EFFECTIVE APPLICATION OF PARTITIONING AND TRANSMUTATION TECHNOLOGIES TO GEOLOGIC DISPOSAL

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Abstract

The present study proposes the total toxicity index of radionuclides that have accumulated in the region exterior to the repository as the environmental impact measure. The measure is quantitatively evaluated by a radionuclide transport model that incorporates the effects of canister-array configuration and the initial mass loading in the waste canister. With the measure, it is demonstrated that the environmental impact of the repository can be effectively reduced by reduction of the initial mass loading and change in the canister-array configuration in the repository. The rate of increase in the environmental impact with the increase in the repository size can be reduced by reducing the initial mass loading of Np and its precursors. Environmental impacts of the mill tailings and the depleted uranium are as important as those from the HLW repository. For a fair comparison of various fuel cycles, the sum of these impacts should be compared.

Introduction

Previous analyses for the impacts of Partitioning-and-Transmutation (P/T) systems on the performance of geologic disposal show that the effects of these P/T systems are likely not justifiable because they provide only modest improvements in radiological safety. This insensitivity of predicted repository performance to fuel cycle schemes occurs because repository performance is currently measured only by radiological exposure dose rates [1].

In the recent study on the repository impact performed at University of California, Berkeley (UCB), the environmental impact measure was defined [2]. With the definition and quantitative analyses, it was shown that the environmental impact of a geologic repository could be significantly different, depending on the repository design and the solidified waste properties, including initial mass loadings of radionuclides in the solidified waste for geologic disposal. The initial mass loading of a radionuclide in the solidified waste is primarily determined by the scheme of the fuel cycle and the conditions for the solidification process. These findings imply possibilities that the repository impacts could be significantly reduced by modifying the waste characteristics, which can be done by modifying the fuel-cycle scheme.

In the present paper, results of an analytical study are presented for the effects of initial mass loading of important contributors on environmental impacts of a geologic repository. The study consists of two major parts.

The first is development of analytical formulae for the environmental impact of a geologic repository as a function of the canister layout in the repository (such as the canister array configuration and the dimensions of engineered barriers), the mass-transport parameters in the repository (such as the groundwater pore velocity, the retardation factors, the diffusion coefficients, and the solubilities), the canister-performance parameters (the canister failure time and the waste-matrix dissolution time), and the initial mass loading of radionuclides in a waste canister. Two formulae have been developed: one for solubility-limited release and one for congruent release of radionuclides from a dissolving waste matrix.

The second is development of the waste-solid conditioning model. The waste conditioning model determines the compositions and initial mass loading in each waste canister by taking into account the constraints imposed on the waste solidification, such as the Na content, the Mo content, and the heat load. This model is considered to be the link between the fuel-cycle mass-flow model and the repository-impact assessment model.

With these, we have obtained the quantitative relationship between the repository capacity and the environmental impact of a geologic repository with high-level wastes (HLW) from light-water reactors (LWR) and fast-breeder reactors (FBR). We have furthermore investigated the effects of reducing the mass of certain radionuclides from HLW on the environmental impact, which leads to a discussion on how the impact from the system can be effectively reduced by applying P/T technologies.

Environmental impact

The environmental impact of a radionuclide included in the waste disposed of in a geologic repository is defined as the peak value of the toxicity index for the radionuclide mass existing in the far field. The toxicity index is defined as the radioactivity of the radionuclide divided by the maximum permissible concentration (MPC) of the radionuclide for drinking.[3] The index represents the volume of water with which the radionuclide solution can be diluted to its MPC. The toxicity index considered here as the impact measure takes into account the mechanisms by which the radionuclides could be released from the repository. In this respect, the environmental impact defined here is remarkably different from the toxicity index of radionuclides in the waste.

A radionuclide-transport model was recently developed [1] [4], where multiple waste-canisters and their spatial configuration were incorporated by considering an array of compartments each containing a waste matrix, a buffer, and a near-field rock (NFR) region (Fig. 1). It is assumed that each canister has identical mass loadings of radionuclides. The repository consists of as many compartments as the number of waste canisters placed in the repository. For more detail about mathematical formulations and assumptions, the reader is referred to [1] [4]. Brief summaries of the model and the findings are given here for readers' convenience.

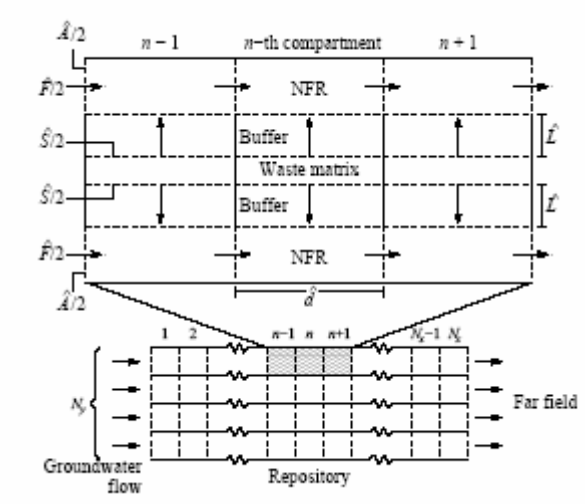


Figure 1: A two-dimensional array configuration of a geologic repository. There are N_y rows, each containing N_x waste canisters in the water-flow direction.

The waste matrix, together with radio nuclides contained in the matrix, starts to dissolve by pore-water in the buffer. The concentration of an actinide species in the pore-water at the waste-matrix dissolution location is limited by its solubility. Soluble species including most of fission products are released congruently with the dissolution of the waste matrix. Radionuclides are assumed to be transported through the pore-water in the buffer region by molecular diffusion, and then released into the NFR region. The buffer and the NFR are assumed to be homogeneously porous.

An array of multiple compartments in the repository is considered to be immersed in the parallel water stream. It is assumed that there is no net mass transport through the interface between two parallel adjacent arrays. Groundwater is assumed to flow through the NFR at a constant, uniform velocity. A radionuclide released from the buffer is transported by advection through the NFR. Hydrodynamic dispersion is treated by an array of multiple compartments in the present model [1]. Linear sorption isotherms between the solid phase and the pore-water phase in the buffer and in the NFR are also assumed.

Uncontaminated water enters the NFR of the first compartment in the array. The water in the pores of the NFR is contaminated by the radionuclide released from the waste matrix and its transport through the buffer by diffusion. Contaminated water in the NFR continues to flow into the NFR of the second compartment in the array, and contacts with the buffer there. This water gets additionally contaminated by radionuclides released from the second canister. In this fashion, the groundwater flowing in the array through the NFR is assumed to be increasingly contaminated before it finally flows out of the repository from the NFR of the end compartment.

By the aforementioned definition, the environmental impact I_i [m^3 of water] due to radionuclide *i* contained in the waste canisters is written as

$$I_i = \left(N_x N_y \hat{M}_i^o \right) P_i C_i , \tag{1}$$

where $N_x N_y$ is the total number of waste canisters placed in the repository. $\hat{M}_i^o$ [kg-nuclide] is the initial mass loading of radionuclide *i* in one canister. Coefficient C_i expresses the toxicity of unit mass of radionuclide *i*, defined as

$$C_i \left[\mathrm{m}^3\text{-water/kg-nuclide} \right] \equiv \frac{\lambda_i \left[/\mathrm{s} \right] \cdot 1000 \left[\mathrm{g/kg} \right] \cdot N_A \left[/\mathrm{mol} \right]}{M_i \left[\mathrm{g/mol} \right] \cdot 3.7 \times 10^{10} \left[\mathrm{Bq/Ci} \right] \cdot \left(\mathrm{MPC} \right)_i \left[\mathrm{Ci/m}^3 \right]} , \tag{2}$$

where λ_i, M_i, and $\left(\mathrm{MPC} \right)_i$ are the decay constant, the molecular mass, and the MPC of nuclide *i*, respectively. N_A is the Avogadro's number.

The factor P_i is the ratio between the total initial mass loading $\left(N_x N_y \hat{M}_i^o \right)$ of radionuclide *i* in the repository to the peak mass of the radionuclide existing in the far field, i.e., the environment. Formulations for the factor P_i have been developed for congruent-release radionuclides and for solubility-limited-release radionuclides, separately. The factor P_i expresses the reduction of the mass of radionuclide *i* due to confinement by the repository. As the repository confinement capability improves, this factor decreases.

Radionuclide transport within the repository region with multiple canisters was already modelled for solubility-limited release in [1] and [2] and for congruent release in [4]. The radionuclide release rates at the downstream edge of the repository, i.e., from the N_x-th compartment in each row to the environment, were formulated for both types of radionuclides. Utilizing the formulae for the release rates derived in [2] and [4], mathematical expressions for the radionuclide mass in the environment have been obtained in this study by integrating the release rates with respect to time while taking into account the radioactive decay loss in the environment. Then, the factor P_i have been formulated by normalising the peak value of the radionuclide mass in the environment released from the repository containing the total of N_xN_y canisters by the total initial mass loading $\left(N_x N_y \hat{M}_i^o \right)$ in N_xN_y canisters.

Waste conditioning model

We have developed a mathematical model which determines the composition of a solidified HLW in a canister and the number of HLW canisters per unit mass of irradiated fuel, in such a way that the mass M_W of waste oxides included in one canister is maximized while satisfying several constraints.

The mass of the solidified HLW in a single canister is the sum of M_W and the mass M_G of matrix material, such as borosilicate glass frit. If the composition vectors $\vec{N}_W$ for the waste oxides and $\vec{N}_G$ for the matrix material are known, the composition vector $\vec{N}_S$ of the solidified HLW is obtained as

$$\vec{N}_S = \frac{M_W}{M_W + M_G}\vec{N}_W + \frac{M_G}{M_W + M_G}\vec{N}_G . \tag{3}$$

The number of HLW canisters for a unit mass of irradiated fuel can be calculated by dividing the mass of the HLW from processing a unit mass of irradiated fuel by M_W.

M_W and M_G can be determined by solving a linear programming problem. The objective function, f, which is the mass M_W, is formulated as

$$\text{Maximize } f = M_W. \tag{4}$$

This objective function is subject to constraints that are imposed on the HLW solidification process. We have developed a set of constraints for borosilicate glass solidification.

The first constraint is on the total mass of solidified HLW. The standard HLW canister contains 400 kg of solidified HLW [5]. Therefore,

$$M_W + M_G \le 400 \ \ [\text{kg}]. \tag{5}$$

The second constraint is on the volume of the solidified HLW. The volume of solidified HLW in a canister is set to be 150 liter [5], yielding

$$\frac{M_W + M_G}{\rho_{WG}} \le 150 \ \ [\text{liter}], \tag{6}$$

where ρ_{WG} is the density of the solidified HLW. From Refs. [6] and [7], the density is formulated as

$$\rho_{WG} = 1230\left(\frac{M_W}{M_W + M_G}\right) + 2419 \quad [\text{kg}/\text{m}^3]. \tag{7}$$

Substituting (7) into (6) yields a quadratic form, which has been approximated as the following linear form:

$$M_W + 1.508 M_G \le 547.4 \ \ [\text{kg}]. \tag{8}$$

The third constraint is on the heat generation. The heat generation from a canister must be smaller than 2300 W [5]. Therefore,

$$\zeta M_W \leq 2300 \quad [\mathrm{W}], \tag{9}$$

where ζ [W/kg] is heat generation per unit mass of waste oxides. This can be calculated by the composition vector $\bar{N}_W$ for the waste oxides, the mean energy release from the decay and the decay constant of each nuclide.

The fourth constraint is on the mass fraction of Mo oxide in the solidified waste. It must be smaller than 2 wt% [8]. Therefore,

$$x_{W,MoO_3} M_W \leq 0.02\left(M_W + M_G\right), \tag{10}$$

where x_{W,MoO_3} is the mass fraction of MoO_3 in waste oxides.

The fifth constraint is on the mass fraction of Na_2O, which must be smaller than 10 wt% [8]. Similar to (10), it is formulated as

$$x_{W,Na_2O} M_W + x_{G,Na_2O} M_G \leq 0.1\left(M_W + M_G\right). \tag{11}$$

Note that Na_2O would be included in the waste oxides as well as in the matrix material.

The sixth constraint is on the waste loading limit in the borosilicate glass to avoid phase separation. The mass fraction of the waste oxides in the solidified waste must be smaller than 25% [9]. Therefore,

$$M_W \leq 0.25\left(M_W + M_G\right) \tag{12}$$

The maximum value of M_W that satisfies the constraints (5), (8), (9), (10), (11), and (12) is determined by a linear programming approach. A graphical solution is shown in Figure 2.

Numerical results and discussions

Results for waste conditioning

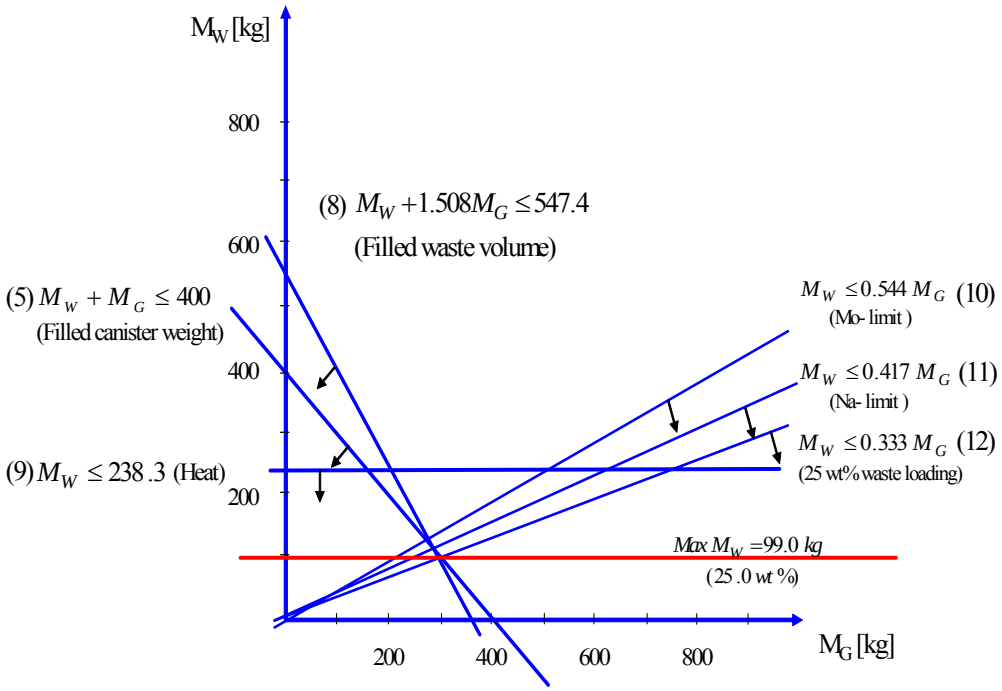
For benchmarking, we have applied the waste conditioning model for the waste oxide that would be generated from reprocessing of PWR fuel with the initial enrichment of 4.0%, the burn-up of 28 GWd-thermal/MTU, the cooling time of 150 days after discharge from the reactor, and the cooling time of 5 years between reprocessing and solidification. The loss fractions of uranium and plutonium into waste oxides are 0.604% and 0.2966%, respectively. Other actinide elements are assumed to be completely transferred to the waste oxide. All fission-product (FP) radionuclides except for noble gas elements are transferred to the waste oxide. No noble gas element is assumed to be included in the waste oxide.

The isotopic composition vector $\vec{N}_W$ of the waste oxide has been numerically calculated by ORIGEN code. With these, the heat generation ζ [W/kg] per unit mass of waste oxides has been calculated as 9.65. Assuming that the radionuclides are all in oxide forms, the mass of waste oxide from 1 MT of the irradiated fuel is calculated to be 78.1 kg. For the waste matrix material, the glass frit PF798 developed by JNC is assumed [6]. The glass frit contains SiO_2 (62.3%), B_2O_3 (19.0%), Al_2O_3 (6.7%), Li_2O (4.0%), CaO (4.0%), and ZnO (4.0%). This gives the vector $\vec{N}_G$.

Figure 2 shows the graphical solution for the linear programming approach. The straight lines represent the constraints (5), (8), (9), (10), (11), and (12). It is observed that the waste-loading limit (12) and the volume constraint (8) determine the maximum for the waste loading, M_W. The heat constraint is not crucial for this case. The optimum is found to be M_w = 99.0 kg and M_G = 301 kg. With these values, the HLW composition vector (3) is written as

$$\vec{N}_S = 0.25\vec{N}_W + 0.75\vec{N}_G . \qquad (13)$$

Figure 2. **Graphical solution for solidification of HLW from LWR spent fuel reprocessing.**



With the optimum waste loading M_w = 99.0 kg and the mass 78.1 kg of the waste oxide from 1 MT of irradiated fuel, the number of waste canisters per MT is calculated to be 78.1/99.0 = 0.79 canister/MT. With the assumed burn-up of 28 GWd-thermal/MTU and the heat-to-electricity conversion efficiency of 33%, the electricity per canister is calculated to be 28/0.79 x 0.33 = 11.7 GWd-e/canister. If a repository that can contain 40,000 canisters is filled with canisters of the loading obtained here, assuming that the reactors were operated with the capacity factor of 90%, the repository capacity is equivalent to 1420 GWyear[1].

Similar calculations have been made for FBR. The number of canisters per MT of reprocessed spent fuel has been obtained as 1.25. Electricity per canister is obtained as 21.3 GWd-e/canister. If the 40,000-canister repository is filled with this type of canisters, assuming the capacity factor of FBR is 90%, the repository capacity of 40,000 canisters is equivalent to 2590 GWyear, which is nearly twice as large as that for the LWR case.

Table I shows the values of the initial mass loadings $\hat{M}_i^o$ per canister for important radionuclides in the solidified HLW from LWR and FBR spent fuels. Initial mass loadings of other radionuclides have also been obtained, but not shown in this table, because their contribution to environmental impact is not significant. Initial mass loadings of precursors to Np-237 and Pu-239 are lumped with initial mass loadings of their short-lived precursors. Pu, Np, and Tc are considered to be released with the solubility-limited mode. Iodine and Cs are released congruently with the waste matrix. For congruent release, the waste-matrix dissolution time is assumed to be 50,000 yeares. Other parameter values related to repository design have been taken from [5].

Table 1. **Initial mass loadings of important radionuclides in a canister of solidified HLW from spent fuel reprocessing, and their parameter values for environmental impact evaluations**

Nuclide		LWR (mol/canister)		FBR (mol/canister)		MPC (Ci/m^3) [10]	λ_i (1/y)	Solubility (mol/m^3) [5]	Retardation factor in buffer [5]	Retardation factor in NFR [5]	Conversion factor C_i (m^3/kg)
			Lumped $\hat{M}_i^o$		Lumped $\hat{M}_i^o$						
Am	243	4.35E-1		3.04E-3							
Pu	239	1.55E-2	4.51E-1	1.93E-1	1.96E-1	2.00E-8	2.84E-5	1.0E-5	49000	2600	3.07E+9
Pu	241	3.67E-3		1.20E-2							
Am	241	9.27E-1		7.23E-3							
Np	237	2.04E+0	2.97E+0	1.12E-3	2.04E-2	2.00E-8	3.23E-7	2.0E-5	4900	2600	3.52E+7
I	129	1.28E+0		2.58E+0		2.00E-7	4.30E-8	---	8.7	1.2	8.60E+5
Cs	135	2.64E+0		1.39E+1		1.00E-5	3.01E-7	---	50	131	1.15E+5
Tc	99	7.06E+0		1.05E+1		6.00E-5	3.28E-6	3.9E-5	490	2600	2.85E+5

[1] In the H12 study for repository performance assessment by JNC [5], 1.25 canisters/MTU is assumed for the burn-up of 45000 MWd/MTU. Assuming the same heat-to-electricity conversion efficiency and the capacity factor, the repository with 40000 canisters is equivalent to 1440 GW-year. This comparison implies that making the burn-up of the fuel greater does not save the repository capacity.

Results for environmental impact evaluation

Based on the initial mass loadings per canister for LWR and FBR, the environmental impact from a repository containing 40,000 canisters of identical loadings has been numerically evaluated. See Figure 3. Repository configuration and layout developed for the H12 study by JNC [5] have been applied. In the formula (1), $N_x N_y = 40{,}000$ is fixed. The values of $\hat{M}_i^o$ are also fixed as shown in Table I.

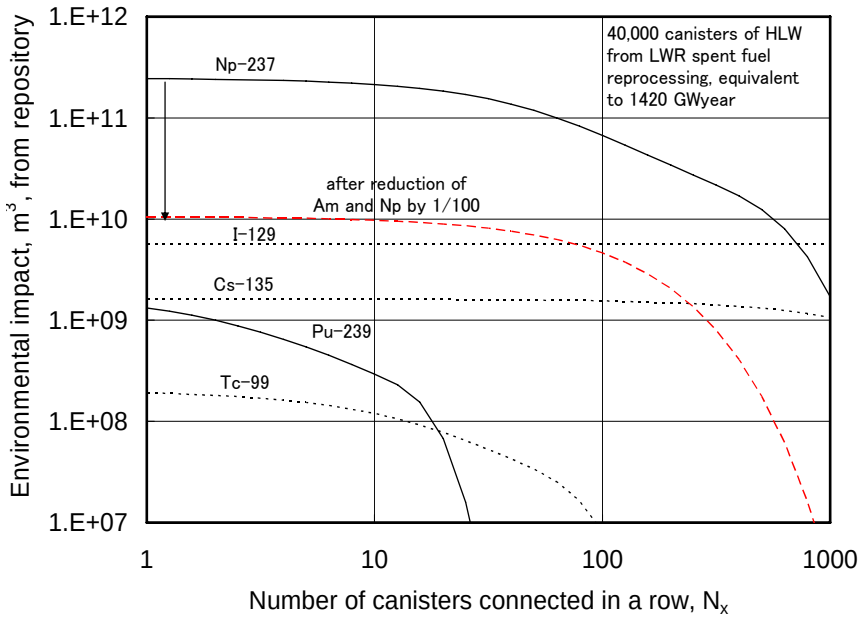
Figure 3 shows that the environmental impact from the repository with the same size for HLW from LWR is 20 times greater than that from FBR, because of the greater mass loading of Np-237 in the LWR canister. This is because, in the assumed FBR system, 99.9 % of Np, Am, and Cm in the spent fuel are recovered and recycled. Note that the total electricity generation supported by the same-size repository is 2590/1420 = 1.8 times greater for FBR than for LWR. If we take the case for $N_x = 1$ for comparison, the environmental impact from the LWR repository is 1.7E8 m^3/GWyear, whereas that from the FBR is 4.4E6 m^3/GWyear, which is about a factor of 40 smaller.

For LWR, the impact from I-129 is 20 times smaller than that from Np-237. For FBR, that is twice as large as from Np-237. Because of this difference, the reduction of the initial mass loading of Np-237 from the HLW canister is more effective in reducing the repository environmental impact for LWR. The same reduction for FBR is not justifiable, however, because it does not change the total impact from the repository, which is primarily determined by I-129 and Cs-135.

For LWR, to reduce the impact due to Np-237, the mass loadings of Pu-241 and Am-241 as well as Np-237 need to be reduced. In the left figure, reduction with a factor of 100 is considered for Np and Am. The initial mass loading of Pu-241 is negligibly small for this case. For the FBR case (the right figure), the mass of Pu-241 is greater than those of Am-241 and Np-237. Therefore, to reduce the mass of Np-237 in a canister for FBR, the mass of Pu in HLW needs to be reduced. Considering that Pu has already been recovered intensively at the stage of reprocessing prior to the HLW conditioning, reduction by a factor of 10 is assumed for Pu.

The two figures below show the cases for LWR and FBR, respectively. The solid curves represent actinide radionuclides, whereas the dashed curves FP radionuclides. The broken curves show the effects of reduction in the initial loadings of selected radionuclides.

Figure 3. **Environmental impact from a repository containing 40000 canisters as a function of the number of canisters connected in the same water flow stream.**



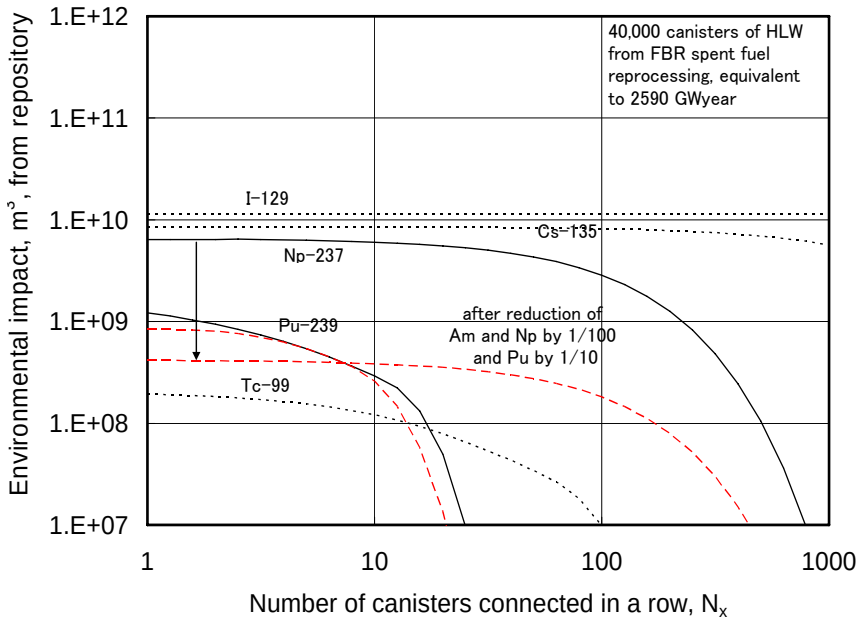
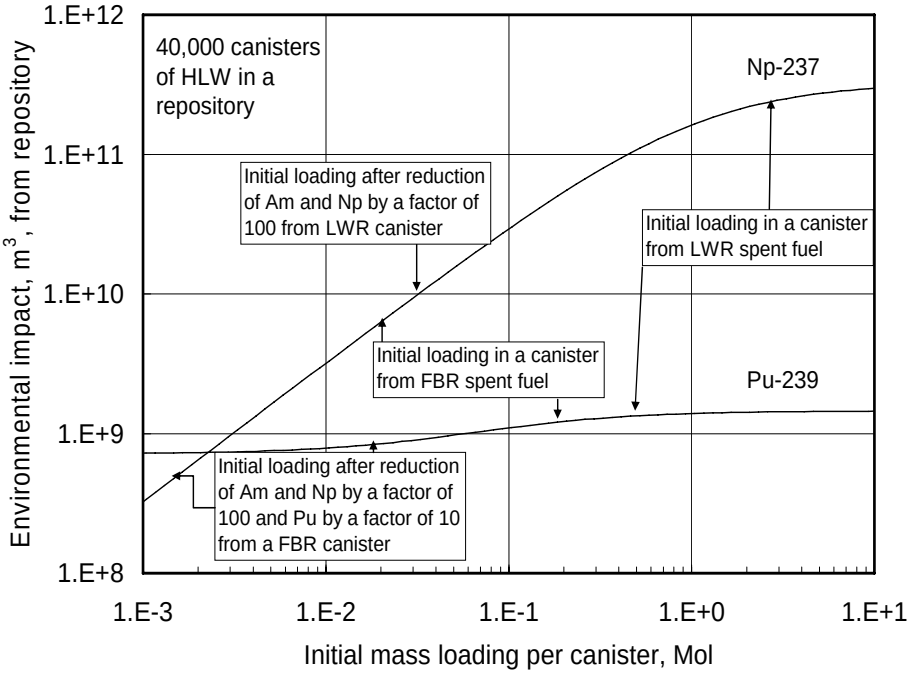


Figure 4 shows the effects of reduction in the initial mass loadings of Pu and Np on the environmental impact from the repository. As the initial mass loading increases, the impact levels off. Because their solubilities are small, a significant amount of these radionuclides decays before they are released into the environment. The greater the mass loading is, the greater mass of the radionuclides decays before they are dissolved into groundwater. As the mass loading of Np decreases, the environmental impact from this radionuclide decreases nearly linearly. This is because decay loss before dissolution becomes negligible, and most of the mass initially existing in the canisters is released to, and accumulates in, the environment.

Figure 4. **Effects of reduction in initial mass loadings per canister on the environmental impact from a 40000-canister repository due to Np-237 and Pu-239. N_x = 1 is assumed**



On the other hand, the environmental impact of Pu is not sensitive to its initial mass loading. This is because Pu-239 is not so long-lived that most of it initially in the canisters decays before they are released from the repository. Confinement by the repository is so effective that the initial mass loading does not affect the environmental impact so much. This would be different if greater mobility is assumed for Pu, such as due to colloid-facilitated transport.

If we do not deploy any new system and keep utilizing the LWR system, the rate of increase of the environmental impact from the repository is 1.7E8 m^3/GWyear.[2] Based on the aforementioned results, there would be two scenarios to reduce the rate of increase in the environmental impact of the repository as the repository size increases. Here we assume N_x = 1.

[2] This can be obtained by dividing the impact, 2.45E11 m^3, of Np-237 by the total electricity generation, 1420 GW/year, for the 40000-canister repository with identical canisters from LWR spent fuel.

We consider two scenarios for impact reduction. In one scenario, we apply a P/T system that can reduce the masses of Am and Np in HLW by a factor of 200. Then, the rate of increase is reduced from 1.7E8 to 4.0E6 m^3/GWyear. In the second scenario, we deploy a full-FBR system. The impact is primarily due to I-129. The rate of increase is similarly calculated as 4.4E6 m^3/GWyear from Figure 3.

While the resulting environmental impact from the same-size repository would be nearly the same, there is a significant difference in the environmental impact between these two systems. With the aforementioned LWR + P/T system, a feed of natural uranium is necessary, resulting in generation of mill tailings and depleted uranium. The environmental impact of these is reported to be 1E10 m^3/GWyear [2]. Therefore, even if the impact from the repository is reduced to 4.0E6 m^3/GWyear, the rate of increase of the total impact from the system would be of the order of 1E10 m^3/GWyear.

With the FBR system, on the other hand, roughly 1 ton of depleted uranium is *consumed* per GWyear. One ton of depleted uranium is equivalent to 5.3E7 m^3 of impact. By deploying the FBR system, depleted uranium is converted to the geologically disposed HLW (4.4E6 m^3/GWyear). Thus, as far as depleted uranium supply lasts, the net rate of *decrease* of the total impact from the FBR system would be 4.9E7 m^3/GWyear.

Conclusions

Environmental impacts from the HLW geologic repository containing 40,000 canisters have been quantitatively evaluated for LWR and FBR. The waste-conditioning model that determines the composition of the solidified HLW and the number of canisters per unit mass of spent fuel has been developed. Mathematical formulae for the peak mass of radionuclides that exist in the environment after released from the repository have been derived. With these mathematical developments, the environmental impact of the repository has been formulated in terms of the repository-design parameters and the isotope vector of the high-level liquid waste from the fuel cycle.

With the developed formulae, the environmental impact from the repository with HLW from LWR operation and from FBR operation has been numerically obtained. From the numerical results, the following observations have been made.

- If a P/T system is applied to the LWR system to reduce the environmental impact from the repository, the target nuclide would be Np-237 and Am-241. The reduction of these nuclides would be meaningful until the environmental impact of Np-237 is reduced to the level of environmental impacts of dominating FP nuclides, such as I-129 and Cs-135.

- The repository filled with 40,000 HLW canisters from FBR operation would result in the environmental impact smaller than that from the LWR repository by a factor of 20. If compared on a per GWyear basis, the advantage of FBR is even greater (a factor of 40). Because the dominating radionuclides are FP nuclides, P/T application for a FBR system to reduce actinides is not attractive.

- The possibility of decreasing the environmental impact from the entire cycle, including legacy depleted uranium, by deployment of the FBR system has been indicated. In contrast, with the LWR + P/T system, depleted uranium will continue to be generated and dominate the environmental impact.

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