

The Role of Solubility as a Barrier to Radionuclide Release

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Abstract – *The release of radionuclides from waste forms is limited by the extent to which the waste form degrades due to interaction with water inside the failed waste package. The release of radionuclides to the environment is reduced further if the radionuclides exhibit limited solubility in the transporting water. Intuitively, it is expected that radionuclides with low solubility would exhibit smaller releases to the environment relative to those radionuclides with high solubility. In this study, performance assessment calculations were conducted using the Total-system Performance Assessment (TPA) Version 4.1 code to determine the parameters that control the solubility limited behavior of radionuclides and the extent to which radionuclide solubility serves as a barrier to radionuclide release to the environment. The results show that the causal relationship between solubility and radionuclide release is not simple. The degree to which a radionuclide is released to the environment depends on the complex interaction between the degree of waste form exposure to water, radionuclide half-life, and radionuclide initial inventory. Under some conditions, radionuclides with low solubility may not experience solubility limited release whereas the release of radionuclides with high solubility can be solubility limited. In addition, the waste form dissolution model, the leaching surface area model, and the length of the analysis period significantly affect the solubility limited behavior of radionuclides. Of the four radionuclides that are major contributors to dose (Cl-36, Tc-99, I-129, and Np-237), only the release of Np-237 is significantly affected by its solubility.*

I. INTRODUCTION

The U.S. Nuclear Regulatory Commission (NRC) and the Center for Nuclear Waste Regulatory Analyses have jointly developed performance assessment tools for conducting a technical review of a potential license application by the U.S. Department of Energy (DOE) for the proposed Yucca Mountain high-level nuclear waste repository. An important application of these tools is the identification of subsystems, processes, barriers, or factors that may significantly affect repository performance. The Total-system Performance Assessment (TPA) Version 4.1 code (a modified version of the TPA 4.0 code [1]), along with its supporting modules and embedded techniques, is one such tool that can be used to estimate the relative importance of various physical features, processes, or repository subsystems in controlling potential human exposure to radionuclides.

The code enhances NRC staff capabilities in performance assessment and license application review [2].

For performance assessment calculations, communication between system and process level analysts can be facilitated if the features, events, and processes that determine repository system behavior are divided into components that could serve as potential barriers to the flow of water or to the transport of radionuclides. One of these potential barriers (or controlling factors) is the radionuclide solubility. The release of radionuclides from the waste form and the waste package may be limited by their solubility in the volume of water contacting the waste form and transporting the radionuclide. In this study, performance assessment calculations were conducted using the TPA 4.1 code to determine the extent to which solubility serves as a barrier to radionuclide release over the regulatory period of interest (10,000 yr).

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II. DESCRIPTION

II.A. Implementation of Solubility Limits in the Total-system Performance Assessment Code

In performance assessments, the approach typically used to bound the aqueous concentration of radionuclides in water is to derive initially the radionuclide concentrations using a waste form dissolution model. In the TPA 4.1 code and its predecessor Version 4.0, a number of parameters that control the release of radionuclides from the spent fuel matrix is included (e.g., fraction of spent fuel that is wet, particle size of the spent fuel, alteration rate of UO_{2+x} , and cladding protection) [1]. After leaching of radionuclides from the spent fuel waste form, a comparison is made between the waste form dissolution-based aqueous concentration of each radionuclide considered and a value for the solubility that is either thermodynamically derived or based on a bounding assumption. If the concentration derived from the waste form dissolution model exceeds the solubility, the aqueous concentration is set to the solubility value and the difference in mass is assumed to precipitate out of the solution. Therefore, solubility can place a constraint on the aqueous concentration of a particular radioelement even if that radioelement is abundant [3].

The solubility of a particular radionuclide will depend on assumptions regarding (i) the likely radionuclide-bearing solid phase (either a solid phase with the radioisotope as the dominant element or a solid phase with trace amounts of the radionuclide, as in coprecipitated species) and (ii) the chemistry of the fluid that reacts with the solid. The stable solid phases depend on temperature, redox conditions, and chemical composition of the groundwater in contact with the waste

form. Because of uncertainty in these variables in the waste package and near-field environment, there is a wide range of possible solubilities. Thus, the ranges in solubility values for some radionuclides are represented in TPA 4.1 in the form of probability distribution functions. Also, some radionuclides are assumed to be highly soluble because no solubility controlling solids for these radioelements are expected to form under geochemical conditions relevant to a Yucca Mountain repository.

II.B. Water Contact Model

The TPA code uses two water contact modes for dissolution of the spent fuel waste form: (i) immersion (bathtub model) and (ii) dripping (flow-through model) (see Fig. 1) [3]. In the bathtub model, water must fill the failed waste package to an assumed overflow height before radionuclides are released from the waste package. In the flow-through model, the fraction of fuel wetted is equivalent to the fraction immersed in water in the bathtub model, but the radionuclides are released after waste package failure without the requirement for water to fill the waste package to a certain level. The bathtub model assumes that the waste package is filled by water entering through a defect and eventually overflows at a future time. The flow-through model assumes liquid enters and leaves the waste package in a continuous stream, but only a small amount of water is continuously in contact with the spent fuel.

II.C. Waste Form Inventory

Radionuclide inventories change with time due to radioactive decay and ingrowth. Most radionuclide

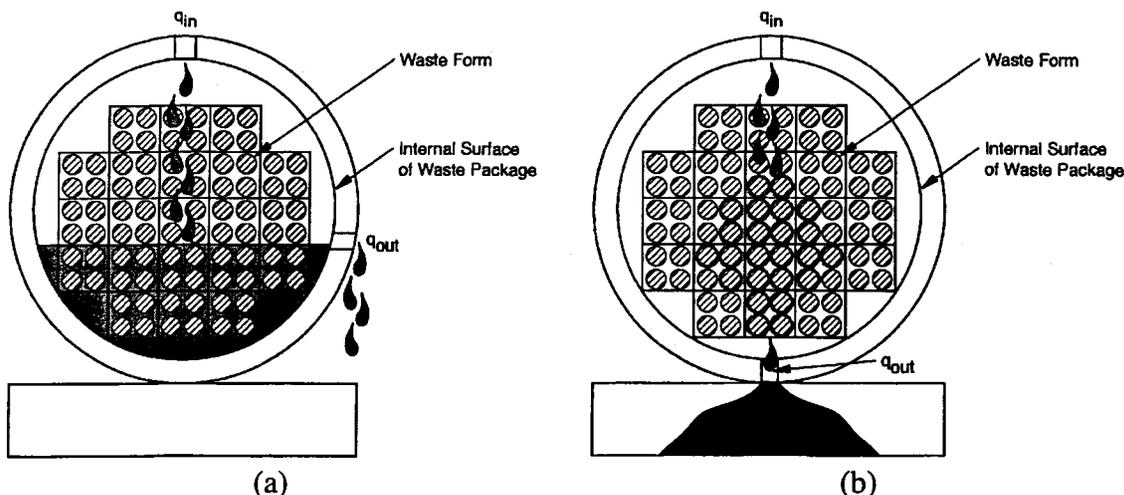


Fig. 1. Schematics showing (a) bathtub and (b) flow-through waste form wetting mode inside a failed waste package

inventories decrease significantly with increasing time, whereas some show a much smaller decrease over long periods of time (those with long half-lives) and others increase with time (daughters in a decay chain). For example, Pu-238 has an 87.7-yr half-life and its inventory quickly decreases with time. On the other hand, U-234 has a 244,500-yr half-life and its inventory decreases by radioactive decay only 25 percent during 100,000 yr. An example of daughter ingrowth can be seen with Th-230, Ra-226, and Pb-210, which are in the Cm-246 decay series of radionuclides. The inventories of these daughter radionuclides increase with time.

For the release and transport calculations, twenty radionuclides in the waste form are modeled in TPA 4.1. These are U-238, U-234, Pu-239, Pu-240, Nb-94, Am-241, Am-243, Np-237, Th-230, Cm-245, Cm-246, Ra-226, Ni-59, Pb-210, Tc-99, Cs-135, I-129, Se-79, Cl-36, and C-14. These radionuclides were identified in a screening process as having the potential to significantly contribute to dose in

the biosphere [1]. The half-life and initial inventory of these radionuclides are listed in Table 1. Four decay chains are also modeled: (i) Cm-246→U-238, (ii) Cm-245→Am-241→Np-237, (iii) Am-243→Pu-239, and (iv) U-234→Th-230→Ra-226→Pb-210.

II.D. Waste Form Dissolution Rate

Four waste form dissolution models are used in the TPA code to compute the source term for unsaturated and saturated zone radionuclide transport [4]. These models represent alternative conceptualizations of waste form release rates and are expected to bound the uncertainties associated with the waste form chemical environment subsequent to waste package failure [1]. Model 1 estimates the spent fuel dissolution rate in waters containing carbonate anions. The waste form dissolution rate in the presence of Ca and Si ions, which are found in Yucca Mountain groundwater, is calculated in Model 2.

Table 1. List of 20 radionuclides considered in groundwater release with their half-lives, initial waste package inventories, and solubilities

Radionuclide	Half-life (yr)	Initial WP Inventory (Ci/MTU at 10 yr)	Solubility (kg/m ³) (Distribution function type and data range)
C-14	5,729	1.44 × 10 ⁰	constant: 1.4 × 10 ¹
Cl-36	301,000	1.15 × 10 ⁻²	constant: 3.6 × 10 ¹
Ni-59	80,000	2.44 × 10 ⁰	constant: 1.1 × 10 ⁻¹
Se-79	64,960	4.58 × 10 ⁻¹	constant: 7.9 × 10 ¹
Nb-94	20,300	8.48 × 10 ⁻¹	constant: 9.3 × 10 ⁻⁷
Tc-99	213,000	1.45 × 10 ¹	constant: 9.93 × 10 ¹
I-129	15,700,000	3.57 × 10 ⁻²	constant: 1.29 × 10 ²
Cs-135	2,300,000	5.36 × 10 ⁻¹	constant: 1.35 × 10 ²
Pb-210	22.3	5.67 × 10 ⁻⁸	constant: 6.6 × 10 ⁻⁵
Ra-226	1,600	4.11 × 10 ⁻⁷	constant: 2.3 × 10 ⁻⁵
Th-230	77,000	1.37 × 10 ⁻⁴	constant: 2.3 × 10 ⁻⁴
U-234	244,500	1.18 × 10 ⁰	constant: 7.6 × 10 ⁻³
Np-237	2,140,000	4.34 × 10 ⁻¹	logtriangular: 1.2 × 10 ⁻³ , 3.4 × 10 ⁻² , 2.4 × 10 ⁻¹
U-238	4,468,000,000	3.15 × 10 ⁻¹	constant: 7.6 × 10 ⁻³
Pu-239	24,060	3.69 × 10 ²	uniform: 2.4 × 10 ⁻⁶ , 2.4 × 10 ⁻⁴
Pu-240	6,537	5.44 × 10 ²	uniform: 2.4 × 10 ⁻⁶ , 2.4 × 10 ⁻⁴
Am-241	432.2	2.08 × 10 ³	uniform: 2.4 × 10 ⁻⁸ , 2.4 × 10 ⁻⁴
Am-243	7380	2.64 × 10 ¹	uniform: 2.4 × 10 ⁻⁸ , 2.4 × 10 ⁻⁴
Cm-245	8499	3.66 × 10 ⁻¹	constant: 2.4 × 10 ⁻⁴
Cm-246	4731	7.62 × 10 ²	constant: 2.4 × 10 ⁻⁴

The solubility is represented by different types of distributions. For the logtriangular distribution, the center value is the most likely value. For the uniform distribution, the minimum and maximum values are shown.

Model 3 utilizes a dissolution rate derived from a natural analog, whereas Model 4 considers the formation of secondary minerals such as schoepite. The mathematical formulations for these models and their associated assumptions are given elsewhere [1,5] and are not in the scope of this paper.

II.E. Waste Form Surface Area

Two models for determining the spent fuel surface area are available in TPA 4.1. The first model determines surface area using fragmented pellets (i.e., particles), whereas the second model uses the waste form grain size criterion. The particle model assumes that the waste form is fragmented into small spherical particles (~1 mm diameter) and that the intergranular porosity does not contribute to the surface area. If subgranular fragmentation of the waste form takes place through fuel conversion from UO_2 to $UO_{2.4}$ and U_3O_8 , a smaller particle size (i.e., equivalent) could be considered in the waste form particle model to represent additional exposed surface area. In the second model, waste form grains are exposed.

The surface area available for leaching conservatively is held constant throughout the leaching period, even though the radius of the unoxidized fuel grains or particles would diminish with time. This simplification is most likely to affect the estimated peak dose. In addition, preferential attack on grain boundaries of fuel particles and the effect of ionizing radiation were not considered. The effect of ionizing radiation is complex and potentially could lead to higher release rates. Factors such as the age of the waste, thickness of water film, cladding protection, and protectiveness of secondary mineral layers on the fuel would have to be considered to be able to account for the effect of ionizing radiation.

II.F. Implementation in the System Model

For the analysis presented in this paper, the model parameters and the associated parametric uncertainties selected for the basecase—defined as the most likely scenario—in the TPA 4.1 code were used. The release of radionuclides from the waste form is assumed to be congruent with the dissolution of the UO_2 spent fuel matrix. The solubility of several radionuclides is set to a constant value while that for other radionuclides is sampled from a distribution function (Table 1 and Fig. 2). For the basecase, waste form dissolution rate model 2 was used in conjunction with the particle size model for surface area determination. It does not include an igneous activity disruptive event, which makes a significant contribution to the estimated risk in 10,000 yr. The

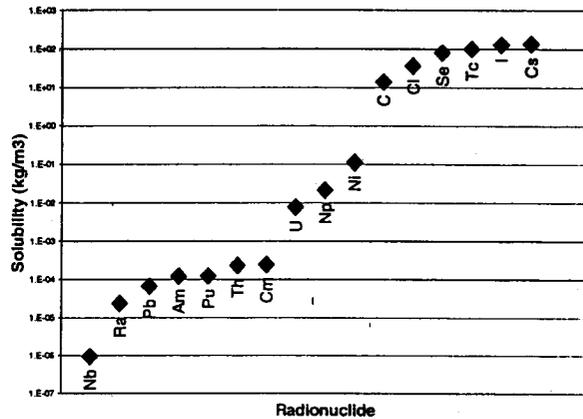


Fig. 2. Mean solubility of radioelements considered in the TPA 4.1 code for groundwater release. The values for Np, Pu, and Am are the mean values of the distributions listed in Table 1.

igneous activity disruptive event gives rise to both ground surface and groundwater releases. Because solubility can be a barrier only to groundwater release, and because the contribution of igneous intrusion to groundwater release is small when appropriately probability weighted, the effect of igneous activity was neglected in this study for computational expediency.

The TPA Version 4.1 code is exercised in probabilistic mode, which implements a Monte Carlo approach to include the full range of uncertainty in the parameters. Values of as many as 330 parameters are sampled randomly from input distributions using the Latin Hypercube Sampling method. The remaining 620 model parameters are assigned constant values. Although some of the sampled parameters in the TPA code are specified as partially correlated to other sampled variables, none of the solubilities are correlated.

To provide a quantitative measure of the solubility limited behavior of the different radionuclides, the following calculations were performed. For a particular radionuclide, if its concentration reaches its corresponding solubility for a time step during which water flows out of the waste package, that time step is marked as solubility limited for that radionuclide. The number of solubility limited time steps divided by the total number of time steps in the analysis period gives the fraction of the simulation period during which release is solubility limited. Furthermore, the number of solubility limited time steps divided by the number of time steps during which there is water flow out of the waste package defines the fraction of the flow period over which radionuclide release is solubility limited.

Simulations were conducted primarily for a 10,000-yr simulation period, which corresponds to the period of regulatory concern. A limited set of results was produced

for a 100,000-yr simulation period so that the long-term effect of solubilities could be evaluated, especially for the time period when many or most of the waste packages are expected to fail by corrosion. A set consisting of 350 Monte Carlo realizations was used in obtaining results for each test case. Results from each realization is a time evolution of mass flux out of the waste packages as a function of time. Although automatic time-stepping is used in the computation of mass flux, the results at 50- and 400-yr time intervals for the 10,000- and 100,000-yr simulation periods, respectively, are reported in this paper.

III. RESULTS

III.A. Simulation Results

For the bathtub waste form wetting mode, the release of 12 out of 20 radionuclides is controlled by solubility. The maximum (realization average) relative time span during the 10,000-yr simulation period over which the release of radionuclides from the waste package is solubility controlled is 11.4% or less. Note that solubility is relevant only during the time frame over which water contacts the waste, accumulates in the waste package, and then flows out of the waste package. The flowing period during the 10,000-yr simulation period over which the release is solubility limited is as much as 99.7% on average. During more than 99% of this flowing period, the release of U-238, U-234, Pu-239, and Pu-240 is controlled by their respective solubility. On average, during more than 85% of the flow period, the release of Nb-94, Am-241, and Am-243 is solubility limited. Solubility controls the release of Np-237, one of the important dose contributors, for at least 63% of the flow period. Th-230, Cm-246, Cm-245, and Ra-226 are solubility limited for less than 50% of the flow period. The solubility limited behavior of Cm-246 is illustrated in Fig. 3 as an example. Tc-99, I-129, and Cl-36 are key dose contributors during the 10,000-yr simulation period, but their release is not solubility controlled at the base case flowrates.

For the flow-through waste form wetting mode, the release of 15 out of 20 radionuclides is controlled by their solubility. The realization average relative time span during the 10,000-yr simulation period over which the release of radionuclides from the waste package is solubility controlled is as much as 19.6% for several radionuclides, which is approximately twice as long compared to the bathtub waste form wetting mode for the corresponding radionuclides, and some radionuclides are solubility limited three to four times longer (e.g., Cm-246, Cm-245, and Ra-226). The radionuclides that were not solubility limited in the bathtub model but are solubility

limited in the flow-through model are Ni-59, Tc-99, and Pb-210. Note that Tc-99, which is one of the major dose contributors, was not solubility controlled for the bathtub waste form wetting mode, but was solubility limited for a short duration under the flow-through waste form wetting mode. On average, during more than 99% of the flow period the release of U-238, U-234, Pu-239, and Pu-240 is solubility limited. In addition, for more than 90% of the flow period, Nb-94, Am-241, and Am-243 release is controlled by the solubility. Np-237 is controlled for at least 78% of the flow period, whereas Th-230 is controlled for 64% of the flow period. Cm-246, Cm-245, Ra-226, Ni-59, Pb-210, Tc-99, and Cs-135 are controlled for less than 50% of the flow period. I-129, Se-79, Cl-36, and C-14 are not solubility limited.

Several radionuclides with low solubility do not experience solubility limited release, whereas others with high solubility do experience solubility limited release. For example, Pb-210 has a low solubility but exhibits no solubility limited release in the bathtub model, while U-238 has a much higher solubility but experiences solubility limited release for most of the time that water flows out of the waste package. Verification calculations were performed to examine this counter-intuitive behavior.

III.B. Verification

Verification studies were conducted on a mean data set to determine why a radionuclide with low solubility did not become solubility limited. The verification study evaluated the effect of flow rate, half-life, radionuclide inventory, and the function of a daughter product as a parent (single member) radionuclide. High, low, and mid-range values of half-life, radionuclide inventory, flow rate, and their combinations were used to determine what drives solubility limited releases. Note that these values were artificially varied to demonstrate that the behavior of

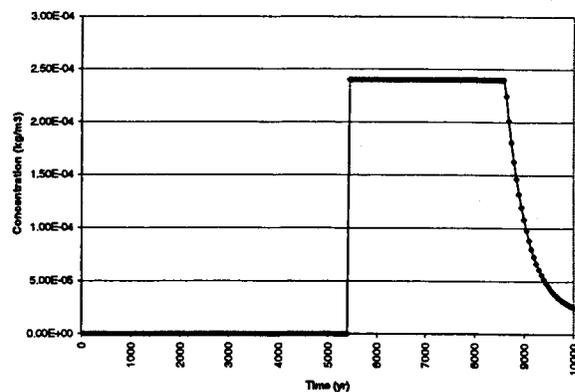


Fig. 3. Cm-246 concentration versus time, showing that the effluent can exit the waste package with a maximum Cm-246 concentration corresponding to its solubility.

the radionuclides is consistent over a range of values.

Initial inventory has a large effect on radionuclide solubility limited release. Two radionuclides, Pb-210 and U-238, are compared in Table 2. For the basecase value of initial inventory, Pb-210 release was not solubility limited in both the bathtub and flow-through models. However, when its initial inventory was increased, as shown in Table 2, Pb-210 changed from non-solubility limited to solubility limited behavior for the same period as U-238.

The verification results also show that changes in reference flow rate had little effect on the solubility limited release of U-238 and no effect on that of Pb-210.

In another study, the daughter products were treated as single member chains in the TPA code. Pb-210 became solubility limited for as long a period as U-238 when inventory and half-life were both set to high values. This result was observed in both the bathtub and flow-through models. Considering radionuclides by themselves, a high inventory by itself, or a long half-life by itself did not result in Pb-210 becoming solubility limited. It took both the high inventory and long half-life to cause this effect.

III.C. Comparative Study

A comparative study was also conducted to identify the effects of the leaching model, analysis period, and

leaching surface area model (grain and particle models) on the solubility limited release of radionuclides. This comparative study used over 350 realizations for each case. The results of this study are presented in Figs. 4-7.

III.C.1. Effect of Waste Form Dissolution Rate

The choice of leaching model has a significant impact on radionuclide solubility limited release. When leaching model 1 was used instead of leaching model 2, all of the radionuclides that were previously solubility limited became solubility limited for longer periods. This result arises from the higher dissolution rate calculated from model 1 relative to model 2. Some became solubility limited for more than twice the percentage of time that they were solubility limited under leaching model 2. For example, Ra-226 was solubility limited 18.6% of the time in leaching model 2, but was solubility limited 53.1% of the time in leaching model 1. In addition, in leaching model 3, all of the radionuclides that showed solubility limited release under leaching model 2 showed less solubility limited release or no solubility limited release at all. Th-230 went from 43.6% solubility limited release under model 2 to no solubility limited release under model 3. This result is due to the lower dissolution rate calculated from model 3 compared to that from model 2. Furthermore, none of the

Table 2. Verification analyses for two radionuclides (Pb-210 and U-238) using a parent-only radionuclide assumption and different values (high, low, mid-range) of flowrates, inventories, and half lives

Parameter		Fraction of the Simulation Period during which the Release is Solubility Limited			
		Bathtub Waste Form Wetting Mode		Flow-through Waste Form Wetting Mode	
		Pb-210	U-238	Pb-210	U-238
Constant reference flow rate [m ³ /yr]	Low: 6.53×10^{-2} [m ³ /yr]	0.00	0.91	0.00	0.99
	Mid: 7.50×10^{-2} [m ³ /yr]	0.00	0.92	0.00	0.99
	High: 8.46×10^{-2} [m ³ /yr]	0.00	0.93	0.00	0.99
Inventory	Low: 3.83×10^{-6} [Ci]	0.00	0.00	0.00	0.00
	Mid: 1.44×10^4 [Ci]	0.18	0.18	0.26	0.26
	High: 2.88×10^4 [Ci]	0.18	0.18	0.26	0.26
Half-life	Low: 2.23×10^1 [yr]	0.00	0.00	0.00	0.00
	Mid: 2.23×10^9 [yr]	0.00	0.18	0.00	0.26
	High: 4.47×10^9 [yr]	0.18	0.18	0.26	0.26
Parent-only radionuclide assumption	Inventory High: 2.88×10^4 [Ci]	0.00	0.18	0.00	0.26
	Half-life High: 1×10^4 [yr]	0.00	0.00	0.00	0.01
	Inventory High: 2.88×10^4 [Ci] Half-life High: 1×10^4 [yr]	0.18	0.18	0.26	0.26
Base Case:	1) Reference flow rate increases over time from 6.53×10^{-2} to 8.46×10^{-2} [m ³ /yr] 2) Inventory for Pb-210 = 3.83×10^{-6} [Ci]. Inventory for U-238 = 2.49 [Ci] 3) Half-life for Pb-210 = 22.3 [yr]. Half-life for U-238 = 4.47×10^9 [yr]				

radionuclides showed solubility limited release in leaching model 4, which gives the lowest waste form dissolution rate of the four models. These results are shown in Fig. 6.

The changes in solubility limited release of radionuclides is directly related to the leach rate, the value of which is either computed (models 1, 2, and 4) or set by the user (model 3). Changes in solubility limited behavior with leaching model indicate that some models may underpredict the potential for solubility limited release of some radionuclides. For example, simulations using the bathtub model and a user-specified leach rate result in Th-230 showing no solubility limited release, whereas Th-230 has significant solubility limited release under models 1 and 2 (82.9% and 43.6% for models 1 and 2, respectively). Other radionuclides such as U-238 show about the same solubility limited release under models 1, 2, or 3.

III.C.2. Effect of Simulation Period

When the analysis period is changed from 10,000 to 100,000 yr, the ranking of radionuclides with respect to solubility limited behavior changes. That is, some radionuclides that are solubility limited longer than others in a 10,000-yr analysis period become solubility limited for a smaller percentage of time in a 100,000-yr analysis period. For example, Cm-245 and Cm-246 are solubility limited for a much lower percentage of time in the 100,000-yr bathtub study than they are in the 10,000-yr bathtub study, whereas Ra-226 is solubility limited for about the same amount of time in the bathtub study regardless of analysis period. A change in analysis period causes the rankings to change among Cm-245, Cm-246, and Ra-226. Cm-245 and Cm-246 become less solubility limited than Ra-226 over the 100,000-yr period. The solubility limited release for Ra-226 is still significant after the 10,000-yr compliance period, whereas solubility limited release of Cm-245 and Cm-246 is minimal after 10,000 yr. This behavior is the result of radioactive decay of Cm-245 and Cm-246 and daughter ingrowth of Ra-226 becoming more evident in 100,000 yr compared to 10,000 yr. The effect of analysis period on simulation results is displayed in Figs. 4 and 5.

III.C.3. Effect of Waste Form Leaching Surface Area

The last test in the comparative study involved a comparison between the leaching surface area grain model and the particle model. As mentioned previously, the particle model assumes the spent fuel is fragmented into small spherical particles, whereas the grain model accounts for the additional exposed surface area from exposed spent fuel grains. All of the radionuclides that showed solubility limited release in the particle model

were solubility limited for a longer period in the grain model. The longer period is due to the greater surface area in the grain model compared to that in the particle model, which provides a larger contact area between water and waste form and a higher rate of leaching. As shown in Fig. 7, all of the radionuclides that showed solubility limited release in the particle model were solubility limited for a longer period of time in the grain model. In addition, the results for the grain model are very similar to those found under leaching model 1.

IV. CONCLUSIONS

Regardless of the waste form wetting mode (i.e., flow-through versus bathtub), most of the key radionuclides in the NRC TPA 4.1 code exhibit solubility limited release. Therefore, radionuclide solubility can be an effective barrier limiting radionuclide release and transport to the environment. It appears that a low solubility limit, by itself, does not necessarily guarantee solubility limited release. Whether the release of a particular radionuclide is solubility limited or not depends on an intricate interplay of the solubility of that radionuclide, the degree of waste form exposure to water, the radionuclide half-life, and the radionuclide inventory. This interplay is particularly apparent for Pb-210. Pb-210 has a lower solubility than other radionuclides but is not solubility limited in either the bathtub or flow-through water contact mode unless its inventory is substantially large. The results of the verification calculations suggest that radionuclides with low solubility may not experience solubility limited release whereas the release of radionuclides with high solubility can be solubility limited, depending on the inventory and decay rate.

In addition, some of the major contributors to dose are solubility controlled over different time durations whereas others are not solubility controlled. For example, Tc-99, I-129, Cl-36, and Np-237 are all major contributors to dose. However, only Np-237 is solubility controlled for a significant period of time. Its release is controlled by solubility for more than 63% of the time that water flows out of the waste package in the bathtub model and for more than 78% of the time in the flow-through model. Therefore, Np-237 would be an even greater contributor to dose if it were not for this significant solubility limited release. Tc-99 release is controlled by solubility in the flow-through model to a much smaller extent, and I-129 and Cl-36 are not solubility limited. Thus, for these three radionuclides, the solubility would have little or no impact on their contribution to dose.

The comparative study reveals that the analysis period, leaching model, and surface area model all have a significant impact on solubility limited release. An

increase in the analysis period makes the effect of radioactive decay more evident. Thus, increasing the analysis period causes some radionuclides to have significant solubility limited release later than other radionuclide. This effect is indicated by a change in radionuclide ranking from most to least solubility limited with an increase in analysis period. In addition, the solubility limited behavior of some radionuclides changes when different leaching models are used. Therefore, different leaching models may have the effect of

underpredicting the solubility limited release of some radionuclides. And finally, in models for which the surface area is greater, radionuclides are solubility limited for a longer percentage of time than they are in models with a smaller surface area because of the higher leaching rate that comes with a larger surface area.

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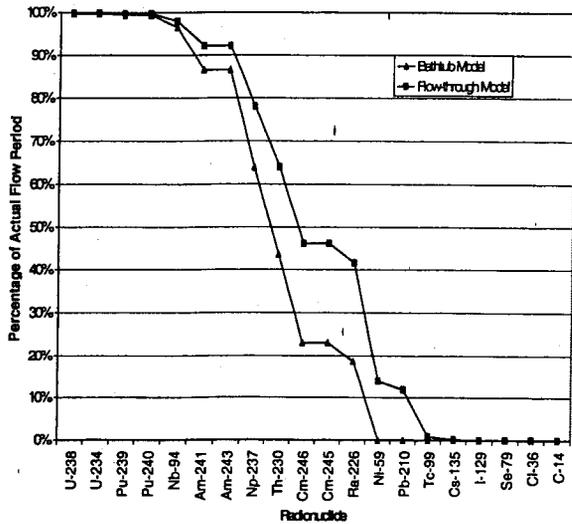


Fig. 4. Percentage of the actual flow period during which the radionuclide is solubility limited (bathtub and flow-through waste form wetting models; 10,000-yr simulation period)

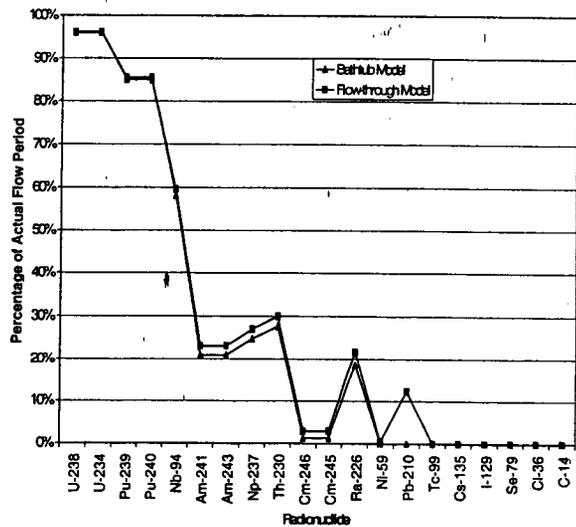


Fig. 5. Percentage of the actual flow period during which the radionuclide is solubility limited (bathtub and flow-through waste form wetting models; 100,000-yr simulation period)

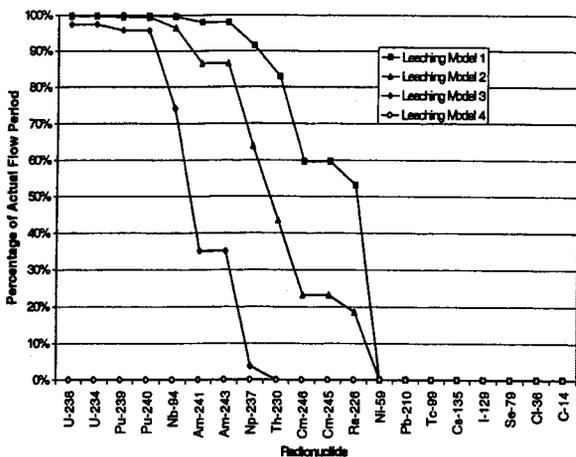


Fig. 6. Percentage of the actual flow period during which the radionuclide is solubility limited (waste form dissolution models 1 to 4; 10,000-yr simulation period)

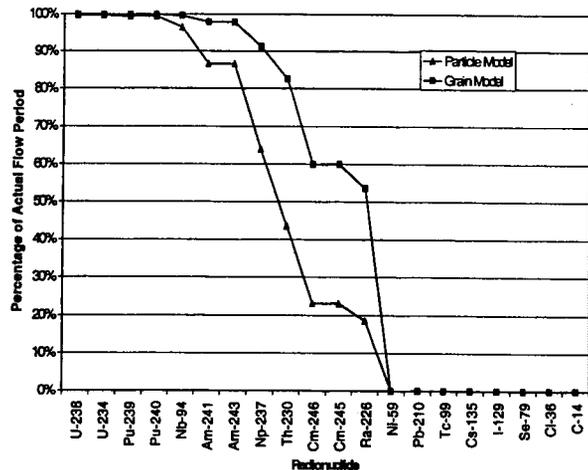


Fig. 7. Percentage of the actual 10,000-yr flow period during which the radionuclide is solubility limited (particle and grain surface area models; 10,000-yr simulation period)

performed by the Center for Nuclear Waste Regulatory Analyses (CNWRA) for the U.S. Nuclear Regulatory Commission (NRC), Office of Nuclear Material Safety and Safeguards, under Contract No. NRC-02-02-012. This paper is an independent product of the CNWRA and does not necessarily reflect the views or regulatory position of the NRC. The reviews by O. Pensado and B. Sagar are gratefully acknowledged.

REFERENCES

[1] S. MOHANTY, T.J. MCCARTIN, and D. ESH. "Total-system Performance Assessment Version 4.0 Code: Module Description and User's Guide (Revised)," Center for Nuclear Waste Regulatory Analyses, San Antonio, Texas (2002).

[2] S. MOHANTY and R.B. CODELL. "A Performance Assessment Review Tool for the Proposed Radioactive Waste Repository at Yucca Mountain, Nevada, USA," Proc. 5th Int. Conf. Probabilistic Safety Assessment and Management. Osaka, Japan, November 27 – December 1, 2000, Vol. 3, p. 1485 (2000).

[3] S. MOHANTY, R.B. CODELL, T.M. AHN, and G. CRAGNOLINO. "An Approach to the Assessment of High-Level Radioactive Waste Containment – II: Radionuclide Releases form an Engineered Barrier System." *Nucl. Eng. Design* **201**, 307 (1999).

[4] R.B. CODELL and S. MOHANTY. "Alternative Conceptual Models for Radionuclide Release," Proc. 9th Int. High-Level Radioactive Waste Management Conf. (CD Publication), Las Vegas, Nevada, April 29 – May 3, 2001.

[5] W.M. MURPHY and R.B. CODELL. "Alternate Source Term Models for Yucca Mountain Performance Assessment Based on Natural Analog Data and Secondary Mineral Solubility," Scientific Basis for Nuclear Waste Management XXII, D.J. Wronkiewicz and J.H. Lee, (eds.), *Mat. Res. Soc. Symp. Proc.* **556**, p. 551 (1999).