MODELING THE ACCUMULATION OF THERMALLY FISSILE MATERIALS IN HLW REPOSITORY

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This study examined the accumulation behavior of thermally-fissile isotopes U-235 and Pu-239 in the far field of an abstracted geological repository consisting of a discrete array of waste canisters that fail simultaneously. A conservative transport model has been applied for two extreme array orientations relative to groundwater flow for various evaluation points in space and time in a homogeneous, saturated porous medium. It was found that the mass of Pu-239 could only contribute significantly to a mass accumulation formed in the near-field of a repository. The enrichment of U-235 is affected by the plutonium plume most noticeably at early times but follows more predictable behavior at longer time periods. At certain distances, enrichment is observed to drop sharply, indicating lower bounds on repository dimensions. However, due to conservatism, enrichment does not vary over a wide scale and restricted the source term to unrealistically narrow limits in order to avoid critical mass formation, suggesting a relaxation of assumptions.

I. INTRODUCTION

A safe repository design for nuclear waste includes the limitation of potential exposure to the biosphere to safe limits throughout geological expanses of time. If multiple waste canisters emplaced into such a repository fail concurrently, it is plausible that groundwater intrusion can result in the dissolution of waste forms and the transport of thermally fissile materials (TFM). Scenarios based on transport phenomena predict that if a critical mass were to accumulate, moderation characteristics in the water-saturated bedrock could result in a significant release of energy.[1] This in turn can cause a radioactive dose to the surrounding environment, the severity of which is evaluated in the context of a total system performance assessment.

The scenario at the focus of this paper is a uranium-driven criticality event in the far field of a saturated granitic repository. Previous simulations have shown that for a given porosity of sandstone or granite, certain masses of uranium compounds in a planar fracture geometry can result in neutron multiplication greater than unity.[2] Such results were obtained for 250 metric ton (MT) of uranium with an enrichment of 2.25% in the case of sandstone. Given the magnitude of spent fuel inventory to be destined to a final geological repository at the end of interim cooling, possibility for such a mass to accumulate in a surrounding host rock cannot be ignored.

The goal of this paper is to provide a bounding analysis on the accumulation of two thermally fissile nuclides, Pu-239 and U-235, in the far field from a discrete array of waste canisters by employing a conservative transport model. Since this study revolves around the safety of an abstract repository concept and the subsequent employment of a dose-based performance metric, conservatism is based on the purposeful overestimation of mass on a worst-case basis. The results are intended to illuminate the combined behavior of multiple sources and explore whether the critical masses obtained in the previous criticality studies are realizable by transport processes in the host rock. Matters of importance include not only the accumulation of TFM but the evolution of uranium enrichment over time and space as well.

II. METHODOLOGY

II.A. Repository Configurations and Inventory

A virtual repository is constructed with non-fractured, homogeneous properties such that porosity ε and transport

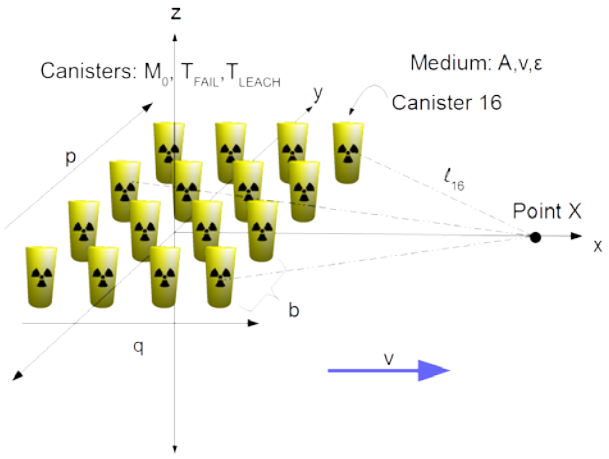

Fig. 1: Repository layout for *p* by *q* array of canisters, which are separated by pitch *b* and lie parallel to groundwater flow with velocity *v*. Accumulation is monitored at point *X* and is based on straight-line pathways, such as l_{16} for canister #16.

characteristics could be held constant. Groundwater was assumed to be flowing in the pore space at a constant velocity *v*, as shown in Fig. 1. A flow area of *A*, common to all canisters, is the cross-sectional area of a hypothetical one-dimensional transport pathway connecting a canister and the accumulation point, and employed later in the nuclide release model.

The initial loading of radionuclides in the repository is discretized into a rectangular grid of $p \times q$ point sources separated by a pitch distance *b*. Two extreme grid orientations are considered: one lying on the YZ plane orthogonal to groundwater flow and one lying on the XY plane parallel to flow (Fig. 1). The Cartesian origin is defined at the center of the grid, and the canisters are indexed to provide an array of linear transport distances to an observation point *X*.

Since this study is meant to provide a bounding analysis, the assumption is made that all canisters fail at the same time t_F and immediately begin dissolving (due to groundwater infiltration) over a period *T*. Each canister contains the same inventory of nuclides, which is detailed in Table 1. These amounts were calculated with the ORIGEN code,[3] which is part of the SCALE package. They correspond to used nuclear fuel cooled for 54 years in interim storage from a high burnup (45000 MWd/MT) PWR after 1184.21-day operation, with an initial fuel enrichment of 4.5 wt%. Amounts are normalized to 1 MT of uranium.

The actinide precursors of the isotope Pu-239 are all assumed to decay before the canisters fail and are therefore lumped into that initial inventory. The initial uranium enrichment is listed as 1.11 wt% but has a theoretical maximum of 1.81 wt% if all Pu-239 isotopes decay to U-235 and there are minimal contributions to the amount of U-238 from Pu-242 decay.

Table 1: Waste Canister Inventory

Nuclide	Amount		Half-Life
	[g/MTU]	[mol]	[yr]
U-235	1.06E4	45.1	7.04E8
Pu-239	6.07E3	25.4	2.41E4
+Np-239 +Am-243 +Cm-243	6.20E3	25.9	6.45E-3 7.38E3 29.09
Total TFM	1.68E4	71.0	
U-238	9.25E5	3890	4.468E9
Pu-242	6.561E2	2.711	3.739E5
Enrichment at emplacement		1.11%	
Maximum enrichment		1.81%	

II.B. Release Model

Upon canister failure and immediate groundwater intrusion, nuclides are assumed to be released congruently from the UO_2 matrix (we call it waste matrix hereafter). This is opposed to a solubility limited release where the solubility of each nuclide acts as the limiting factor for dissolution and necessitates the modeling of precipitation in the vicinity of the waste form. This approach has not been employed in the present study as it would have removed conservativeness and simplicity from the model.

In a congruent release, during the dissolution period of the waste form *T*, nuclides enter the water phase, as the nuclides are all assumed to be readily soluble. The stylized concentration of a nuclide at the time of package failure in the water phase in the pore at the location of waste form dissolution can thus be described by

$$C^0 = \frac{M^0}{A\epsilon v T}\left[\frac{mol}{m^3}\right] \qquad (1)$$

where M^0 is the mass of the radionuclide in moles at the time of package failure, and $A\varepsilon v$ describes the volumetric flow rate (m^3/yr) of water allocated for each canister through the pore space in host rock. Since each canister has the same inventory, *T* is made common to all canisters. Using parameters from Tables 1-2 in Eqn (1), the initial concentration is C_1^0 =6.48E-5 mol/m^3 for Pu-239 and C_2^0=1.13E-4 mol/m^3 for U-235.

Samples of fracture groundwater obtained from drilled crystalline bedrock at the Palmottu natural analogue in Finland demonstrate uranium concentrations ranging from 6E-6 mol/m^3 for reducing, alkaline conditions 240 m underground to 4E-4 mol/m^3 for an oxidative environment closer to the surface.[4] The observed groundwater concentration of uranium at the Cigar Lake natural analogue in Canada ranges from 6E-6 mol/m^3 for the reduced zone to 2E-3 mol/m^3 in the oxidized zone.[5] For plutonium, the maximum observed concentration here is 1E-6 mol/m^3. (Ref. 6) Given that spent fuel has a greater proportion of plutonium than such an ore deposit, and considering that TFM transport will be driven by departures in the reducing environment of granite, the concentrations derived from Eqn. (1) are reasonable as conservative upper bounds.

II.C. Transport Model

The governing equation for the transport of an *n*-member chain of radionuclides is written as:

$$K_i \frac{\partial C_i}{\partial t} + v\frac{\partial C_i}{\partial l} + \lambda_i K_i C_i - \lambda_{i-1} K_{i-1} C_{i-1} = 0 \qquad (2)$$

where $t>0$ and $i=1,2,3\ldots n$. K_i is the retardation factor for *i*-th nuclide, which is a function of porosity ε and the nuclide's sorption distribution coefficient, C_i is the concentration (mol/m^3) of nuclide *i* in water in pores of the host rock, *v* is the groundwater pore velocity (m/yr), *l* is the coordinate along the one-dimensional transport pathway (m), and λ_i is the decay constant of the *i*-th nuclide (yr^{-1}). Note that the hydrodynamic dispersion term is not included to obtain a conservative estimate for mass accumulation in the far field. In the present study, we consider the case for

n = 2. Subscripts "1" and "2" denote Pu-239 and U-235, or Pu-242 and U-238, respectively.

The initial condition requires that no concentrations of nuclides exist beyond the repository origin:

$$C_i(l,0) = 0,\ l > 0,\ i = 1, 2 \tag{3}$$

The boundary condition is specified for every member of the decay chain. For the first member:

$$C_1(0,t) = C_1^0 f_1(t) \tag{4}$$

where $\boldsymbol{f_i(t)}$ defines the manner in which the nuclide is released from the waste form. Taking into account the decay of the nuclide in the waste form, for the congruent release, we can write:

$$f_1(t) = e^{-\lambda_1 t}\{h(t - t_F) - h(t - t_F - T)\} \tag{5}$$

where h is the Heaviside function.

Applying these side conditions results in the following solution for the first member:

$$C_1(l,t) = C_1^0 e^{-\lambda_1 t}\left\{h\left(t - t_F - \frac{K_1 l}{v}\right) - h\left(t - T - t_F - \frac{K_1 l}{v}\right)\right\} \tag{6}$$

where the previously mentioned concentration profile is further defined in terms of the nuclide travel time $K_1 l/v$. The distance l is dependent on the position of the canister of interest relative to the observation point. For the canisters at the extreme periphery of the repository, this travel time will have noticeable effects on overall accumulation behavior.

For the second member of the decay chain, the boundary condition is written as:

$$C_2(0,t) = \left[C_2^0 e^{-\lambda_2 t} + \frac{\lambda_2 C_1^0}{\lambda_2 - \lambda_1}\left(e^{-\lambda_1 t} - e^{-\lambda_2 t}\right)\right] * \{h(t - t_F) - h(t - t_F - T)\} \tag{7}$$

The solution for $\boldsymbol{C_2(l,t)}$ is characterized by bands defined in terms of the canister failure time, the dissolution time, the travel time of the parent nuclide, and the travel time of the daughter nuclide. Now that the evolution of concentration for both the parent and daughter nuclides is defined, their accumulation masses can be formulated and evaluated.

II.D. Accumulation Model

The equation for accumulation is written as:

$$\frac{dM_i}{dt} = A\epsilon v C_i(l_k,t) - \lambda_i M_i + \lambda_{i-1} M_{i-1}\left[\frac{mol}{yr}\right] \tag{8}$$

where M_i is the mass of nuclide i deposited at the accumulation location, contributed by a single canister, k. l_k indicates the distance between canister k and the accumulation location. Analytical solutions for this problem has been obtained, but are omitted for conciseness. The contribution from each canister has been summed up.

The solutions are employed for both the Pu-239→U-235 decay chain and Pu-242→U-238 decay chain. The observation point is placed on the x-axis for symmetry, and analyses involve incrementally increasing the distance of X from the repository in the $+\hat{\boldsymbol{x}}$ direction for certain moments in time. The evaluated quantities will then be normalized to the initial spent fuel inventory (corrected for decay using the Bateman equations).

With the amounts of U-235 and U-238 numerically evaluated as functions of time and the location of the accumulation location, the enrichment of the deposited uranium, can be calculated by

$$e(l,t) = \frac{M_2^{235}(l,t)}{M_2^{235}(l,t) + M_2^{238}(l,t)} \tag{9}$$

where the subscript "2" is meant to show that both nuclides of interest employ the second-member model. This relationship will be valuable in comparing the enrichment employed in simulation with the initial loading. Quantities will be normalized to the initial enrichment of 1.11 wt%.

II.E. Parameter Space

The parameter values in Table 2 are used based on conditions expected for a granitic repository. Due to the use of normalized quantities, the size of the array is not of utmost importance and was chosen arbitrarily to be a square array of 100 canisters. Although some repository designs have much larger pitch distances, such as the deep borehole concept with a 200 m pitch (Ref. 7), 20 m was considered representative for a mined repository, and chosen to emphasize the combined effects from individual sources at any X.

The failure time of 1000 years was chosen to be highly conservative, as some copper-based canisters are technically designed to last for up to 10^5 years,[8] and also to observe effects of release and transport of Pu isotopes (the first member nuclide) with relatively short half-lives. The dissolution time of 10,000 years is also conservative. Although this period could be extended, this value was chosen to readily show the behaviors of accumulation upon the end of dissolution.

Table 2: Parameter Values for Saturated Granite

Parameter	Value
Porosity of the host rock (ε)	0.1
Cross sectional area per canister (A) [m^2]	400
Velocity (v) [m/yr]	1
Flow Rate (V) [m^3/yr]	40
Retardation factor for Plutonium (K_1)	15000
Retardation factor for Uranium (K_2)	2600
Package failure time (t_F) [yr]	1000
Dissolution time (T) [yr]	10000
Array ($p \times q$)	10×10
Canister pitch (b)	20 m

The retardation factor of plutonium is assumed to be about an order of magnitude greater than that of uranium. This is based on the observation that the granitic host rock is expected to have reducing properties, which inhibits dissolution and leads to low oxidation states of plutonium that exhibit such high values of retardation.[9] Uranium undergoes solute-driven transport and retardation can vary in magnitude from 1 to 10^3 depending on measured sorption distribution coefficients for a host rock of interest.[10] we chose a value which was observed for a granitic rock.

Groundwater velocity varies from one rock type to another, and can be highly heterogeneous. For example, a highly fractured rock can exhibit velocities on the order of 100-1000 m/yr.[1] In this paper, a velocity of 1 m/yr was chosen as a representative value.

III. RESULTS

III.A Effect of Pitch

The most direct impact that multiple discrete sources have on accumulation is through pitch distance, which is largely a geometric analysis (using the assumption of no transverse effects). Fig. 2 shows plots of normalized accumulation versus pitch at various locations away from the origin. For the orthogonal YZ layout, as the observation distance increases, the trend in accumulation is depressed until no accumulation is achieved at all. That is, the contributions from peripheral canisters are less and less realized as pitch increases since larger and larger transport distances are required of them. Therefore, intuitively, for minimal accumulation, canisters need to be spaced as far apart as possible.

For the parallel (XY) layout, this depression effect is realized more drastically. However, for larger observation distances, accumulation does not drop to zero but instead follows a $\sqrt{b}$ trend. Geometrically, since the repository is on the plane with the x-axis, expanding pitch size merely extends canisters into the direction of groundwater flow. Therefore, accumulation is realizable regardless of how far part the canisters are arranged in the repository.

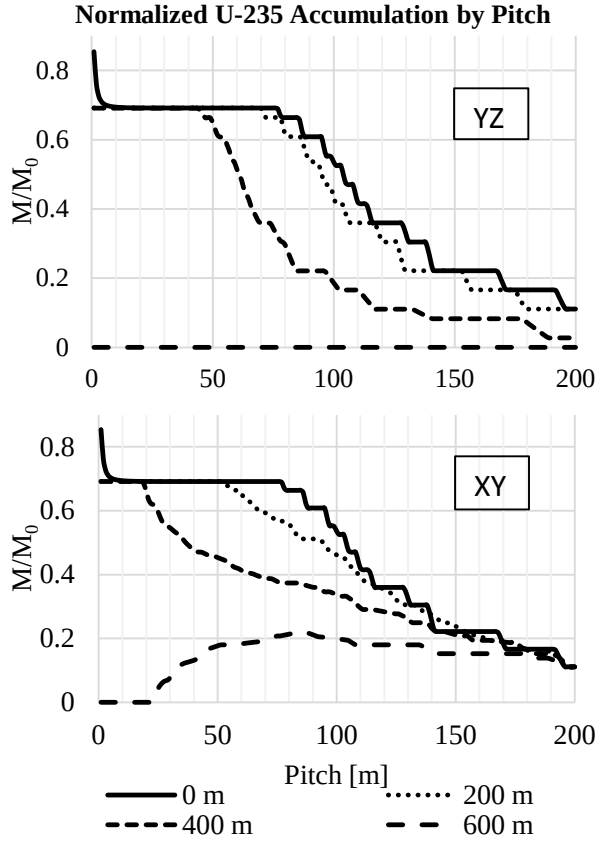

Fig. 2: The variation in U-235 accumulation over increasing pitch distance between individual sources. Plots given for YZ (top) and XY (bottom) layouts at four different distances from the origin. Data for t=5E5 years.

III.B. Accumulation over Distance

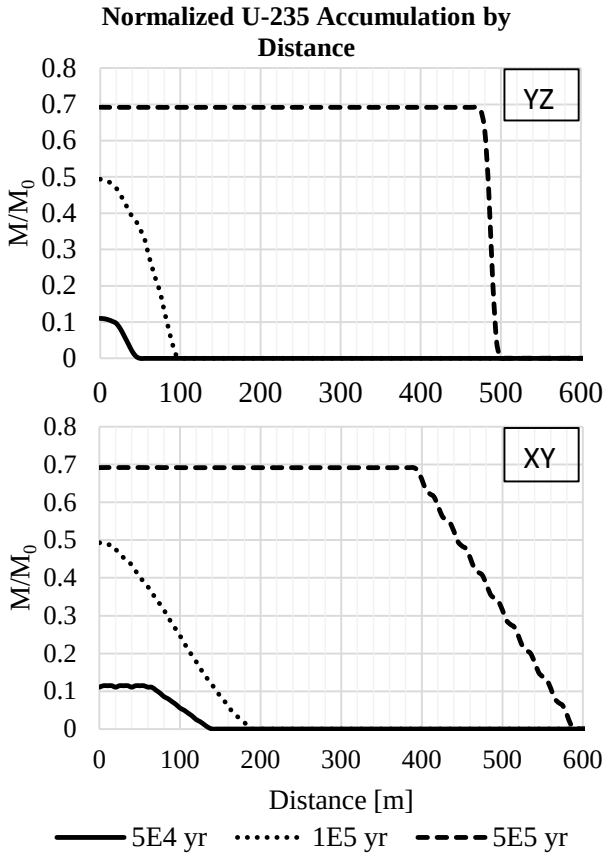

Fig. 3: Accumulation of U-235 as a function of the distance between the accumulation location and the center of the repository for three different snapshots in time for the YZ (top) and XY (bottom) plane.

When designing a repository, considerations are made with respect to the proximity of natural resources and locales. The model was thus used to evaluate the accumulation of Pu-239, U-235, and U-238 at various points on the x-axis for different moments in time.

Fig. 3 shows the U-235 results for the YZ layout, where it is evident that for given time periods, no accumulation is found to occur at a certain point (termed "drop-off" point) and beyond. The drop-off point gets farther as the time period is lengthened due to increased transport time. Due to the long half-life of U-235, the general behavior at long times is consistent due to negligible losses from decay. Indeed, this behavior is confirmed well beyond the range of the plot for snapshots in the millions of years, where the drop-offs are more like step transitions than the more gradual decreases at earlier times.

From Fig. 3, for the XY layout, more or less the same behavior is observed except for farther and more gradual drop-offs. This effect is the result of canisters in the $+\hat{x}$ anterior periphery bringing in the last contributions while those from the $-\hat{x}$ exterior periphery are dying off. This is

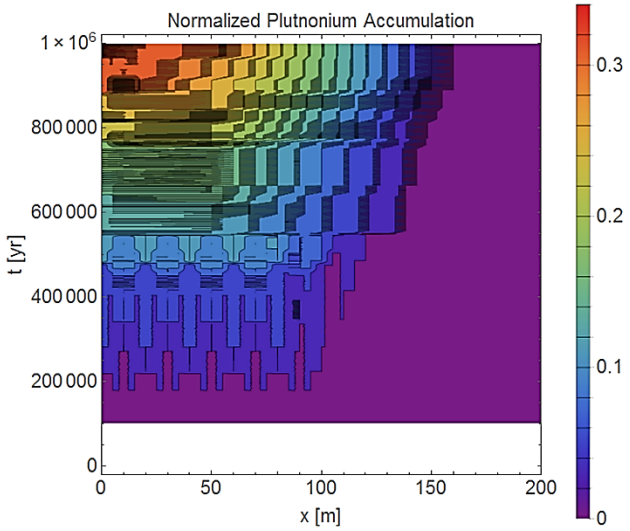

Fig. 4: Contour plot for the normalized plutonium mass over time and distance between the center of the repository and the accumulation location for the XY layout.

shown clearly in the staggered line comprising the data for t=5.0E5 years. For either layout, the maximum M/M_0 does not exceed ~0.7, which is consistent for longer periods beyond those included.

For Pu-239, the same general observations are made, except that the relatively short half-life of the isotope makes accumulation for long periods of time (10^6 years or more) negligibly small. Due to retardation, the drop-off points occur at smaller values of x for a given moment in time relative to U-235. This behavior is plotted in contours in Fig. 4 for the XY configuration. Here, the effects of the anterior canisters is evident through the humps in the lower left portion for the first 110 meters (representing the portion of the repository jutting onto the x axis). The contours indicate that the most significant plutonium contributions occur within the first 30 to 50 meters of the repository, confirming that plutonium-driven criticality events would be restricted to the "near-field" of the repository consisting of the engineered barrier system. Similar behavior occurs for the orthogonal layout, such that these XY contours can be thought of as a superposition of the YZ contours.

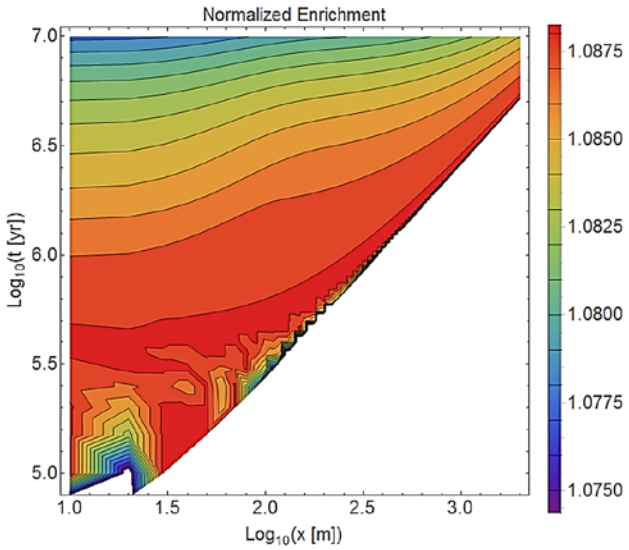

Fig. 5: Contour plot of normalized enrichment over log-scale time and the distance between the center of the repository and the accumulation location for the YZ repository. The effects of plutonium contributions are noticeable in close proximity to the origin.

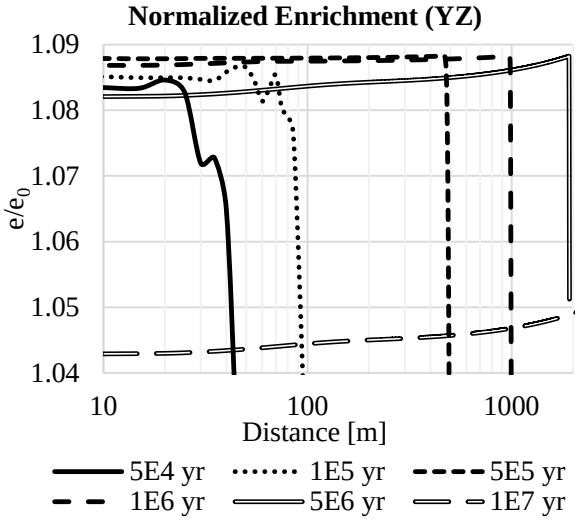

Fig. 6: Snapshots of normalized enrichment over distance. The Pu-239 contributions are evident at early times.

III.C. Evolution of Enrichment

For various cases in time and the distance of the accumulation location, Eqn. (9) was used to evaluate uranium enrichment relative to the initial inventory. Due to the contributions from Pu-239 and the long half-life of U-235, normalized values remain above unity for long periods of time (up to 10^8 years). Yet, there is a limited range of variation over time and space, peaking at approximately 1.09.

Fig. 5 shows a contour plot for enrichment over a 10^7 year time scale for the orthogonal layout, where a base-10 logarithmic scale is used to illustrate most characteristics. (Once again, the XY plot is essentially a superposition of the YZ plot, and is therefore omitted.) Early on, enrichment behaves irregularly in close proximity to the repository plane and then proceeds to follow more general behavior as time goes on. This is due to the short transport distance of plutonium. In fact, correlated behavior is observed in the normalized Pu-239 mass contours of Fig. 4. It is clear that the behavior at ~5.8E5 years is caused by the final significant contributions of U-235 from the Pu-239 plume. Snapshots of the data in Fig. 5 are shown in Fig. 6. Drop-offs are observed in the same manner as accumulation, with a peak of ~1.09 e/e_0 observed at 2000 m from the YZ plane. Beyond 5E7 years, behavior is essentially at or below unity.

IV. DISCUSSION

The contributions of plutonium in the accumulation of TFM are limited to the near field of the repository array. Consideration for potential criticality phenomena in roughly the first 50 meters away from the repository should revolve around plutonium-driven scenarios, for which analyses should be specific to the heterogeneous engineered barrier system likely to constitute this region, including the waste canister components, bentonite clay buffer and rocky backfill. The decay of Pu-239 has an additive effect on the plume of U-235 and affects the enrichment for early times and close proximity. Transport modeling with more liberal assumptions may closely resemble data obtained from natural analogues, particularly the Oklo, Gabon ore deposit. In such a case, where fission product migration is observed to remain in the near vicinity of the critical mass, the plutonium contribution to U-235 is likely to be of great importance in determining criticality scenarios at early observation times.

For a given time, drop-off distances have been observed beyond which TFM accumulation will not occur. The drop-off distance becomes less pronounced for the parallel layout due to the contributions from canisters in the anterior. While this step-like behavior is favorable to repository design since it establishes boundaries on nuclide transport, it is of consideration that this clear drop-off distance occurred in the present numerical exploration because of the advection-only transport model. Dispersive processes applied to a three-dimensional analysis would spread nuclides longitudinally and transversally (depending on the host rock) and eliminate such defined boundaries.

The pitch distance between individual canisters is highly indicative of the extent of accumulation for a given time and distance from the center of the repository. The larger the pitch, the less mass accumulates relative to the initial inventory. However, for the parallel configuration, due to canisters extending downstream of groundwater flow, accumulation does not approach zero with distance for closely packed sources. Therefore, coupled with the drop-off behavior, the parallel XY layout is generally unfavorable. It should be noted that without acknowledgment of transverse transport between canisters, the effect of pitch on near-field accumulation on the array is still unknown.

The long half-life of U-235 allows for consistent accumulation behavior over geological periods. Normalized accumulation does not exceed 70% of the initial inventory and normalized enrichment does not exceed ~1.09. Enrichment behaves with the drop-off characteristics of accumulation and includes a pronounced peak, particularly at long times. The plutonium contributions take effect at close proximity and early times, which introduces somewhat spiked aberrations. In contrast, long-term behavior is more-or-less predictable and encompasses a wider range of variation. Different enrichment behavior would be expected for higher proportions of plutonium or perhaps in an oxidizing environment where plutonium retardation is drastically reduced. Fractured bedrock could also enhance plutonium movement by exploiting colloid-facilitated transport. Nonetheless, the relatively short half-life of Pu-239 presents universal limitations.

With accumulation and enrichment behavior known, the machinery exists to analyze the applicability of critical mass scenarios. Suppose a performance assessment requires the prevention of critical mass formation within 5 million years and 1000 meters from the center of the repository. It is known for these coordinates that e/e_0=1.086 and M_{U235}/M^0_{U235} =0.692 or an orthogonal layout of canisters. The utilized porosity of 0.1 implies a void ratio of 0.11, and previous data suggests that for 250 MT uranium at 2.25% enrichment, with a similar void ratio, that a heavy metal volume faction of ~0.075 is needed for a critical mass. Therefore, correcting for decay and assuming a proportional initial amount of Pu-239, the original repository inventory can be no larger than 232 MTU with 2.07% enrichment by mass at the time of emplacement.

V. CONCLUSIONS

The accumulation of Pu-239 is limited to the near field region of a repository due to transport characteristics, restricting its role in criticality events to what would likely comprise the engineered barrier system. Its contributions to the plume of U-235 are most noticeable at early times and close proximity to the repository array. Afterwards, U-235 enrichment follows a predictable pattern with a limited scope of variation, and there are certain drop-off distances where no uranium can accumulate for a given time duration. A layout orthogonal to groundwater flow reduces these drop-offs and thus decreases the lower bounds on repository dimensions.

The highly conservative nature of the assumptions employed in these analyses has presented strict limitations for the repository source term in order to avoid criticality. The following measures (in order of increasing effect) can be used to suppress the amount of material accumulated at a given point:

- Increasing interim cooling time
- Employing higher burnup in the reactor or using a twice-through fuel assembly shuffling scheme
- Using larger pitch distances by utilizing as much of the repository footprint as possible
- Using reprocessing to separate as much TFM as possible for reuse and depositing only the residual HLW in a geochemically stable vitrified form, which would also impose solubility-limitations
- Relaxing the assumptions in the model to include transverse dispersion effects
- Using a solubility-limited release mode allowing for the precipitation of TFM in the near field
- Including the transport of a certain number of mobile, long-lived neutron poisons present in the spent fuel, which would increase the critical mass per given accumulation of TFM

This study has illustrated certain general behaviors for a discrete array of canisters containing LWR spent fuels. However, the methodology permits the use of different source terms for examination, such as the reprocessing scheme mentioned above. For example, mixed-oxide fuel will have a different nuclide inventory in the waste form, which may enhance the contributions of precursors to accumulation over expanded points in space. Furthermore, distributions of the package failure time and dissolution time as well as other model parameters should be taken into account, as opposed to the uniform and fixed values used in this paper. Such statistical analyses will indicate uncertainties associated with the results shown in this study.

Employment of the analytical tools developed in this study in critical mass simulations, including the more sophisticated model that is suggested, will be dependent on the geometry of deposition and site-specific geological measurements. Rather than using an accumulation point *X*, geometric considerations would involve fractures distributed in either a regular, heterogeneous manner or a random distribution. Natural analogues can be used to develop the parameter space, in particular the redox conditions, host rock heterogeneity, and manner of heavy metal deposition within the fractures.

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REFERENCES

1. J. AHN, E. GREENSPAN, and P. CHAMBRÉ. “A Preliminary Consideration for Underground Autocatalytic Criticality by Vitrified High-Level Waste in Water Saturated Geologic Repository,” *J. Nuc Sci & Tec*, **37**(5) (1999).

2. X. LIU, J. AHN, and F. HIRANO, "Conditions for Criticality by Uranium Deposition in Water-Saturated Geological Formations," *J. Nuc Sci & Tech*, ahead-of-print (2014).

3. Oak Ridge National Laboratory. ORIGEN-ARP Isotopic Depletion and Decay Analysis System, part of SCALE software package (Version 6.1). Available at rsicc.ornl.gov (2013).

4. L. AHONEN, H. ERVANNE, T. RUSKEENIEMI, T. JAAKKOLA, and R. BLOMQVIST, “Uranium Mineral – Groundwater Equilibration at the Palmottu Natural Analogue Study Site, Finland,” *Symposium V – Scientific Basis for Nuclear Waste Management XVI*, **294** (1992).

5. J. BRUNO and I. CASAS. "Spent fuel dissolution modelling." *Final report of the AECL/SKB Cigar Lake Analog Study*, SKB 94-04 (1994).

6. J. FABRYKA-MARTIN et al. “Nuclear reaction product geochemistry: Natural nuclear products in the Cigar Lake deposit,” *Final Report of SKB/AECL Cigar Lake Analog Study*, J.J. CRAMER and J.A.T. SMELLIE, Eds., SKB-(AECL) TR 94-04 (10851). (1994).

7. E.A. BATES, et al., "Plug Design for Deep Borehole Disposal of High-Level Nuclear Waste," *Nuclear Technology*, **188**(3) (2014).

8. Swedish Nuclear Fuel and Waste Management Company, “SKB Annual Report 1995,” Tech. Rep. 95-37, Svensk Kärnbränslehantering AB (1996).

9. D.I. DEMIRKANLI, et al., “Modeling Long-Term Plutonium Transport in the Savannah River Site Vadose Zone,” *Vadose Zone Journal*, **6**(2), 344 (2007).

10. J. S. CONTARDI, D. R. TURNER, and T. M. AHN, "Modeling Colloid Transport for Performance Assessment," *Journal of Contaminant Hydrology*, **47**(2), 323-333 (2001).

11. T. HARTER, *Groundwater Hydrology*, p. 10. L. ROLLINS and P. SUYAMA, Ed., University of California Agricultural Extension Service and the California Department of Health Services, Davis, CA (2001).