CRITICALITY SAFETY OF GEOLOGIC DISPOSAL FOR HIGH-LEVEL RADIOACTIVE WASTES

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A review has been made for the previous studies on safety of a geologic repository for high-level radioactive wastes (HLW) related to autocatalytic criticality phenomena with positive reactivity feedback. Neutronic studies on geometric and materials configuration consisting of rock, water and thermally fissile materials and the radionuclide migration and accumulation studies were performed previously for the Yucca Mountain Repository and a hypothetical water-saturated repository for vitrified HLW. In either case, it was concluded that it would be highly unlikely for an autocatalytic criticality event to happen at a geologic repository. Remaining scenarios can be avoided by careful selection of a repository site, engineered-barrier design and conditioning of solidified HLW. Thus, criticality safety should be properly addressed in regulations and site selection criteria.

The models developed for radiological safety assessment to obtain conservatively overestimated exposure dose rates to the public may not be used directly for the criticality safety assessment, where accumulated fissile materials mass needs to be conservatively overestimated. The models for criticality safety also require more careful treatment of geometry and heterogeneity in transport paths because a minimum critical mass is sensitive to geometry of fissile materials accumulation.

KEYWORDS : Autocatalytic, Positive Feedback, Geologic Repositories, Thermally Fissile Materials, Radionuclide Transport

1. INTRODUCTION

The possibility of a large energy release by autocatalytic criticality events caused by thermally fissile materials (TFM) buried in a deep geologic repository was pointed out by Bowman and Venneri [1] in 1995. While original emplacement of fissile material in repositories will be made sub-critical by design, incorporating neutron-absorbing materials and by controlling the quantity and geometry of fissile material in each canister, chemical and hydrological processes could reconfigure TFM into a critical or super-critical configuration, where on average one or more of the neutrons released by fission of a nucleus go on to cause another fission. Rock and groundwater function as moderators to slow fast neutrons to thermal velocities by collisions with light nuclei, which facilitates fission of the thermally fissile isotopes ^{235}U and ^{239}Pu.

Possibility of sustainable fission chain reactions in geologic formations was first theoretically studied by Kuroda in 1956 [2]. His theory was proved by the natural reactor found in Gabon [3] in 1972. Clayton [4] in 1979 and Gore, et al [5]. in 1981 studied criticality phenomena for the case of burial of spent nuclear fuel. In the Swedish KBS-3 study [6] in 1983, criticality safety for their concept of geologic disposal for spent nuclear fuel in granitic rock with cupper canisters was studied. What was new about scenarios postulated by Bowman and Venneri was explosive energy release due to positive feedback mechanisms, where the neutron multiplication factor increases as the system temperature increases. They called it autocatalytic criticality.

Like ordinary performance assessments focusing on the radiological safety of geologic repositories, where the exposure dose to the public is usually evaluated as the performance measure, the assessment for criticality safety of geologic repositories should also consist of four stages: (1) development of scenarios leading to TFM accumulation in an autocatalytic critical configuration, (2) development of a set of deterministic models for a performance measure for criticality safety, based on the scenarios developed, (3) a statistical analysis for uncertainties associated with the performance measure obtained by the established deterministic models and (4) judgment of repository safety by comparing the obtained value of the performance measure (with the uncertainty) with a safety standard.

First, careful scenario development needs to be made. For the scenarios postulated by Bowman and Venneri, careful reviews were made by various researchers. As discussed in the Section 2., two main categories of scenarios were identified in the previous studies [7,8]: scenarios for under-moderated (dry) autocatalytic criticality occurring

at the original emplacement location of a waste canister, and scenarios for over-moderated (wet) autocatalytic criticality either at the original emplacement location or at a location in the far field involving transport of TFM from multiple canisters. These scenarios were, then, screened out by preliminary considerations, and scenarios that need further detailed analyses were identified.

Second, for the remaining scenarios, deterministic models were developed to obtain quantitative estimates for accumulation of TFM. Models developed previously for radiological safety assessment could be utilized after some modifications (as discussed in Section 4.) because radionuclide release from failed waste canisters and transport in geologic formations are essential components also for this purpose. Careful considerations would be necessary, however, when we utilize those models developed for radiological safety in criticality safety assessment. Because a critical mass of TFM is significantly different for differing configuration and geometry of accumulated TFM, geometry and spatial distribution of waste canisters in a repository would be more important than in the case of a radiological performance assessment. Conservatism included in the radiological-safety assessment models should be carefully checked because conservative cases for criticality safety, where mass accumulation of TFM is overestimated, might not be conservative for radiological safety. For example, if significant mass accumulation occurs in the middle of a transport path between the repository and the biosphere, radiological exposure dose to the public would remain small while possibility for a criticality event to occur could be increased. The assessment models would consist of different components. For example, criticality safety assessment does not require radionuclide transport in the biosphere, whereas conservative radiological safety assessment does not include accumulation of radionuclides, which requires more detailed information for heterogeneity in geochemical and hydrological conditions in the far field.

Regarding the third and fourth stages, minimum critical masses can be calculated for various configurations of TFM and used as safety standards for criticality safety. Minimum critical masses can also be used to screen scenarios. That is if a minimum critical mass is so large that a critical configuration is not achieved even if all TFM in a repository accumulates in one location, that scenario can be excluded from the list for further investigations.

In the present paper, the previous studies performed at University of California, Berkeley for the scenario development and safety assessment for criticality safety are summarized and reviewed for the following two cases: (1) disposal of commercial spent nuclear fuel and defense wastes including weapons-grade plutonium from dismantled nuclear weapons and highly enriched uranium from spent fuels owned by the US Department of Energy at the Yucca Mountain Repository (YMR) [7,8,9], and (2) disposal of vitrified high-level waste (HLW) from reprocessing of commercial spent nuclear fuel at a water-saturated repository [10,11,12]. Discussions are made for comparison between safety assessments for radiological safety and for criticality safety.

2. SCENARIO ANALYSES FOR CRITICALITY SAFETY

Detailed scenario analyses were made in Ref. 7 for YMR. The United States is considering four classes of materials for potential geologic disposal (see Figure 1 [7]). Because of significant differences in the quantities and environmental transport behavior of TFM, the potential routes to criticality differ for each nuclide. The diagram includes four types of materials to be disposed of in the repository: commercial spent fuel with typical effective enrichment (^{235}U and ^{239}Pu) of ~2%, vitrified military HLW that contains fission products, minor actinides, and trace quantities of plutonium and uranium, 50 metric ton (MT) or more of separated excess weapons plutonium, which may be immobilized in glass or ceramic, and 210 MT or more of highly enriched uranium (HEU) from research and naval reactors.

Seven events were identified to cause autocatalytic criticality with rapid energy release, sufficient to vent radioactivity above ground, as shown in Figure 1. For a given scenario and waste form, if engineered or natural features prevent any of the seven events from occurring, venting of radioactivity become impossible. The scenario can be divided into two subsets: (1) the one at original waste emplacement location (paths 2-5-10-14 and 3-7-12-14) and (2) the one involving transport of TFM in solution to the far field away from emplacements (1-8-13, 2-4-9-13 and 3-6-11-13).

In the first subset, the resulting under-moderated configuration is driven critical by dispersion of the plutonium into surrounding rock, which effectively added more moderator to the initially under-moderated system and increased reactivity. Vaporized plutonium would vent through fractures in the surrounding dry rock, which would provide a positive feedback mechanism. Sanchez et al. [13] calculated the minimum ^{239}Pu critical mass for reflected homogeneous spheres with tuff rock as moderator to be 86 kg. A typical package for commercial spent fuel would contain 186 moles [14] (44 kg) of ^{239}Pu with a comparable mass of ^{240}Pu. Considering realistic moderating materials, heterogeneity and non-spherical geometry, this mass is not sufficient to support dry criticality. For weapons plutonium vitrified for geologic disposal, neutron absorbers with solubilities comparable to plutonium can be incorporated into waste-form design to prevent criticality. Thus, subset (1) could be screened out without detailed calculations or analyses [7]. Actually, while previous reviews [15,16,17,18] confirmed the neutronics calculations performed by Bowman and Venneri, none supported their estimates of high probability for the formation of critical deposits or for a significant release of energy.

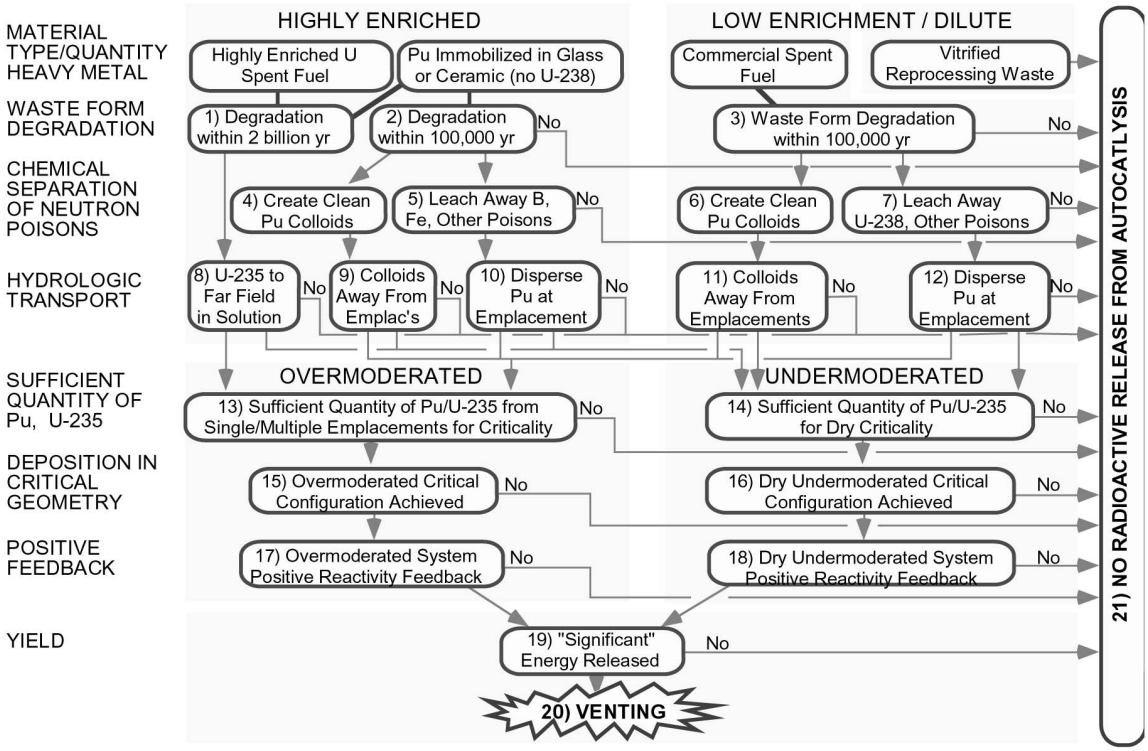


Fig. 1. Autocatalytic Event Tree [7]. The Arrow from "Vitrified Reprocessing Waste" is Directly Going to 21). For a Water Saturated Repository this in not Necessarily so, as Discussed in the Present Paper

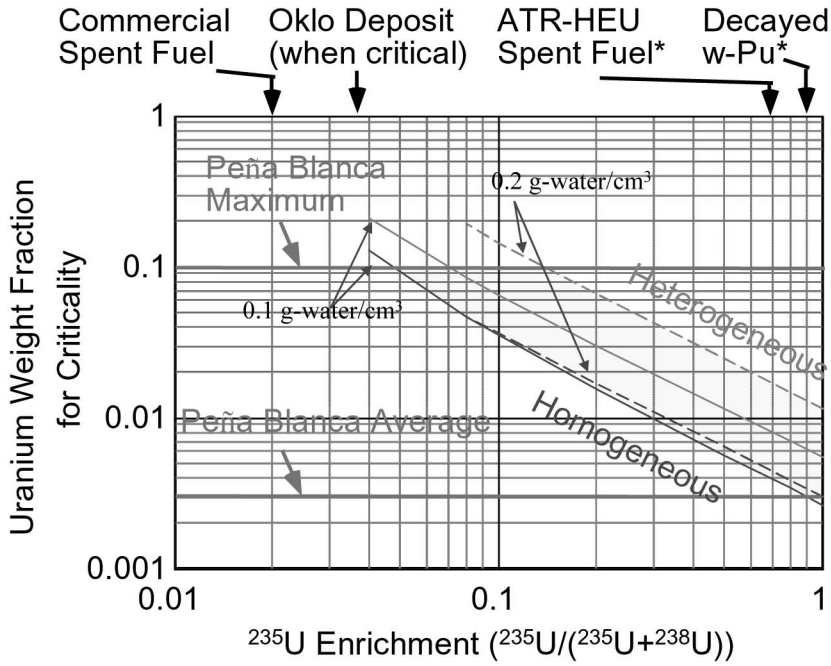
In subset (2), over-moderated (wet) criticality scenarios were considered. Water absorbs neutrons more effectively than many types of rock. Hence, in a geologic setting, a system is over-moderated when the neutron absorbing effect of water exceeds its moderating contribution, so that water expulsion from rock pores increases reactivity. If water is further expelled away from the system as the system temperature increases, then the neutron moderating contribution will become insufficient, resulting in quenching chain reactions. But, if other positive feedbacks such as neutron-spectrum hardening and homogenization of heterogeneous TFM deposition exceed the effects of water, the system can be autocatalytic. This will be discussed in Section 3. Over-moderated criticality in a geologic setting was recognized in the 1970s but generated less interest then because no plans existed to dispose of HEU and it was difficult to postulate credible chemical mechanisms to separate plutonium from the large quantities of neutron absorbing ^{238}U in commercial spent fuel.

At YMR [7], while the path 3-6-11-13 involving Pu in commercial spent fuel was screened out because the mass of ^{238}U is significantly large to avoid criticality events, two waste forms were considered to yield HEU for this scenario: HEU spent fuel (path 1-8-13) and weapons-grade plutonium immobilized in glass or ceramic (path 2-4-9-13). For a water-saturated repository containing vitrified HLW from reprocessing of commercial spent fuel where more than 99% of uranium is removed, the path 1-8-13 was also considered important because the mass fraction of ^{235}U in uranium could be as high as 12% after decay of ^{243}Am and ^{239}Pu (Ref. 10).

For these scenarios involving transport of TFM away from emplacements, the potential quantity of TFM is greater than the inventory in a single canister. The aforementioned paths could not be eliminated by preliminary consideration and detailed analyses were performed (Refs. 8 and 9 for YMR; Refs. 10, 11 and 12 for water-saturated repositories). For both cases, first neutronic evaluations were performed to evaluate minimum critical masses with ^{239}Pu or HEU in heterogeneous geologic settings for autocatalytic criticality. Then, transport models were established to evaluate if such mass can be supplied to by far-field transport from multiple waste canisters.

3. CRITICAL MASSES AND POSITIVE FEEDBACK MECHANISMS

For critical mass for uranium, Figure 2 [7] shows the uranium mass fraction for criticality as a function of the



- Heterogeneous system has parallel U-filled fractures spaced 20 cm
- Actual uranium deposits likely lie between homogeneous and heterogeneous cases shown here

Fig. 2. Average Uranium Density Required to Achieve Criticality in Tuff Rock in an Infinite System [7]. Red Curves Show Heterogeneous Mixtures for High Porosity (0.2 g-water/cm^3 – dashed) and Low Porosity (0.1 g-water/cm^3 – solid). Blue Curves (dashed and solid) for Homogeneous Mixture Show Negligible Effects of Porosity

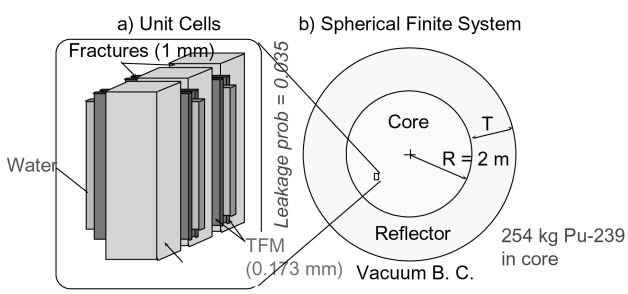


Fig. 3. Schematic Parallel Fracture Lattice and the Spherical Finite System for Static Neutronic Calculation to Determine Minimum Critical Mass of TFM

uranium enrichment for an infinite system. This shows that uranium from commercial spent fuel is not sufficiently enriched for criticality configuration even in an infinite system where neutron leakage from the system does not have to be considered. But, with decayed weapons plutonium or HEU spent fuel, if heterogeneous depositions of uranium is homogenized and water is removed, the minimum uranium mass fraction for criticality decreases significantly, implying possibility of positive reactivity feedback.

In Ref. 7, static calculations were performed to determine minimum critical mass for plutonium dioxide deposited on surfaces of parallel fractures in tuff rock at YMR as shown in Figure 3. To make the spherical homogenized core critical, for example, one must have a 2-meter core contain 254 kg of ^{239}Pu surrounded by 1.2 m of rock. For various radii, the minimum Pu surface concentration for maintaining criticality in the parallel-fracture configuration was obtained as ~ 1 kg/m^2 depending on the geometrical and material parameters such as fracture spacing and rock compositions.

In Ref. 10, the minimum mass of 12%-enriched uranium in granite with various values of rock porosity was obtained. The same configuration as Figure 3 was used for evaluation. Granite was assumed instead of tuff. Table 1 summarizes the critical masses of ^{235}U and the radii of the corresponding spherical cores. The over-moderated critical mass is significantly greater than the under-moderated critical mass. Both kinds of critical masses are sensitive to the rock porosity and composition. While with a deposit in rock with a porosity of 0.1 or lower, no critical configuration

Table 1. Summary of Critical Masses of ^{235}U and Core Radii of Homogeneous Spherical Reflected UO_2, Rock and Water

Medium/Porosity	Minimum critical mass of U-235, Core radius	
	under-moderated criticality	over-moderated criticality
Water	1.5 kg, 19 cm	—
SiO_2 / 10%	17 kg, 71 cm	48 kg, 130 cm
Granite / 10%	60 kg, 90 cm	—
Granite /30%	7.0 kg, 37 cm	35 kg, 90 cm

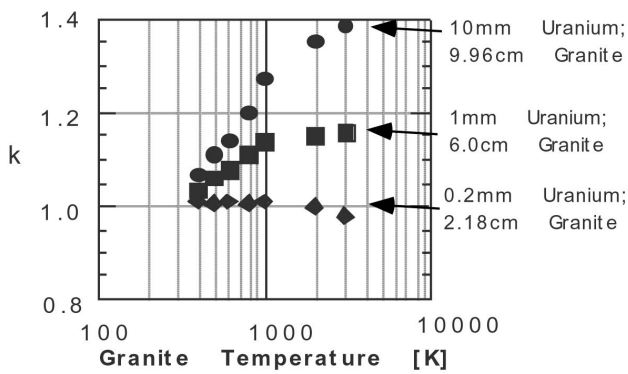


Fig. 4. Variations of the Neutron Multiplication Factor, k_∞, due to Temperature Elevations in Granite for the Case of $U(12)O_2$ Deposition on Fracture Surfaces in Granite with 30% Porosity

can be conceived of, high porosities such as 0.3 or greater are observed in highly fractured zones in granitic formations. While such highly fractured regions will be excluded from candidate sites for a repository, this result implies that high-porosity regions in rock formations surrounding the repository site would also be of concern for repository safety.

Figure 4 shows variations of the neutron multiplication factor with the temperature in granite. It is observed that the neutron multiplication factors increase with the rock temperature, because the neutron spectrum is hardened and the self-shielding effects by the thin layer of uranium on the fracture surfaces decrease.

Table 2 summarizes the effects of temperature elevations in rock and in the UO_2 layer on the neutron multiplication factor. By comparing the values in a vertical column, negative feedback due to Doppler broadening is observed, while on the horizontal rows, positive feedback due to spectrum hardening is observed. The values on the diagonal line can be considered to show the coupled effects of these two feedback mechanisms, indicating positive feedback with elevating system temperatures.

Table 2. Change in the Neutron Multiplication Factor due to Temperature Elevations in Rock and in $U(12)O_2$ Layer

Temperature in UO_2 layer [K]	Rock Temperature [K]		
	293	587	881
293	1.000	—	—
587	0.997	1.068	—
881	0.989	—	1.090

Table 3. Positive Feedback due to Homogenization of Rock, Water and $U(12)O_2$

Porosity of granite [%]	$U(12)O_2$ Layer Thickness [mm]		
	0.2	1.0	10
10	1.0535	1.2654	1.3872
20	1.0706	1.3193	1.4911
30	1.0841	1.3470	1.5264

Table 3 shows positive feedback due to homogenization of the UO_2 layer with surrounding rock matrix. It shows that with a greater porosity, i.e., with a greater moisture content in the rock matrix, the positive feedback is greater due to homogenization.

Thus, it was confirmed that uranium with 12% enrichment deposited heterogeneously in rock formations can be made in an over-moderated criticality configuration, if the rock porosity is 30% or higher. Because of these positive reactivity feedback mechanisms due to temperature elevation in addition to removal of water from the system, the over-moderated system with heterogeneous depositions of $U(12)O_2$ in granite formations can be autocatalytic.

4. TRANSPORT MODELS FOR CRITICALITY SAFETY ASSESSMENT

From the aforementioned scenario analyses, it was concluded that geometric and material configurations of TFM with rock and water that are critical and have positive feedback are actually conceivable both for YMR and water-saturated repositories. Then, the next question was, can these configurations occur in geologic, hydrologic and

geochemical setting of a repository? To answer this question, models for TFM transport and accumulation were developed by utilizing those developed for radiological safety assessment readily available at that time.

4.1 Yucca Mountain Repository

As discussed in Section 2., two potential scenario subsets were pointed out for YMR, leading to deposition of Pu and HEU in a critical configuration away from multiple canisters, transported by groundwater flow. To simplify the analysis, parallel planar fractures were assumed to intersect the repository plane (Figure 5). It was assumed that water flows down through vertical fractures steadily in a liquid column with a constant velocity, and that water in the pores of the rock matrix is stationary. Materials released from a failed package that exist as solutes are transported through fractures by advection, and diffuse into the rock matrix by molecular diffusion. As the pores in the rock matrix are partially filled with water, diffusion in the rock matrix occurs only through the interconnected water phase in the pores. The solutes in the pores of the rock matrix are in sorption equilibrium with the solid phase of the rock, resulting in retardation of radionuclide transport.

Weapons plutonium would be solidified in borosilicate glass for geologic disposal. After the package has failed, plutonium and uranium generated by decay of plutonium isotopes are released into groundwater flow by glass-matrix alteration and dissolution. For the glass dissolution time after package failure, 10^5 years was assumed.

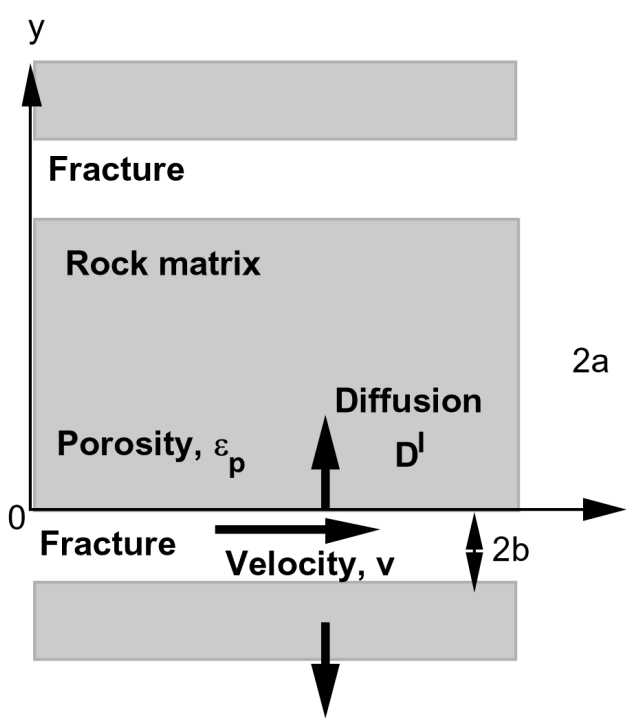


Fig. 5. Conceptual Configuration of Parallel Fractures in the Host Rock [24]. Radionuclides Released from the Engineered Barriers Enter from the Left

Plutonium would be transported in a colloidal form as well as in a solute form. Plutonium that exists as a true solute in the water phase (solute plutonium) in the fractures and in the pores of the rock surrounding the fractures are in equilibrium with the sorbed phase on the rock (sorbed plutonium). Plutonium also attaches to colloids (colloid plutonium) in the water present in the fractures, and detaches from colloids by soluble complexing agents such as carbonate. Colloid plutonium may accumulate onto the surfaces of the fractures by agglomeration, flocculation, and/or precipitation. In Yucca Mountain conditions, colloids are observed to have positive surface charge, and so would be expected to attach to tuff with negative surface charge. Thus, to evaluate deposition of plutonium on the fracture surface, two bounding models were developed: (1) the pure-colloid model and (2) the pure-solute model. In the pure-colloid model, it was assumed that all the plutonium released from glass logs is carried away in a colloidal form. In the pure-solute model, plutonium transport was modeled with the planar fracture configuration with matrix diffusion and sorption retardation taken into account.

4.1.1 Pure-colloid Model

In the pure-colloid model, it was assumed that plutonium release continues for the glass dissolution time, and that the colloid plutonium cannot enter the rock matrix pores. The governing equation for the colloid plutonium deposition $P_9^C(t;z)$ [kg-Pu/m] in the fracture is written as

$$\frac{dP_9^C(t;z)}{dt} = 2bLN_9^C(z,t)\Lambda_9 - \lambda_9 P_9^C(t;z), \quad t>0,\ P_9^C(t=0)=0, \tag{1}$$

where $N_9^C(z,t)$ is the concentration of the colloid plutonium in the fracture water, governed by

$$\frac{\partial N_9^C}{\partial t} + v\frac{\partial N_9^C}{\partial z} + (\Lambda_9+\lambda_9)N_9^C = 0,\ t>0,\ z>0, \tag{2}$$

subject to

$$N_9^C(z,0) = 0,\ z>0, \tag{3}$$

and

$$N_9^C(0,t) = N_9^{C0}\exp(-\lambda_9 t)\left[h(t)-h(t-\tau)\right],\ t>0. \tag{4}$$

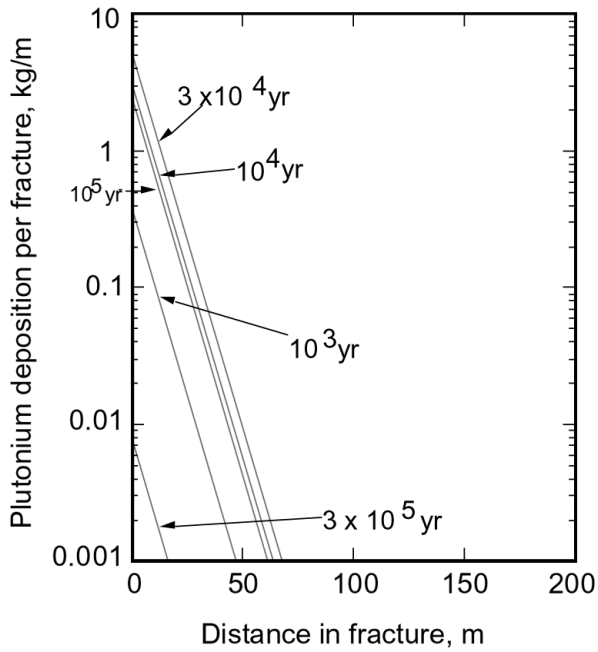


Fig. 6. Colloid Plutonium Immobilization on the Fracture Surfaces for v = 40 m/yr, Λ_9 = 0.0139 day^{-1}, L = 100 m [9]

Λ_9 is defined as a rate constant for flocculation. z is the distance from the entrance of the fracture. $2b$ is the aperture of the fracture. v is the water velocity in the fracture. λ_9 is the radioactive decay constant. N_9^{C0} is the initial colloid concentration determined by the initial inventory of ^{239}Pu, the water flow rate through a fracture, and the glass dissolution time, τ. The width L of the fracture opening allocated for one canister also needs to be determined from the canister-array configuration in the repository. The function $h(t)$ is a Heaviside step function.

The solution is illustrated in Figure 6. The amount of plutonium deposited on the fracture walls increases with time at an early time. The accumulation becomes as large as 5 kg/m in the vicinity of the fracture entrance at 30,000 years. If we define the cut-off accumulation arbitrarily as 0.01 kg/m, the plutonium deposition extends to 50 m downward from the fracture entrance. After 30,000 years, due to radioactive decay, the amount of accumulated ^{239}Pu decreases with time. Thus, the significant accumulation can occur only in the vicinity of the fracture entrance at a relatively early time. From 5 kg/m, a maximum surface concentration of Pu is calculated as 5 [kg/m] / (2 $\times$ 100 [m]) = 0.025 kg/m^2. Compared with the minimum Pu surface concentration 1 kg/m^2 (see Section 3.) for maintaining criticality in the same parallel-fracture configuration, the maximum accumulation obtained here is too small for criticality.

4.1.2 Pure-solute Model with Two-member Decay Chain

For transport of pure solutes, governing equations are established for solute concentrations in groundwater in the fracture and in the pores in the rock matrix. For this type of radionuclide migrations, previous researchers developed analytical solutions for variety of cases [19,20,21, 22]. Among them, the Laplace-transformed solutions given in Ref. 22 are considered as one of the most generalized cases with hydrodynamic dispersion in the fracture, type-III boundary condition for solute concentrations at the entrance of the fracture and a multiple-member decay chain. The radionuclide transport equations are shown here for the reader's convenience for the concentrations C_k (z,t) [mol/m^3] of radionuclide k in the fracture and C_k^P $(y,t;z)$ in the pores of the rock matrix the porosity ε_p :

$$R_{e(k)}\frac{\partial C_k}{\partial t}+v\frac{\partial C_k}{\partial z}-D^L\frac{\partial^2 C_k}{\partial z^2}-\frac{D^I_{e(k)}}{b}\cdot\frac{\partial C_k^P}{\partial y}\bigg|_{y=0}$$
$$+R_{e(k)}\lambda_k C_k-R_{e(k-1)}\lambda_{k-1}C_{k-1}=0, \qquad (5)$$
$$t>0,\ z>0,$$

and

$$\alpha_{e(k)}\frac{\partial C_k^P}{\partial t}=$$
$$D^I_{e(k)}\frac{\partial^2 C_k^P}{\partial y^2}-\alpha_{e(k)}\lambda_k C_k^P+\alpha_{e(k-1)}\lambda_{k-1}C_{k-1}^P, \qquad (6)$$
$$t>0,\ 0<y<a,\ z>0.$$

Here k = 1, 2, …, i, with λ_0 = 0, and λ_k [yr^{-1}] is the decay constant of radionuclide k. In the present case, ^{239}Pu and ^{235}U are treated as a 2-member decay chain (i =2).

The radionuclides released at the outer surface of the Engineered Barrier System (EBS) are assumed to enter the fracture at z = 0. y is the coordinate into the rock matrix surrounding the fracture. y = 0 is located at the interface between the fracture and the rock matrix. The boundary at $y = a$ is considered as the mid-plane between two adjacent planar fractures. Through this boundary, the diffusive mass flux is assumed to be zero. t is the elapsed time since the beginning of the radionuclide release from the failed canister. The longitudinal hydrodynamic dispersion coefficient

D^L [m^2/yr] is assumed to be constant with time and uniform in the fracture and common for all radionuclides. Molecular diffusion of radionuclides from water flowing in the fractures into the pores of the rock matrix is an important retardation mechanism. $D^I_{e(k)}$ [m^2/yr] is the diffusion coefficient for radionuclide k of element e in the rock. Subscript $e(k)$ indicates that the k-th member nuclide in a decay chain is an isotope of element e. For sorption equilibrium in the porous rock matrix, we define the capacity factor $\alpha_{e(k)}$ for radionuclide k of element e as

$$\alpha_{e(k)} = \varepsilon_p + \rho_p \left(1-\varepsilon_p\right) K^e_{dp} \ , \tag{7}$$

where ρ_p [kg/m^3] is the density of the porous rock matrix, and K^e_{dp} [m^3/kg] the sorption distribution coefficient of element e for the rock matrix. The retardation coefficient, $R_{e(k)}$, for advection and longitudinal dispersion in the fracture is defined as

$$R_{e(k)} = 1 + \frac{1-\varepsilon_f}{\varepsilon_f} \rho_f K^e_{df} \ , \tag{8}$$

where ρ_f [kg/m^3] and ε_f are the density and the porosity of the material filling the fracture. K^e_{df} [m^3/kg] is the sorption distribution coefficient of element e for the material filling the fracture.

The initial and boundary conditions for the governing equations are

$$C_k(z,0) = 0, \ z > 0, \tag{9}$$

$$C^P_k(y,0;z) = 0, \ 0 < y < a, \ z > 0, \tag{10}$$

$$S_f \varepsilon_f \left[v C_k(0,t) - D^L \left. \frac{\partial C_k}{\partial z} \right|_{z=0} \right] = S_2 Q_k(t), \ t > 0, \tag{11}$$

$$C_k(\infty,t) = 0, \ t > 0, \tag{12}$$

$$C^P_k(0,t;z) = C_k(z,t), \ t > 0, \ z>0, \tag{13}$$

$$\left. \frac{\partial C^P_k}{\partial y} \right|_{y=a} = 0, \ t > 0, \ z > 0. \tag{14}$$

The time dependent function $Q_k(t)$ [mol/m^2·yr] is the mass flux of radionuclide k that is released from the engineered barrier and enters the fracture system at $z = 0$. For this, mechanisms assumed for the release of radionuclides from a failed waste canister and for the transport in the EBS need to be modeled. An expression given in Ref. 24 was developed for the EBS in a water-saturated repository. Expressions for boron, ^{239}Pu and ^{235}U at YMR were also developed in Ref. 9. S_2 [m^2] is the surface area of the EBS containing one failed canister. S_f [m^2] is the area of the fracture opening, through which radionuclides released from the EBS enter water flow in the fracture.

Analytical solutions were given in the form of recursive Laplace transforms in Ref. 22. The inverse Laplace transform was performed numerically by the Talbot method [23]. Computer codes were developed for this calculation and reported in Ref. 24.

Matrix diffusion is an important mechanism for retardation of radionuclide transport. The matrix diffusion coefficient and the sorption retardation coefficient are the two principal parameters for this mechanism. From the reported ranges of the sorption distribution coefficients, the largest values were selected as the conservative case for the criticality safety assessment because it leads to greater accumulation of TFM. For the same reason, no sorption is assumed for boron because boron is an effective neutron absorber, and with no sorption assumed, boron will be quickly removed from TFM accumulation. This consideration is a good example of difference in conservatism between radiological safety and criticality safety.

The mass of ^{239}Pu in the repository can be calculated, based on the mass balance equations, considering losses by radioactive decay and release by glass-matrix dissolution. By solving them, the residence time t^*_{Pu} of ^{239}Pu in the repository is obtained as

$$t^*_{Pu} = \frac{1}{\lambda_9} \ln\left(1 + \frac{m^0_9 \lambda_9}{Q N^*_{Pu}}\right), \tag{15}$$

where m^0_9 is the initial inventory of ^{239}Pu in a waste canister, Q the volumetric water flow rate washing the canister, and N^*_{Pu} the solubility of Pu. Because of this relationship, even if we assume two orders of magnitude variations for the water infiltration rate and the solubility, the amount of ^{239}Pu taken away from the repository by advection into the fractures is still likely to be small, and most ^{239}Pu decays to ^{235}U while still in the repository. For example, with the solubility of 10 ppb for Pu and an extremely high water flow

rate of 1000 m^3/yr, 3% of the initial inventory of ^{239}Pu is carried away by advection from the repository as plutonium; the rest will be carried out as ^{235}U.

Uranium-235 is generated by decay of ^{239}Pu in the waste package. The detailed analysis was also done for the residence time of uranium. The residence time is determined by the rate of generation by decay of ^{239}Pu and the rate of removal by water flow from the canister. Boron is readily soluble, so that what is released from the glass logs can entirely dissolve in the water infiltrating the repository.

Figure 7 depicts the time evolution of spatial distributions of ^{239}Pu, ^{235}U and boron. In each rectangle, the fracture is located on the left vertical side. The failed waste package is located at the top-left corner of each rectangle. The right vertical side represents the mid plane between two adjacent fractures. The dimensions of the rectangle are 200 m (vertical) and 0.37 m (horizontal). The water velocity in the fractures is assumed to be 40 m/yr. Numerical results indicate that solute ^{239}Pu stays within a distance of 10 m from the repository, and decays there to a negligible level in 200,000 years.

Uranium-235 spreads much farther from the repository than ^{239}Pu because of its weaker sorption and greater solubility. The maximum concentration, which is equal to the uranium solubility, is observed at the fracture entrance as long as ^{235}U remains in the repository. After that, because water with no uranium contamination enters the fractures, ^{235}U in the rock matrix diffuses back to the fractures due to the reversed concentration gradient at the rock/fracture interface [21]. Uranium stored in the rock matrix is slowly washed by the water flowing in the fractures, and by 1 million years, ^{235}U remaining in the rock matrix becomes negligible. Boron spreads in the medium during the glass leaching period, and quickly disappears after the end of the leach time.

4.1.3 Interpretation of Results

All the ^{239}Pu that originally exists in the repository reaches the bottom as ^{235}U. Practically no ^{239}Pu is observed at the bottom. Because plutonium in either solute or colloidal form exists in only a small travel distance, uranium is the principal TFM of concern that might create accumulation

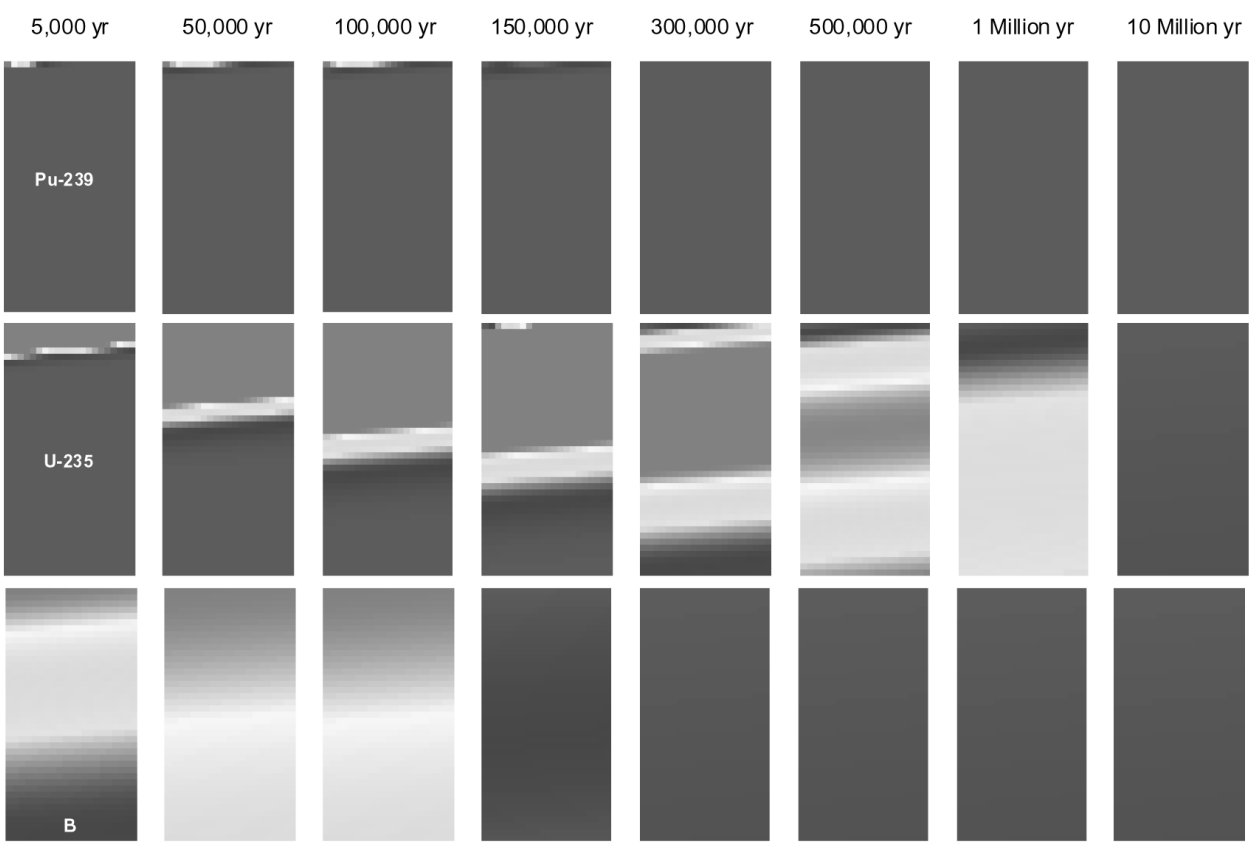


Fig. 7. Evolution of Concentration Isopleths of ^{239}Pu (top), ^{235}U (middle) and Boron (bottom). For Pu and U, Concentrations are Normalized by Their Solubilities. The Glass Dissolution Time is 100,000 Years

in the far field in a critical configuration.

The critical concentration of ^{235}U in infinite homogeneous UO_2-tuff-water system with 10% porosity and pure ^{235}U is obtained as 6×10^{-3} g/cm^3 (Ref. 7). With the uranium solubility (10^{-6} g/cm^3), the porosity (0.1), water saturation (65%), the tuff density (2.2 g/cm^3) and the sorption distribution coefficient (5 cm^3/g), the maximum uranium concentration in the tuff is calculated as $[0.1 \times 0.65+(1- 0.1 \times 0.65) \times 2.2 \times 5] \times 10^{-6} = 10^{-5}$ g/cm^3, which is more than two orders of magnitude smaller than the critical concentration.

Thus, while we can conceive of geometric and material configurations of TFM with rock and water, which are critical and have positive feedback, it is unlikely that plutonium will travel large distances to form these configurations. Plutonium is more likely to be re-deposited close to its original emplacement. Uranium 235 can move large distances as a solute, but it is unlikely to precipitate into critical configurations because of a lack of reducing agents in the mountain. So, without further detailed probabilistic studies, it was concluded that autocatalytic criticality events at YMR would be unlikely [8].

4.2 Water-saturated Repositories for Vitrified HLW

4.2.1 Background and Outline for Analyses

The path 1-8-13 in Figure 1 for vitrified HLW from reprocessing of commercial spent fuel was studied for a water-saturated repository. After reprocessing, more than 99% of uranium and plutonium are separated, leaving all fission products and minor actinides (Np, Am and Cm) as high-level liquid wastes, which are solidified with borosilicate glass. Because more than 99% of uranium (mostly ^{238}U, a strong neutron absorber) in spent fuel is removed by reprocessing, and because ^{243}Am, ^{239}Pu, ^{245}Cm, ^{241}Am and ^{237}Np decay to fissile uranium isotopes (^{235}U or ^{233}U), the fraction of fissile uranium isotopes in the total actinide mass can reach 46%. If we do not include the contribution by ^{233}U because of its relatively short half-life, the fraction of fissile uranium isotopes can be 12%.

To evaluate contribution of failed canisters for accumulation of TFM as a function of time and the transport distance between the failed canister and the accumulation location, an existing computer code [24] for the release and transport of radionuclides from a single failed canister in the water-saturated geologic formation was utilized and modified. The mass accumulation obtained by this model was considered the upper bound for the mass that a single failed canister can create [10]. Then, contribution of multiple failed canisters in a repository to TFM accumulation in the far field was evaluated by considering different transport distances from failed canisters to the accumulation location and by superposing results of single canister results. By parametric studies, it was found thet for the case where high mobility and solubility were assumed for uranium isotopes decay loss in the course of transport to the accumulation location is negligible, so that possibility of accumulation of large mass of uranium could not be eliminated. But, there was no logic to determine how many failed canisters would contribute to accumulation. Therefore, to remove too much conservatism, a model was developed by applying compartmentalization to take into account interference of adjacent canisters, i.e., non-linear effects of canister multiplicity in a repository [12]. In Section 4.2.2, the result from the single canister model is briefly summarized. In Section 4.2.3, the multi-compartment model is discussed.

4.2.2 Single Canister Model and Linear Superposition

The model for transport of a multi-member decay chain in a planar fracture, for which the formulation is shown in Section 4.1.2, was utilized and modified to evaluate mass accumulation of radionuclides in the far field. All the radionuclide of interest arriving at a specified point a distance l [m] from the surface of the EBS was assumed to accumulate there. The governing equation for the mass $\Omega_k(t)$ [mol] of the k-th radionuclide in a decay chain of length i, accumulated at $z = l$ is written as

$$\frac{d\Omega_k(t)}{dt} = -\lambda_k \Omega_k(t) + \lambda_{k-1}\Omega_{k-1}(t) + \varepsilon_f S_f j_k(l,t), \quad t > 0,\ k = 1, 2, \ldots, i, \text{ with } \lambda_0 = 0, \tag{16}$$

subject to

$$\Omega_k(0) = 0, \text{ where} \tag{17}$$

where

$$j_k(l,t) = vC_k(l,t) - D^L \left.\frac{\partial C_k}{\partial z}\right|_{z=1}, \quad t > 0, \tag{18}$$

is the mass flux of radionuclide k flowing into the accumulation location at $z = l$.

The Laplace transform $\tilde{\Omega}_k$ for the mass accumulation of radionuclide k was obtained by solving the above equations [10]. The formula for $\tilde{\Omega}_k$ contains the Laplace-transformed mass flux $\tilde{j}_k(l,p)$ of radionuclide k at a distance l from the fracture entrance. A computer code was developed to obtain the time-dependent mass accumulation $\Omega_k(t)$ of radionuclide k in the host rock by modifying the code developed and reported in Ref. 24.

The accumulated masses of radionuclides from a single failed canister were evaluated as a function of time at the locations 10, 100, 1000 m away from the surface of EBS

containing the failed canister. For conservative evaluations, it was assumed that the radionuclides released from the failed canister migrate toward the accumulation location, and those reaching the accumulation location accumulate at that location.

If mobility of uranium isotopes is assumed to be significantly smaller than those of other actinide elements, only ^{233}U can contribute to a significant accumulation after a transport of more than 100 m, because ^{237}Np with a half-life as long as 2.14 million years generates ^{233}U. ^{233}U is accompanied by ten times or more mass of ^{237}Np, a strong neutron absorber. Therefore, this case can be excluded from further analyses of criticality effects.

If mobility of uranium isotopes is significantly greater than those of other actinide elements, it was found that the precursors to uranium isotopes decay out before they accumulate in the host rock. At the accumulation location, only uranium isotopes are observed. Approximately one mole of uranium could be accumulated at the 1000 m location in the host rock, contributed from one failed canister. The fissile-isotope fraction (^{235}U/[^{238}U + ^{235}U]) is about 12%, but decreases significantly with time after 10^7 yr. Considering that a repository would contain tens of thousands of canisters, for this case, possibility of criticality events cannot be eliminated by this single-canister model, where accumulation from a single canister is superposed linearly. A model that takes into account canister-array configuration in a repository and interference of adjacent canisters needs to be developed.

4.2.3 Compartment Model with Canister Multiplicity Effects

To take into account the effects of interference, a model is developed, depicted in Figure 8, where a row of canisters is divided into compartments, each containing a canister, the diffusion buffer around the canister, and the near-field host rock. The compartment has an areal extent of $d \times d$, where d [m] is the pitch between two adjacent canisters. The cylindrical waste form is transformed into slab geometry with the same interfacial area. The slab source is assumed to have a width d. Between the slab source and the near-field rock, there are assumed to be the bentonite regions on both sides of the slab source. The bentonite region has the thickness of L_b.

Mass balance equations are obtained for radionuclide k existing in the canister:

$$\frac{dm_k^n(t)}{dt} = -\lambda_k m_k^n(t) + \lambda_{k-1} m_{k-1}^n(t) - q_k^n(t), \qquad (19)$$
$$0 < t < t_k^n,\ n = 1, 2, \ldots, N,\ \lambda_0 \equiv 0,$$

where $m_k^n(t)$ is the mass of radionuclide k in the canister in the n-th compartment located relative to the upstream side of the repository. $q_k^n(t)$ is the release rate of radionuclide k from the canister into the buffer. $q_k^n(t)$ is written as the diffusive mass flux based on the solution to Eq. (24). After t_k^n, $q_k^n(t)$ is set equal to zero. N is the total number of compartments in a row. t_k^n is the time when radionuclide k depletes completely from the canister in compartment n, and is obtained numerically.

For the near field rock region, the concentration of radionuclide k is homogenized to be $C_k^n(t)$ [mol/m^3] in the pore water of the near-field rock of compartment n. The governing equation for $C_k^n(t)$ is written as

$$\alpha_{e(k)} V \frac{dC_k^n(t)}{dt} =$$
$$-\lambda_k V \alpha_{e(k)} C_k^n(t) + \lambda_{k-1} V \alpha_{e(k-1)} C_{k-1}^n(t)$$
$$+ F C_{k-1}^n(t) - F C_k^n(t) + Q_k^n(t), \qquad (20)$$
$$C_k^0 = 0,\ t > 0,\ n = 1, \ldots, N.$$

The volumetric flow rate F [m^3/yr] of water through the interface between two adjacent compartments, is defined as

$$F = vHd\varepsilon_p, \qquad (21)$$

where v [m/yr] is the pore velocity of groundwater. ε_p is the porosity of rock. V [m^3] is the volume of the near-field rock in a compartment. Sorption equilibrium between the solid phase and the porewater phase is assumed. Then, $\alpha_{e(k)}$ is the capacity factor for element e, and is defined by Eq. (7). $Q_k^n(t)$ [mol/yr] is the release rate of radionuclide k at the buffer/rock interface. $Q_k^n(t)$ is also written as the diffusive mass flux based on the solution to Eq. (24).

The initial conditions are:

$$C_k^n(0) = 0,\ n = 1, \ldots N, \text{ and} \qquad (22)$$

and

$$m_k^n(0) = M_{k(o)},\ n = 1, \ldots N, \qquad (23)$$

where $M_{k(0)}$ is the initial mass of radionuclide k in a canister, and is assumed to be identical for all the compartments.

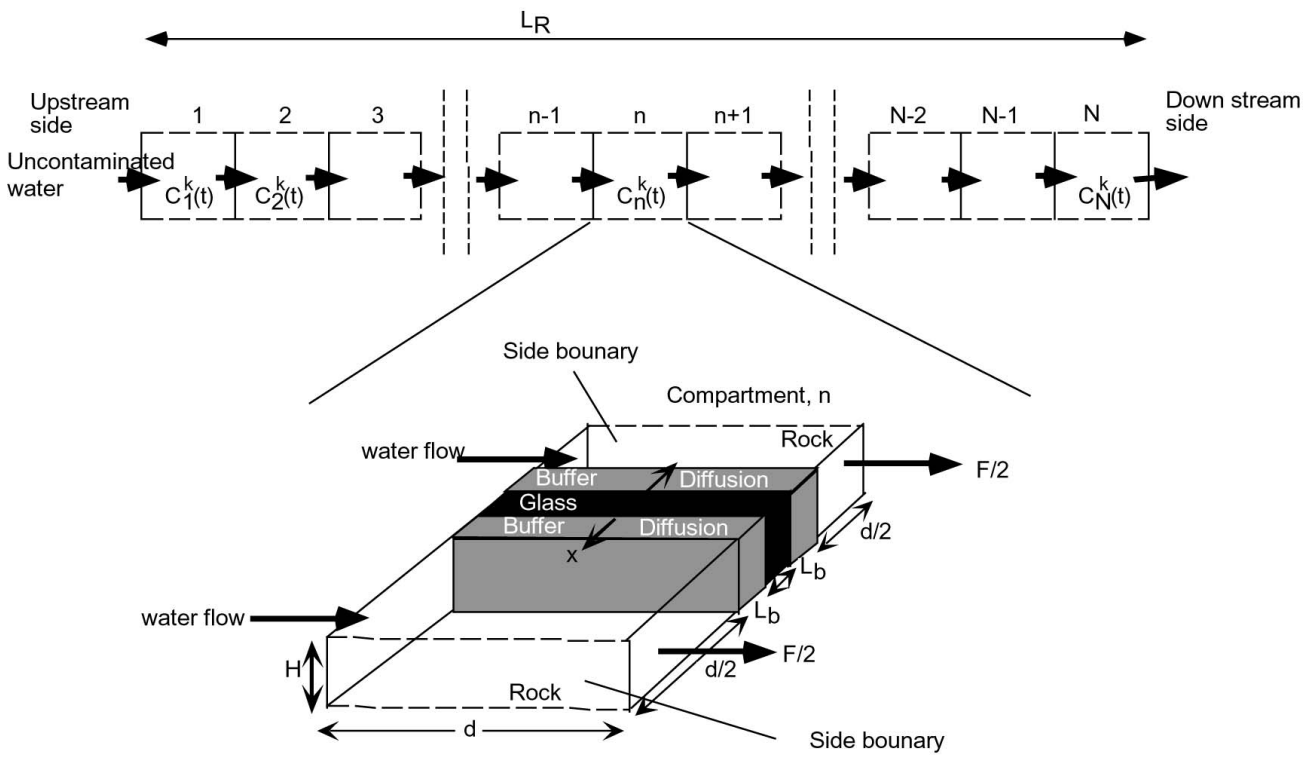


Fig. 8. Schematic Representation of Compartments and Diffusive-advective Transport Path

Radionuclides released from the canister are assumed to diffuse through the buffer into the near-field rock. The diffusion of radionuclides through the buffer region is considered to be one-dimensional. Radioactive decay during the diffusion is included. Then, the concentration, $N_k^n(x;t)$, of radionuclide k in the buffer is governed by the following equation:

$$K_{e(k)}\frac{\partial N_k^n}{\partial t} = D_{e(k)}\frac{\partial^2 N_k^n}{\partial x^2} - K_{e(k)}\lambda_k N_k^n, \quad t>0, 0<x<L_b,\ n=1,\ 2,\ldots,N, \tag{24}$$

where $D_{e(k)}$ is the molecular diffusion coefficient [m^2/yr] for element e. Sorption equilibrium between the solid phase and the pore-water phase is assumed. $K_{e(k)}$ is the retardation factor.

At the buffer/rock interface, the following boundary condition is imposed,

$$N_k^n(L_b,t) = C_k^n(t),\ 0<t<t_k^n,\ n=1,2,\ldots,N. \tag{25}$$

At the canister surface, the following boundary condition is imposed:

$$N_k^n(0,t) = N_e^*\gamma_k^n,\ 0<t<t_k^n,\ n=1,2,\ldots,N, \tag{26}$$

where N_e^* is the solubility of element e. The factor γ_k^n is the solubility appropriation for isotope k, if element e has multiple isotopes. ^{235}U and ^{238}U are assumed to share the uranium solubility.

From the downstream side of the repository, i.e., from compartment N, the radionuclide k is released into the far field. The total mass $\overline{M}_k(t)$ of the radionuclide k existing in the entire far field at time t gives the theoretical upper bound for the mass of radionuclide k that can accumulate in a single location. For a fissile radionuclide such as ^{235}U, the most conservative assumption for criticality safety assessment is that all of the radionuclide released from the repository region to the far field accumulates at a single location. Because the radionuclide that is released from the

repository region is dispersed in the far field, the situation considered will never happen; the mass of accumulated radionuclide in a single location will always be smaller than the total mass existing in the far field. To quantify the actual accumulation, the detailed mechanisms and conditions for the mass transport and accumulation in the far field must be known.

To calculate the mass $\overline{M}_k(t)$ of the radionuclide k, existing in the far field, the mass balance equation can be written. Radioactive decay loss is taken into account.

$$\frac{d\overline{M}_k}{dt} = -\lambda_k \overline{M}_k + \lambda_{k-1}\overline{M}_{k-1} + FC_k^N(t), \quad t > 0, \qquad (27)$$

subject to $\overline{M}_k(0) = 0$. The term $FC_k^N(t)$ represents the mass release rate of radionuclide k from the down stream side of the repository to the far field. $\overline{M}_k(t)$ includes the details of mass transport in the repository through the term $C_k^N(t)$.

For the analysis of accumulation of uranium isotopes, as mentioned above, transport of two uranium isotopes, ^{235}U and ^{238}U with no precursors from multiple canisters is of interest. For this particular case, as discussed in Ref. 12, steady-state diffusion can be assumed in the buffer for further simplification. Then, analytical formulae for $N_k^n(x;t)$, $Q_k^n(t)$ and $q_k^n(t)$ could be obtained.

4.2.4 Uncertainty Evaluation and Interpretation of Results

With this multi-compartment model, a statistical analysis was performed to determine uncertainty associated with the mass of ^{235}U accumulation in the far field resulting from 40,000 canisters in a water-saturated repository and from uncertainties with the pore velocity, the near-field rock porosity, the buffer porosity, the diffusion coefficient of uranium in the buffer, the sorption distribution coefficients in the buffer and in the near field rock, and uranium solubility. The total mass of ^{235}U in the far field before 100 million years is less than 40 kg with a 90% confidence level. Considering that the minimum mass of ^{235}U for 12%-enriched uranium for autocatalytic criticality in over-moderated condition is 35 kg (Table 1), the result shows that almost all ^{235}U existing in the far field at that time must accumulate at a single location for ^{235}U existing in the far field to be critical in an over-moderated configuration.

5. DISCUSSIONS

5.1 So, Is Geologic Disposal Safe?

From the previous studies summarized here, we can consider the followings regarding the criticality safety of geologic disposal of HLW.

First, we can theoretically conceive of a large number of geometric and material configurations of TFM with various types of rocks and water, some of which are critical and have positive reactivity feedback. It was pointed out that effects of rock porosity are important; the greater the porosity is, the smaller the minimum critical mass is. The effects of neutron absorption in constituent elements in rock are also important. The minimum critical mass tends to be significantly greater if these are taken into account. While heterogeneity in deposition of uranium in rock formations is another essential factor in realizing positive reactivity feedback, Pu deposition can have positive feedback even if it is homogeneous, because of the 0.3-eV resonance peak in ^{239}Pu [8].

Second, by the radionuclide transport analyses, it was pointed out that it is highly unlikely at YMR that plutonium will travel large distances as a solute or as a colloid to form such critical configurations. Pu-239 will decay to ^{235}U before most of a waste form is dissolved, or while it stays within the vicinity of the failed waste package due to strong sorption with rocks and due to re-deposition of colloids. For a hypothetical water-saturated repository with vitrified HLW, minor actinides would decay to ^{235}U before most of a waste form is dissolved, creating approximately 12%-enriched uranium. The enrichment is dependent on the recovery efficiencies of uranium and plutonium. The better the recovery efficiencies of U and Pu are, the greater the fraction of ^{235}U in the accumulation in geologic formations would be.

Thus, the question was boiled down to "will ^{235}U be deposited heterogeneously in rock formations with a sufficiently large mass for criticality?" At YMR, the water chemistry is conducive to high uranium solubility; hence, ^{235}U can move large distances as a solute. Once uranium is in solution, it is unlikely to precipitate into these critical configurations because of a lack of reducing agents in the surrounding geologic formations. In a water-saturated repository, detailed information to make similar conclusion is yet to be obtained. However, by the bounding analyses and the statistical uncertainty analyses summarized above, it was also pointed out that accumulation of sufficient mass of $U(12)O_2$ originating from tens of thousands of failed waste packages funneling into a single location is highly unlikely. From the analyses available, it is safely concluded that possibility of autocatalytic criticality events is negligibly small.

5.2 Engineering Strategies, Regulations and Licensing

Though the probability of forming critical deposits of TFM appears exceedingly low, engineered systems can further reduce the probability and may also provide a simpler basis for meeting licensing criteria. An engineered strategy is threefold: (1) to provide neutron absorbers, (2) to reduce mass of TFM in a waste package, and (3) to prevent accumulation in the far field.

First, the neutron absorbers must have similar chemical behaviors when they are transported in geologic formations and when they are released by dissolution of waste matrix from failed packages. Boron is not suitable because it easily dissolves in groundwater. Utilizing gadolinium or depleted uranium generated at the uranium enrichment plant was pointed out as effective measures in Ref. 8. When reprocessed HLW is disposed of, the amount of ^{238}U in the vitrified HLW can be increased by deliberately keeping the separation efficiency of uranium low at the reprocessing.

Second, the amount in the HLW of transuranic elements (^{243}Am, ^{239}Pu) that decay to ^{235}U can be decreased by recycling these as fuel and by burning them in a reactor. This will also beneficial to decrease concern for proliferation of nuclear materials [25] and environmental impacts [14] from geologic repositories and interim storages for HLW.

Third, accumulation of TFM in the far field can be avoided by careful site selection and improved design for a repository. From the previous studies, the rock porosity, groundwater flow rates, and existence of reducing agents for TFM precipitation were pointed out as major factors for consideration. At YMR, the lack of reducing agents for uranium precipitation was the key to finally conclude that the criticality event is unlikely. At a water-saturated repository, the bentonite buffer around a waste package will play an important role to filter plutonium colloids and prevent plutonium accumulation by multiple canisters. Small porosity and slow water flow by having the buffer also assures negligibly low probability for criticality accident due to plutonium isotopes. At YMR, the capillary barrier concept would be ideal for this purpose [8,26].

From the viewpoint of regulations and licensing applications, the previous studies imply the followings:

First, a site for a geologic repository should be selected in such a way that the concern for criticality accident is minimized. A candidate site should be considered in a host rock with sufficiently small porosities or with no reducing agents for uranium precipitation. It is important to confirm that high porosity regions around the repository do not contain high concentrations of reducing agents. These considerations should be included in site-selection criteria for future repository siting activities.

Second, the previous studies for a water-saturated repository were performed only for a generic repository concept. As was done for YMR, similar analyses in detail should be made once a repository site has been decided, based on detailed site-specific conditions for that repository site.

Third, impacts and benefits of advanced fuel cycles should be carefully considered. Actinides will be effectively eliminated by such advanced fuel cycles with highly efficient separation processes. In reality, however, some spent fuel from an advanced fuel cycle might be disposed of directly in a geologic repository for some reasons. Transition from the present fleet of light-water reactors to advanced fuel cycles means more variety of materials in a repository. Criticality safety assessment should be made to check influences of newly added materials in a repository.

5.3 Comparison with Conventional Performance Assessment Models

From previous experiences of performance assessment of geologic repositories for criticality safety, the following observations can be made in comparison with conventional performance assessment focusing on the exposure dose rate to the public.

First, conservatism in the models for criticality safety assessment is different from that in those for radiological safety. An example was shown above in the case of YMR, where greater sorption was considered conservative for criticality safety because it overestimates TFM deposition in the rock. For radiological safety, usually low sorption is considered conservative because it gives higher concentrations in the downstream, resulting in higher doses.

Second, configuration of canister array in a repository and the flow field through the repository would be important for criticality safety assessment. This is more so because for criticality safety assessment geometries of transport path and of TFM depositions are key information. For radiological safety, while the groundwater flow field and geological heterogeneity are also essentially important because they determine dispersion of radionuclides in the geosphere, detailed information in the repository such as canister array configuration is not always an essential factor that determines radionuclide concentrations at the downstream points because of greater dispersal in the far field by hydrodynamic dispersion.

Third, these differences arise because masses of radionuclides released from the repository and accumulating in the far field are the basis for criticality safety assessment, while concentrations of radionuclides in groundwater in the far field are the basis for radiological safety assessment [14]. The criticality safety can be improved effectively by decreasing TFM mass initially included in waste canisters. For radiological safety, reduction of toxic radionuclides in a waste canister is not always effective, especially for solubility-limited radionuclides. This leads to a thought that, for comprehensive understanding of repository performance and safety, not only radiological safety measured by the exposure dose rate to the public but also other aspects of repository performance would be important. Criticality safety, proliferation resistance, and environmental impact of a geologic repository are such aspects, which are measured principally by the mass of radionuclides to be disposed of in the repository.

6. CONCLUSIONS

A review of the previous studies for autocatalytic criticality phenomena due to a geologic repository for high-level radioactive wastes has been made, based on those performed

for Yucca Mountain Repository and for a hypothetical water-saturated repository for vitrified HLW. The previous studies focused on two points. One was the neutronic study to investigate geometric and materials configuration consisting of rock, water and thermally fissile materials that exhibit positive reactivity feedback with increasing system temperatures. The other was radionuclide migration and accumulation study, which was necessary because some subsets of scenarios could not be screened out by the neutronic study. It was concluded that in either case it is highly unlikely that an autocatalytic criticality event would happen at a geologic repository.

The autocatalytic criticality from over-moderated criticality configurations in the far-field contributed by multiple canisters in a repository is the remaining concern, which can be avoided by careful siting and engineered-barrier design. Thus, this aspect of a geologic repository should be properly addressed in regulations and siting criteria.

In criticality safety assessment, models developed for conventional performance assessment for radiological safety to the public can be utilized. Because those models for radiological safety were developed in such a way that the exposure dose to the public is conservatively overestimated, they may not be used directly in the criticality safety assessment, where accumulated TFM mass needs to be conservatively overestimated. The models for criticality safety also require more careful treatment of geometry and heterogeneity in transport paths because a minimum critical mass is sensitive to geometry of TFM accumulation.

REFERENCES

[1] C. D. Bowman and F. Venneri, Underground Autocatalytic Criticality from Plutonium and Other Fissile Material, Los Alamos National Laboratory, LA-UR-94-4022A, March (1995).

[2] P. K. Kuroda, On the nuclear physical stability of the uranium minerals, *J. Chem. Phys.*, **25**, 781 (1956), *ibid*, On the infinite multiplication constant and the age of the uranium minerals, **25**, 1295 (1956).

[3] G. A. Cowan, A Natural Fission Reactor, *Scientific American*, **235**, 36 (1976).

[4] E. D. Clayton, Anomalies of Nuclear Criticality, Pacific Northwest Laboratory, Richland, WA, PNL-SA-4868 Rev. 5, June (1979).

[5] B. F. Gore, U. P. Jenquin and R. J. Serne, Factors Affecting Criticality for Spent-Fuel Materials in a Geologic Setting, Pacific Northwest Laboratory, PNL-3791, April (1981).

[6] "Final Storage of Spent Nuclear Fuel – KBS-3," Volume IV, Chapter 21, Swedish Nuclear Fuel Supply Co., Division KBS (SKBF/KBS) (May, 1983).

[7] W. E. Kastenberg, P. F. Peterson, J. Ahn, J. Burch, G.Casher, P. L. Chambré, E. Greenspan, D. R. Olander, J. Vujic, B. Bessinger, N. G. W. Cook, F. M. Doyle, and B. Hilbert : Mechanisms for Autocatalytic Criticality of Fissile Materials in Geologic Repositories, UCB-NE-4214 (1996).

[8] W. E. Kastenberg, P. F. Peterson, J. Ahn, J. Burch, G.Casher, P. L. Chambré, E. Greenspan, D. R. Olander, J. Vujic, B. Bessinger, N. G. W. Cook, F. M. Doyle, and B. Hilbert : Consideration of Autocatalytic Criticality of Fissile Materials in Geologic Repositories, *Nuclear Technology*, **115**, 298–310 (1996).

[9] J. Ahn, Transport of Weapons-Grade Plutonium and Boron Through Fractured Geologic Media, *Nuclear Technology*, **117**, 316 (1997).

[10] J. Ahn, E. Greenspan and P. L. Chambré, A Preliminary Consideration for Underground Autocatalytic Criticality by Vitrified High-Level Waste in Water-Saturated Geologic Repository, *Journal of Nuclear Science and Technology*, **37**(5), 465 (2000).

[11] J. Ahn, E. Greenspan and P. L. Chambré, Mechanisms and Possibility of Autocatalytic Criticality by High-Level Wastes Buried in Water-Saturated Geologic Formation, *Journal of Nuclear Fuel Cycle and Environment*, **4**(2), 3 (1998).

[12] J. Ahn, Criticality Safety Assessment for a Conceptual High-Level-Waste Repository in Water-Saturated Geologic Media, *Nuclear Technology*, **126**, 303 (1999).

[13] R. Sanchez et al., "Criticality Characteristics of Mixtures of Plutonium, Silicon Dioxide, Nevada Tuff, and Water," LA-UR-95-2130, Los Alamos National Lab., 1995.

[14] J. Ahn, Environmental Impact of Yucca Mountain Repository in the Case of Canister Failure, *Nuclear Technology*, in press.

[15] G. H. Canavan, et al., "Comments on 'Nuclear Excursions and Criticality Issues,' " LA-UR-95-0851, Los Alamos National Laboratory (Mar, 1995).

[16] P. B. Parks, M. L. Hyder and T. G. Williamson, "Final Issue of 'Consequences of the Bowman-Venneri Nuclear Excursion Thesis on the Prospects for Placing Vitrified Plutonium Canisters in Geologic Repositories'(U)," PD1-SPP-95-0023, Westinghouse Savannah River Company (June, 1995).

[17] R. A. van Konynenburg, "Comments on the Draft Paper 'Underground Supercriticality for Plutonium and Other Fissile Material,' " UCRL-1D-120990 COM, Lawrence Livermore National Laboratory (May, 1995).

[18] M. J. Bell, "NRC Comments on the Report 'Underground Supercriticality from Plutonium and Other Fissile Material,' " U. S. Nuclear Regulatory Commission (Aug., 1995).

[19] P. L. Chambré, T. H. Pigford, A. Fujita, T. Kanki, H. Lung, A. Kobayashi, D. Ting, Y. Sato and S. J. Zavoshy, "Analytical Performance Models for Geologic Repositories," LBL Report, LBL-14842 (1982).

[20] E. A. Sudicky and E. O. Frind, "Contaminant Transport in Fractured Porous Media: Analytical Solution of a Two-Member Decay Chain in a Single Fracture," *Water Resources Research*, **20**, 2021 (1984).

[21] J. Ahn, "Mass Transfer and Transport of Radionuclides in Fractured Porous Rock," Ph.D. Dissertation, University of California, Berkeley (1988).

[22] D. P. Hodgekinson and P. R. Maul, "1-D Modelling of Radionuclide Migration Through Permeable and Fractured Rock for Arbitrary Length Decay Chains Using Numerical Inversion of Laplace Transform," *Ann. Nucl. Energy*, **15**(4), 175 (1988).

[23] A. Talbot, "The Accurate Numerical Inversion of Laplace Transforms," *J. Inst. Math. Appl.*, **23**, 97 (1979).

[24] J. Ahn, "Integrated Radionuclide Transport Model for an HLW Repository in Water-Saturated Geologic Formations," *Nuclear Technology*, **121**(1), 24–39 (1998).

[25] J. Ahn, Possibility of Safety Improvement for Vitrified HLW Geologic Disposal by Partitioning and Transmutation, Global

'99, International Conference on Future Nuclear Systems, August 29-September 3, 1999, Jackson Hole, Wyoming (1999).

[26] L. J. Carter and T. H. Pigford, "NUCLEAR WASTE: Proof of Safety at Yucca Mountain," *Science*, **310(5747)**, 447 (October 21, 2005).