform	1 (min); 80 (max)	
Carbon, chlorine, Selenium & Tin	Constant	0 (no sorption)	

Note: Kds for all other radionuclides not listed in the table were assumed zero (no sorption on the marker bed filling medium) because data was not available for the marker bed filling medium.

TABLE 4. Far-Field Model Parameters for Overlying Carbonate Aquifer for the Disturbed Case

Parameter	Distribution Type	Parameter Value	Source
Aquifer thickness	Constant	4 m	Ref. 14 (Table E-6); Ref. 18
Matrix porosity	Uniform	0.07 (min); 0.3 (max)	
Bulk density	Constant	2800 kg/m^3	
Matrix Tortuosity	Uniform	0.03 (min); 0.5 (max)	
Brine flow rate upward through borehole (m^3/yr)	Uniform	0.1 (min); 5.0 (max)	Ref. 1
Aquifer water flow rate (m/yr)	Log-uniform	3.15E-03 (min); 3.15E+01 (max)	Ref. 1 (Fig. 12.1.1)
Longitudinal Dispersivity	Constant	10% of flow conduit length	
Kd for Radioelements (ml/g) :			
Uranium	Uniform	0.03 (min); 20 (max)	Refs. 16-19
Plutonium	Log-uniform	20 (min); 1.0E+04 (max)	
Neptunium	Log-uniform	1 (min); 200 (max)	
Americium	Uniform	20 (min); 400 (max)	
Thorium	Log-uniform	7.0E+02 (min); 1.0E+04 (max)	
Technetium	Triangular	0 (min); 50 (mode); 100 (max)	
Cesium	Triangular	40 (min); 500 (mode); 3000 (max)	
Strontium	Triangular	5 (min); 13 (mode); 4.0E+04 (max)	
Iodine	Uniform	0.01 (min); 100 (max)	
Carbon, chlorine, Selenium & Tin	Constant	0 (no sorption)	

Note: Kds for all other radionuclides not listed in the table were assumed zero (no sorption on the marker bed filling medium) because data was not available for the aquifer medium.