Cumulative production of transuranium isotopes in a multi-recycling fast reactor system†

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A simple numerical calculation method based on ORIGEN-II to evaluate cumulative production of TRUs in multi-recycling fast reactor system was proposed. By combining the results of some basic repeated irradiation calculation of TRUs by ORIGEN-II, the cumulative amount of isotopes of Pu, Np, Am and Cm were obtained for fast reactor systems having different breeding ratios. Their cumulative amounts were given as simple numerical functions. By using these results, the time dependent characteristics of the accumulation of TRU isotopes were studied both in terms of weight and radiotoxicity. Some characteristics of the TRU accumulation in Pu-Recycling and Actinide -recycling FBR system were revealed and both systems are compared.

Keywords:transuranium, TRU, actinide recycle, fast reactor system, FBR, fuel cycle, cumulative production of TRU, ORIGEN, radiotoxicity

多重リサイクリングを基本とした高速炉燃料サイクルにおけるTRU核種の累積生産量を評価するために、ORIGEN-IIを用いた簡単な計算手法を提案した。ORIGEN-II を用いた基本繰り返し計算の結果を組み合わせることによって、異なる増殖比を持つ高速炉燃料サイクルにおける、Pu, Np, Am, Cm の同位体の累積生産量を得た。これらの累積生産量を簡単な関数形に表して紹介した。これらの結果を用いて、時間依存のTRUの累積生産量について、それらの重量及び放射線毒性の二つの指標を用いて評価した。プルトニウムリサイクル型の高速炉サイクルと、TRUをリサイクルする高速炉サイクルの特性が明らかになり、両者を比較評価した。

Keywords: 超ウラン元素, TRU, アクチニドリサイクル, 高速炉システム, FBR, 燃料サイクル, TRUの累積生産量, ORIGEN, 放射線毒性

1 Introduction

Nuclear power generation, though more efforts are needed for obtaining public's understanding on its safety, is still expected as only realistic candidate of the post-fossil energy source. However, its utilization over long future period and its substantial prominence against fossil energy can not be obtained without recycling the fuel materials in the system. On the other hand, during the long-term operation of nuclear recycling system, taransuranium (plutonium as major element and other minor elements; TRUs hereafter) will be produced and accumulated, and their quantity and quality will strongly depend on the types of the recycling systems[1,2]. The amount and isotopic composition of the accumulated TRUs in the future nuclear recycling system are indispensable information for the analysis of the TRU-bearing wastes in the future, and the strategic analysis on the handling of massive cumulated man-made toxicity. Generally for this purpose, detailed reactorphysical calculations are needed for which sophisticated tools and personnel skills are needed. However, for the purpose of obtaining a rough prospect on the future fuel cycle options, a rather simple calculation will be enough and preferred. Thus in this study, for the purpose of calculating the accumulation of TRU nuclides in multi-recycling fast reactor systems, authors pro-pose a simple calculation method using well-known burnup calculation code ORIGEN-II.

By using this new method, in order to characterize the massive accumulation of TRUs in the future society which will depends on the fast reactor recycling system, authors analyzed the isotopic composition and quantity of the TRUs accumulated in multi-recycling fast reactor systems.

2 Assumption and Condition for the Analysis

Two types of the fast reactor systems are treated , (1)fast breeder reactor fuel cycle in which plutonium is repeatedly recycled, and (2)fast breeder reactor system in which all the TRUs (other TRUs together with plutonium) are repeatedly recycled[3,4]. The latter is often called actinide recycling system, thus is abbreviated "An-R system" in this paper. The former is called plutonium recycling system abbreviated by "Pu-R system" in this paper. In both systems, as the repeated recycling proceeds, the composition of every TRU isotope changes from the initial state and attains a certain equilibrium state which is a nature of the system. In this paper, a simple calculation method was applied to obtain numerical formula expressing the inventories of TRU isotopes.

2.1 Basic Calculation and Conditions

For the derivation of numerical formula providing the cumulative amount of isotopes in a multi-recycling systems, some basic irradiation calculation results of the starting materials in the core and blanket of the fast reactor are used. For obtaining these, irradiation calculation tool ORIGEN-II was applied[5]. Fast Reactor specifications assumed are summarized in Table 1 the basic parameters of which are cited from the literature[6] but some details are estimated. Applied conditions are summarized in Table 2. The starting fuel materials supplied from LWRs to fast reactor system are also calculated by the ORIGEN-II with the conditions listed in Table 3.

The TRU isotopes treated in this paper are thirteen

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Table 1 Fast breeder reactor specifications assumed

Reactor Power	GWe	1	
Thermal Efficiency	%	38.5	
Plant Capacity Factor	%	85	
Equilibrium Core HM Inventory	t	23.13	
Core U Inventory	t	18.78	
Core Pu Inventory	t	4.39	
Equilibrium Blanket HM Inventory	t	32.10	
Refueling Batches		3	
		Core fuel	Blanket fuel
Average Burnup	MWD/t	90000	5198
Average Specific Power	MW/t	82	4.7
Refueling Time	d	1097.6	1097.6
Loaded Fuel Composition	per tHM	Core fuel	Blanket fuel
U-235	kg	2.4	3.0
U-238	kg	812.4	997.0
Pu-238	kg	4.2	0.0
Pu-239	kg	100.6	0.0
Pu-240	kg	43.9	0.0
Pu-241	kg	24.6	0.0
Pu-242	kg	11.9	0.0

Table 2 ORIGEN-II Calculation conditions for FBR

	Core	Blanket
Cross Section Library	LMFBR Core Recycle Pu/U/U/U	LMFBR A.Blanket Recycle Pu/U/U/U
Average Flux (n/s/cm^2)	3.38 X 10^{15}	9.2 X 10^{14}
Irradiation Period (d)	1097.6	1097.6
Cooling Time (d)	1097.6	1097.6

Table 3 LWR specifications assumed

Reactor Power	GWe	1.18
Thermal Efficiency	%	35
Plant Capacity Factor	%	85
Core HM Inventory	t	86
Anual Spent Fuel Discharge	tHM	24.16
Average Burnup	MWD/t	44000
Specific Power	MW/t	39.8
LoadedFuel Enrichment	%	4.1(^{235}U)
Discharged Fuel Composition (per tHM)		
Pu-238	kg	2.41E-01
Pu-239	kg	5.76E+00
Pu-240	kg	2.51E+00
Pu-241	kg	1.41E+00
Pu-242	kg	6.78E-01
Np-237	kg	6.54E-01
Am-241	kg	2.67E-01
Am-242m	kg	1.01E-03
Am-243	kg	1.50E-01
Cm-242	kg	1.83E-04
Cm-244	kg	4.70E-02
Cm-245	kg	2.33E-03
Cm-246	kg	3.05E-04

isotopes shown in Table 4. These are selected for their significance in weight (mass) and radiotoxicity at relatively long time after irradiation. All the calculations are done in terms of weight, and converted to radiotoxicity as needed. For evaluating the amount of TRUs as radiotoxic materials, isotope's Annual Limitation on Intake (ALI) for the workers is adapted. Specific radiotoxicity of isotope j (SRT_j) is given by the next equation.

$$SRT_j(1/g) = S_j(Bq/g)/ALI_j(Bq) \qquad (1)$$

where S_j is a specific radioactivity of isotope j and ALI_j is Annual Limitation on Intake (ingestion) recommended by the ICRP[7] which are listed in Table 6. Multiplication of SRT_j to the weight of isotope j gives the corresponding radiotoxicity index.

3 Numerical Modeling of the Multi-Recycling System

In order to precisely calculate the cumulative amount of TRUs in a multi-recycling system, numbers of detailed and

Table 4 Transuranium isotopes analyzed

TRU Isotopes	Half Life(y)	Decay Constant(1/y)
Pu-238	8.77E+01	7.90E-03
Pu-239	2.41E+04	2.87E-05
Pu-240	6.56E+03	1.06E-04
Pu-241	1.44E+01	4.83E-02
Pu-242	3.73E+05	1.86E-06
Np-237	2.14E+06	3.24E-07
Am-241	4.32E+02	1.60E-03
Am-242m	1.41E+02	4.92E-03
Am-243	7.37E+03	9.40E-05
Cm-242	4.46E-01	1.56E+00
Cm-244	1.81E+01	3.83E-02
Cm-245	8.50E+03	8.15E-05
Cm-246	4.73E+03	1.47E-04

Table 5 Fissile equivalence in the FBR core

Isotopes	σf [1] barn	σa [1] barn	neutron emission per fission	$\nu\sigma f - \sigma a$ (fissile equivalence)	Relative fissile equivalence
U235	1.87E+00	2.40E+00	2.42	2.12E+00	7.21E-01
U238	5.13E-02	3.45E-01	2.80	-2.01E-01	-6.85E-02
Np-237	3.85E-01	1.82E+00	3.01	-6.66E-01	-2.27E-01
Pu-238	1.18E+00	1.88E+00	2.83	1.47E+00	5.00E-01
Pu-239	1.82E+00	2.29E+00	2.88	2.94E+00	1.00E+00
Pu-240	4.19E-01	9.10E-01	3.14	4.03E-01	1.37E-01
Pu-241	2.44E+00	2.89E+00	2.93	4.28E+00	1.46E+00
Pu-242	3.02E-01	7.19E-01	3.28	2.73E-01	9.27E-02
Am-241	3.40E-01	2.00E+00	3.28	-8.84E-01	-3.01E-01
Am-242m	3.91E+00	4.28E+00	3.16	8.08E+00	2.75E+00
Am-243	2.68E-01	1.28E+00	3.36	-3.78E-01	-1.29E-01
Cm-242	1.95E-01	5.03E-01	3.75	2.29E-01	7.78E-02
Cm-244	4.80E-01	1.28E+00	3.73	5.10E-01	1.73E-01
Cm-245	2.60E+00	2.90E+00	3.83	7.05E+00	2.40E+00
Cm-246	3.15E-01	5.37E-01	3.86	6.78E-01	2.30E-01

1) cited from ORIGEN-II Library : LMFBR-PU/U/U/U:CORE

Table 6 Annual Limitation on Intake for workers

Isotopes	Ingestion ALI(Bq)	Specific Activity Bq/kg	Specific Radiotoxicity 1/kg
Np-237	3.00E+03	2.61E+10	8.69E+06
Pu-238	3.00E+05	6.33E+14	2.11E+09
Pu-239	2.00E+05	2.29E+12	1.15E+07
Pu-240	2.00E+05	8.39E+12	4.20E+07
Pu-241	1.00E+07	3.82E+15	3.82E+08
Pu-242	3.00E+05	1.46E+11	4.88E+05
Am-241	5.00E+04	1.27E+14	2.54E+09
Am242m	5.00E+04	3.88E+14	7.75E+09
Am243	5.00E+04	7.38E+12	1.48E+08
Cm242	2.00E+06	1.23E+17	6.13E+10
Cm-244	9.00E+04	2.99E+15	3.33E+10
Cm-245	5.00E+04	6.35E+12	1.27E+08
Cm-246	5.00E+04	1.14E+13	2.27E+08

Reference [7]

repeated reactor physical calculations have to be performed. This requires specific tool, skill and tremendous calculation time. However, for the purpose to know rough profiles of the future systems in terms of a first-order approximation, especially for relative comparison among different systems, a simpler method is sufficiently convenient and sometimes useful. The method proposed in this paper can provide a rough tendency of the isotope accumulation by rather easy calculations.

In this method, the calculation is simplified by the following approximation.

(1) Same cross section library of ORIGEN-II is used over long period of multi-recycling. This means that the change of the fuel specification and resultant shift of neutron spectrum is neglected in this method.

(2) Same cross section library is used for the core calculation of An-R system and Pu-R system on a recognition that the spectrum change by the addition of other TRUs does not make a large influence. This is basically justified by the fact that the relative amount of the TRU added are quite smaller than Pu isotopes which have similar nuclear properties to TRUs.

3.1 Method to Express Multi-Recycling

The method proposed in this paper expresses the effect of multi-recycling of TRUs by combining the results of repeated irradiation calculations of three different starting materials. The concept of this method is indicated in Fig. 1. In this model, initially loaded TRU elements to the fast reactor are hypothetically considered to have life-through multiple-irradiation over consecutive irradiation cycles. For example, initially loaded plutonium having specific isotopic composition of the spent fuels from LWRs is irradiated by the first irradiation cycle, then after 3 years cooling, the residual plutonium having changed isotopic composition is reloaded and irradiated. And then, this manner is repeated till the cumulative time of irradiation cycles reaches the objective period of the analysis. The decrease of isotope j along these

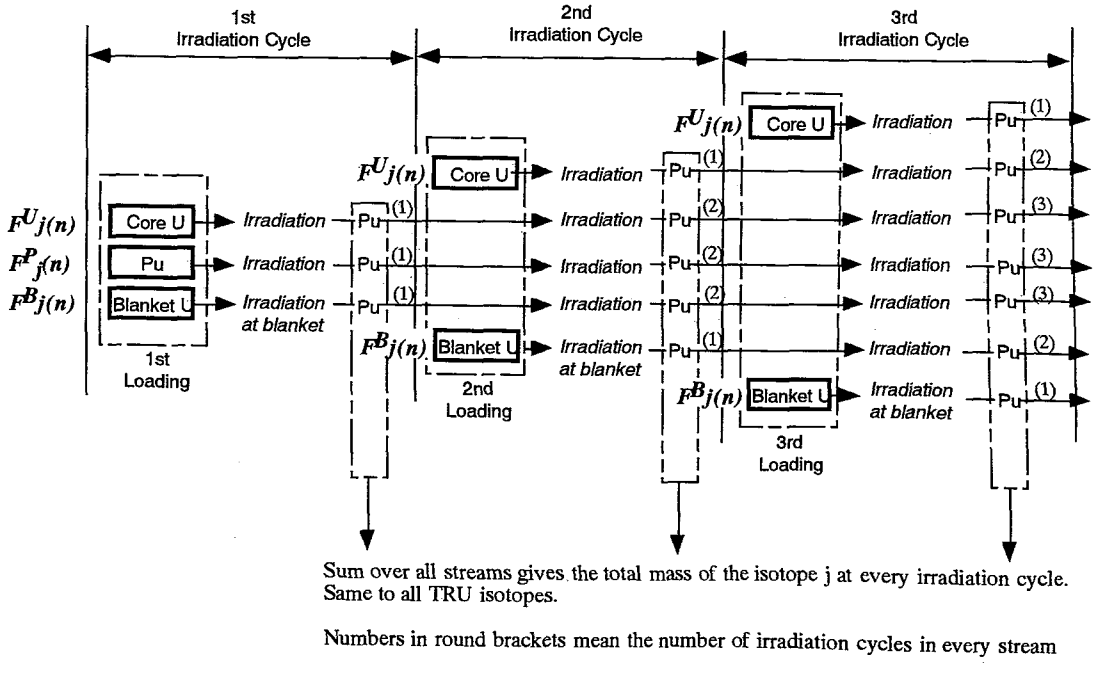


Fig.1 Concept of the calculation method for TRU isotopes

repeated irradiations is denominated as "multi-irradiation function (MIF hereafter)". MIF is defined for every isotope by the repeated irradiation calculation with one run of ORIGEN-II. MIF is a row matrix consisting of isotope's inventories at every consecutive irradiation number. Therefore, MIF gives the life-through trend of the decrease of isotopes throughout the repeated irradiations.

MIFs for isotopes are obtained for three different streams; (1) initially loaded TRUs supplied from LWRs (e.g. plutonium (Pu-R), all TRUs (An-R) supplied from LWRs), (2) core-loaded uranium as matrix of mixed oxide fuel, and (3) uranium loaded in axial and radial blanket. These MIFs for isotope j are denominated F^P_j, F^U_j, and F^B_j, respectively, and are standardized to appropriate unit amount. The first irradiation to obtain F^B_j is done by the cross section library for blanket (axial blanket), but after the second irradiations, the cross section library for the core is used because the TRUs are recycled to the core. Both F^P_j and F^U_j, are obtained by the cross section library for the core.

Three MIFs start from the very beginning of the fast reactor system operation. However, at every new irradiation cycle, because blanket uranium and core-matrix uranium are newly loaded and irradiated, two additional new F^P_j and F^U_j join. Thereby, at the n-th irradiation cycle, the total inventory of isotope j can be expressed as the sum of n-th entry of F^P_j, 1st to n-th entry of F^P_j and F^U_j. It should be noted that coefficients representing mass of the loaded materials are multiplied to all MIFs because they are standardized to a unit amount of the starting materials.The coefficient for F^U_j represents the mass of uranium loaded with plutonium, and the coefficient for F^B_j represents the mass of blanket. These amounts of the loaded uranium relate to the breeding ratios of plutonium. The breeding ratio in this paper is defined on the fissile equivalence which is defined in the next section. As the irradiation cycles proceed, the isotopic composition of plutonium isotopes changes, hence, the reactor-physical reactivity of the system changes. In general cases, it increases by the breeding of plutonium, therefore, the effect of the capacity expansion should be included too. In an actual fast reactor system, it can not continuously grow because the gain of fuel material by the breeding has to be stocked till minimum necessary amount for next unit is attained. However in this model, it is assumed that the capacity of the system is large enough to enable its continuous growth. Thus, the capacity of the system is considered to be proportional to the reactorphysical reactivity possessed in a system. The index for the reactorphysical reactivity is given by "fissile equivalence" which is defined in the next section.

The composition of the starting materials of F^P_j for Pu-R and An-R systems is summarized in Table 7. The starting materials for F^P_j and F^U_j are commonly tail-uranium having 0.2 % enrichment.

Table 7 Composition of loaded TRUs[1)]

Isotopes	Pu-R	An-R
Pu-238	22.7	22.7
Pu-239	543.4	543.4
Pu-240	236.8	236.8
Pu-241	133.1	133.1
Pu-242	64	64
Np-237	0	61.74
Am-241	0	25.12
Am-242m	0	0.10
Am-243	0	14.1
Cm-242	0	0.02
Cm-244	0	4.43
Cm-245	0	0.22
Cm-246	0	0.03
total	1000	1105.75

unit: kg

1)Composition of the starting material for multi-irradiation calcularion for F^P_j

3.2 Numerical Treatment

The sum of the MIFs for isotope j by the manner of Fig.1 is denominated as "growth function (GF hereafter)" which is a row matrix giving the change of the inventory of isotopes along the irradiation cycles. MIF is also defined for the fissile equivalence in the three streams ($F^P_f(n)$, $F^U_f(n)$, $F^B_f(n)$). The fissile equivalence of isotope j is defined by equation (2)

$$\text{fissile equivalence} = \nu\sigma_f - \sigma_a \qquad (2)$$

where, σ_f is a fission cross section, ν is an average neutron emissions per fission, and σ_a is a neutron capture cross section. Fissile equivalence is a factor to convert the atom density to the value as fissile materials, but in this paper as an approximation, this factor is applied to isotope's weights. Relative fissile equivalence (RFE) is defined by their ratio to that of Pu-239. MIF for the fissile equivalence is defined by the ratio of them equations (3) to (5) as the sum of the fissile equivalence in the corresponding streams (Table 5). GF for the fissile equivalence, $g(n)$) is defined by the equation (6) which gives the total fissile equivalence of the system starting from capacity of 1 GWe. Dividing $g(n)$ by the C^P_0 which is the fissile equivalence necessary for the reference core (1 GWe: 3306.11 kg) gives the capacity of the system at n-th cycle. C^P_0 3306.11 kg is a total relative fissile equivalence of Pu (Pu-239 standard) in the core which is under equilibrated burning cycles. $g_j(n)$ which is a growth function of the inventory of isotope j is defined by equation (7), and it gives the cumulative amount of isotope j in the system which started from capacity of 1 GWe.

$$F_f^P(n) = \sum RFE_j F_j^P(n) \quad (3)$$

$$F_f^B(n) = \sum RFE_j F_j^B(n) \quad (4)$$

$$F_f^U(n) = \sum RFE_j F_j^U(n) \quad (5)$$

$$g(n) = \frac{C_0^U}{C_0^P}\sum_{i=0}^{i=n-1} g(i)F_f^U(n-i) + \frac{C_0^B}{C_0^P}\sum_{i=0}^{i=n-1} g(i)F_f^B(n-i) + C_0^P F_f^P(n) \quad (6)$$

$$g_j(n) = \frac{C_0^U}{C_0^P}\sum_{i=0}^{i=n-1} g(i)F_j^U(n-i) + \frac{C_0^B}{C_0^P}\sum_{i=0}^{i=n-1} g(i)F_j^B(n-i) + C_0^P F_j^P(n) \quad (7)$$

Nomenclature is the followings.

$F^P{}_j(n)$, $F^U{}_j(n)$, $F^B{}_j(n)$):
MIF for the amount of isotope j starting from core-loaded plutonium, core-loaded uranium, and blanket-loaded uranium. Standardized to initially loaded unit fissile equivalence of plutonium (kg/kg), to unit amount of core-uranium (kg/kg), and to unit amount of blanket-uranium (kg/kg), respectively.

$F^{P(n)}_f$, $F^{U(n)}_f$, $F^{B(n)}_f$:
MIF for fissile equivalence starting from core-loaded plutonium, core-loaded uranium, and blanket-loaded uranium. Standardized to initially loaded unit fissile equivalence of plutonium (kg/kg), to unit amount of core-uranium (kg/kg), and to unit amount of blanket-uranium (kg/kg), respectively.

$g(n)$:Growth function of the fissile equivalence at n-th cycle. Standardized to the initial 1GWe system.

$g_j(n)$:Growth function of the amount of isotope j at n-th cycle. Standardized to the initial 1GWe system.

$C^P{}_0$:Fissile equivalence of plutonium firstly loaded to the reference core (kg)

$C^U{}_0$:Amount of core uranium loaded to the reference FBR core (kg)

$C^B{}_0$:Amount of blanket uranium loaded to the reference FBR (kg)

RFE_j:Relative fissile equivalence of isotope j

The length of one irradiation cycle is the sum of residence time in the core and that in the fuel cycle facility. In this paper, because the residence time in the fuel cycle facility is assumed to be same as the core residence time. Thus, n-th irradiation cycle corresponds to the double of the core residence time (1097.6 d). Thereby, growth function $g(n)$ and $g_j(n)$ can be expressed as functions of time t. The growth function $g_j(t)$ gives the amount of isotope j resides in a core initially started from 1 GWe fast reactor. Because the total inventory of isotope j in the system is the sum of the core inventory and fuel cycle inventory, $2g_j(n)$ gives the total inventory of the fuel cycle system. $C^P{}_0$, $C^U{}_0$ and $C^B{}_0$ for the reference 1 GWe FBR is 3306.11 kg, 18781.6 kg and 32100 kg, respectively.

3.3 Formula for Various Multi-Recycling System

The MIFs for Pu-R system is obtained by the repetition calculation of ORIGEN-II in which plutonium isotopes are recovered and multi-irradiated. The discharge of Pu isotopes at every irradiation directly gives their inventory in the system. But, the discharge of Np, Am, and Cm at every irradiation cycle only gives the amount generated at every irradiation cycle, thus does not give the cumulative inventory. Therefore, their cumulative inventory was obtained by accumulating the discharge of every irradiation cycle and used as the MIF for Pu-R system. Adequate decay-corrections were performed for these out-core inventory. For the An-R system, MIFs were directly obtained by repeated irradiation of all TRUs.

In the An-R system, Np, Am, and Cm supplied from LWR spent fuels are loaded to the starting fuel together with plutonium. In order to compare the two different systems with same initial condition, same amount of Np, Am, and Cm from LWR is added to the Pu-R system too as out-core inventory to which newly generated TRUs by the operation of FBR are added. Decay correction is performed for this initial amount. This manner is illustrated in Fig.2. In the Pu-R system, Pu-238 and Pu-240 are produced outside the core by the decay of Cm isotopes. However, this is neglected because they are as low as 1 % of their core inventory.

The resultant $g_j(t)$ are standardized to initially loaded 1 kg of fissile equivalence and plotted in Fig.3 for three different breeding ratios (1.05, 1.10, and 1.20) of the system. Growth functions standardized to initial 1 kg fissile equivalence is denominated as $g'(t)$ and $g'_j(t)$ .For accomplishing the numerical modeling, $g'(t)$ and $g'_j(t)$ were fit to appropriate function forms. Least square fitting to appropriate function forms are performed on the $g'(t)$ and $g'_j(t)$ determined by the above procedures. Because Pu-239 and Pu-240 simply increase along the time, they can successfully be fit to a single exponential form. On the other hand, because Pu-238, Pu-241 and Pu-242 decrease at the early stage and then turns to increase, combination of two exponential functions showed better fitting results. For the other TRUs whose behavior is much more complex, combination of three exponentials are applied. Some of the TRU isotopes were successfully fit to single or double exponential form. The lines indicated in Fig.3 show the fit functions.

General function form is given by equation (8) with the coefficients listed in the appendix tables.

$$g'_j(t) = Ae^{at} + Be^{bt} + Ce^{ct} + D \quad (8)$$

Because the least square fittings given in appendix tables are performed with heavier weights on the time longer than 100 years, coefficients satisfactorily represents the isotopic

composition only beyond 100 years. For earlier period, especially before 75 years, these functions do not always give good approximation. For the assessment over a period longer than 100 years, these formula are quite useful for obtaining cumulative amount of the isotopes. These formula will enable us to perform mathematical treatment or modeling of more complex systems consisting of fast reactor systems. Their simple function form as linear combination of exponentials enables easy solution of integral and differential equations involving these as components. The $g_j(t)$ of Pu for Pu-R and An-R systems are shown in Figs.3(1) and (2), and that of Np, Am and Cm isotopes for Pu-R and An-R systems are shown in Figs,3(3) and (4), respectively.

Multiplying 3306.11 to $g'_j(t)$ gives the inventory in the core starting from 1 GWe capacity, and twice of this gives the total inventory in the entire fuel cycle (core plus fuel cycle facilities).

4 Results and Discussion

The characteristics of the accumulation of TRUs in different fast reactor systems are discussed in this section according to the results of the above calculation. The analysis was done over 200 years from the start of the systems. The result includes the effect of the growth of the systems by breeding. The capacity of the system at 200 years is about 3.7, 14.5 and 191.4-fold of the initial capacity by the breeding ratio 1.05, 1.10 and 1.20, respectively.

4.1 Plutonium isotopes

As can be seen in every case of Figs.3(1) and (2), the behavior of plutonium isotopes are separated into two different regions of period, first region before 75 years and second region beyond 75 years. In the first region which should be denominated as transient period, a rapid change of the isotopic composition from the initial level proceeds asymptotically to the equilibrium composition. In the second region, because the generation and consumption of isotopes are almost balanced, isotopic composition attains a kind of equilibrium and it becomes to show simple increase governed by the breeding of the system.

In both Pu-R system and An-R system, the rapid decreases of Pu-241 and Pu-242 are noticeable, which suggests that the equilibrium composition of these which are determined by the neutron spectrum of FBR is quite lower than that of LWR. This is attributed to the higher production of Pu-241 in the LWR owing to the high thermal neutron capture of Pu-240. The decrease of Pu-242 owes to the decrease of Pu-241. This is the reason why Pu-242 shows delayed minimum compared with the Pu-241. The trend of Pu-238 in An-R system is noticeable. Contrary to the rather low content of Pu-238 in Pu-R system, it is drastically increased in An-R system. This can be attributed to both reactions of $^{237}Np(n, \gamma)\ ^{238}Np(\beta^-)^{238}$ Pu and $^{241}Am(n, \gamma)^{242}Am(\beta^-)^{242}Cm(\alpha)\ ^{238}$Pu.

These trends are obviously shown in the change of the isotopic percentage of the plutonium isotopes as illustrated in the Fig.4 of the case of breeding ratio 1.05. It can be seen that the isotopic composition converges within ca. 75 years in both systems and that quite high content of Pu-238 is shown in An-R system. The equilibrated isotopic composition of the plutonium in various systems is summarized in the Table 8. Higher content of Pu-239 than that of LWR spent fuel is obvious. The specific fissile equivalence of the equilibrated system is shown in the Table 9. The initially loaded plutonium which is supplied from the LWR spent fuel has specific fissile equivalence of 78.7 kg/100kg HM. 19.38 out of 78.7 is of Pu-241. The fissile equivalence under equilibrium of Pu-R are 74.3, 76.7 and 80.3 in the systems having breeding ratio 1.05, 1.10 and 1.20, respectively. In the fast reactor systems, the contribution of Pu-241 is as small as 4. The decrease of specific fissile equivalence by recycling other TRUs in An-R is as small as 0.3, which makes us expect that loading minor actinides to the MOX core does not lead to serious loss of the reactivity of plutonium.

4.2 Np, Am and Cm

As can be seen in every isotope of Fig.3(3) of the case of Pu-R systems, TRUs gradually increase along the time except for Cm-244 which decreases first then show a plateau due to its short life. In Pu-R system, there is no large difference in the patterns of their increase between the different breeding ratios, except that Np-237 slightly increases in higher breeding ratio (1.20). This can be attributed to the loading of large amount of U-238 as blanket which causes the increase of Np-237 by (n,2n) reaction.The largest one in mass of TRUs is Am-241which exceeds the amount of minor plutonium isotopes like Pu-241, 242, 238. The resultant cumulative amount of Am-241 over 200 years is ca. 0.7 kg/kg fissile at breeding ratio 1.05, which corresponds to 2314 kg in a system starting from 1 GWe. Am-242m is negligible in terms of its weight compared with other americium isotopes but it should

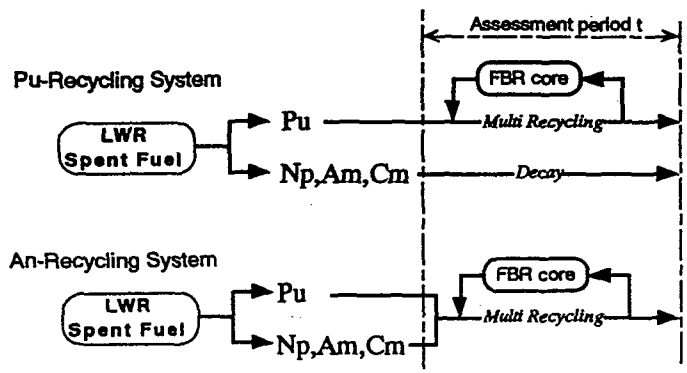
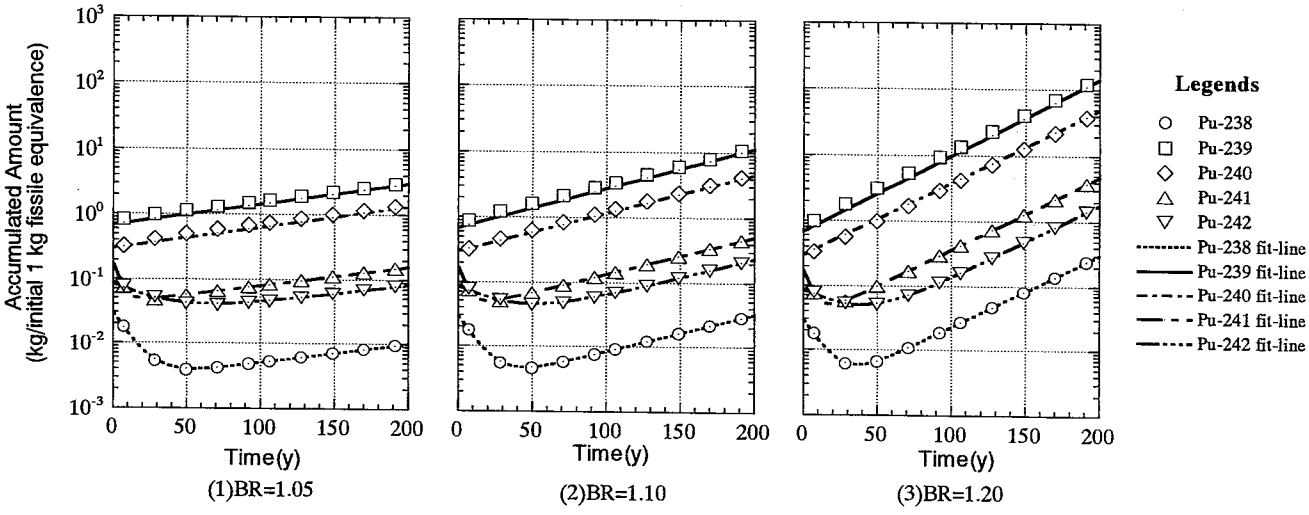


Fig.2 Pu-Recycling and An-Recycling system

be carefully watched for its prominence as a most fissile nucleus, accumulates up to ca. 20kg during the period of 200 years.

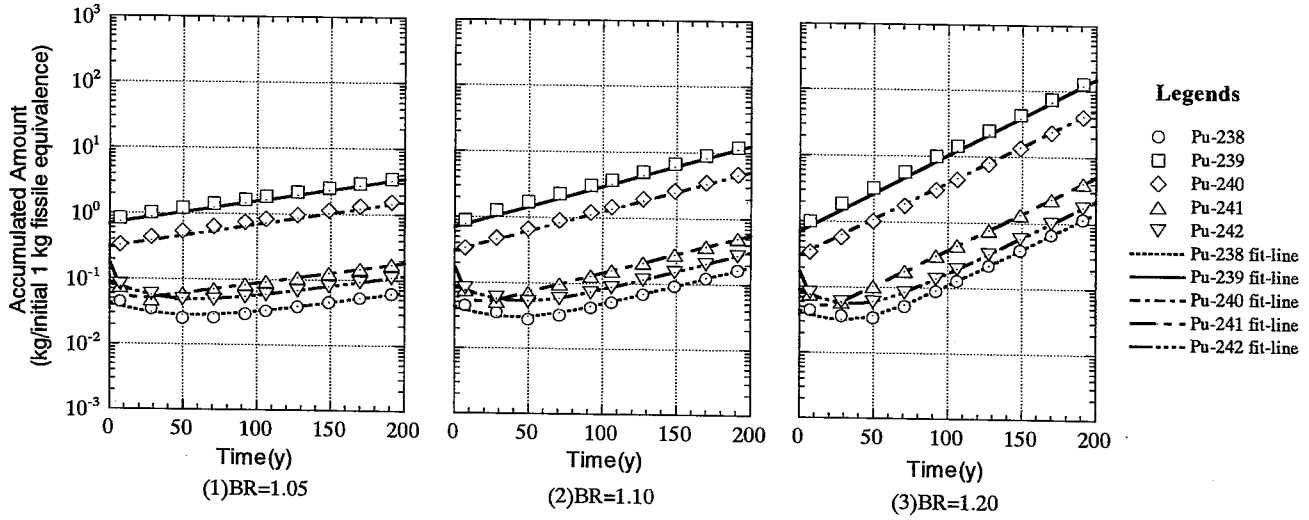
In the case of An-R system, the trend can also be separated to first transient period and second equilibrated period. The end of the first period is about 75 years from the start. In the first region, Am-241 and Np-237 whose conversion rate is quite high rapidly decrease and attain equilibrated level. On the contrary, heavier curium isotopes (Cm-245, Cm-246) increase owing to the loading of their fertile isotopes Cm-244 and Am-243. As the result, in the An-R system, the equilibrated amount of Cm-245 and Cm-246 is much higher than the Pu-R system. The major three isotopes Am-241, Np-237 and Am-243 are reduced to about one tenth of these in Pu-R system, which suggests actinide-recycling is effective for the reduction of the mass of these isotopes. On the other hand, the relative weight of the Cm-244, 245 and 246 are quite increased in the An-R than Pu-R. There is no large difference among the different breeding ratios.



BR : Breeding Ratio

The ordinate is standarized to 1 kg of fissile equivalence at the start of the system. Multiplying 3306.11 kg(fissile equivalence necessary for starting up 1 GWe FBR system) gives cumulative amount of isotopes in the system starting from 1GWe.

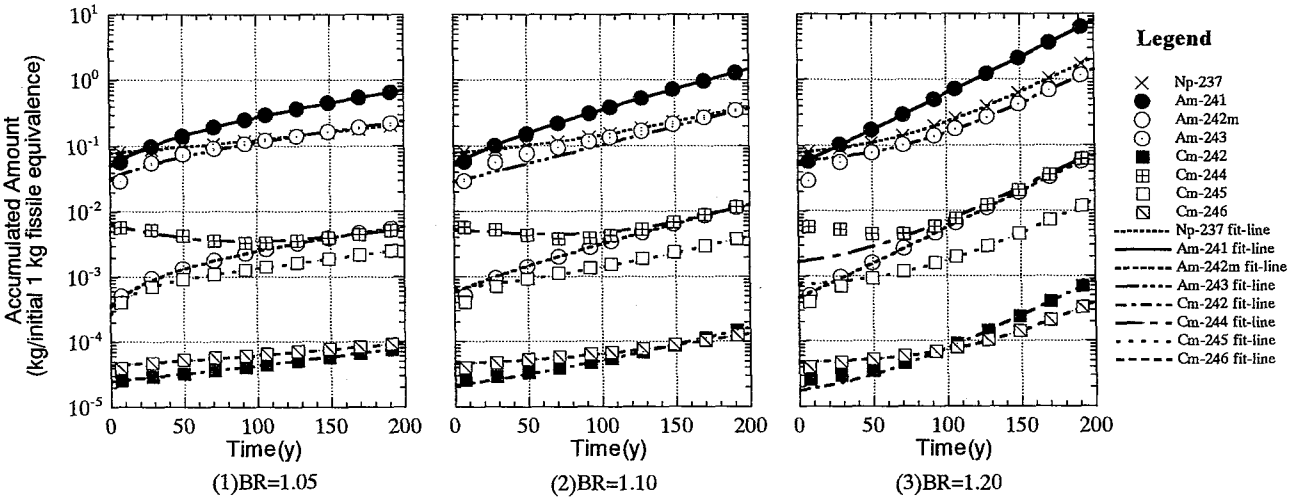
Fig.3(1) Accumulation of Pu isotopes in the Pu-Recycling system



BR : Breeding Ratio

The ordinate is standarized to 1 kg of fissile equivalence at the start of the system. Multiplying 3306.11 kg(fissile equivalence necessary for starting up 1 GWe FBR system) gives cumulative amount of isotopes in the system starting from 1GWe.

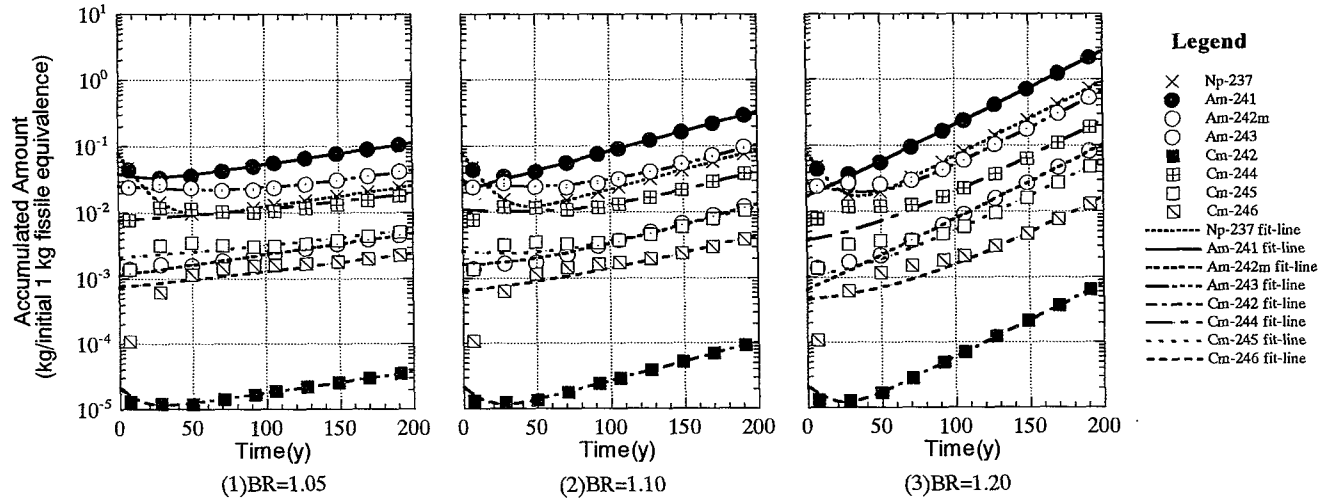
Fig.3(2) Accumulation of Pu isotopes in the An-Recycling system



BR : Breeding Ratio

The ordinate is standarized to 1 kg of fissile equivalence at the start of the system. Multiplying 3306.11 kg(fissile equivalence necessary for starting up 1 GWe FBR system) gives cumulative amount of isotopes in the system starting from 1GWe.

Fig.3(3) Accumulation of Np, Am and Cm isotopes in the Pu-Recycling system



BR : Breeding Ratio

The ordinate is standarized to 1 kg of fissile equivalence at the start of the system. Multiplying 3306.11 kg(fissile equivalence necessary for starting up 1 GWe FBR system) gives cumulative amount of isotopes in the system starting from 1GWe.

Fig.3(4) Accumulation of Np, Am and Cm isotopes in the An-Recycling system

The difference of the accumulation rate of TRUs between the different systems is an interesting point. The growth of the cumulative amount of TRUs is generally given by the function form of equation (8). However, in the time region beyond 100 years, because 2nd, 3rd and 4th term of (8) becomes relatively smaller than the 1st term, the amount can be approximated by only the first term as equation (9). The growth rate of the cumulative amount is given by the differential of (9) as equation (10). Because the exponential term of the equation (10) represents the effect of expansion of the system by the breeding, thus, the growth rate standardized to initial capacity can be simply given by the term Aa in equation (10), and is constant along the time. The ratio of the growth rate of two systems having a pair of coefficient A and a, and A' and a', is then given by $Aa/A'a'$. Table 10 shows the comparison of Aa between An-R system and Pu-R system using the obtained A and a for BR=1.05 which are shown in Appendix Table 2.

$$g'_j(t) \approx Ae^{at} \qquad (9)$$

$$\frac{dg'_j(t)}{dt} = Aae^{at} \qquad (10)$$

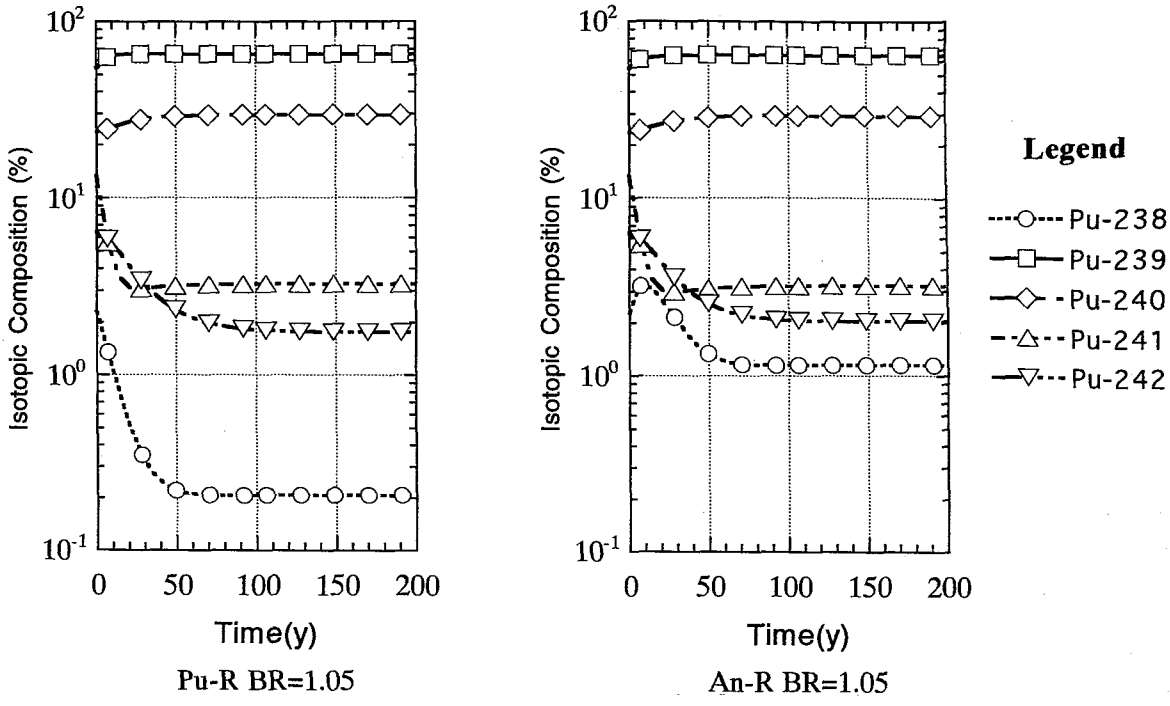


Fig.4 Change of the isotopic composition of Pu in the Pu-R and An-R system

Table 8 Isotopic composition of plutonium converged in various systems

System[1)]	Pu-238	Pu-239	Pu-240	Pu-241	Pu-242
An-R 1.05	1.2	64.4	29.2	3.2	2.0
An-R 1.10	0.9	67.9	26.7	2.8	1.6
An-R 1.20	0.7	73.2	22.9	2.3	1.0
Pu-R 1.05	0.2	65.2	29.6	3.3	1.7
Pu-R 1.10	0.2	68.6	27.0	2.9	1.3
Pu-R 1.20	0.1	73.7	23.0	2.3	0.9
LWR Spent Fuel 3y[2)]	2.3	54.3	23.7	13.3	6.4

unit : %

1) numbers mean breeding ratio
2) Spent fuel of LWR 3 years cooled

Table 9 Comparison of the fissile equivalence converged in various systems

System[1)]	Pu-238	Pu-239	Pu-240	Pu-241	Pu-242	total
An-R 1.05	0.58	64.43	3.99	4.69	0.19	73.88
An-R 1.10	0.47	67.95	3.66	4.15	0.14	76.36
An-R 1.20	0.33	73.21	3.13	3.33	0.09	80.08
Pu-R 1.05	0.10	65.19	4.05	4.77	0.16	74.28
Pu-R 1.10	0.09	68.60	3.69	4.20	0.12	76.71
Pu-R 1.20	0.07	73.69	3.15	3.35	0.08	80.34
LWR Spent Fuel 3y[2)]	1.13	54.34	3.24	19.38	0.59	78.70

unit : kg/100 kg HM

1) numbers mean breeding ratio
2) Spent fuel of LWR 3 years cooled

Table 10 shows that the growth rates of Np-237, Am-241, Am-243, Cm-242 in An-R system are ca. 0.13-fold to 0.27-fold of those in Pu-R system, which suggests that these

Table 10 Comparison of the growth rate of TRUs

Isotopes	Aa (An-R)	$A'a'$ (Pu-R)	$Aa/A'a'$ [1)]
Np-237	4.42E-05	3.28E-04	0.13
Am-241	1.92E-04	1.58E-03	0.12
Am-242m	7.22E-06	1.12E-05	0.65
Am-243	4.74E-05	1.73E-04	0.27
Cm-242	3.15E-08	1.41E-07	0.22
Cm-244	2.42E-05	9.26E-06	2.61
Cm-245	7.24E-06	4.67E-06	1.55
Cm-246	3.70E-06	1.26E-07	29.49

1)An-R/Pu-R

production rates in the An-R system are around 0.2 of Pu-R system. On the other hand, production rates of curium isotopes are quite accelerated, especially in the case of Cm-246.

4.3 Cumulative Radiotoxicity of the TRUs

In order to assess the short-term risk as radiological impact owing to the unexpected release of the accumulated TRUs by the generation of TRU-wastes or accidents, the radiotoxicity of TRUs will be an important index for quantitative evaluation. Applying the conversion coefficients listed in the Table 6 to the $g_j(t)$ above, we can easily get the relative level of radiotoxicity. Fig.5 compares the two systems having breeding ratio 1.05 in terms of radiotoxicity index. In the Pu-R system, when compared in terms of the weight, Pu-239 and Pu-240 account for the major part of the TRUs, but, when compared in terms of the radiotoxicity index, Am-241 accounts for the greatest part of the total radiotoxicity. Cm-244 which is negligible in the weight scale

is the second largest in the radiotoxicity scale.

In the radiotoxicity scale, by in An-R system, Am-241 becomes lower than Cm-244 owing to both the reduction of Am-241 and the increase of Cm-244. The increase of Cm-244, 245 and 246 in radiotoxicity by actinide recycling is obvious, which is attributable to their accelerated formation by the irradiation of Am-243 as their fertiles. Pu-238 as the most minor plutonium in the weight scale in Pu-R system becomes the most important one in the radiotoxicity scale in An-R.

4.4 Comparison of Pu-R and An-R system

In Fig.6, results of both systems over 200 years are compared in weight and radiotoxicity. In the weight scale, the drastic reduction of the total mass of Np, Am and Cm by the actinide recycling is obvious due to the reduction of Am-241, Am-243 and Np-237. The relative increase of curium isotopes is remarkable, but it does not make large impact to the reduction of the total mass of TRUs. Thus, by actinide recycling, the total mass of minor actinide isotopes is reduced to about one tenth of that in Pu-R system at 200 years. This effect is not powerful enough, if we see the actinide-recycling FBR system as a transmutation system. However, in a sense that as low production of TRUs as possible is favored for future FBR system, this may be quite satisfactory.

The reduction of the radiotoxicity of Am by actinide recycling is remarkable, but it is compensated by the increase of Cm. Consequently, the reduction effect on non-plutonium isotopes in radiotoxicity is as small as 50 % of the level of Pu-R system, which is not satisfactorily high. If we focus on the life of radiotoxicity, the increase of Cm-244 whose life is short (18 years) may be considered less important, thus effect of the reduction of Am-241 will be much appreciated. The relative reductions of Am-243 and Np-237 in radiotoxicity are large, but their significance in the total radiotoxicity is relatively low.

If we focus on the all radiotoxicity contained by the system including plutonium isotopes and other TRUs, the balance between plutonium and others is interesting. In the Pu-R system, total radiotoxicity of Np, Am and Cm is ca. ten times as much as that of plutonium isotopes. However, in the An-R system, because the radiotoxicity of Pu increase by the formation of Pu-238, the discrepancy between Pu and others becomes smaller than in the Pu-R system. Therefore, actinide recycling can be characterized by "more radiotoxicity of Pu with less radiotoxicity of non-plutonium TRUs", which should be compared with the "less radiotoxicity of Pu with more radiotoxicity of non-plutonium TRUs" of Pu-R system. This is obvious in the summary shown as Table 11. For evaluating the potential risks of two systems, it should be noted that the more toxic non-plutonium in Pu-R system are kept in a deep geologic site, in contrast, all the TRUs are in the cycle in An-R system.

The importance of the recycling of Np-237 and Am-243 is rather less in radiotoxicity reduction compared with that of Am-241. However, regardless of the meaning as radiotoxicity, we have to see their property as burnable materials. Fig.7 graphically shows the relative fissile equivalence of the TRUs in which both Np-237 and Am-243 have slightly negative numbers. This means that these are as less-burnable as U-238. However, by n-capture, Np-237 becomes Pu-238 which is a

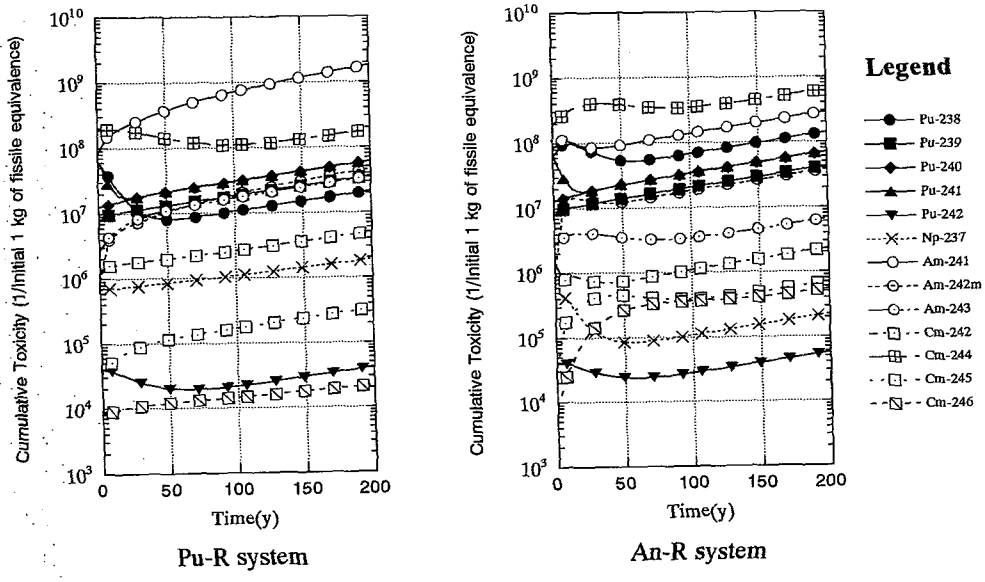


Fig.5 Accumulation of radiotoxicity in the systems with breeding ratio 1.05

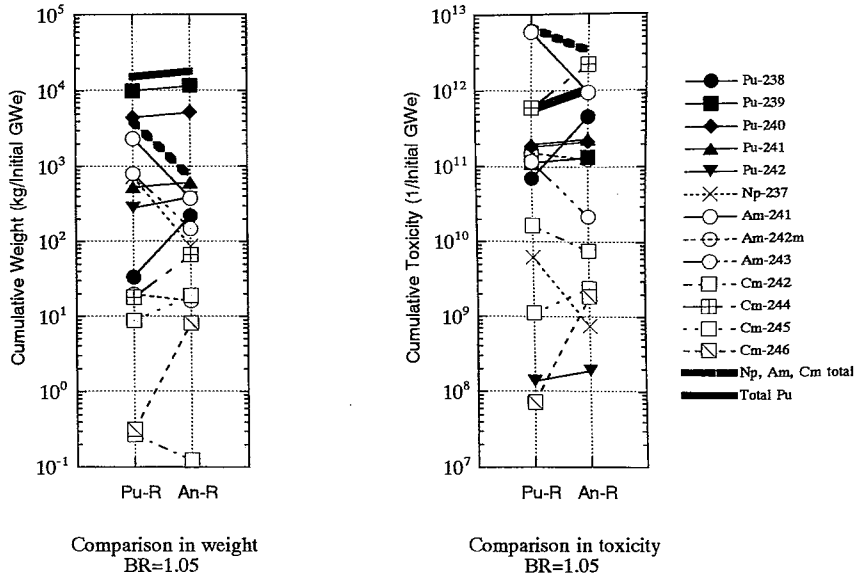


Fig.6 Comparison of Pu-R and An-R system in their cumulative mass and toxicity at 200 years

Table 11 Result of the actinide-recycling[1)]

	Weight	Radiotoxicity
Np,Am,Cm Total	18%	49%
Total Pu	118%	183%
Total TRUs	98%	59%

1)Numbers in this table are relative amounts of the elements to those of Pu-R system. Comparison at 200 years.

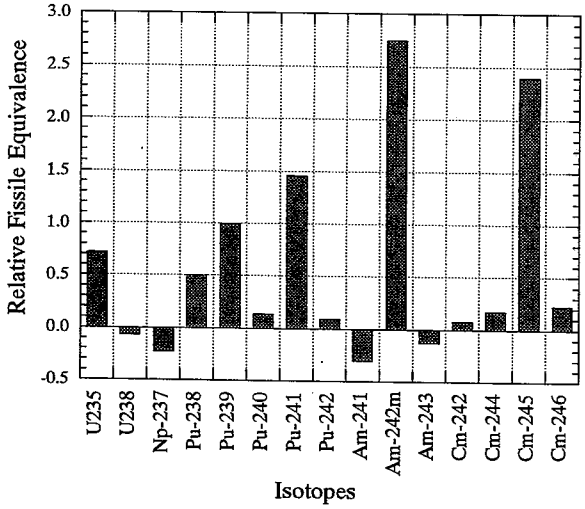


Fig.7 Comparison of relative fissile equivalence of TRU nuclides

fissionable material. Therefore, Np-237 actually can be recognized as a fertile material which has a similar meaning as the relation between U-238 and Pu-239. Loading Np-237 raises up the equilibrium amount of Pu-238 causing increased specific radiotoxicity of plutonium. But, if its handling in the fuel cycle facilities is technologically acceptable, recycling Np-237 is quite a natural choice for utilizing out the manmade materials. Am-243 has similar position as Np-237 because its n-capture product Cm-244 and Cm-245 are burnable by rapid decay to fissionable Pu-238 and very high fissionability.

5 Conclusion

A new easy and simple numerical method based on ORIGEN-II to calculate cumulative production of the TRUs in multi-recycling fast reactor system was proposed. The cumulative amount of isotopes of Pu, Np, Am and Cm were given as simple numerical functions. By using these functions, rough profile of the amount and composition of TRU isotopes can be obtained. The time dependent characteristics of the accumulation of isotopes were obtained both in terms of weight and radiotoxicity. The accumulation of Am-241 in Pu-R system is quite large, especially in terms of radiotoxicity. Regarding with the effect of the actinide recycling, it was found that (1) the total weight of the Np, Am and Cm can be reduced up to ca. one tenth of Pu-R system, (2) the reduction of total radiotoxicity of Np, Am and Cm is as small as 50 % of the Pu-R system.

The accumulation of man-made radiotoxic materials in the closed cycle will have to be carefully treated including the possibility of the disposal or transmutation. In addition, the loss of the isotopes to the waste from the closed cycle during its long term operation will have to be well assessed for the

environmental effect. Utilizing the above results, we can roughly estimate the level and characteristics of the source term possessed in the multi-recycling system.

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Appendix Table 1 Coefficients for the fit functions (Pu)[1),2)]

	BR	Isotope	*A*	*a*	*B*	*b*
Pu-R	1.05	Fis.Eq	1.00E+00	6.53E-03	0.00E+00	0.00E+00
		Pu-238	2.58E-03	6.80E-03	2.65E-02	-8.32E-02
		Pu-239	6.91E-01	7.37E-03	0.00E+00	0.00E+00
		Pu-240	3.01E-01	7.51E-03	0.00E+00	0.00E+00
		Pu-241	4.09E-02	6.80E-03	1.28E-01	-2.06E-01
		Pu-242	2.17E-02	6.81E-03	6.50E-02	-3.37E-02
	1.10	Fis.Eq	1.00E+00	1.31E-02	0.00E+00	0.00E+00
		Pu-238	2.24E-03	1.33E-02	2.68E-02	-8.25E-02
		Pu-239	6.91E-01	1.40E-02	0.00E+00	0.00E+00
		Pu-240	3.01E-01	1.36E-02	0.00E+00	0.00E+00
		Pu-241	3.56E-02	1.33E-02	1.34E-01	-1.92E-01
		Pu-242	1.65E-02	1.33E-02	7.06E-02	-3.32E-02
	1.20	Fis.Eq	1.00E+00	2.56E-02	0.00E+00	0.00E+00
		Pu-238	1.76E-03	2.58E-02	2.73E-02	-8.12E-02
		Pu-239	6.91E-01	2.66E-02	0.00E+00	0.00E+00
		Pu-240	3.01E-01	2.55E-02	0.00E+00	0.00E+00
		Pu-241	2.76E-02	2.58E-02	1.42E-01	-1.74E-01
		Pu-242	1.04E-02	2.58E-02	7.73E-02	-3.22E-02
An-R	1.05	Fis.Eq	1.00E+00	7.28E-03	0.00E+00	0.00E+00
		Pu-238	1.51E-02	7.39E-03	2.80E-02	-2.78E-02
		Pu-239	7.10E-01	8.01E-03	0.00E+00	0.00E+00
		Pu-240	3.09E-01	8.14E-03	0.00E+00	0.00E+00
		Pu-241	4.26E-02	7.36E-03	1.31E-01	-2.11E-01
		Pu-242	2.68E-02	7.36E-03	6.29E-02	-3.33E-02
	1.10	Fis.Eq	1.00E+00	1.36E-02	0.00E+00	0.00E+00
		Pu-238	1.22E-02	1.37E-02	3.11E-02	-2.88E-02
		Pu-239	7.10E-01	1.44E-02	0.00E+00	0.00E+00
		Pu-240	3.09E-01	1.41E-02	0.00E+00	0.00E+00
		Pu-241	3.72E-02	1.37E-02	1.37E-01	-1.97E-01
		Pu-242	2.03E-02	1.37E-02	7.00E-02	-3.26E-02
	1.20	Fis.Eq	1.00E+00	2.59E-02	0.00E+00	0.00E+00
		Pu-238	8.21E-03	2.59E-02	3.55E-02	-2.89E-02
		Pu-239	7.10E-01	2.68E-02	0.00E+00	0.00E+00
		Pu-240	3.09E-01	2.57E-02	0.00E+00	0.00E+00
		Pu-241	2.88E-02	2.59E-02	1.45E-01	-1.77E-01
		Pu-242	1.25E-02	2.59E-02	7.86E-02	-3.13E-02

1) **Function Form:** $Aexp(a\,t)+Bexp(b\,t)$ where t is time (year).

2) Numerical result gives cumulative amount of isotopes(kg) per 1 kg of initial fissile equivalence at the start of the system. Multiply 3306.11 for obtaining amounts per initial 1 GWe system.

Appendix Table 2 Coefficients for the fit functions (Np, Am and Cm)[1),2)]

System	BR	Isotope	*A*	*a*	*B*	*b*	*C*	*c*	*D*
Pu-R	1.05	Np-237	4.84E-02	6.79E-03	0.00E+00	0.00E+00	0.00E+00	0.00E+00	3.04E-02
		Am-241	2.40E-01	6.58E-03	0.00E+00	0.00E+00	0.00E+00	0.00E+00	-1.88E-01
		Am-242m	1.64E-03	6.81E-03	-1.28E-03	-5.56E-03	0.00E+00	0.00E+00	0.00E+00
		Am-243	2.35E-02	7.37E-03	-9.34E-02	-4.03E-03	2.35E-02	7.37E-03	7.86E-02
		Cm-242	2.09E-05	6.76E-03	5.19E-02	-2.89E-07	0.00E+00	0.00E+00	-5.19E-02
		Cm-244	1.34E-03	6.91E-03	5.12E-03	-1.75E-02	0.00E+00	0.00E+00	0.00E+00
		Cm-245	6.91E-04	6.76E-03	-4.23E-04	-4.21E-02	0.00E+00	0.00E+00	0.00E+00
		Cm-246	1.91E-05	6.56E-03	0.00E+00	0.00E+00	0.00E+00	0.00E+00	2.53E-05
	1.10	Np-237	2.44E-02	1.33E-02	0.00E+00	0.00E+00	0.00E+00	0.00E+00	5.43E-02
		Am-241	1.07E-01	1.33E-02	0.00E+00	0.00E+00	0.00E+00	0.00E+00	-5.31E-02
		Am-242m	9.32E-04	1.33E-02	0.00E+00	0.00E+00	0.00E+00	0.00E+00	-3.48E-04
		Am-243	1.27E-02	1.18E-02	1.59E-02	1.36E-02	0.00E+00	0.00E+00	0.00E+00
		Cm-242	1.08E-05	1.34E-02	0.00E+00	0.00E+00	0.00E+00	0.00E+00	1.07E-05
		Cm-244	1.31E-03	1.25E-02	8.96E-02	-4.41E-04	0.00E+00	0.00E+00	-8.55E-02
		Cm-245	2.67E-04	1.33E-02	0.00E+00	0.00E+00	0.00E+00	0.00E+00	3.98E-04
		Cm-246	6.73E-06	1.33E-02	0.00E+00	0.00E+00	0.00E+00	0.00E+00	3.95E-05
	1.20	Np-237	1.22E-02	2.58E-02	0.00E+00	0.00E+00	0.00E+00	0.00E+00	6.65E-02
		Am-241	4.62E-02	2.58E-02	0.00E+00	0.00E+00	0.00E+00	0.00E+00	6.36E-03
		Am-242m	4.20E-04	2.58E-02	0.00E+00	0.00E+00	0.00E+00	0.00E+00	5.57E-05
		Am-243	8.05E-03	2.58E-02	0.00E+00	0.00E+00	0.00E+00	0.00E+00	5.08E-02
		Cm-242	5.03E-06	2.58E-02	0.00E+00	0.00E+00	0.00E+00	0.00E+00	1.26E-05
		Cm-244	4.55E-04	2.58E-02	0.00E+00	0.00E+00	0.00E+00	0.00E+00	1.22E-03
		Cm-245	8.28E-05	2.58E-02	0.00E+00	0.00E+00	0.00E+00	0.00E+00	6.61E-04
		Cm-246	2.10E-06	2.58E-02	0.00E+00	0.00E+00	0.00E+00	0.00E+00	4.59E-05
An-R	1.05	Np-237	6.01E-03	7.35E-03	7.47E-02	-8.22E-02	0.00E+00	0.00E+00	0.00E+00
		Am-241	2.62E-02	7.35E-03	1.37E-02	-6.28E-02	0.00E+00	0.00E+00	0.00E+00
		Am-242m	9.65E-04	7.49E-03	1.80E-04	6.25E-03	0.00E+00	0.00E+00	0.00E+00
		Am-243	6.94E-03	6.82E-03	2.68E-02	-8.12E-03	6.94E-03	6.77E-03	-1.47E-02
		Cm-242	4.32E-06	7.29E-03	1.28E-05	-8.65E-02	4.32E-06	7.42E-03	0.00E+00
		Cm-244	2.94E-03	8.24E-03	0.00E+00	0.00E+00	0.00E+00	0.00E+00	4.88E-03
		Cm-245	8.93E-04	8.10E-03	0.00E+00	0.00E+00	0.00E+00	0.00E+00	1.25E-03
		Cm-246	4.92E-04	7.52E-03	0.00E+00	0.00E+00	0.00E+00	0.00E+00	2.68E-04
	1.10	Np-237	5.71E-03	1.37E-02	7.50E-02	-8.23E-02	0.00E+00	0.00E+00	0.00E+00
		Am-241	1.09E-02	1.36E-02	9.64E-03	-3.25E-01	1.09E-02	1.36E-02	0.00E+00
		Am-242m	1.04E-03	1.33E-02	9.08E-02	-1.25E-04	0.00E+00	0.00E+00	-9.03E-02
		Am-243	9.14E-03	1.31E-02	2.88E-01	-7.08E-04	0.00E+00	0.00E+00	-2.70E-01
		Cm-242	6.90E-06	1.37E-02	1.46E-05	-7.71E-02	0.00E+00	0.00E+00	0.00E+00
		Cm-244	3.63E-03	1.31E-02	1.92E-01	-4.11E-04	0.00E+00	0.00E+00	-1.85E-01
		Cm-245	8.00E-04	1.36E-02	1.80E-03	-1.26E-02	0.00E+00	0.00E+00	0.00E+00
		Cm-246	2.63E-04	1.38E-02	0.00E+00	0.00E+00	0.00E+00	0.00E+00	3.81E-04
	1.20	Np-237	5.19E-03	2.59E-02	7.55E-02	-8.22E-02	0.00E+00	0.00E+00	0.00E+00
		Am-241	1.53E-02	2.59E-02	0.00E+00	0.00E+00	0.00E+00	0.00E+00	2.06E-03
		Am-242m	5.80E-04	2.59E-02	0.00E+00	0.00E+00	0.00E+00	0.00E+00	7.98E-05
		Am-243	3.76E-03	2.59E-02	2.44E-02	-2.42E-02	0.00E+00	0.00E+00	0.00E+00
		Cm-242	4.58E-06	2.59E-02	1.68E-05	-6.37E-02	0.00E+00	0.00E+00	0.00E+00
		Cm-244	1.35E-03	2.59E-02	0.00E+00	0.00E+00	0.00E+00	0.00E+00	2.34E-03
		Cm-245	3.41E-04	2.59E-02	0.00E+00	0.00E+00	0.00E+00	0.00E+00	6.87E-04
		Cm-246	9.23E-05	2.59E-02	0.00E+00	0.00E+00	0.00E+00	0.00E+00	3.68E-04

1) Function Form: $A\exp(a\,t)+B\exp(b\,t)+C\exp(c\,t)+D$ where t is time (year).
2) Numerical result gives cumulative amount of isotopes(kg) per 1 kg of initial fissile equivalence at the start of the system. Multiply 3306.11 for obtaining amounts per initial 1 GWe system.