

Understanding the Impact of Model Assumptions on Estimating Constituent Leaching during Disposal of Cementitious Waste Forms – 14382

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ABSTRACT

Cementitious waste forms, such as Salt Stone, Cast Stone and grouted wastes, are being used or considered for use for on-site disposal of low activity wastes at Savannah River and Hanford sites. Similar types of waste forms (e.g., solidified/stabilized waste) also are widely used for remediation and hazardous waste management. Waste form qualification and disposal facility waste acceptance criteria is often determined based on estimated release of radionuclides or other waste constituents under anticipated disposal conditions. However, the selection of the model and boundary conditions assumed for constituent release can impact the estimated release of constituents of concern by several orders of magnitude. Important conceptual model factors include representation of (i) constituent mass transport within the waste form (e.g., simple diffusion or coupled chemical speciation based mass transport), (ii) water composition, amount and conditions at the waste form interface (i.e., deionized water vs. percolate composition, infinite dilution vs. limited volume, annual average or intermittent infiltration), (iii) the extent of preferential flow pathways (e.g., engineered or cracking), and (iv) mechanisms of ingress of atmospheric gas phase reactants (i.e., oxygen and carbon dioxide). Use of a simplified leaching index representation as an evaluation basis for waste form performance can result in substantial misrepresentation of physical-chemical release mechanisms and anticipated leaching rates. This paper will provide an overview of impact of different key model assumptions by comparing release estimates for several radionuclides and inorganic species derived from a range of model assumptions.

INTRODUCTION

Cementitious or grouted materials have been used historically for a wide variety of applications in nuclear waste management including waste forms, containment structures, entombments, and environmental remediation for near-surface engineered disposal systems and also as structural concrete components for nuclear facilities (spent-fuel pools, dry spent-fuel storage units, and recycling facilities such as fuel fabrication, separations processes). Performance assessments often indicate that engineered barriers and treated wastes are needed to help limit hazardous and radioactive constituent releases from near-surface facilities into the environment protecting human health and biota.

Cementitious or grouted waste forms (e.g., Salt Stone at the Savannah River Site or Cast Stone proposed for the Hanford Site) are being or have been proposed for treating the low activity fraction of high-level wastes at these DOE sites and secondary waste at the Hanford Site [1]. Similar stabilized and solidified waste forms are in wide application internationally for remediation and hazardous waste management. Because of the long time frames that waste management applications are judged, reasonable predictions of waste form performance are needed. Without adequate predictive tools, assessments cannot reliably judge the effectiveness of cement barriers and waste forms used in containment or the waste zone.

The selection of assumptions, models, scenarios, and initial and boundary conditions to represent constituent release from cementitious materials can influence the predicted release of constituents of

potential concern by several orders of magnitude. Important conceptual factors for modeling cementitious materials include consideration and representation, where necessary, of the

- (i) constituent mass transport within the waste form (e.g., simple diffusion or coupled chemical speciation based mass transport),
- (ii) water composition, amount and conditions at the waste form interface (i.e., deionized water versus percolate composition, infinite dilution versus limited volume, annual average or intermittent infiltration),
- (iii) extent of preferential flow pathways (e.g., engineered or cracking), and
- (iv) mechanisms of ingress of atmospheric gas phase reactants (i.e., oxygen and carbon dioxide).

Furthermore, the common (and often required) use of a simple leaching index (Toxicity Characteristic Leaching Procedure) as the basis for waste form performance assessment often results in substantial misrepresentation of physical-chemical release mechanisms and resulting release and leaching rates. This paper provides an overview of the impact of different key model assumptions by comparing release predictions for several radionuclides and inorganic species based on a range of model assumptions.

DESCRIPTION OF THE MODELS USED

Two different prediction models available in LeachXS™ are examined in this paper: 1) cementitious monolith with diffusion and constituent leaching and 2) percolation (column) with radial diffusion and leaching. These models employ significantly different assumptions for very different scenarios involving cementitious materials. LeachXS is an expert system tool facilitating the characterization and evaluation of constituent leaching from a wide range of materials (e.g., secondary or recycled materials, stabilized waste and construction materials) under various conditions based on leaching test results [2,3]. LeachXS can also be used to evaluate field results for various scenarios based using either empirical relationships or mass transfer and geochemical speciation models to provide a source term for constituent leaching.

LeachXS includes a geochemical/reactive transport model (ORCHESTRA [4]) used to model liquid phase diffusion in the pore space of a concrete and the resulting reactions, constituent leaching, and diffusion [2]. This geochemical model can be used to evaluate diffusion from a monolith where system size, material layers, time frames, water contact and composition at the boundary can be varied to represent field scenarios. The model can also be used to model sulfate attack [5] as well as unsaturated conditions including gas phase transport of CO₂ and O₂ and the resulting carbonation of the concrete [6]. Other important degradation mechanisms (e.g., alkali-silica reaction) and scenario configurations will be considered in future studies.

Cementitious Monolith with Diffusion and Constituent Leaching

The first model considers a segmented monolithic solid (i.e., a true monolithic form or a compacted granular material compacted to act like a monolith) as a series of layers (cells) from the external boundary to the interior core as illustrated in Fig. 1 [2]. Within each layer or cell, the monolith segment is divided into gaseous, aqueous, and solid phases. Local equilibrium among phases is calculated in each cell at each time step to account for changes in pH and local composition based on dissolved constituent mass transport between the cells by diffusion through the liquid phase. As shown in Fig. 1, the external surface of one face of the monolith is considered in contact with an external well-mixed bath of finite volume (user defined) that is either refreshed at selected time intervals or continuously. This model can be used to evaluate impacts to release rates from changes in saturation in the monolith, eluate volume, eluate chemistry (e.g., influx of groundwater), and layering of material composition and properties within a monolith (e.g., a carbonated surface layer with an un-carbonated core).

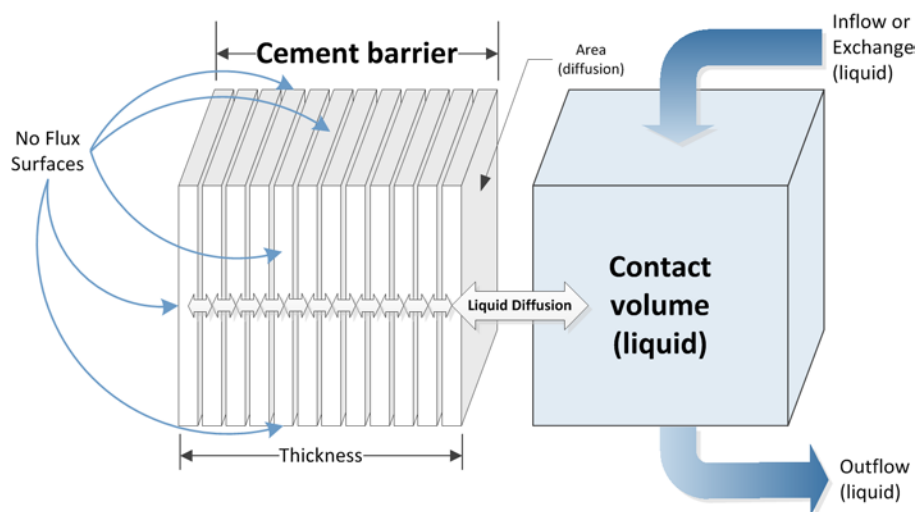


Fig. 1. 1-Layer mass transport prediction scenario (saturated or unsaturated with no gas interaction) [2].

The one-layer LeachXS/ORCHESTRA (LXO) model (Fig. 1) was used to simulate one-dimensional diffusion and resulting leaching from a cementitious monolith. The extent of soil that is considered in the model is 1-m based on previous experience (resulting in a 1 m³ soil cube assuming a 1 m² contact area for this study [6]). The soil boundary condition is represented by a single well-mixed cell that contains the volume, V_R , of pore water (accounting for soil saturation and porosity) assumed to be in near equilibrium with the pore water in the boundary cell of the monolith. For this study, the soil porosity (ϕ_{soil}) and saturation (S_{soil}) are assumed to be 0.38 (representing Hanford soil [7,8]) and 50% (to allow movement of soil gas), respectively. These values translate to a contact fluid volume of $V_R = (0.38) \times (0.50) \times (1 \text{ m}^3) = 0.190 \text{ m}^3$.

For the conceptual model represented in Fig. 1, the fluid in contact with the pore solution of the monolith is refreshed either at a selected refresh interval or continuously. For this study, a continuous refresh scheme based on the rate of infiltrating water is assumed. For example, the assumed infiltration rate at the Hanford Site can vary from 4 to 80 mm/yr (0.004 to 0.08 m/yr) [9] that translates into a continuous refresh rate of 2.41×10^{-10} to $4.82 \times 10^{-11} \text{ m}^3/(\text{m}^2 \cdot \text{s})$ based on V_R calculated above and referenced to a 1 m² contact area. Cases intended to represent infinite dilution were also considered. The model simulates transport at both saturated and a 60% saturated monolith to evaluate the impact of saturation on model predictions.

Percolation with Radial Diffusion and Leaching

The conceptual model for percolation with radial diffusion from porous solid particles (Fig. 2) consists of two zones segmented along the flow path, with one zone containing a mobile fluid phase in local equilibrium with the solid phase (percolation zone) and the second zone (matrix zone) containing porous spheres with an immobile fluid phase [2]. Mass transport occurs within the spheres via diffusion through the fluid phase with a boundary condition of equal fluid composition at the interface between the sphere surface and the percolation zone, and no diffusion at the center of the spheres [10,11]. Thus, constituents in the fluid phase can diffuse into and out of the spheres, and within the spheres where local solid-liquid equilibrium is maintained at each radial layer within the sphere. Within each column segment, each of the phases in the percolation zone is well mixed and in local equilibrium between the solid and liquid phases. This approach can be used to reflect systems where the diffusion gradients within the aggregate or matrix

(fractured) zone control release to the percolation zone. Thus, this model is sensitive to overall percolation flow rate and can be used to reflect the impact of fast infiltration that does not reach complete equilibrium between percolation and matrix zones. This approach can also be used to evaluate leaching under the effects of preferential flow, cracking in monoliths, varying flow conditions (e.g., intermittent flow, different flow rates), and solution chemistry [2].

The percolation model with radial diffusion in LeachXS/ORCHESTRA (LXO) (Fig. 2) has been used to simulate a cracked cementitious material. The behavior of the cracked material is controlled by a set of parameters including crack spacing (R), mobile fraction (w), percolation zone porosity (ϕ_m), matrix zone porosity (ϕ_{im}), and total porosity (ϕ_t). The column was designed to represent the conditions similar to those in the monolith diffusion scenario. However, due to space limitations in this paper, only the results from the monolith diffusion scenario will be the focus of this paper. It should be noted that due to the much larger contact area (including cracks) of the material in the percolation column, the leaching results tend to be significantly higher than those in the monolith cases that will be presented.

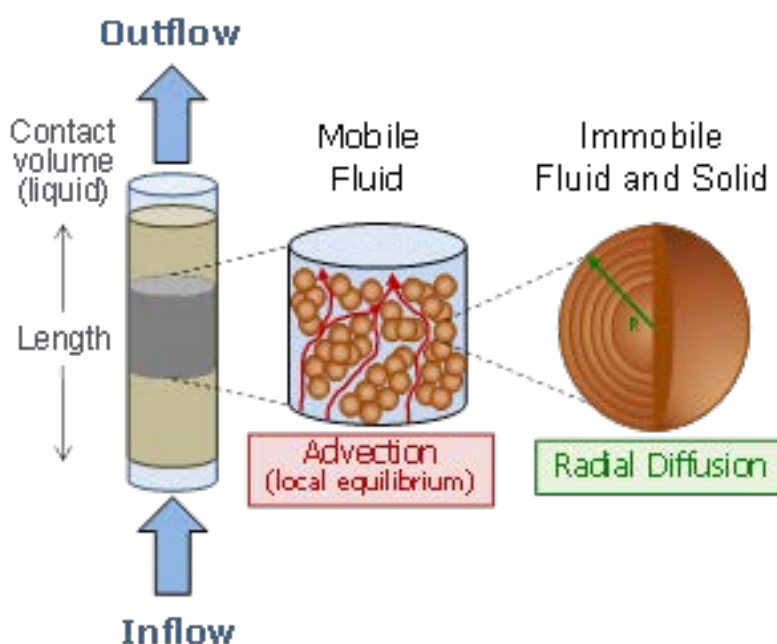


Fig. 2. Conceptual model of percolation with radial diffusion for an up-flow column [2].

Reactions Databases Used in this Study

A reference database for cementitious materials primarily incorporating the reactions from the MINTEQV4 [12] and CEMDATA07 [13] databases is available for LeachXS users from the Cementitious Barriers Partnership (CBP) program (cementbarriers.org) with support from DOE Office of Environmental Management. A database has recently been extended (in a separate “patch” file) that implements the Nuclear Energy Agency (NEA) Thermochemical Database (TDB) Project (<https://www.oecd-neo.org/dbtdb/>) reactions. The NEA database includes equilibrium reactions for various radionuclides (i.e., U, Pu, Np, Am, Tc, and I) important for nuclear waste disposal. Both reaction databases (denoted “REF” and “NEA”) are used in this paper; the minerals selected from these databases are presented in Table I.

TABLE I. Mineral Phase Reactions Considered in this Study

REF Reactions [12,13]		REF [12,13] + NEA Reactions (https://www.oecd-nea.org/dbtdb/)		
AA_Fe[OH]3[am]	Fluorite	AA_Fe[OH]3[am]	Cem07_CAH10	Schoepite
BaSrSO4[50%Ba]	Ni[OH]2[s]	AA_Magnesite	Cem07_Calcite	SiO2[a]
Ba[SCr]O4[96%SO4]	Pb2V2O7	Am2[CO3]3[am]	Cem07_Gypsum	Strengite
Ca4Cd[PO4]3OH	Pb3[VO4]2	Am[OH]3[am]	Cem07_Portlandite	Tc2O7_H2O[s]
CaSeO4:2H2O	PbMoO4[c]	BaSrSO4[50%Ba]	Cem07_Syngenite	TcO2[cr]
Ca[Sb[OH]6]2[sAJ]	Pb[OH]2[C]	Ba[SCr]O4[96%SO4]	Cr[OH]3[A]	Tenorite
Ca[Sb[OH]6]2[sC]	Strengite	Ca3[AsO4]2:2.25H2O	Fe_Vanadate	ThF4:2.5H2O
Cd[OH]2[C]	Tenorite	Ca4Cd[PO4]3OH	Fluorite	Th[OH]4[s]
Cem07_Al[OH]3[am]	ThF4:2.5H2O	CaSeO4:2H2O	Manganite	Thorianite
Cem07_Anhydrite	Willemite	Ca[Sb[OH]6]2[sAJ]	NP2O5[cr]	UO3[C]
Cem07_Brucite	alpha-TCP	Ca[Sb[OH]6]2[sC]	Na3NpO2[CO3]2[cr]	Uranophane
Cem07_C2AH8 ^a	beta-TCP	Cd[OH]2[C]	Ni[OH]2[s]	Willemite
Cem07_C2ASH8	Cem07_C4AsH12 ^b	Cem07_Al[OH]3[am]	NpO2CO3[s]	alpha-TCP
Cem07_C2FH8	Cem07_C4FsH12 ^b	Cem07_Anhydrite	NpO2[OH]2[cr]	beta-TCP
Cem07_C3AH6	Cem07_SiO2[am] ^b	Cem07_Brucite	Pb2V2O7	Cem07_C4AsH12 ^b
Cem07_C3FH6	Cem07_Tob_I ^b	Cem07_C2AH8	Pb3[VO4]2	Cem07_C4FsH12 ^b
Cem07_C4AH13	Cem07_Jenn ^b	Cem07_C2ASH8	PbMoO4[c]	Cem07_C6As3H32 ^b
Cem07_C4FH13	Cem07_Tob_II ^b	Cem07_C2FH8	Pb[OH]2[C]	Cem07_C6Fs3H32 ^b
Cem07_CAH10		Cem07_C2FSH8	Pu2O3[cr]	Cem07_SiO2[am] ^b
Cem07_Calcite		Cem07_C3AH6	PuO2CO3[s]	Cem07_Tob_I ^b
Cem07_Gypsum		Cem07_C3FH6	PuO2[OH]2.H2O[cr]	Cem07_Jenn ^b
Cem07_Portlandite		Cem07_C4AH13	Pu[OH]3[cr]	Cem07_Tob_II ^b
Cem07_Syngenite		Cem07_C4FH13	Sb[OH]3[s]	

a. Some of the CEMDATA07 (denoted here with the “Cem07_” prefix) used cement notation: C = CaO (lime), S = SiO₂ (silica), A = Al₂O₃ (alumina), F = Fe₂O₃ (iron oxide), and H = H₂O (water),

b. Solid solution phase

Materials Used in this Study

A material denoted the Analogous Waste Form (AWF) was used as the basis for the hypothetical material considered in this study. This material represents a well-characterized reference cementitious formulation for a typical DOE salt waste solution stabilized with a reference premix (ordinary portland cement, blast furnace slag, and fly ash) [14]. The waste water contribution to AWF consists of various sodium and aluminum salts (NaOH, NaNO₃, NaNO₂, Na₂CO₃, Al(NO₃)₃•9H₂O, Na₂SO₄, and Na₃PO₄•12H₂O) representative of Savannah River low-activity waste. The AWF is a highly porous material ($\phi_{AWF} = 0.45$).

Two leachates are used in this study. The first is deionized water (where all non-water constituents are considered to have concentrations of 10⁻¹³ mol/kg). The second leachate used in this study has a composition (Table II) representing Hanford ground water.

TABLE II. The Hanford Ground Water Composition Used in this Study (mol/m³)

Ag ⁺	Al[OH] ₄ ⁻	Ba ⁺²	Br ⁻	Ca ⁺²	Cd ⁺²	Cu ⁺²	Fe[OH] ₄ ⁻	F ⁻
7.79E-11	1.00E-04	3.64E-08	2.42E-10	2.11E-05	4.46E-11	1.57E-06	3.58E-07	3.48E-10
K ⁺	Li ⁺	Mg ⁺²	Na ⁺	Ni ⁺²	Pb ⁺²	SO ₄ ⁻²	Sr ⁺²	Zn ⁺²
6.39E-05	7.20E-06	8.56E-06	4.35E-06	8.52E-07	2.42E-10	5.69E-10	3.62E-08	7.65E-08

MODELING RESULTS

A series of simulations were run in LeachXS/ORCHESTRA to examine the sensitivity of the monolith diffusion model shown in Fig. 1 to different key model assumptions and parameters. Cumulative release predictions were compared for several radionuclides and inorganic species derived from a range of model assumptions. The simulations run are summarized in Table III.

TABLE III. Simulations Run for the Monolith Diffusion Model
(Material AWF, S_{soil} 50%, ϕ_{soil} 0.38, V_R 0.19 m³, Tortuosity 15, Area 1 m², Width 1 m, ϕ_{AWF} 0.45)

Case	Database	IR (m/y) ^a	F (mL/m ² /h)	F (m/s) ^b	Leachate ^c	S_{AWF} (%)	pH+pe ^d
00	REF	13.83	300	8.33E-08	DI	100	14.5
00a	REF	138.3	3000	8.33E-07	DI	100	14.5
01	REF	0.08	1.735	4.82E-10	DI	100	14.5
02	REF	0.004	0.087	2.41E-11	DI	100	14.5
04	REF	0.08	1.735	4.82E-10	DI	60	14.5
04a	REF	0.004	0.087	2.41E-11	DI	60	14.5
07	REF	13.83	300	8.33E-08	HGW	100	14.5
08	REF	0.08	1.735	4.82E-10	HGW	100	14.5
08a	REF	0.004	0.087	2.41E-11	HGW	100	14.5
10	REF	0.08	1.735	4.82E-10	HGW	60	14.5
10a	REF	0.004	0.087	2.41E-11	HGW	60	14.5
13	REF+NEA	13.83	300	8.33E-08	DI	100	14.5
14	REF+NEA	0.08	1.735	4.82E-10	DI	100	14.5
15	REF+NEA	0.004	0.087	2.41E-11	DI	100	14.5
17	REF+NEA	0.08	1.735	4.82E-10	DI	60	14.5
17a	REF+NEA	0.004	0.087	2.41E-11	DI	60	14.5
20	REF+NEA	13.83	300	8.33E-08	HGW	100	14.5
21	REF+NEA	0.08	1.735	4.82E-10	HGW	100	14.5
22	REF+NEA	0.004	0.087	2.41E-11	HGW	100	14.5
24	REF+NEA	0.08	1.735	4.82E-10	HGW	60	14.5
24a	REF+NEA	0.004	0.087	2.41E-11	HGW	60	14.5
28	REF+NEA	13.83	300	8.33E-08	HGW	100	11
29	REF+NEA	0.004	0.087	2.41E-11	HGW	100	11
30	REF+NEA	0.004	0.087	2.41E-11	HGW	60	11

- I_R (m/yr) is the selected infiltration rate for the scenario.
- The area used to convert is 1 m².
- DI is deionized water and HGW is Hanford Ground Water.
- More oxidized conditions represent a pH+pe of approximately 14.5; whereas, mildly reduced conditions represent a pH+pe of approximately 11.

Fig. 3 shows the impact of leachate, flow, and saturation on the predicted pH in the external fluid in contact with the AWF material for the first 100 years. The contacting fluid pH values for the Hanford ground water cases are generally higher than those when deionized water is used for the leachate. For the deionized water cases, there is at most approximately 1 pH unit (representing an order of magnitude) variation among the cases. For Hanford ground water, there is a difference of approximately 1.5 pH units. The corresponding pH versus time relationships in the AWF material pore solution directly in contact with the external fluid for the various flow cases is shown in Fig. 4. Note the variation in the pore solution is generally on the same order of magnitude as that in the contacting fluid (Fig. 3).

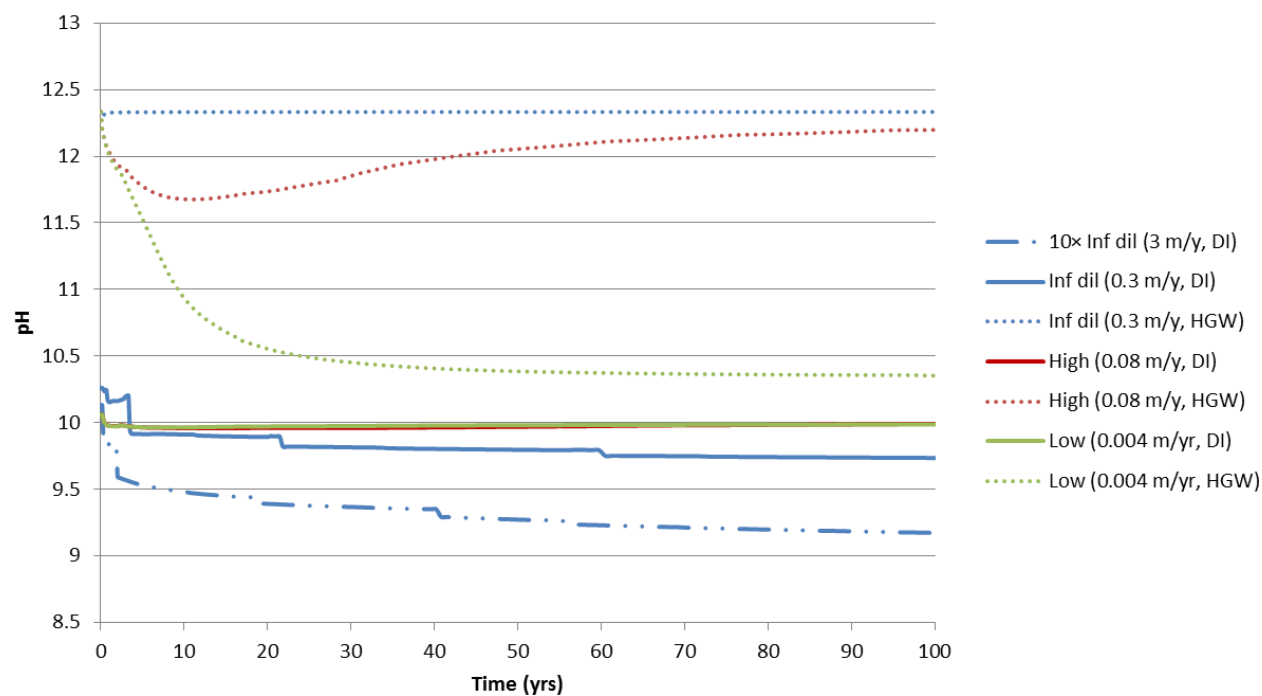


Fig. 3. Comparison of pH results in the external fluid in contact with the material showing the impact of flow and leachate (deionized water versus Hanford ground water) on AFW using the reference database (Table I). Note material saturation had little impact on the results for the pH in contacting fluid.

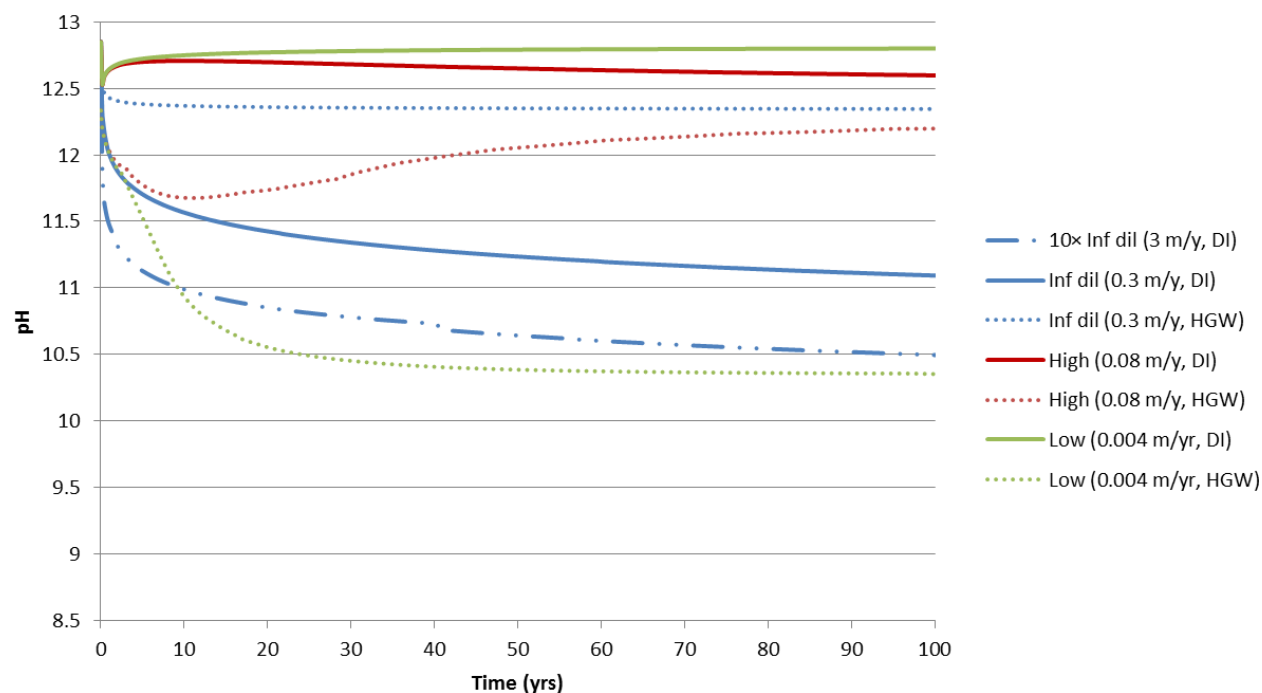


Fig. 4. Comparison of pH results in the material pore solution in (just inside the material directly in contact with the external fluid) showing the impact of flow and leachate (deionized water versus Hanford ground water) on the AFW material using the reference (REF) database.

Fig. 5 illustrates the impact of flow and saturation on the predicted cumulative leach results (using the reference database and deionized water as the leachate) for a major non-reactive constituent (namely Na⁺) in the external fluid in contact with the AWF material for the first 100 years. Note that the difference between the low and high flow cases results in approximately a 50% increase in the cumulative Na⁺ in the contacting fluid. The impact of the AWF saturation (from 100% to 60% saturation) decreases the cumulative Na⁺ in the contacting fluid by more than one-third for the higher flow case and one-fourth in the lower flow case.

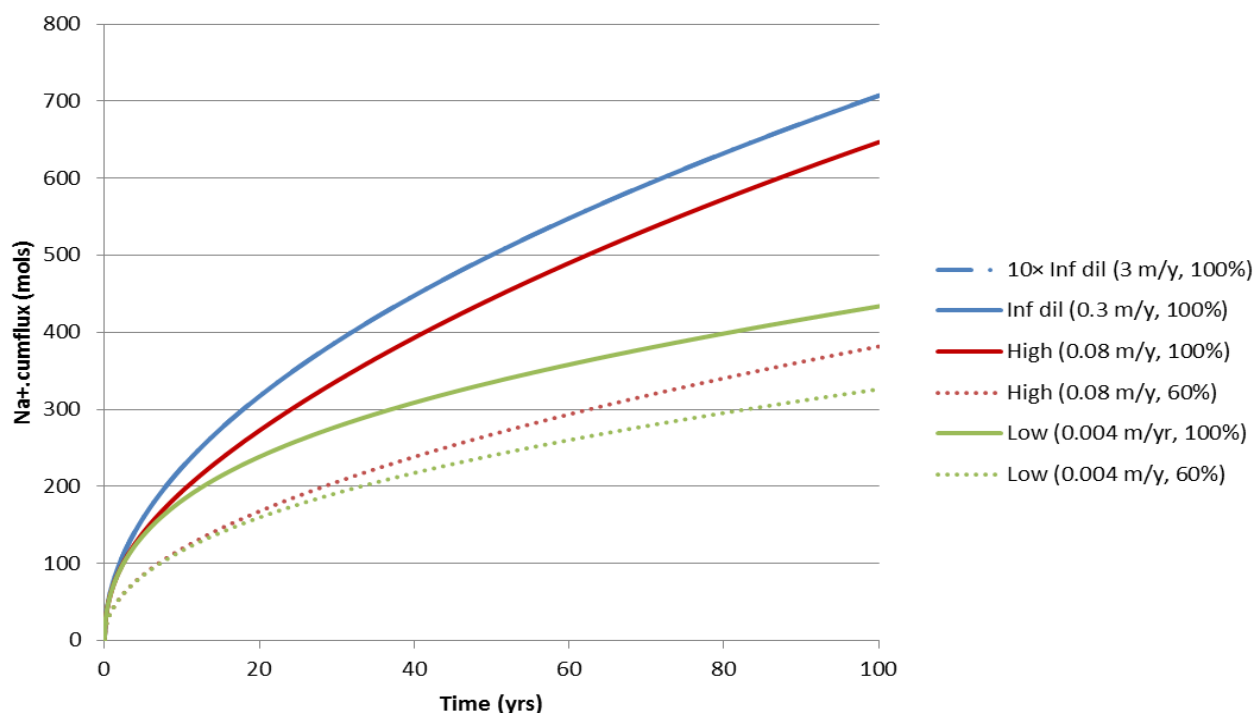


Fig. 5. Comparison of cumulative leach results for a non-reactive constituent (Na⁺) showing the impact of flow and saturation on the AFW material using the reference (REF) database. Note that the 10× infinite dilution case is coincident with the infinite dilution results.

Fig. 6 illustrates the impact of flow and saturation on the predicted cumulative leach results (using the reference database and deionized water as the leachate) for a major reactive constituent (namely Ca²⁺) in the external fluid in contact with the AFW material over the first 100 years. Note that the difference between the low and high flow cases results in approximately a four-fold increase in the cumulative Ca²⁺ in the contacting fluid (with a similar impact between the high flow and infinite dilution cases). The solubility of calcium is dependent on pH. The impact of AFW material saturation (from 100% to 60% saturation) has only a minor impact on the cumulative release results for Ca²⁺.

Fig. 7 illustrates that the leachate used in the model can have a large impact on the cumulative release for a reactive constituent such as Ca²⁺. The influence of the leachate used can increase leaching by approximately a factor of two for higher flow conditions and may even result in surface precipitation or reverse the diffusion (into the solid material) at lower flow conditions over the first 100 years. These results (i.e., decreasing cumulative amounts) for the cases using Hanford ground water (Table II) as the leachate likely reflect the reprecipitation of constituents (e.g., Ca²⁺) at the material interface with the contacting fluid (Fig. 1).

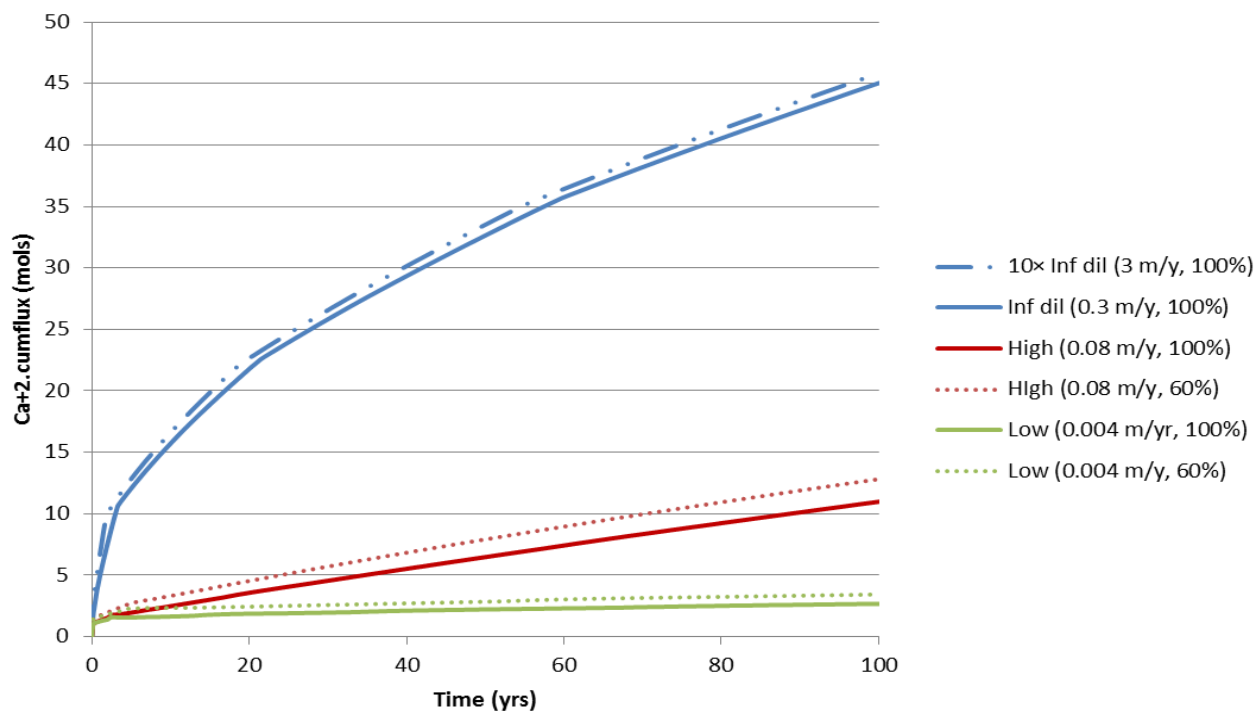


Fig. 6. Comparison of cumulative leach results for a reactive constituent (Ca^{+2}) showing the impact of flow and material saturation on AFW leaching using the reference (REF) database and deionized water.

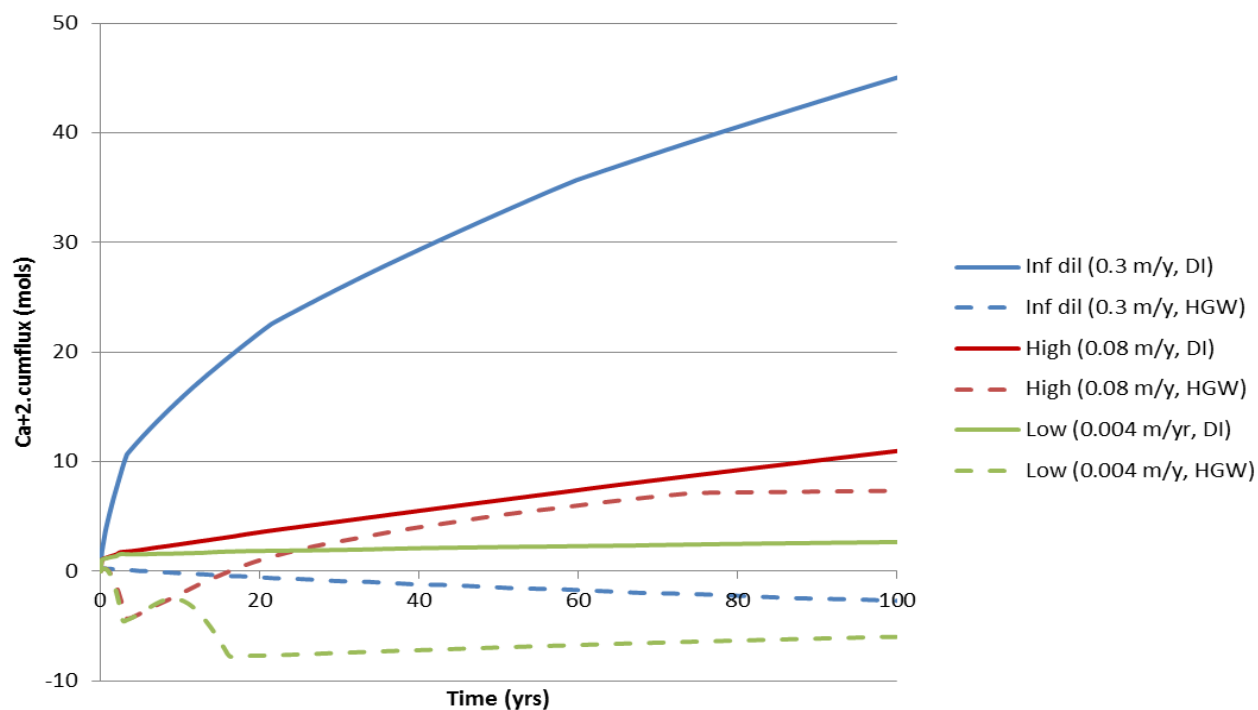


Fig. 7. Comparison of cumulative leach results for a reactive constituent (Ca^{+2}) showing the impact of leachate (deionized water versus Hanford groundwater (Table II)) on the AFW material using the reference (REF) database at 100% saturation.

Fig. 8 indicates that the thermodynamic database used had little impact on the cumulative release of Ca^{+2} (for the conditions studied). However, a much larger impact can be found depending on whether or not mineral reactions are selected for a constituent. As shown in Fig. 9, the differences between cumulative releases for uranium (as OU^{2+}) vary by up to eight orders of magnitude depending on whether or not mineral reactions (in this case, $\text{UO}_3[\text{C}]$ and uranophane) are selected (Table I). Additional investigation of the potential impacts of selected minerals and reaction databases is underway.

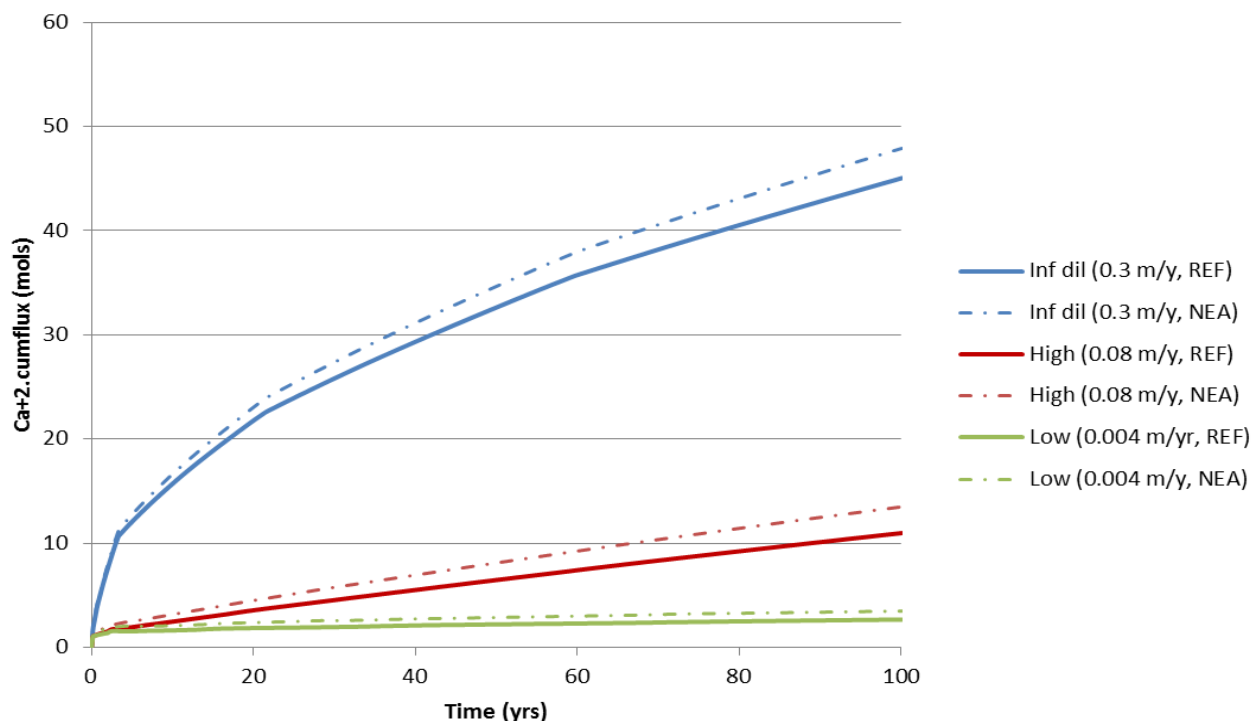


Fig. 8. Comparison of cumulative leach results for a reactive constituent (Ca^{+2}) showing the impact of the thermodynamic database used (Reference versus NEA) on the AFW material using the deionized water at 100% saturation.

The final comparisons discussed in this paper evaluate the impact of the redox state of the material on leaching results. A hypothetical reduced version of the AFW material was developed by “adding” a small amount of slag (sulfur) and changing the pH+pe from approximately 14.5 for the more oxidized case to approximately 11 for the reduced case (Table III). The effect was expected to result in the formation of Cr-containing mineral species at the reduced conditions, which would reduce the availability of chromium for leaching and release. This impact can be observed in Fig. 10 where the cumulative release from the AFW decreased by more than an order of magnitude. The leaching characteristics of other important constituents (including Tc) are strongly influenced by the redox state of the material. The result that the cumulative releases are not generally increasing (or flat) for the case using Hanford ground water (as the leachate) at the lower Hanford flow rate (0.004 m/yr) likely again reflects the reprecipitation of constituents at the material interface with the contacting fluid (Fig. 1).

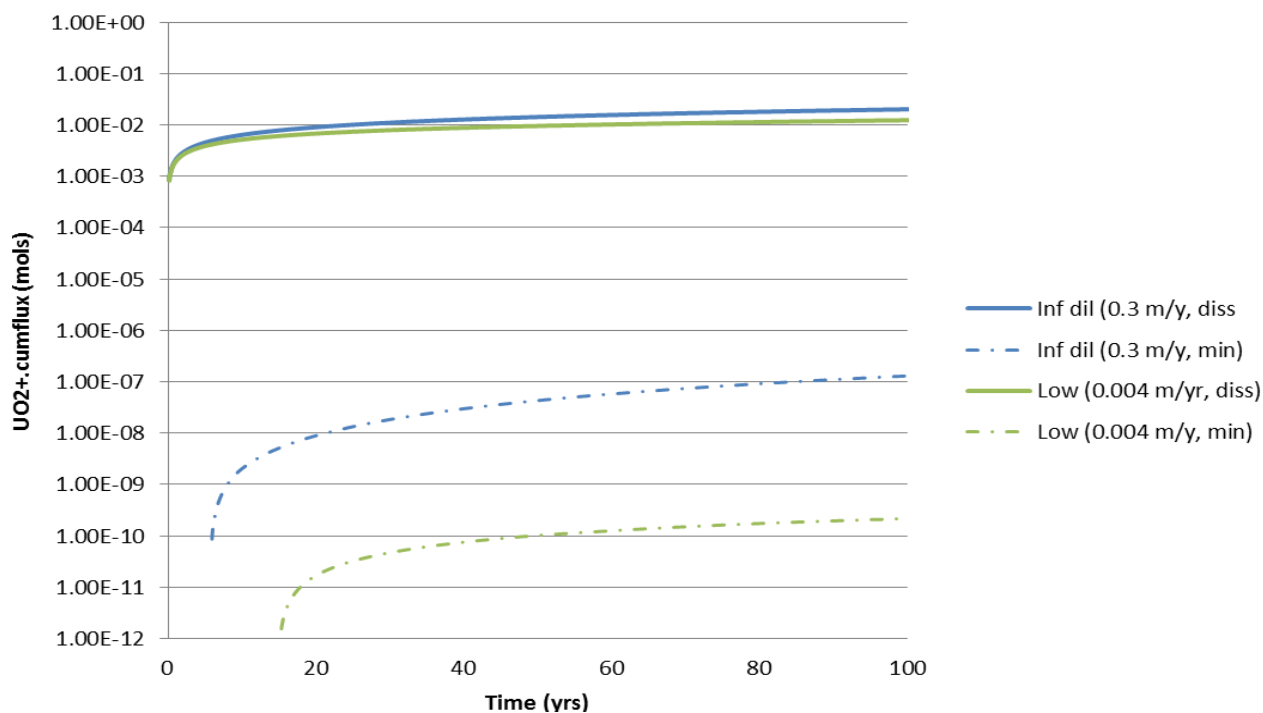


Fig. 9. Comparison of cumulative leach results for a reactive, radioactive constituent (e.g., a U isotope as UO_2^+) showing the impact of not including mineral reactions (diss) versus including mineral reactions (min) on the AFW material using the deionized water at 100% saturation and oxidized conditions.

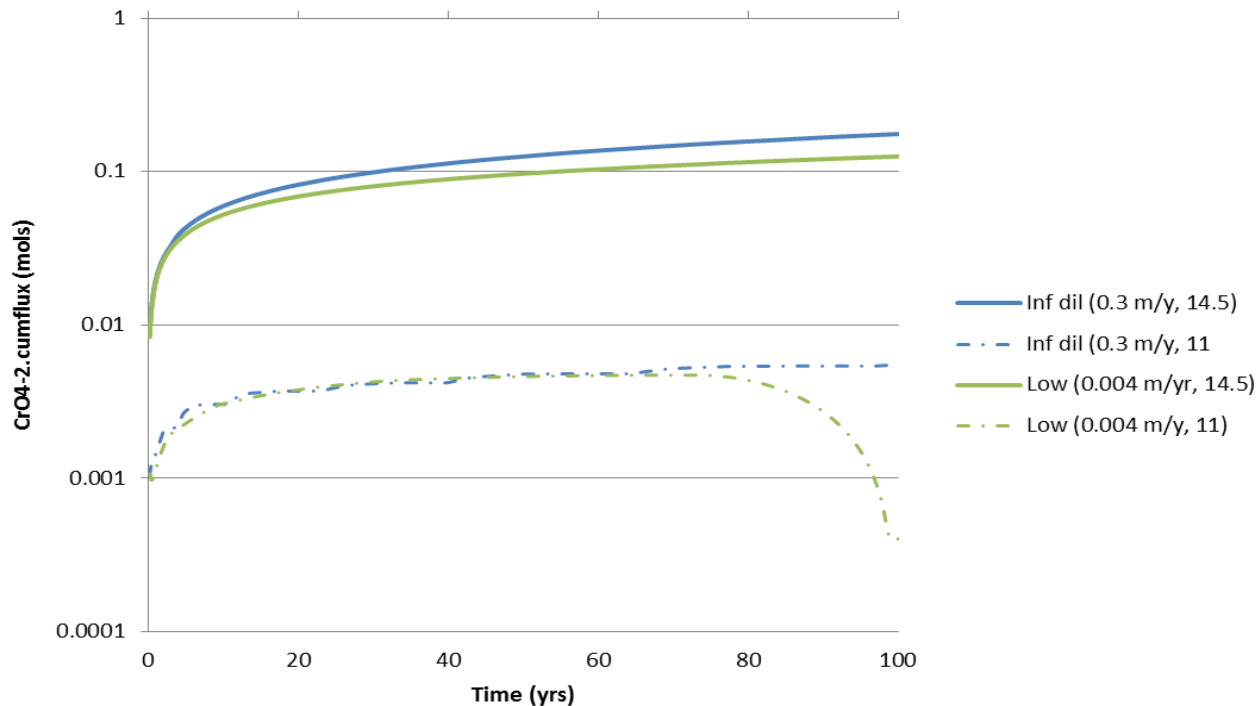


Fig. 10. Comparison of cumulative leach results for a reactive, non-radioactive constituent (Cr as CrO_4^{2-}) showing the impact of redox conditions (pH+pe 14.5 versus 11) on the AFW material using the NEA database and Hanford ground water (Table II) at 100% saturation.

CONCLUSIONS

The results presented in this paper provide an overview of the potential order of magnitude impacts of different key model assumptions by comparing release estimates for several radionuclides and inorganic species derived from a range of model assumptions for a waste monolith. The results indicate that the impact of differing flow rates (continuous) can decrease constituent release by up to a factor of four over the first 100 years. The influence of the leachate used can approximately halve leaching for higher flow conditions and may result in near surface precipitation at lower flow conditions. The selection of mineral reactions indicated a predicted decrease in cumulative flux for uranium of up to eight orders of magnitude. A simple evaluation of the impact of the material redox state on chromium release indicated a potential decrease in the cumulative release by an order of magnitude over the 100-yr simulation period. Thus it is often critical to understand the sensitivity of leaching to selected mineral reactions and the material redox state.

Thus the selection of the model and boundary conditions assumed for constituent release impact the estimated cumulative release of constituents of concern by up to several orders of magnitude. Similar results, when corroborated by laboratory and field measurements, could have a dramatic impact on the decision making process for selected constituents of potential concern. These results highlight that the common use of a simple leaching index as the basis for waste form performance assessments can often result in substantial misrepresentation of physical-chemical release mechanisms and resulting release and leaching rates.

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