

Selection of Key Radionuclides for P&T Based on Radiological Impact Assessment for the Deep Geological Disposal of Spent PWR/CANDU/DUPIC Fuels

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Abstract

When it is assumed that PWR, CANDU and DUPIC spent fuels are disposed of in deep geological repository, consequent annual individual doses are calculated, and it is shown that doses meet the regulatory limit. From these results, the hazardous radionuclides applicable to partitioning and transmutation are selected. These selected radionuclides such as Tc-99, I-129, Cs-135 and Np-237 are then reviewed in terms of partitioning and transmutation. Separation of I-129, Np-237 and Tc-99 from spent fuels is considered desirable, and transmutation of these radionuclides results in remarkable hazard reduction. However, it is concluded that separation and transmutation of Cs-135 may be ineffective although it is classified into a hazardous radionuclide.

Key Words : transmutation, partitioning, P&T strategy, radiological impact assessment, deep geological disposal, spent fuel, annual individual dose

1. Introduction

Since nuclear power plants have been operated, spent fuels and radioactive wastes from the nuclear power generation have been accumulated year after year. Disposal of those has been a big issue of all countries operating nuclear power plants. As a result, the methods such as reprocessing, transmutation, and geological

disposal have been suggested to deal with the radioactive wastes and spent fuels. In recent years, transmutation has been dreamed of progressive idea and feasible method to support a safe disposal of the radioactive wastes. However, there are many limits to be overcome and it still remains at a stage of R&D. Therefore, the deep geological disposal of spent fuels is considered to be a practical and feasible method than other proposed

methods at present, but there are also many problems to be solved.

Under the assumption that the deep geological disposal is primary policy to deal with spent fuels and transmutation is a cooperative and ancillary method, environmental impact assessment for the deep geological disposal of spent fuels should be performed first to know the extent of impact. Then, the selection of radionuclides to be transmuted should be done on the basis of environmental impact of each radionuclide in order to reduce the impact after the disposal. Annual individual doses from the performed assessment will be more reasonable criteria to decide the radionuclides for transmutation than a simple decision based on the amount of radionuclides.

For the PWR spent fuels, the selection of hazardous radionuclides by using a kind of toxicity index approach was previously studied without further comments related with P&T (Partitioning and Transmutation)[1]. A study has proposed the key radionuclides and P&T strategies on the basis of repository performance assessment, but it is irrelevant to domestic circumstances[2]. Therefore, the selection of key radionuclides for P&T, based on environmental impact from domestic spent fuel disposal concept, was studied in this work.

2. Theoretical Background

2.1. Source Term

When spent fuels are disposed of in the deep geological repository, groundwater plays an important role to release radionuclides. As groundwater infiltrates repository, canister continues to be degraded, and radionuclides begin to dissolve in groundwater.

If all amount of radionuclides dissolve in pore

water within the repository, the initial concentration of i^{th} radionuclide will be like

$$N_i^0 = \frac{M_i}{V \epsilon} \quad (1)$$

where M_i is inventory of i^{th} radionuclide, V is volume of repository and ϵ is porosity of medium[3]. This means that all amount of i^{th} radionuclide is dissolved into only pore water within repository homogeneously. However, in realistic situation, sorption of geological medium will reduce the initial concentration up to the sorption equilibrium state. Equation of the initial concentration for sorption equilibrium is

$$N_i^0 = \frac{M_i}{V(\epsilon + \rho K_d)} \quad (2)$$

where ρ is the density of geological medium and K_d is sorption distribution coefficient[3]. However, if solubility of a radionuclide is lower than the initial concentration decided by sorption equilibrium, then solubility of the radionuclide should be the initial concentration.

2.2. Geosphere

Radionuclide migration in geosphere is a very complex process related with many factors, e.g., hydrogeology, chemistry, radioactive decay and so on. It needs much effort and computing time to precisely predict the behavior of radionuclides in geosphere. The objective of this study is not an exact prediction of radionuclides migration in geosphere but a decision of radionuclides that have significant contribution to the annual individual dose. Therefore, simplified one-dimensional concentration transport equation is used, and it will suffice our purpose.

In the semi-infinite one-dimensional porous medium with flow along the z-direction, canonical system, considering decay chain, is expressed as

$$\frac{R_i}{D_i} \frac{\partial N_i}{\partial t} + \frac{v}{D_i} \frac{\partial N_i}{\partial z} + \nu_i N_i = \frac{\partial^2 N_i}{\partial z^2} + \nu_{i-1} N_{i-1}, \quad i=1,2,\dots \quad (3)$$

where

$$\nu_0=0, \quad \nu_i = \frac{R_i \lambda_i}{D_i}, \quad \nu_{i-1} = \frac{R_{i-1} \lambda_{i-1}}{D_{i-1}} \quad (4)$$

Eq(3) is to be solved for $N_i = N_i(z, t)$, the concentration of the i^{th} member, in a one dimensional domain for time $t > 0$.

On account of the linearity of (3), the solution for the individual chain member can be represented as a sum of functions, which satisfy (3). For an arbitrary i^{th} member with its j^{th} precursor,

$$\frac{R_l}{D_l} \frac{\partial N_l^{(j)}}{\partial t} + \frac{v}{D_l} \frac{\partial N_l^{(j)}}{\partial z} + \nu_l N_l^{(j)} = \frac{\partial^2 N_l^{(j)}}{\partial z^2} + \nu_{l-1} N_{l-1}^{(j)}, \quad \nu_0 = 0, \quad l = 1, 2, \dots, i, \quad j \leq l$$

$$N_{l-1}^{(j)} = 0, \quad \text{for } l \leq j$$

According to this decomposition of the problem, the functions $N_l^{(j)}(z, t)$ must satisfy the following system of equation for $t > 0$

The functions are subject to

$$N_l^{(j)}(z, 0) = 0 \quad (7)$$

$$-D_l \epsilon \left. \frac{\partial N_l^{(j)}}{\partial z} \right|_{z=0} + \nu_l N_l^{(j)}(0, t) = \delta_{ij} \nu_l N_l^0 \phi_j(t), \quad j \leq l \quad (8)$$

where $\phi_j(t) = 0$ for $t < 0$.

The boundary concentration at $z = 0$ obeys the Bateman equation

$$N_i(0, t) = \sum_{j=1}^i B_j e^{-\lambda_j t}, \quad (9)$$

$$B_j = \sum_{m=1}^i N_m^0 \left(\frac{1}{\lambda_i} \prod_{r=m}^i \lambda_r \right) \left/ \prod_{\substack{l=m \\ l \neq j}}^i (\lambda_l - \lambda_j) \right. \quad (10)$$

where N_m^0 is the initial concentration of the m^{th} nuclide and the product term in the denominator is defined as unity when $m = j = i$.

The exact solution[4] is

$$N_i(z, t) = e^{\frac{vz}{2D}} \frac{D}{R_i \pi} \left\{ \sum_{k=1}^i B_k e^{-\lambda_k t} \int_0^\infty \frac{p \sin(pz)}{\Delta_k} (1 - e^{-\Delta_k t}) dp + \sum_{j=1}^{i-1} C_j^{(i)} \sum_{k=1}^j B_k e^{-\lambda_k t} \times \sum_{n=j}^i \int_0^\infty \frac{p \sin(pz)}{\prod_{\substack{r=j \\ r \neq n}}^i (\Gamma_{nr} p^2 + \gamma_{nr})} \frac{(1 - e^{-\Delta_{nk} t})}{\Delta_{nk}} dp \right\},$$

$z > 0, \quad t > 0, \quad i = 1, 2, \dots$

$$\Delta_{nj} = \frac{D}{R_n} (p^2 + q_{nj}^2), \quad q_{nj}^2 = \left(\frac{v}{2D} \right)^2 + \frac{R_n}{D} (\lambda_n - \lambda_j) \quad (12)$$

$$\gamma_m = \left[(\lambda_n - \lambda_r) + \left(\frac{v}{2D} \right)^2 \right] \Gamma_m, \quad \Gamma_m = D \left(\frac{1}{R_n} - \frac{1}{R_r} \right)$$

and

$$C_i^{(j)} = \prod_{n=j}^{i-1} \lambda_n \quad (13)$$

This solution is applicable to semi-infinite one-dimensional porous medium with Bateman boundary condition. There are slight changes for other situations such as constant boundary concentration, finite domain and so on.

2.3. Biosphere

The annual dose, resulting from groundwater containing dissolved radionuclides, can be calculated by the general equation

$$R_{aipj} = 10^6 C_{ip} U_{ap} D_{aipj}, \quad (14)$$

where

R_{aipj} : Annual dose [Sv/yr],

C_{ip} : Concentration of radionuclide i ([Bq/l],

Table 1. Inventory, Half Life and K_d Value of Radionuclides

Nuclide	Half life [yr]	Kd [m^3/kg]		Inventory [g/canister/ m^2]		DUPIC ^[7]
		Backfill ^[10]	Hostrock ^[11]	PWR ^[11]	CANDU ^[11]	
Tc-99	2.13E5	1E-1	7.5E-4	5.646	4.995	2.049
I-129	1.57E7	1E-2	0	1.309	1.008	0.4882
Cs-135	2.3E6	4.82E-2	6.025E-4	2.197	0.3941	0.3906
Np-237	2.14E6	3	7.5E-4	3.276	0	2.447
U-233	1.585E5	2.96E-8	7.4E-11	2.08E-4	0	0
Th-229	7.34E3	3	1.75E-2	0	0	0

[Bq/kg] or [Bq/ m^2],

D_{aipj} : Dose conversion factor ([Sv/yr / Bq/ m^2] or [Sv/yr / Bq/l]),

U_{ap} : Ingestion rate or human activity rate ([kg/yr] or [hr/yr]),

10^6 : Unit conversion factor [μ Sv/Sv],

p, i, a and j denote pathway, radionuclide, age group and target organ respectively in this equation[5].

3. Numerical Calculation

Three different types of spent fuels are considered: PWR, CANDU and DUPIC. A conceptual repository design is adopted because a site for the repository is not specified yet[6]. Fig. 1 shows the repository design. The YGN 3&4 spent fuel is chosen for PWR spent fuel, which has a 33,000MWD/MTU specific burnup. The WSN 2 spent fuel is chosen for CANDU spent fuel, which is assumed to have a 7,200MWD/MTU specific burnup. It is assumed that PWR spent fuels with a 35,000MWD/MTU specific burnup are made into DUPIC fuel, and then cooled for 10 years after being burned up to 19,000 MWD/MTU within CANDU reactor[7]. The previous study shows that one canister can contain 4 PWR spent fuel assemblies and 333 CANDU spent fuel bundles respectively, which considered thermal loading limit(<100°C) of geological medium[8]. According

to this, one canister can contain 1.76MTU for PWR spent fuel and 6.327MTU for CANDU spent fuel, respectively.

Since no commercial operation is established yet, DUPIC remains at a conceptual study. However, from a recent DUPIC fuel study, the amount of fuels in a disposal container is assumed to be 60 bundles (1.095MTU) to achieve the 90 °C temperature design limit because of its high heat output[9].

Data such as inventory, K_d value and half life of major radionuclide for each fuel type are listed in Table 1[1,7,10-11]. To know the initial concentration of each radionuclide, sorption equilibrium values are calculated and compared with solubility of each radionuclide. Backfill material adopted in the conceptual repository design is a compacted bentonite. Density and porosity of the bentonite in this work is 2,500kg/ m^3 and 0.25[10]. Lower value between solubility and sorption equilibrium is adopted for initial concentration.

It is assumed that groundwater containing dissolved radionuclide migrates about 1,870m through geosphere. Then, the groundwater is discharged to biosphere through the surface water body. As described previously, one-dimensional radionuclide transport equation through porous medium is used in this study. UCBNE 41, programmed according to a numerically modified

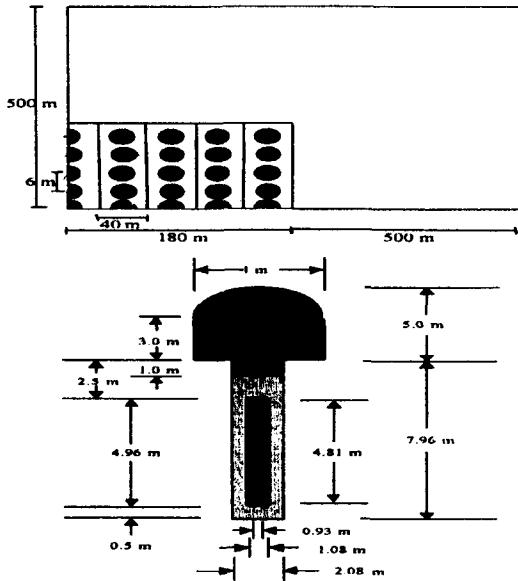


Fig. 1. Conceptual Repository Design [6]

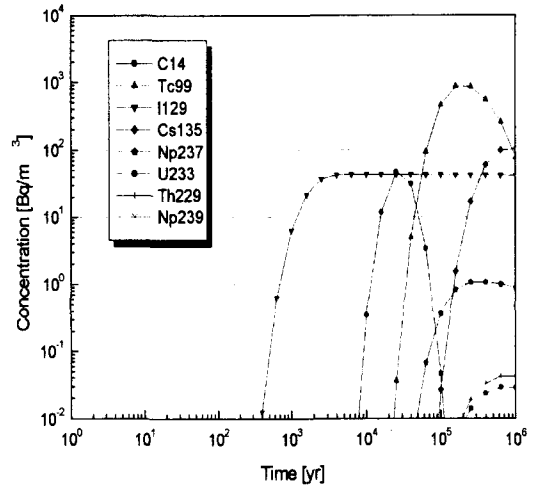


Fig. 3. Concentration Profile, CANDU

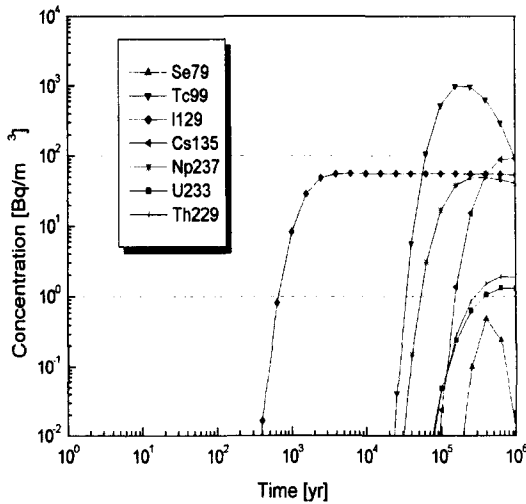


Fig. 2. Concentration Profile, PWR

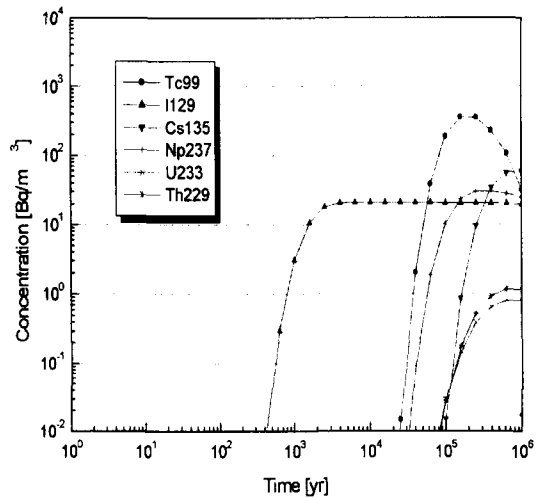


Fig. 4. Concentration Profile, DUPIC

form of equation (11), is used for geosphere calculation. Average groundwater velocity and dispersion coefficient used in this work are 1.08 m/yr and 202.8 m²/yr, respectively, for all radionuclides[1]. Density and porosity of hostrock are 2,000 kg/m³ and 0.025 each[1]. The results in terms of the concentration dissolved in

groundwater [g/m³] at a discharge point are represented in Figs 2~4. These figures correspond to those of calculation for PWR, CANDU and DUPIC, respectively.

Groundwater released into biosphere through the surface water body is used for many purposes such as drinking, cultivation, livestock breeding

Table 2. Maximum Individual Data [5]

Category	Adult
Drinking [l/yr]	196.3
Crops [kg/yr]	190
Leaf vegetable [kg/yr]	225
Milk [l/yr]	63
Meat [kg/yr]	55

and so on. To calculate the annual individual dose that can be caused by groundwater containing dissolved radionuclides, INDAC(Integrated Dose Assessment Code Package for KINS)[5] is used in this work. However, to include various exposure pathways, it needs much information as to biosphere. The biosphere data used in this study are from the default values within INDAC. Various biosphere data used are summarized in Table 2, which shows the maximum individual data related to the human habitat. Drinking, livestock breeding, cultivation and domestic usage were considered.

The results of INDAC calculation are concentration-to-dose factors. Its dimension is annual individual dose [mSv/yr] per unit concentration of radionuclide [Bq/m³]. It means how much dose is induced by 1 Bq/m³ of radionuclide in biosphere. These calculated factors are listed in Table 3 separately. Since these are based on 1 Bq/m³ of radionuclide concentration, actual annual individual dose can be acquired by combining these with the results of the previous section.

4. Results and Discussion

4.1. Annual Individual Dose

The annual individual dose resulting from the disposal of PWR spent fuels corresponds to Fig. 5. Figs 6 and 7 show the results for CANDU and DUPIC spent fuels, respectively. For PWR, a small contribution from Se-79, which does not appear

Table 3. Concentration-to-dose Conversion Factor

Nuclide	Concentration-to-dose factor [mSv/yr per Bq/m ³]
Tc-99	1.62E-10
I-129	2.79E-08
Cs-135	5.47E-10
Np-237	2.79E-08
U-233	1.27E-08
Th-229	1.54E-08

in other cases, is observed. For CANDU, except I-129, other radionuclides have lower contributions than those to PWR and DUPIC cases. Especially, contributions of actinides are low because of small initial inventories due to low burnup. C-14 is noticeable radionuclide in disposal of CANDU spent fuels but it does not have significant effect on the dose though it may not be neglected. I-129 and Tc-99 have contribution to the dose as high as in other cases. Disposal of DUPIC spent fuels has similar tendency to the results for PWR as shown in Fig. 7.

From these results, it is clear that I-129 has the greatest contribution to the annual individual dose. I-129, Cs-135, and Tc-99 are key radionuclides among fission products. Although they have slightly different contribution, results show that these are major contributor to the dose at early time. Long-lived I-129 has a low sorption distribution coefficient and high solubility. These properties imply that most of I-129 can reach biosphere without significant loss through geosphere.

Another significant feature is the effect of 4n+1 decay chain to the annual individual dose. Np-237 and its daughters, namely U-233 and Th-229, have considerable contribution to the dose after twenty thousand years. Np-237 is a key radionuclide among actinides. Its impact on the dose is as high as that of I-129 especially in the disposal of PWR and DUPIC spent fuels. In

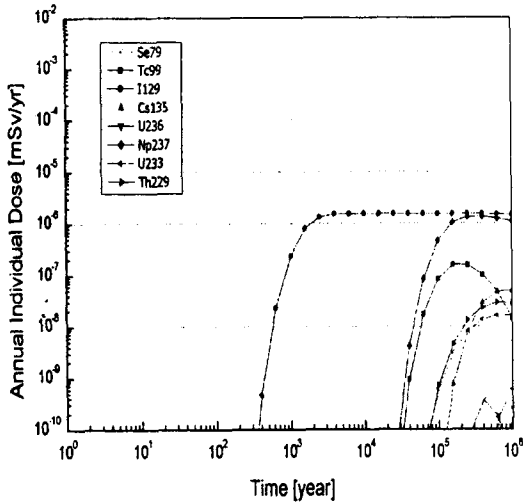


Fig. 5. Annual Individual Dose, PWR

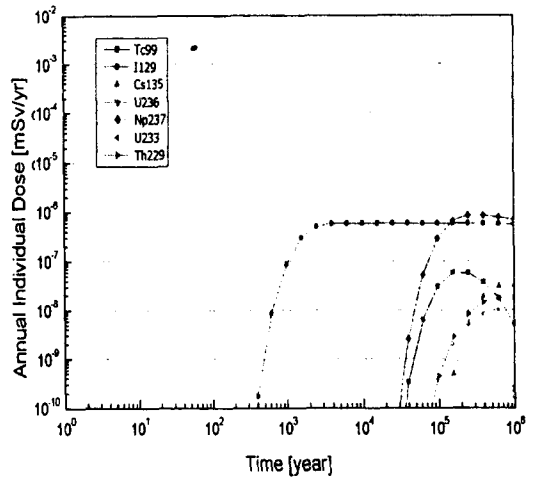


Fig. 7. Annual Individual Dose, DUPIC

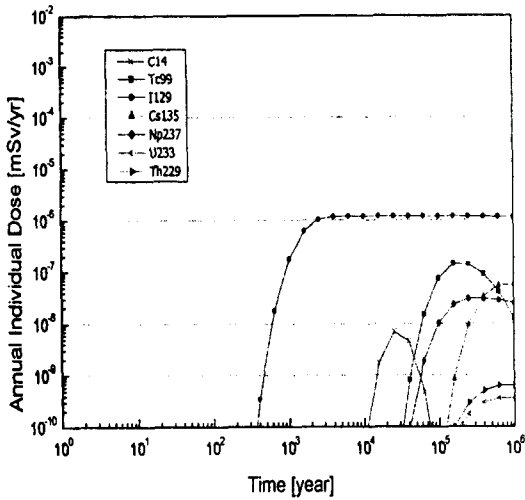


Fig. 6. Annual Individual Dose, CANDU

addition to the radiotoxicity itself, its decay has also an important meaning.

4.2. P&T

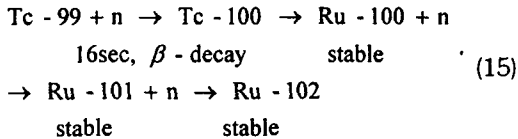
On the basis of these results for the groundwater release pathway, the radionuclides, which should be removed from spent fuels as a

part of a P&T strategy with an aim to reduce the environmental impact of waste disposal, are Tc-99, I-129, Cs-135, and Np-237. Other actinides such as U-233 and Th-229 are not included because they are strongly dependent on the presence of Np-237. If Np-237 could be removed from spent fuel, these daughter radionuclides would have no significant contribution to the dose. The capability of the envisaged transmutation technology and separation processes to deal with the key radionuclides identified in this study is assessed.

4.2.1. Technetium-99

Technetium is divided into two process streams in the dissolution of irradiated fuel into nitric acid. 10~30% of it is associated with the insoluble residues and the remainder remains in the solvent extraction feed solution. The behavior of the soluble fraction during solvent extraction is complex, but it has been shown that virtually all the dissolved technetium can be converged on a separate stream by using an auxiliary high nitric acid scrub, although this leaves the insoluble

technetium to be dealt with[2]. The only isotope of technetium present in cooled fuel is Tc-99, and this can be readily transmuted to Ru-100, via the very short-lived Tc-100 with a thermal neutron capture cross section of about 17b. The capture steps are



The irradiated products are stable but separation of Ru from Tc may require some complex chemistry[12]. Thus, Tc-99 could be transmuted with several cycles of partitioning and transmutation.

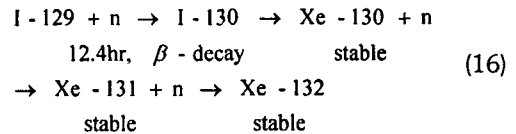
4.2.2. Cesium-135

Cesium is a very soluble fission product and not extracted by TBP(Tributylphosphate). It will therefore pass to the HLW(High Level Waste) in reprocessing plants. Since Cs-133 can be transmuted into Cs-135 by neutron capture, it is necessary to remove other cesium isotopes in the reprocessing before transmutation. But, it is very difficult process to extract the only Cs-135 from cesium isotopes because of strong gamma ray caused by Cs-137.

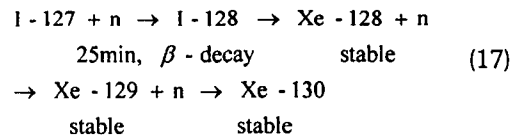
The transmutation of cesium-135 is very difficult at present. Although Cs-135 can be transmuted to the short-lived Cs-136, Cs-133 and Cs-134 are also present and their activation will lead to the production of more Cs-135. In addition, Cs-137 will make cesium target highly active, making it hard to handle in the processes. Therefore, transmutation of cesium could probably be contemplated only if it is preceded by an isotopic separation[12].

4.2.3. Iodine-129

I-129 is mainly converged on ILW(Intermediate Level Waste) streams in reprocessing and can be separated if required. The thermal neutron cross section for neutron capture of I-129 is similar to that of Tc-99. An additional factor is the formation of Xe-130 from decay of the product I-130, which will lead to pressurization of the target unless released. There will also be significant problems of iodine release in case of target failure. The related transmutation sequence is



Stable I-127 is also present in cooled spent fuel. To transmute I-129, it is necessary to verify the unexpected changes of I-127 as



From these results, iodine could be transmuted with several cycles of partitioning and transmutation. However, the problems of corrosion and gaseous product would be encountered[12].

4.2.4. Neptunium-237

The behavior of neptunium in the PUREX process is complicated because the element can exist, in nitric acid solutions, in three different oxidation states(IV, V, VI) which are interconvertible under process conditions and also have different solvent extraction characteristics. To prevent neptunium from entering HLW stream, it is necessary to

Table 4. P & T Strategy of Key Radionuclides

Nuclide	Partitioning	Transmutation
Tc-99	Soluble fraction can be separated, but there is insoluble fraction to be dealt with.	Could be transmuted with several cycles including intermediate reprocessing.
I-129	Mainly diverted into separated streams. Could be largely separated.	Could be transmuted with several cycles including intermediate reprocessing. Corrosion and gaseous product could be problems.
Cs-135	Difficult to extract only Cs-135 from cesium isotopes. Methods for separation of Cs from HLW have been developed.	Not feasible because of transmutation of other cesium isotope, forming more Cs-135. Isotopic separation needed.
Np-237	Could be separated during conventional reprocessing.	Could be transmuted, but significant amounts of Pu-238 would be formed.

adjust the redox conditions in the first cycle to keep the neptunium in the oxidation state VI, which is extractable. Neptunium then passes either to the uranium product or to the plutonium product. If it goes to the plutonium product in the inextractable oxidation state V, neptunium could remain with plutonium during the production and incineration of new plutonium fuel. Alternatively, neptunium may be deliberately reduced to neptunium(IV), so that it follows uranium product. After suitable conditioning, it could be removed as a separate product in the uranium purification cycles. This would provide a separated product for transmutation. Therefore, neptunium could be separated in the partitioning process following conventional reprocessing[2].

In thermal reactors, neutron capture dominates and almost all the Np-237 will react to form Np-238. In a very high neutron flux, Np-238 is fissionable since it has a very high fission cross section of 1,286 b, but, in normal flux levels it will preferentially decay with a 51-hour half-life to Pu-238, which is an alpha emitter of a very high specific activity. It also has a low fission cross section, being transformed into Pu-239 by a further neutron capture. In a fast neutron flux, the

situation is not much different. It is inevitable that considerable intermediate formation of Pu-238 will occur, and this will cause severe handling problems during reprocessing to remove fission products[12]. Consequently, Np-237 could be transmuted but significant amounts of Pu-238 would be formed.

5. Conclusions

Annual individual doses from the groundwater release pathway were found to be low. The calculated results show that the annual individual doses are below the order of 10^{-6} mSv/yr. However, the P&T application to selected key radionuclides will make doses much lower, leading to improved safety. Although the contributions of each radionuclide to doses are slightly different according to the fuel types, the key radionuclides are Tc-99, Cs-135, I-129, and Np-237, regardless of fuel types. From the results in this work, it is revealed that these radionuclides have most contribution to the dose, and their removal from spent fuel by applying P&T will lead to remarkable reduction of the dose.

Separation of Cs-135 from HLW may be the

last choice because radiological impact through normal evolution scenario would be at least one order of magnitude lower than other key radionuclides. However, its separation and transmutation are not so feasible at present that P&T of cesium would be ineffective. Separation of Tc-99 from HLW still has a problem to deal with insoluble portion. Nevertheless, the separation of Tc-99, if possible, may be desirable from the standpoint of hazard reduction.

Np-237 and I-129 are proved to be certainly removed from spent fuels and HLW in any conditions. Np-237 needs to be removed from wastes destined for deep geological disposal and to be diverted into a stream suitable for P&T. Np-237 can be removed in the partitioning process. Separation of Np-237 for transmutation would be most desirable. Finally, I-129 is one of the most significant contributors of radiological hazard resulting from deep disposal of spent fuels and HLW. I-129 has also a few problems as described previously, but removal and transmutation of I-129 could be justified to reduce the environmental impact significantly. These conclusions for selected key radionuclides are summarized in terms of P&T in Table 4.

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