

Characteristics of Several Equilibrium Fuel Cycles of PWR

Abdul WARIS and Hiroshi SEKIMOTO*

Research Laboratory for Nuclear Reactors, Tokyo Institute of Technology, 2-12-1 O-okayama, Meguro-ku, Tokyo 152-8550

(Received January 16, 2001)

This paper evaluated the influence of neutron spectrum on characteristics of several equilibrium fuel cycles of pressurized water reactor (PWR). In this study, five kinds of fuel cycles were investigated. Required uranium enrichment, required natural uranium amount, and toxicity of heavy metals (HMs) in spent fuel were presented for comparison. The results showed that the enrichment and the required amount of natural uranium decrease significantly with increasing number of confined heavy nuclides when uranium is discharged from the reactor. On the other hand, when uranium is totally confined, the enrichment becomes extremely high. The confinement of plutonium and minor actinides (MA) seems effective in reducing radio-toxicity of discharged wastes. By confining all heavy nuclides except uranium those three characteristics could be reduced considerably. For this fuel cycle the toxicity of HMs in spent fuel become nearly equal to or less than that of loaded uranium.

KEYWORDS: *neutron spectrum, equilibrium fuel cycle, importance, uranium enrichment, required natural uranium, heavy metals, PWR type reactors, spent fuels, plutonium, minor actinides, radiotoxicity,*

I. Introduction

Recently, sustainable development has come to a headline. This comes with the so-called “trilemma” issues, which are sustainable economic growth, resources and energy supply, and environmental deterioration. In the light of the explosive growth in world population, massive consumption of energy and resources are required for the sustainable economic growth, which may cause damage to global environment. We must simultaneously overcome these problems in order to ensure sustainable development of the world. As a “smoke free” energy source, nuclear power will play a key role to do it.^{1,2)} The nuclear power can produce enough energy for long period, even for a million years of mankind’s utilization by using fast breeder reactors (FBR) and uranium from seawater.³⁾ However, since commercial operation of FBR will not be realized in near future, in the present study we consider light water reactors (LWR) with enriched uranium. Furthermore, there are global common important concerns in utilization of nuclear energy. The major three are to secure “Nuclear Safety,” “Nuclear Non-proliferation,” and “Radioactive Waste Management.” As for nuclear safety, estimated risk of severe accident of reactor becomes presently very low, compared with any risk in usual civilized life. This progress is obtained by major efforts for enhancing reactor safety technology and safe management. Nuclear non-proliferation should be secured not only by technical measures but also by all kinds of international political endeavors.⁴⁾ Although the amount of radioactive waste per unit electricity generation from nuclear power plants is rather small, the toxicity of the radioactive waste should be reduced as low as possible in order to get higher public acceptance. For studying the long-term future nuclear energy utilization, a nuclear equilibrium state model has been introduced, which may become the most probable condition in the far future.⁵⁾

Nuclear equilibrium state analysis by using enriched uranium fuel for simple one-group microscopic cross-sections data has been performed.⁶⁾ In this basic study, the same neutron spectrum and one-group microscopic cross-sections were employed for all evaluated fuel cycles. These data were generated by using SRAC code for the fresh fuel of pressurized water reactor (PWR) whose fuel enrichment is 3.5% of ²³⁵U. The results showed that the enrichment of charged fuel decreases significantly with increasing number of confined heavy nuclides when uranium as well as fission products (FPs) are discharged from the reactor. The amount of required natural uranium also decreases as well. On the other hand, when uranium is perfectly confined, the enrichment becomes unacceptably high (more than 70% enrichment). The toxicity of the discharged heavy metals (HMs) also decreases with increasing number of confined heavy nuclides.

However, each fuel cycle has its own neutron spectrum and therefore its own one-group microscopic cross-sections. The present paper evaluates the characteristics of several equilibrium fuel cycles of PWR by using its own neutron spectrum. Three characteristics are investigated for several fuel cycle cases. They are uranium enrichment required for reactor criticality, required natural uranium amount, and the toxicity of HMs in spent fuel.

II. Scope of Fuel Cycles and Reactor Core Design

The following five typical fuel cycle cases are investigated, where all FPs and final products of HMs natural decay chain (Tl-Fr) are discharged from the reactor at a certain rate.

Case 1: All HMs are discharged from the reactor.

Case 2: All HMs except Pu are discharged from the reactor. Plutonium is discharged at the rate of one-half of the other HM discharge constant.

Case 3: All HMs except Pu are discharged from the reactor. Plutonium is confined in the reactor.

Case 4: All HMs except U are confined in the reactor. Uranium is discharged from the reactor.

*Corresponding author, Tel. +81-3-5734-3066, Fax. +81-3-5734-2959, E-mail: hsekimot@nr.titech.ac.jp

Table 1 Discharge rates of HMs for each fuel cycle case^{a)}

Case	U	Pu	Other HMs
1	r	r	r
2	r	r/2	r
3	r	0	r
4	r	0	0
5	0	0	0

^{a)} r is chosen to be 33%/yr in the present study.

Table 2 Reactor core design parameters of studied PWR

Power output	3,000MWth
Average power density of pellet	280 W·cm ⁻³
Average power density of cell	100 W·cm ⁻³
Fuel pellet diameter	8.0 mm
Pin diameter	9.6 mm
Pin pitch	11.8 mm
Materials	
Fuel pellet	Oxide
Cladding	Zircaloy-4
Coolant	Light water
Volume fraction	
Fuel pellet	36%
Cladding	16%
Coolant	48%

Case 5: All HMs are confined in the reactor.

In this study we use the discharge constant of 33%/yr. This discharge constant corresponds to the three batches operation of PWR.⁷⁾ **Table 1** summarizes the discharge rate of HMs for all investigated fuel cycles cases.

In the present paper, 3,000MWt PWR systems are studied. **Table 2** shows the basic reactor core design parameters of the investigated PWR for this study. These core design parameters are the same as those of the reference.⁶⁾ The average power density of fuel pellet was fixed to 280 W·cm⁻³ to satisfy 100 W·cm⁻³ of cell average power density.

III. Calculation Method

1. Equilibrium State Model

The nuclear equilibrium-state model in the present study

$$k = \frac{\sum_{j \in \text{HM,FP}} \nu \sigma_{f,j} n_j \phi_{fp}}{\sum_{j \in \text{HM,FP}} \sigma_{a,j} n_j \phi_{fp} + \sum_{k \in \text{cladding}} \sigma_{a,k} n_k \phi_{cl} + \sum_{l \in \text{coolant}} \sigma_{a,l} n_l \phi_{co}}, \quad (4)$$

where ϕ_y is the neutron flux. Subscript y denotes the fuel cell region (fp , cl , and co correspond to fuel pellet, cladding, and coolant, respectively). The ν represents the number of neutrons produced in each fission reaction. The actual calculation is performed by SRAC95 code.

For calculating the number density of each nuclide in the fuel pellet and the total supply rate of fuel nuclides, we perform an equilibrium burnup calculation. In the nuclear equi-

librium state analyses, h -value, which is defined by

- Refueling process is a continuous process.
- Number density of each nuclide in the reactor is constant.

In these conditions the number density of i -th nuclide, n_i , in the reactor core should satisfy the following equation:⁵⁾

$$\frac{dn_i}{dt} = -(\lambda_i + \phi \sigma_{a,i} + r_i) n_i + \sum_j \lambda_{j \rightarrow i} n_j + \phi \sum_j \sigma_{j \rightarrow i} n_j + s_i = 0, \quad (1)$$

where

- ϕ : Neutron flux
 - λ_i : Decay constant of i -th nuclide
 - r_i : Discharge constant of i -th nuclide
 - $\lambda_{j \rightarrow i}$: Decay constant of j -th nuclide to produce i -th nuclide
 - $\sigma_{j \rightarrow i}$: Microscopic transmutation cross-section of j -th nuclide to produce i -th nuclide
 - s_i : Supply rate of i -th nuclide
 - $\sigma_{a,i}$: Microscopic absorption cross-section of i -th nuclide.
- Here, the absorption cross-section includes fission, capture, (n , $2n$) and other nuclear transmutation cross-sections. The formation of fission products can be estimated by substituting $\sigma_{j \rightarrow i}$ in Eq. (1) with the following equation:

$$\sigma_{j \rightarrow i} = \sigma_{f,j} \gamma_{j \rightarrow i}, \quad (2)$$

where

- $\sigma_{f,j}$: Microscopic fission cross-section of j -th nuclide
- $\gamma_{j \rightarrow i}$: Yield of i -th nuclide from j -th fissile nuclide.

In a matrix form Eq. (1) can be written as follows:

$$\mathbf{Mn} = \mathbf{s}, \quad (3)$$

where the elements of matrix \mathbf{M} comprise all the coefficients in Eq. (1), and \mathbf{n} and \mathbf{s} are the vectors of n_i and s_i , respectively.

The one-group microscopic cross-sections for Eq. (1) are calculated by using SRAC95⁸⁾ code with nuclear data prepared from JENDL-3.2.⁹⁾

2. Uranium Enrichment Calculation

For criticality judgment of the system, infinite multiplication factor, k , which is defined by the following equation, is commonly used:

$$h = \frac{\sum_{j \in \text{HM,FP}} \nu \sigma_{f,j} n_j \phi}{\sum_{j \in \text{HM,FP}} \sigma_{a,j} n_j \phi} = \frac{(\nu \sigma_f, \mathbf{n})}{(\sigma_a, \mathbf{n})}, \quad (5)$$

is commonly used. The h -value is a ratio of the number of neutrons produced by fission to the number of absorbed neu-

trons in the fuel pellet. By using nuclide importance vector, Eq. (5) can be written as the following equation:¹⁰⁾

$$h = \frac{(f, s)}{(a, s)}, \quad (6)$$

where the importance vectors f and a are calculated from the following adjoint equations:

$$\begin{aligned} M^t f &= \phi \nu \sigma_f, \\ M^t a &= \phi \sigma_a, \end{aligned} \quad (7)$$

where M^t is the adjoint matrix of M . We have called f and a as fission neutron importance and absorbed neutron importance, respectively. The fission neutron importance represents the number of neutrons produced from fission of one nucleus of the studied nuclide and its family members (reaction products) during its existence in the reactor. While the absorbed neutron importance represents the number of neutrons absorbed by one nucleus of the studied nuclide and its family members during its presence in the reactor.

The uranium enrichment calculation is performed iteratively until the one-group cross-sections and nuclide number

densities are converged. The control parameter for this iterative calculation is α -value, which is the parameter for evaluating the ratio between the neutron absorption of nonfuel materials and the neutron absorption of fuel pellet, and defined as the following equation:

$$\alpha = \frac{h}{k} = 1 + \frac{\sum_{k \in \text{cladding}} \sigma_{a,k} n_k \phi_{cl} + \sum_{l \in \text{coolant}} \sigma_{a,l} n_l \phi_{co}}{\sum_{j \in \text{HM,FP}} \sigma_{a,j} n_j \phi_{fp}}. \quad (8)$$

In order to judge the criticality of the system, the neutron leakage from the system should be evaluated. For current PWR, the neutron leakage is estimated about 2% of produced neutrons. Then the following condition is employed for the criticality condition in the present paper:

$$k = 1.02 \equiv k_c. \quad (9)$$

The uranium enrichment to satisfy the criticality condition for each case is determined as follows. The equilibrium burnup calculation is performed to determine the flux level and

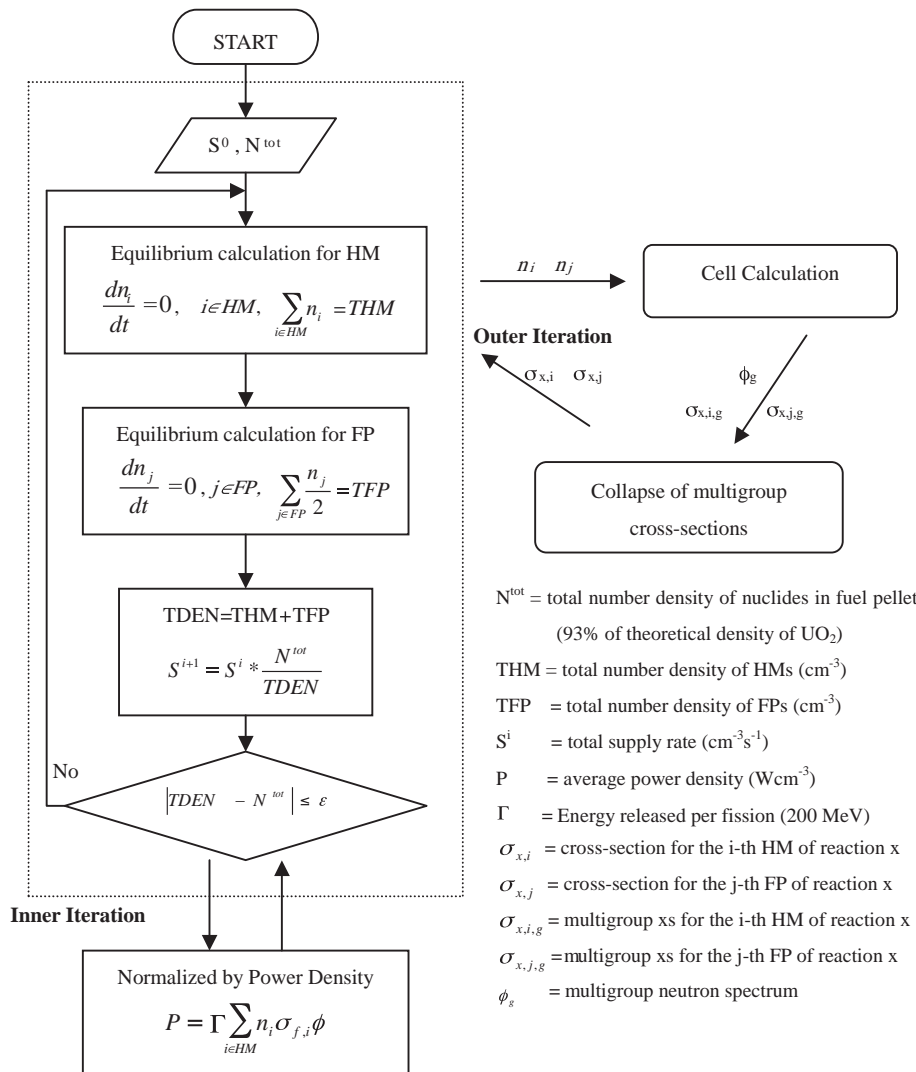


Fig. 1 Flowchart of the equilibrium cell iterative calculation system

the number density of each nuclide in the fuel pellet. In this calculation, 129 heavy nuclides and 1,238 fission products are employed. This equilibrium burnup calculation is coupled with SRAC95 cell calculation code in order to get the neutron spectrum and the one-group microscopic cross-sections of each investigated case as shown in Fig. 1. We have called this coupling calculation as an equilibrium cell iterative calculation system (ECICS).¹¹⁾ In the cell calculation, 26 heavy metals and 66 fission products and 1 pseudo FP are employed. From this coupling calculation procedure we evaluate the value of the infinite multiplication factor, k . If k equal to k_c , the calculation is finished and then we choose the initial enrichment input as the required uranium enrichment for the criticality of the investigated case.

In case k differs from k_c , then we calculate the fission neutron importance and the absorbed neutron importance of fuel nuclides ^{234}U , ^{235}U and ^{238}U by solving Eq. (7). We can evaluate the h -value of each system from the supply densities of fuel nuclides and their importance's. By using this value the control parameter for the iterative calculation, α , can be determined from Eq. (8). Finally, the uranium enrichment for criticality is determined by solving the following three linear equations:

$$(f_{24} - \alpha k_c a_{24})s_{24} + (f_{25} - \alpha k_c a_{25})s_{25} + (f_{28} - \alpha k_c a_{28})s_{28} = 0, \quad (10)$$

$$s_{24} + s_{25} + s_{28} = 100, \quad (11)$$

$$100s_{24} - 0.9937s_{25} = -0.1925, \quad (12)$$

where s_x is an atomic percent of uranium isotopes (^{234}U , ^{235}U and ^{238}U) in the supplied fuel. Equation (10) is derived from Eqs. (6) and (8). Equation (12) shows the relationship between ^{234}U isotope and ^{235}U isotope in the enriched uranium. This equation was derived from data given in the reference.¹²⁾ These calculations are repeated until the value of the infinite multiplication factor, k is equal to k_c . The flow diagram of all calculations procedure to obtain the uranium enrichment for each fuel cycle case is shown in Fig. 2.

3. Natural Uranium Supply

The required amount of natural uranium is calculated by means of two different enrichment processes where the concentration of ^{235}U in the tail is chosen to be 0.3 wt% and 0.1 wt%, respectively. The amount of required natural uranium, S_0 is given by the following equation:⁶⁾

$$S_0 = \frac{(e_1 - e_2)}{(e_0 - e_2)} S_1, \quad (13)$$

where S_1 is the amount of required enriched uranium, which is determined from the equilibrium burnup calculation as already mentioned before. Here e_0 , e_1 , and e_2 are the ^{235}U abundance in natural uranium (0.711 wt%), the concentration of ^{235}U in the produced enriched uranium and the concentration of ^{235}U in the tail of enrichment plant, respectively.

4. Toxicity Evaluation

According to the International Commission on Radiological Protection (ICRP) recommendation on ICRP Publication 60, for toxicity evaluation we used the annual limit on intake

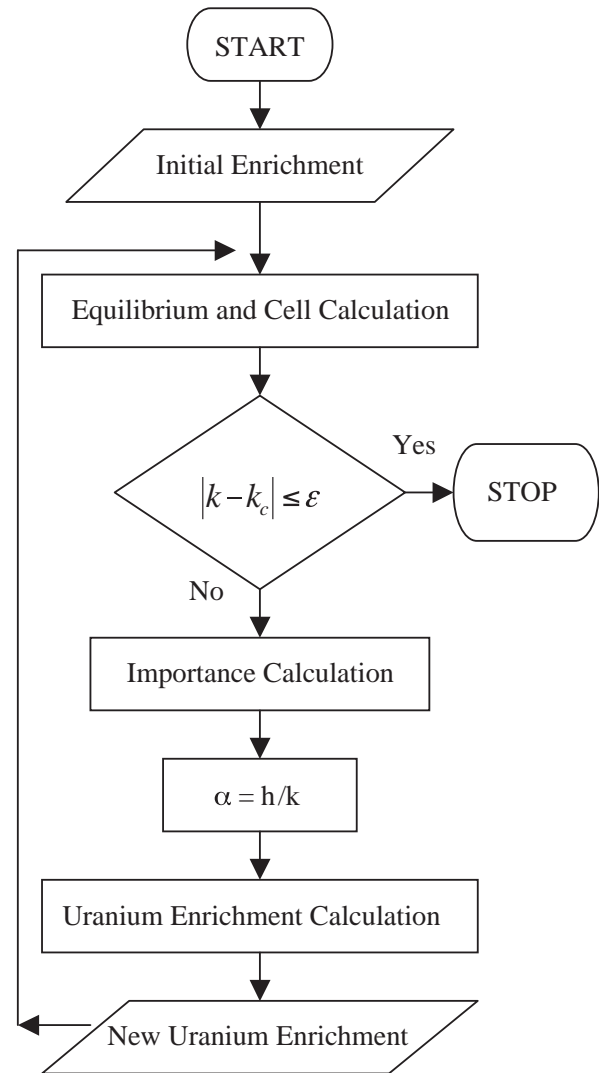


Fig. 2 Flowchart of the whole uranium enrichment calculation process

(ALI) for member of public, which was defined as the following equation:¹³⁾

$$ALI = \frac{0.001}{e(50)}, \quad (14)$$

where $e(50)$ is an effective dose coefficient ($\text{Sv}\cdot\text{Bq}^{-1}$). The value 0.001 in Eq. (14) expresses the 1 mSv limited dose per year for the member of public. In the present study, only $e(50)$ values for ingestion were employed because most of heavy metals are in solid form. The ICRP has published some set of data of $e(50)$, such as on ICRP Publication 68¹⁴⁾ and ICRP Publication 72.¹⁵⁾ Even though the $e(50)$ from ICRP Publication 72 are the newest one, in this paper we employed the data from the former one. There are two reasons for this choice. First, $e(50)$ for the member of public in the ICRP Publication 72, is grouping into 6 groups of age. It makes the treatment too complicated, but the present work does not require such a detail information. The second, the effective dose coefficient of the Publication 68 has been used in the previous work.

The toxicity of i -th heavy nuclide is given by the following equation:

$$\text{Toxicity}_i = \frac{\lambda_i N_i}{\text{ALI}_i}, \tag{15}$$

where ALI_i is the annual limit on intake of the i -th nuclide (Bq), and N_i is the total number of the i -th nuclide discharged per year.

The toxicity along the decay time after discharge from the reactor without cooling process is evaluated up to one million years by employing the fourth-order Runge-Kutta method.

IV. Numerical Results and Discussion

1. Neutron Spectra and Microscopic Cross Sections

As already mentioned above, each fuel cycle has its own neutron spectrum, since neutron spectrum depends sensitively on its operating conditions (*e.g.*, fuel loading, isotopic composition, temperature, and coolant conditions).¹⁶⁾ **Figure 3** shows the neutron spectra of all investigated fuel cycle cases. As shown in this figure, the neutron spectrum becomes harder with increasing number of confined nuclides in the reactor core. This change is attributed to the presence of plutonium, especially fissile plutonium (^{239}Pu , ^{241}Pu) in the recycled nuclides. In thermal energy region microscopic absorption cross-section of fissile plutonium is much higher than that of ^{235}U , therefore by recycling Pu in PWR the flux depression occurs, and the neutron spectrum becomes harder. From cases 1 to 3 the neutron spectrum becomes harder gradually due to significant increasing of the number density of Pu isotopes in the core, as shown in **Fig. 4**.

In thermal reactors, transplutonium nuclides with odd number of neutrons like ^{242}Am , $^{242\text{m}}\text{Am}$, ^{244}Am , ^{243}Cm , and ^{245}Cm have absorption cross-sections larger than ^{235}U .¹⁷⁾ For the fuel cycle cases 4 and 5, these nuclides together with Pu are confined, which results in the increment of number density of Pu and MA in the reactor, as shown in **Fig. 4**. Consequently, the neutron spectrum for these latter cases becomes much harder.

One-group microscopic fission and absorption cross-sections of important actinides are tabulated in **Table 3**. This table shows that along the change of cases from 1 to 5 the microscopic fission cross-section of fissile nuclides decreases, but the microscopic fission cross-section of fertile nuclides except ^{238}Pu increases. It is attributed to the hardening of the neutron spectrum. The microscopic absorption cross-section of these all actinides except ^{236}U decreases with increasing number of confined nuclides in the reactor.

As noted in the reference,¹⁸⁾ because of its higher microscopic cross-section, an equal concentration of ^{239}Pu offers more competition for thermal neutrons than ^{235}U . This results in a reduction of the absorption cross-section of the non-fuel materials (cladding and coolant) with increasing number of confined nuclides in the core as shown in **Table 4**. The α -value is also shown in this table. The α -value becomes smaller with increasing number of confined nuclides in the reactor. This means that though by confining Pu and MA in the reactor the absorption cross-sections become smaller, the neutron absorption of nuclides in the fuel pellet still more competitive than that of nonfuel materials.

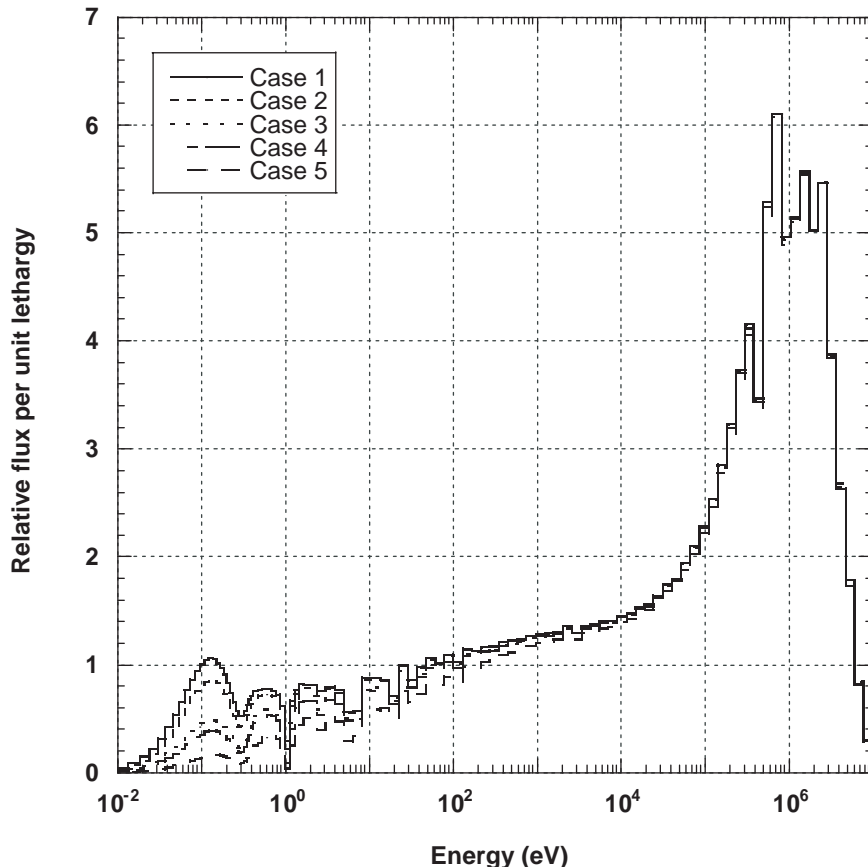


Fig. 3 Neutron spectrum of all fuel cycle cases

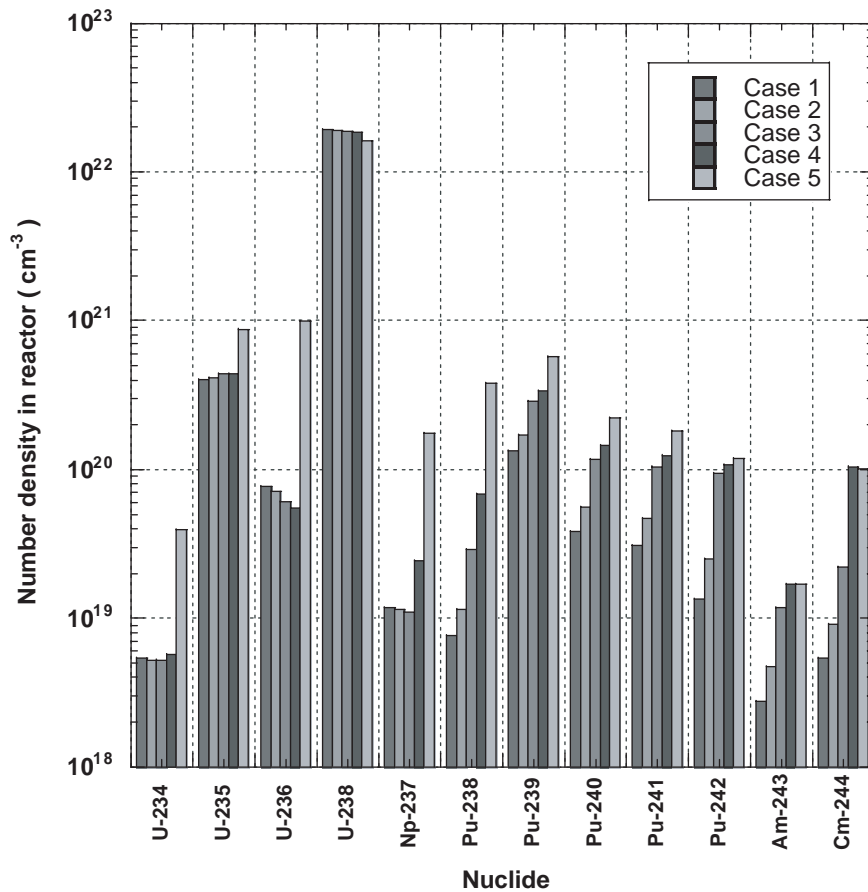


Fig. 4 Number density of important actinides in the reactor

Table 3 One-group microscopic cross-sections of important actinides (barns)

Cross-section	Case	²³⁴ U	²³⁵ U	²³⁶ U	²³⁸ U	²³⁷ Np	²³⁸ Pu	²³⁹ Pu	²⁴⁰ Pu	²⁴¹ Pu	²⁴² Pu	²⁴³ Am	²⁴⁴ Cm
Fission	1	0.503	25.991	0.289	0.100	0.544	2.126	61.609	0.578	64.530	0.433	0.438	0.808
	2	0.512	22.598	0.293	0.102	0.553	2.063	52.232	0.585	55.301	0.441	0.444	0.812
	3	0.532	16.158	0.302	0.107	0.575	1.947	34.232	0.602	37.775	0.458	0.455	0.820
	4	0.540	14.223	0.305	0.109	0.583	1.910	28.905	0.610	32.568	0.466	0.459	0.810
	5	0.567	9.365	0.267	0.115	0.613	1.807	16.265	0.640	19.912	0.492	0.473	0.828
Absorption	1	17.544	32.517	6.908	0.960	27.545	20.328	96.591	95.394	86.899	25.127	45.905	17.138
	2	16.917	28.539	6.962	0.950	26.024	17.680	81.950	78.350	74.405	22.497	44.024	16.429
	3	15.437	20.953	6.940	0.929	22.428	12.702	53.790	48.987	50.643	14.601	38.418	14.312
	4	14.685	18.652	6.849	0.919	20.851	11.160	45.449	41.597	43.597	13.428	35.344	10.205
	5	9.919	12.676	2.888	0.897	15.398	7.279	25.592	25.708	26.454	10.755	27.010	8.881

Table 4 α -value and one-group microscopic cross-sections of cladding and coolant materials (barns)

Case	α	Cladding (Zircaloy-4)				Coolant	
		Zr	Sn	Fe	Cr	H	O
1	1.025	0.0447	0.4277	0.1357	0.1593	0.0173	0.0022
2	1.021	0.0439	0.4141	0.1192	0.1392	0.0150	0.0023
3	1.011	0.0424	0.3859	0.0875	0.1003	0.0107	0.0024
4	1.010	0.0419	0.3756	0.0777	0.0883	0.0093	0.0025
5	1.006	0.0406	0.3378	0.0534	0.0584	0.0059	0.0026

2. Required Uranium Enrichment and Natural Uranium Supply

Table 5 shows the calculation results for the required enrichment, the amount of charged fuel per year, and the burnup. Here, the burnup means the energy produced from the unit amount of discharged fuel. This table also shows the annually required amount of natural uranium, which has been calculated for two different tail concentrations of uranium enrichment. For cases 1 to 4, where uranium is not confined, the enrichment decreases considerably with increasing number of confined nuclides. The quantity of charged fuel reduces a little. Then, the amount of required natural uranium de-

Table 5 Required enrichment and amount of charged uranium fuel per year

Case	Enrichment (wt%)	Charged fuel (t/yr) ^{a)}	Flux (cm ⁻² ·s ⁻¹)	Burnup (GWd/t) ^{b)}	Required natural U(t/yr)	
					0.1 wt% tail	0.3 wt% tail
1	4.2	29.0	3.83 × 10 ¹⁴	37.9	195.4	276.3
2	4.0	28.8	3.80 × 10 ¹⁴	38.1	185.3	261.4
3	3.8	28.1	3.73 × 10 ¹⁴	39.0	169.2	237.9
4	3.6	27.6	3.71 × 10 ¹⁴	39.8	158.9	222.8
5	41.6	1.2	3.46 × 10 ¹⁴	952.7	79.4	117.5

^{a)}tons per year, ^{b)}GWd per ton discharged fuel

Table 6 Required enrichment and amount of charged uranium fuel calculated with the same spectrum (for fresh fuel) case

Case	Enrichment (wt%)	Charged fuel (t/yr) ^{a)}	Flux (cm ⁻² ·s ⁻¹)	Burnup (GWd/t) ^{b)}	Required natural U (t/yr)	
					0.1 wt% tail	0.3 wt% tail
1	4.0	29.2	3.43 × 10 ¹⁴	37.7	188	265
2	3.7	29.0	3.19 × 10 ¹⁴	37.9	170	238
3	3.4	28.8	2.87 × 10 ¹⁴	38.2	156	218
4	3.2	28.5	2.71 × 10 ¹⁴	38.6	144	201
5	70.2	1.2	4.29 × 10 ¹⁴	1,026	134	199

^{a)}tons per year, ^{b)}GWd per ton discharged fuel

creases considerably with increasing number of confined nuclides. This reduction is attributed significantly to the change of importance values.

The large changes of the importance values are attributed to the change of the neutron spectrum. To confirm the effect of the change of the neutron spectrum employed in the calculation, the same parameters of the results for the same spectrum case⁶⁾ are shown in **Table 6**. As can be seen from this table for cases 1 to 4, where uranium is not confined, the same spectrum case showed the underestimated values for the uranium enrichment, the amount of required natural uranium, the neutron flux, and the burnup compared with the present results. For case 5, the same spectrum case gave the overestimated results for the all mentioned parameters compared with the present study. These facts show the significance and necessity of the present study.

By confining the transuranium nuclides in the reactor, the neutron spectrum becomes harder, which generally results in an increasing of the importance values. The fission neutron importance and absorbed neutron importance of important actinides are tabulated in **Table 7**. For the cases 1 through 4, where uranium is discharged from the reactor, fission neutron importance of ²³⁶U, ²³⁸U, ²³⁷Np, and plutonium isotopes increase with increasing number of confined nuclides, but those for ²³⁴U and ²³⁵U decrease regularly with increasing number of confined nuclides. The fission neutron importance of these all actinides become close to the ν -value, the number of neutrons produced in one fission reaction, when uranium is confined together with these nuclides (case 5). This importance value of ²⁴³Am and ²⁴⁴Cm decrease with increasing number of confined Pu, when these MAs are discharged from the reactor.

The absorbed neutron importance values show the same

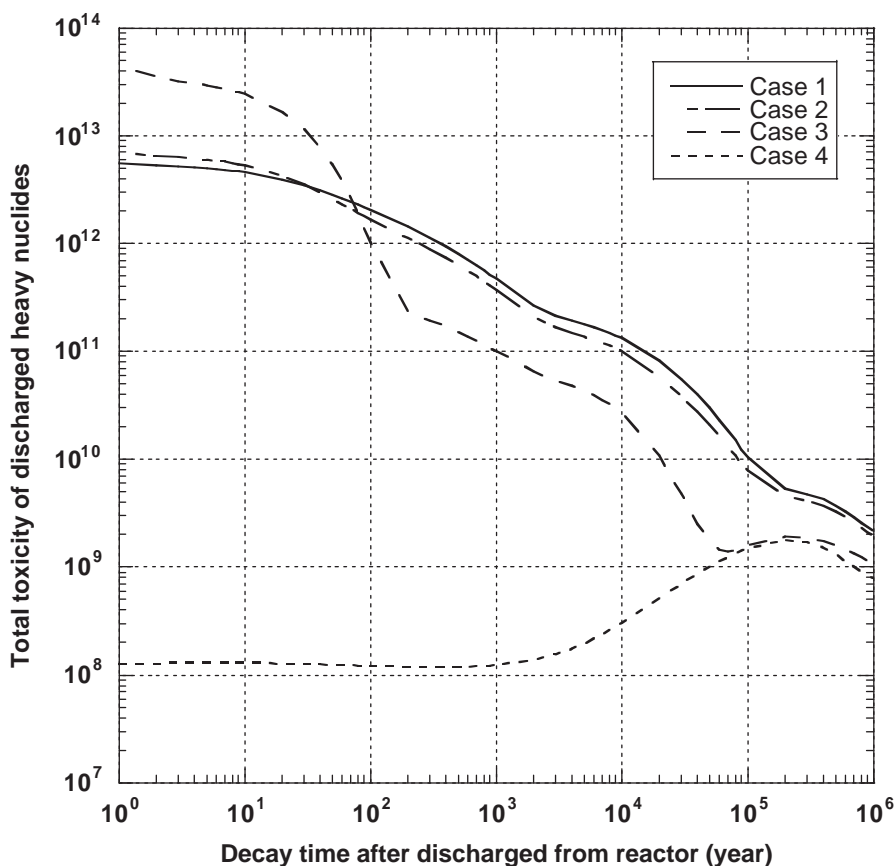
trend as that of the fission neutron importance. **Table 7** also shows a net neutron importance, a measure directly estimating neutron economy.¹⁹⁾ The net neutron importance can be evaluated simply by calculating the fission neutron importance minus the absorbed neutron importance. In general, this value becomes positive for fissile nuclides and opposite value for fertile ones. In thermal energy region, capture cross-section of ²⁴⁴Cm is about ten times larger than fission cross-section.¹⁷⁾ Interestingly, the net neutron importance of this nuclide is positive with the highest value is 0.863 for fuel cycle case 4. This fact was also mentioned in the reference.¹⁹⁾ Significantly, the net neutron importance of ²³⁸U is very small, which means that most of absorption reactions of ²³⁸U nuclide finally results in the fission reaction of its produced fissile nuclides. The absolute value of this importance for ²³⁸U becomes smaller along the change of cases from 1 to 4. Consequently, the required uranium enrichment decreases with increasing number of confined nuclides in the reactor.

In case of fuel nuclides (²³⁴U, ²³⁵U and ²³⁸U) we may define a modified net neutron importance as $(f - \alpha k_c a)$ importance, which also tabulated in **Table 7**. According to this table, even though the modified net neutron importance of ²³⁵U decreases along the change of cases 1 to 4, the absolute value of those for ²³⁴U and ²³⁸U also decrease significantly. Clearly, the required uranium enrichment decreases with increasing number of confined nuclides in the reactor from cases 1 to 4.

The present study shows that recycling of Pu and MA can reduce the required natural uranium. Moreover, this reduction will become significant when uranium is perfectly confined in the reactor. However, the required enrichment becomes extremely high. It is attributed to large accumulation of ²³⁶U. Uranium-236 is produced mostly from (n, γ) reaction of ²³⁵U. According to **Fig. 4**, the number density of ²³⁶U accumulated

Table 7 Importance values of the important actinides

Importance	Case	²³⁴ U	²³⁵ U	²³⁶ U	²³⁸ U	²³⁷ Np	²³⁸ Pu	²³⁹ Pu	²⁴⁰ Pu	²⁴¹ Pu	²⁴² Pu	²⁴³ Am	²⁴⁴ Cm
<i>f</i>	1	0.437	1.071	0.106	0.065	0.440	0.819	1.804	1.326	1.697	0.374	0.736	0.936
	2	0.399	0.998	0.139	0.072	0.612	1.211	2.073	1.621	1.897	0.469	0.705	0.901
	3	0.316	0.827	0.239	0.086	1.157	2.602	2.669	2.303	2.294	0.703	0.620	0.805
	4	0.294	0.791	0.539	0.093	2.809	2.807	2.946	3.062	3.063	3.450	3.459	3.458
	5	2.566	2.562	2.908	2.932	2.933	2.938	2.954	3.071	3.071	3.432	3.443	3.442
<i>a</i>	1	0.652	0.686	0.389	0.077	0.975	0.962	1.410	1.686	1.175	1.110	1.339	0.734
	2	0.618	0.642	0.419	0.082	1.132	1.362	1.640	2.002	1.363	1.427	1.325	0.727
	3	0.540	0.540	0.502	0.094	1.616	2.726	2.175	2.774	1.797	2.241	1.281	0.703
	4	0.519	0.536	0.886	0.099	3.752	2.847	2.399	3.412	2.449	4.453	3.579	2.595
	5	3.102	2.230	4.328	3.047	3.671	2.797	2.350	3.378	2.439	4.426	3.592	2.611
<i>(f - a)</i>	1	-0.216	0.384	-0.283	-0.012	-0.536	-0.143	0.394	-0.360	0.522	-0.736	-0.603	0.202
	2	-0.219	0.355	-0.280	-0.011	-0.520	-0.151	0.433	-0.381	0.533	-0.958	-0.620	0.174
	3	-0.224	0.287	-0.263	-0.008	-0.458	-0.124	0.493	-0.471	0.497	-1.538	-0.661	0.102
	4	-0.225	0.254	-0.347	-0.006	-0.943	-0.040	0.547	-0.350	0.614	-1.003	-0.120	0.863
	5	-0.536	0.332	-1.420	-0.115	-0.738	0.141	0.604	-0.307	0.632	-0.994	-0.150	0.831
<i>(f - αk_ca)</i>	1	-0.245	0.353	-0.301	-0.016	-0.580	-0.187	0.330	-0.437	0.468	-0.787	-0.664	0.169
	2	-0.245	0.329	-0.298	-0.014	-0.567	-0.208	0.365	-0.464	0.477	-1.017	-0.675	0.144
	3	-0.241	0.271	-0.278	-0.011	-0.509	-0.210	0.426	-0.558	0.441	-1.608	-0.701	0.080
	4	-0.241	0.238	-0.374	-0.009	-1.057	-0.126	0.474	-0.453	0.540	-1.137	-0.228	0.785
	5	-0.617	0.273	-1.533	-0.195	-0.834	0.068	0.542	-0.395	0.569	-1.109	-0.244	0.763

**Fig. 5** Total toxicity of discharged HMs in the spent fuel

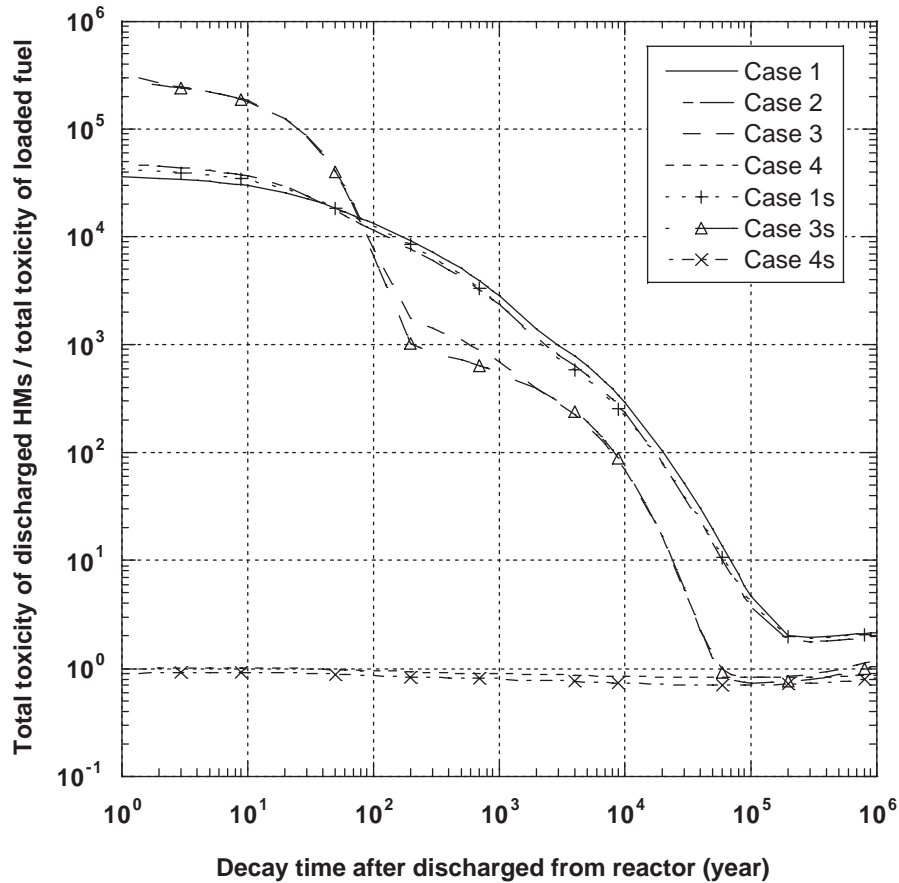


Fig. 6 Ratio of total toxicity of discharged HM to loaded fuel (s indicates the same spectrum results.)

in the reactor core is very large for case 5, more than that of ^{235}U . In other words, by confining uranium in the reactor, larger fraction of ^{235}U undergoes (n, γ) reaction to produce ^{236}U . The latter is also a strong absorber. This characteristic is clearly shown in Table 7 with the value of the absorbed neutron importance of about 4.33. As a result, we need much higher uranium enrichment to overcome this problem.

3. Toxicity of Heavy Metals

The radio-toxicity change for different fuel cycle case is shown in Fig. 5. They are the total toxicity of discharged HMs along the time after discharged from the reactor without cooling process. Figure 6 shows the ratio of total toxicity of discharged HMs to total toxicity of loaded fuel along the time after discharged from the reactor without cooling process.

Figure 6 also shows the ratio of total toxicity of discharged HMs to total toxicity of loaded fuel of some investigated cases (Case 1s, Case 3s, and Case 4s) for the same spectrum case⁶) as a matter of comparison with the present results. There are some discrepancies between the results for the present study and for the same spectrum case.

As shown in these figures, the confinement of Pu and MA seems effective in reducing the radio-toxicity of discharged wastes. However, to realize these incineration performances, many technical challenges should be confronted such as the need of higher plutonium inventory and extremely high decontamination factor. The waste is zero for case 5, and the graph is not shown in these figures. The results for cases

1 and 2 show the similar values each other, while case 3 shows the higher value up to 70 years after discharged and then becomes lower. The higher toxicity ratio in case 3 during first 90 years comes from middle-lived MA, especially, ^{244}Cm (half-life=18.11 yr). The toxicity ratio of case 4 is always less than unity along decay time after 20 years discharge from the reactor.

V. Conclusions

The characteristics of several equilibrium fuel cycles of PWR have been studied. In this study, five kinds of fuel cycles were investigated. The required uranium enrichment, the required natural uranium amount and the toxicity of heavy metals (HMs) in spent fuel were presented for comparison. The numerical results showed that the uranium enrichment decreases considerably with increasing number of confined heavy nuclides when uranium is discharged from the reactor. The amount of required natural uranium also decreases as well. This scenario also seems effective in reducing the radio-toxicity of discharged wastes. However, when uranium is perfectly confined in the reactor, the enrichment becomes extremely high, but the toxicity of HMs in the spent fuel is zero.

By confining all heavy nuclides except uranium those three characteristics can be reduced considerably. For this fuel cycle the toxicity of HMs in the spent fuel become nearly equal to or less than that of loaded uranium.

References

- 1) S. Yoda, *Proc. GLOBAL'97.*, Vol. 1, p. 2 (1997).
 - 2) A. E. Waltar, *Proc. GLOBAL'97.*, Vol. 1, p. 8 (1997).
 - 3) H. Sekimoto, *Proc. Int. Conf. on Reactor Physics and Reactor Computation*, Tel Aviv, Israel, p. 515 (1994).
 - 4) S. Matsuura, *Nucl. Phys.*, **A654**, 417c (1999).
 - 5) H. Sekimoto, N. Takagi, *J. Nucl. Sci. Technol.*, **28**, 941 (1991).
 - 6) A. Waris, H. Sekimoto, *Ann. Nucl. Energy*, **28**, 153 (2001).
 - 7) N. E., Todreas, M. S. Kazimi, *Nuclear Systems I, Thermal Hydraulic Fundamentals*, 1st Ed., Taylor & Francis, Bristol, 30 (1990).
 - 8) K. Okumura, *et al.*, JAERI-Data/Code 96-015, (1996).
 - 9) T. Nakagawa, *et al.*, *J. Nucl. Sci. Technol.*, **32**, 1259 (1995).
 - 10) H. Sekimoto, A. Nemoto, *Proc. GLOBAL'97.*, Vol. 1, p. 198 (1997).
 - 11) A. Mizutani, H. Sekimoto, *J. Nucl. Sci. Technol.*, **34**, 596 (1997).
 - 12) G. E. Hansen, H. C. Paxton, *Nucl. Sci. Eng.*, **72**, 230 (1979).
 - 13) ICRP, *Ann. ICRP*, **21**, 67 (1990).
 - 14) ICRP, *Ann. ICRP*, **24**, 25 (1995).
 - 15) ICRP, *Ann. ICRP*, **26**, 25 (1996).
 - 16) J. J. Duderstadt, L. J. Hamilton, *Nuclear Reactor Analysis*, 3rd Ed., John Wiley & Sons, New York, 291 (1976).
 - 17) OECD, NEA/WPEC-8, (1999).
 - 18) H. W. Graves, *Nuclear Fuel Management*, 1st ed., John Wiley & Sons, New York, 298 (1979).
 - 19) H. Sekimoto, A. Nemoto, *Nucl. Sci. Eng.*, **135**, 84 (2000).
-