DESIGN OF ACCELERATOR-DRIVEN SYSTEM CONSISTENT TO PARTITIONING TECHNOLOGY

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Abstract

Partitioning and transmutation technology will be a promising technology to reduce the burden of high-level waste disposal problem. The Japan Atomic Energy Agency (JAEA) has investigated accelerator-driven system (ADS) to transmute minor actinides (MAs) in the transmutation cycle and obtained various results. On the other hand, the neutronics design of the ADS had used the ideal fuel composition which did not include impurities such as rare earth elements or uranium.

This study aims to investigate a new ADS fuel composition based on a separation process called SELECT (Solvent Extraction from Liquid-waste using Extractants of CHON-type for Transmutation) process developed for extracting U, Pu, and MAs from dissolution solution of spent nuclear fuels and/or high-level liquid waste. By performing the neutronics calculation of the ADS with the new fuel composition, it is confirmed that the new fuel composition based on the SELECT process is acceptable. These results also indicate that the decontamination factors for RE in the reprocessing process are also adequate.

1. INTRODUCTION

The Japan Atomic Energy Agency (JAEA) has performed research and development of the partitioning and transmutation (P&T) technology to reduce the burden of high-level waste (HLW) disposal problem. JAEA has proposed the double-strata strategy which will introduce a dedicated minor actinide (MA) transmutation cycle using an accelerator-driven system (ADS) [1]. The transmutation cycle consists of a partitioning of MA from HLW, MA fuel fabrication, transmutation by the ADS and reprocessing of spent fuel discharged from the ADS.

In the previous studies [2-3], the ideal fuel condition was supposed in the neutronics design of the ADS. For example, impurities such as rare earth (RE) nuclides were not assumed. However, in the real condition, RE will accompany with MA because the chemical behaviour of RE is similar to those of americium and curium. It is also supposed that uranium from the partitioning process will accompany with Pu and it will be included in the ADS fuel. The capture reaction of these nuclides deteriorates the neutron economy of the ADS core.

This study investigates a new fuel composition based on the SELECT (Solvent Extraction from Liquid-waste using Extractants of CHON-type for Transmutation) process proposed by JAEA. By the SELECT process, a certain amount of uranium and RE will be contained in the ADS fuel. By performing the neutronics calculation of the ADS with the new fuel composition, a feasibility of the new fuel composition will be investigated. An investigation for RE from the reprocessing will be also carried out to know whether the decontamination factor is suitable or not.

2. SEPARATION PROCESS

2.1. Outline of separation process

Extensive studies on recovering U, Pu, and MAs from spent nuclear fuels have been carried out, and many separation processes by hydrometallurgical method have been proposed. However, some of the proposed process use extractants containing phosphorus, and this could be one of reasons for increasing the volume of secondary waste. Thus, extractants in accordance with CHON principle, that is, reagents consisting of carbon, hydrogen, oxygen, and nitrogen are desirable in that they can be decomposed into gases by incineration, and we

have proposed a hydrometallurgical process called SELECT (Solvent Extraction from Liquid-waste using Extractants of CHON-type for Transmutation) to recycle nuclear materials and separate actinides for transmutation [4].

This process consists of the following four steps: selective extraction of U and co-extraction of U and Pu (Step IA and IB), recovery of Am, Cm, and REs from high level waste (Step II), separation of REs from Am and Cm (Step III), and mutual separation of Am and Cm (Step IV). An outline of the SELECT process is shown as Fig. 1. *N*,*N*-Dialkylamides, diglycolamides, nitrilotriacetamides, and alkyl diamide amines have been proposed as the extractants for the steps of IA, IB, II, III, and IV respectively. Continuous counter-current experiments have already been carried out to demonstrate the validity of SELECT process [4]-[6].

Many of the previous studies on the development of separation processes focused on the separation performance, and further discussion to evaluate the feasibility of transmuting minor actinides provided by the separation processes was limited. Thus we focused on the steps of IA, IB, II, and III, and the separation performance of these steps for some of REs (Y, La, Ce, Pr, Nd, Pm, Sm, Eu, and Gd), nuclear materials (U and Pu), and minor actinides (Np, Am, and Cm) was calculated using a simulation code to evaluate the validity of the proposed process from the viewpoint of an ADS design. The present study did not treat Step IV in that the fundamental data required for evaluating the separation performance of this step were insufficient.

A calculation code called PARC was used for simulating the behaviour of metal elements in separation processes using liquid-liquid extraction. The concentration profiles of the metal elements in mixer-settlers and/or pulsed columns are obtained by this code. A further explanation of PARC code can be found in references [7] and [8]. Calculation by PARC code was carried out on the assumption that a spent PWR fuel of 45 GWd/t burn-up after 7-year cooling dissolved in nitric acid was fed to Step IA as the feed. The composition of the spent fuel was calculated by ORIGEN2, and the concentrations of acid and metal elements in the feed are shown in Table 1. The valence state of Np in nitric acid was assumed to be pentavalent or hexavalent state, and the ratio of Np(VI) to Np(V) was set as 1.5. The extractants used in the steps IA, IB, II, and III were *N*,*N*-di(2-ethylhexyl)-2,2-dimethylpropanamide(DEHDMPA),*N*,*N*-di(2-ethylhexyl)butanamide(DEHBA),*N,N,N',N'*-tetradodecyldiclycolamide (TDdDGA), and *N,N,N',N',N'',N''*-hexyaoctyl nitrilitriacetamide (HONTA), respectively.

TABLE 1. ACID AND METAL CONCENTRATIONS IN THE FEED SUBJECTED TO THE CALCULATION

Constituent	Conc. [mol/dm3]
H+	4.0
U	8.4×10^{-1}
Pu	9.9×10^{-3}
Np(V)	2.3×10^{-4}
Np(VI)	3.4×10^{-4}
Am	6.1×10^{-4}
Cm	4.3×10^{-5}
Y	1.5×10^{-3}
La	2.5×10^{-3}
Ce	4.9×10^{-3}
Pr	2.3×10^{-3}
Nd	8.1×10^{-3}
Pm	5.4×10^{-5}
Sm	1.6×10^{-3}
Eu	2.6×10^{-4}
Gd	2.3×10^{-4}

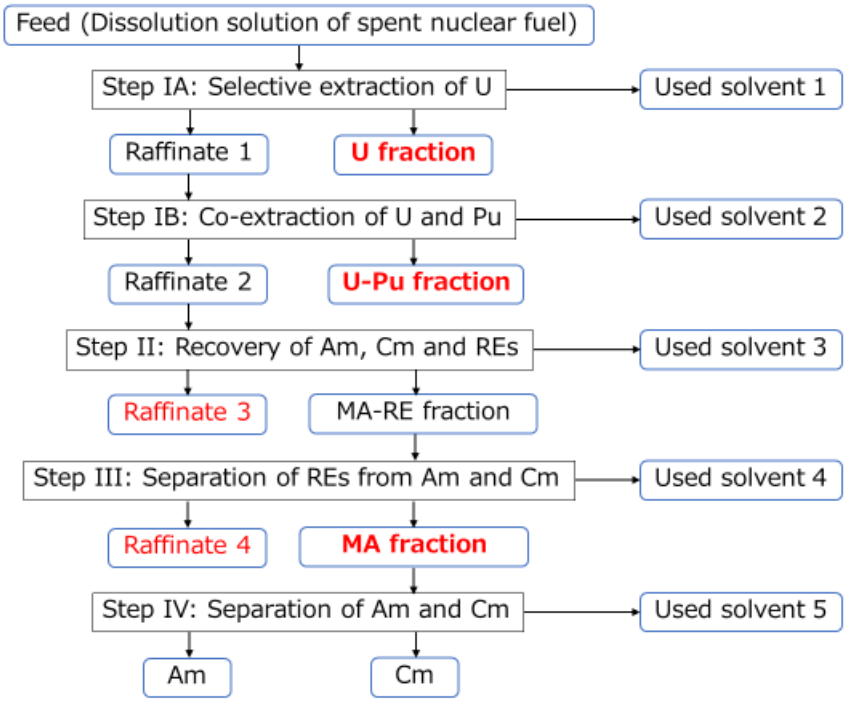

FIG. 1. An outline of SELECT process.

2.2. Separation performance

Table 2 shows the ratio of each element distributed to each outgoing stream. Almost all U and Pu were recovered in the U fraction or U-Pu fraction, respectively, indicating that Step IA and Step IB were effective for recovering U and Pu. It should be noted that part of U, 0.21%, was distributed to the U-Pu fraction, and this process did not isolate Pu. The present calculation showed that Np(V) was distributed to various fractions, and Np(VI) was distributed to the U fraction and the U-Pu fraction. Valence state of Np should be adjusted by using reductants or oxidants for gathering Np in a fraction. Americium, Cm, and REs were barely extracted in Steps IA, IB, and II. As a result, the ratios of Am, Cm, and REs except La entered into Step III were less than 0.01%. Most of Am and Cm were separated from REs in Step III, and the ratios of Am and Cm recovered in MA fraction were 99.86% and 89.28%, respectively.

Table 3 shows the decontamination factors of each metal element in Steps IA, IB, II, and III calculated by eqs. (1), (2), (3), and (4), respectively.

$$\mathrm{DF}_{\mathrm{M,IA}} = \left(Q_{\mathrm{feed}} \cdot C_{\mathrm{M,feed}}\right)/\left(Q_{\mathrm{raf1}} \cdot C_{\mathrm{M,raf1}}\right) \quad (1)$$
$$\mathrm{DF}_{\mathrm{M,IB}} = \left(Q_{\mathrm{raf1}} \cdot C_{\mathrm{M,raf1}}\right)/\left(Q_{\mathrm{raf2}} \cdot C_{\mathrm{M,raf2}}\right) \quad (2)$$
$$\mathrm{DF}_{\mathrm{M,II}} = \left(Q_{\mathrm{raf2}} \cdot C_{\mathrm{M,raf2}}\right)/\left(Q_{\mathrm{MA-RE}} \cdot C_{\mathrm{M,MA-RE}}\right) \quad (3)$$
$$\mathrm{DF}_{\mathrm{M,III}} = \left(Q_{\mathrm{MA-RE}} \cdot C_{\mathrm{M,MA-RE}}\right)/\left(Q_{\mathrm{MA}} \cdot C_{\mathrm{M,MA}}\right) \quad (4)$$

In these eqs., subscript M indicates metal element, and Q_{feed}, Q_{raf1}, Q_{raf2}, $Q_{\mathrm{MA\text{-}RE}}$, and Q_{MA} represent the flow rates of Feed, Raffinate 1, Raffinate 2, MA-RE fraction, and MA fraction, respectively. The terms of $C_{\mathrm{M,feed}}$, $C_{\mathrm{M,raf1}}$, $C_{\mathrm{M,raf2}}$, $C_{\mathrm{M,MA\text{-}RE}}$, and $C_{\mathrm{M,MA}}$ represent the concentrations of metal element M in Feed, Raffinate 1, Raffinate 2, MA-RE fraction, and MA fraction, respectively.

Total decontamination factors calculated by eq. (5) are also shown in Table 3

$$\mathrm{DF}_{\mathrm{M,Tot.}} = \left(Q_{\mathrm{feed}} \cdot C_{\mathrm{M,feed}}\right)/\left(Q_{\mathrm{MA}} \cdot C_{\mathrm{M,MA}}\right) \quad (5)$$

The low decontamination factors of Am, Cm, and REs observed in the steps of IA and IB indicate that U fraction and U-Pu fraction were hardly contaminated with these elements, and this result was favorable from the viewpoint of recycle use of nuclear material. The values of $\mathrm{DF}_{\mathrm{Am,III}}$ and $\mathrm{DF}_{\mathrm{Cm,III}}$ were 1.0 and 1.1, respectively,

while $DF_{M,III}$ (M: Y, La, Ce, Pr, Nd, Pm, Sm, Eu, and Gd) were on the order of 10^2 or higher. Part of REs were distributed to MA fraction, and the ratio of total REs concentration with respect to total REs and minor actinides (Np, Am, and Cm) concentration was 5.8%.

Although further works for optimizing the separation performance would be necessary, the present results supported the validity of the proposed process from the viewpoint of the extraction and separation behavior of U, Pu, MAs, and REs.

TABLE 2. RATIOS OF ELEMENTS DISTRIBUTED TO EACH OUTGOING STREAM [%]

	Step IA		Step IB		Step II		Step III		
	U frac.	Used solv. 1	U-Pu frac.	Used solv. 2	Raf. 3	Used solv. 3	Raf. 4	MA frac.	Used solv. 4
U	99.79	<0.01	0.21	<0.01	<0.01	<0.01	<0.01	<0.01	<0.01
Pu	<0.01	<0.01	>99.99	<0.01	<0.01	<0.01	<0.01	<0.01	<0.01
Np(V)	1.2	<0.01	7.19	<0.01	72.33	<0.01	19.27	<0.01	<0.01
Np(VI)	96.13	<0.01	3.87	<0.01	<0.01	<0.01	<0.01	<0.01	<0.01
Am	<0.01	<0.01	<0.01	<0.01	<0.01	<0.01	0.13	99.86	<0.01
Cm	<0.01	<0.01	<0.01	<0.01	<0.01	<0.01	10.72	89.28	<0.01
Y	<0.01	<0.01	<0.01	<0.01	<0.01	<0.01	>99.99	<0.01	<0.01
La	<0.01	<0.01	<0.01	<0.01	0.02	<0.01	99.98	<0.01	<0.01
Ce	<0.01	<0.01	<0.01	<0.01	<0.01	<0.01	99.89	0.11	<0.01
Pr	<0.01	<0.01	<0.01	<0.01	<0.01	<0.01	99.89	0.11	<0.01
Nd	<0.01	<0.01	<0.01	<0.01	<0.01	<0.01	99.31	0.69	<0.01
Pm	<0.01	<0.01	<0.01	<0.01	<0.01	<0.01	99.89	0.11	<0.01
Sm	<0.01	<0.01	<0.01	<0.01	<0.01	<0.01	99.89	0.11	<0.01
Eu	<0.01	<0.01	<0.01	<0.01	<0.01	<0.01	99.98	0.01	<0.01
Gd	<0.01	<0.01	<0.01	<0.01	<0.01	<0.01	99.89	0.11	<0.01

TABLE 3. DECONTAMINATION FACTOR

M	$DF_{M,IA}$	$DF_{M,IB}$	$DF_{M,II}$	$DF_{M,III}$	$DF_{M,Tot.}$
U	4.7×102	1.5×106	1.6	9.8×1016	1.1×1026
Pu	1.0	9.0×10^4	1.0	9.8×10^{16}	8.8×10^{21}
Np(V)	1.0	1.1	4.8	6.8×10^6	3.5×10^7
Np(VI)	2.6×10	6.4×10^3	1.0	2.9	4.8×10^5
Am	1.0	1.0	1.0	1.0	1.0
Cm	1.0	1.0	1.0	1.1	1.1
Y	1.0	1.0	1.0	2.1×10^8	2.1×10^8
La	1.0	1.0	1.0	1.7×10^7	1.7×10^7
Ce	1.0	1.0	1.0	8.8×10^2	8.8×10^2
Pr	1.0	1.0	1.0	8.8×10^2	8.8×10^2
Nd	1.0	1.0	1.0	1.5×10^2	1.5×10^2
Pm	1.0	1.0	1.0	8.8×10^2	8.8×10^2
Sm	1.0	1.0	1.0	8.8×10^2	8.8×10^2
Eu	1.0	1.0	1.0	6.9×10^3	6.9×10^3
Gd	1.0	1.0	1.0	8.8×10^2	8.8×10^2

3. NEUTRONICS DESIGN OF ADS CORE

3.1. New fuel composition

Based on the results in Section 2, a new fuel composition for the ADS was investigated. The conventional investigation for the ADS in JAEA has employed (Pu+MA)+Zr nitride fuel which was U-free fuel [2-3]. Both Pu and MA are provided from the partitioning process at the first cycle. After the burnup and the cooling, fission products (FPs) are removed in the reprocessing process. After that, only MAs from the partitioning process are provided and a fresh fuel for the next cycle is fabricated. Based on this flow, the following cases are considered in this study.

- CASE 1: Np, Am and Cm are loaded as the MA fuel. It means Raffinate 3 and MA fraction (shown in Fig. 1) are used for the MA fuel fabrication. Raffinate 4 will be discarded.
- CASE 2: Only Am and Cm (MA fraction in Fig. 1) are loaded as the MA fuel. In this case, almost all Np are discarded as Raffinate 3 and 4.

In both cases, U fraction is reused for fast reactors (FRs) in the commercial cycle. U-Pu fraction is employed as Pu for the ADS or FRs.

Tables 4 and 5 show the new fuel compositions of Pu, MA-CASE1 and MA-CASE2, respectively. For the Pu, the accompaniment of 15at.%U is observed. For the MA, the accompaniment of about 4at.%RE is confirmed. In CASE 1, about 20at.% Np is contained. In CASE2, Am is dominant (about 90%) in the MA.

TABLE 4. COMPOSITION OF Pu (at.%)

Nuclide	Pu
U-235	0.18%
U-236	0.09%
U-238	14.71%
Np-237	0.25%
Pu-238	2.12%
Pu-239	48.88%
Pu-240	19.18%
Pu-241	9.42%
Pu-242	5.17%

TABLE 5. COMPOSITION OF MA (at.%)

Nuclide	MA-CASE1	MA-CASE2
Np-237	18.95%	0.00%
Am-241	57.10%	71.11%
Am-242m	0.11%	0.14%
Am-243	15.05%	18.75%
Cm-243	0.05%	0.06%
Cm-244	4.23%	5.26%
Cm-245	0.39%	0.49%
Cm-246	0.03%	0.04%
RE	4.09%	4.14%

3.2. Calculation condition

Lead-bismuth cooled ADS with 800MWth [9] is employed for this study. For the neutronics calculation, the ADS3D code [10] developed by JAEA was utilized. It can calculate the transport of neutrons by the

deterministic method, the burnup calculation, and the fuel exchange in two- and three-dimensional calculation models. The energy group for the transport of neutrons is the 73-group structure. This calculation code performs the following flow. After the geometry preparation, an eigen mode calculation is carried out to get the k_{eff} value. Then, the fixed-source calculation is performed with the use of the external neutron source data and the flux distribution is obtained. Based on the flux distribution, the burnup calculation for each fuel region is performed and after the burnup calculation, the fuel exchange is carried out. In this study, two-dimensional RZ calculation model shown in Fig. 2 was employed. Two MA fuel zones, inner core (IC) and outer core (OC), were considered to ease the power distribution. The following factors, 1.10 and 0.89 for IC and OC, were considered to the ZrN ratio. Other details for the calculation model are described in reference [9]. As the nuclear data library, JENDL-4.0 was used.

The burnup calculation was performed with 800MW thermal power and 600 EFPDs (effective full power days) operation. After the burnup and 4-year cooing, the discharged fuels are reprocessed at the reprocessing facility. The previous studies [2-3] assumed that all REs would be recovered by the reprocessing and only MAs were recycled for the next fresh fuel. On the other hand, this study considers DF for REs in the reprocessing. It means MAs from the reprocessing include REs. Table 6 shows the DF values in the reprocessing. These values were based on the process flow diagram of the pyrochemical reprocessing proposed in reference [11]. By this process, 99.96% TRU is recovered and DF=3.7 is achieved for REs.

TABLE 6. DF VALUES FOR RE ELEMENTS IN REPROCESSING

Element	DF
Y	400
La	9.4
Ce	3.4
Pr	2.8
Nd	3.0
Pm	3.0
Sm	43000
Eu	190000
Gd	13

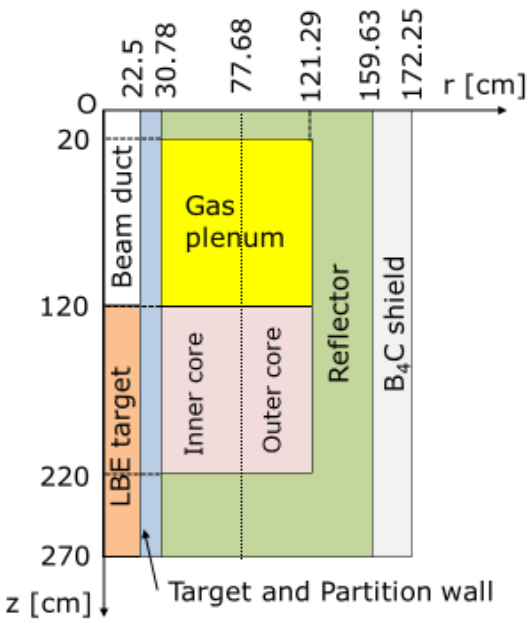

FIG. 2. RZ calculation model.

3.3. Results and discussion

The changes of the k_{eff} value and the proton beam current are presented in Figs. 3 and 4, respectively. The k_{eff} values at the beginning of cycle (BOC) are almost maintained at 0.98 although the maximum k_{eff} value became 0.988 in CASE2. In the previous study [9], the discussion of the maximum k_{eff} value was performed and it was confirmed the upper limit of the criticality was 0.989 in this ADS design. Although re-examination for the upper limit of the criticality is required due to the change of the fuel composition, these calculation results are acceptable from the viewpoint of the criticality. In these results, the maximum proton beam current became 14 mA in both cases. Table 7 summarizes the fuel parameters at each burnup cycle. This table also represents the value of relative lattice parameter difference (RLPD) [12]. This value is used as the limitation of TRU ratio for the solid solution formation between ZrN and TRU nuclides. This value should be less than 8.6%. From Table 7, it was observed that all RLPD values were less than 8.6%, so there was no problem from the viewpoint of the MA fuel fabrication. Through these results, there was no big difference between CASE1 and CASE2.

The amounts of each heavy metal element at BOC are illustrated in Figs. 5 and 6. It is found that the amounts of Pu and Cm slightly increase and those of Np (CASE1) and Am decrease and converge on the constant. Table 8 shows the transmutation amount at 10th cycle. The transmutation amount dw_i was calculated as:

$$\mathrm{dw}_i = w_i^{cooling} - w_i^{BOC} \tag{6}$$

where i is each element, w_i^{BOC} is the weight of element i at BOC and $w_i^{cooling}$ is the weight of element i after burnup and cooling. In CASE1, 100 kg of Np and 400 kg of Am can be transmuted during 600 EFPDs. In CASE2, 500 kg of Am can be reduced. In the previous studies, the MA transmutation amount was about 500 kg/600 EFPDs, so there was no deterioration by including U and RE.

TABLE 7. FUEL PARAMETERS AT EACH BURNUP CYCLE (UNIT: %)

Cycle	ZrN ratio*1 (CASE1)	RLPD (CASE1)	ZrN ratio (CASE2)	RLPD (CASE2)
1 (Pu ratio) *1	51.0 (30.9)	8.20	51.0 (31.0)	8.47
2	61.8	8.17	62.6	8.43
3	66.7	8.08	66.2	8.33
4	66.7	8.05	66.2	8.29
5	66.7	8.03	66.2	8.26
6	66.7	8.02	66.2	8.25
7	66.7	8.01	66.2	8.23
8	66.7	8.01	66.2	8.22
9	66.7	8.00	66.2	8.22
10	66.7	8.00	66.2	8.21

*1: Volume ratio

TABLE 8. TRANSMUTATION AMOUNT AT 10TH CYCLE (UNIT: kg)

Element	CASE1	CASE2
U	8.2	5.5
Np	-104.3	-0.5
Pu	1.6	5.2
Am	-399.4	-500.9
Cm	-19.2	-24.3
MA(Np+Am+Cm)	-522.9	-525.8

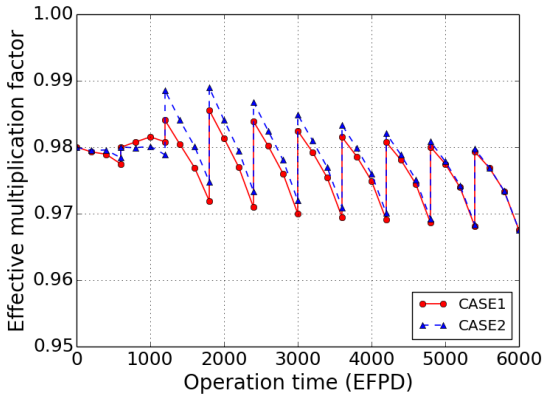

FIG. 3. Changes of the k_{eff} value.

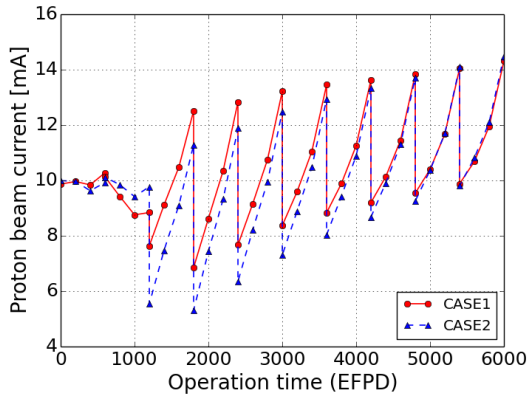

FIG. 4. Changes of the proton beam current.

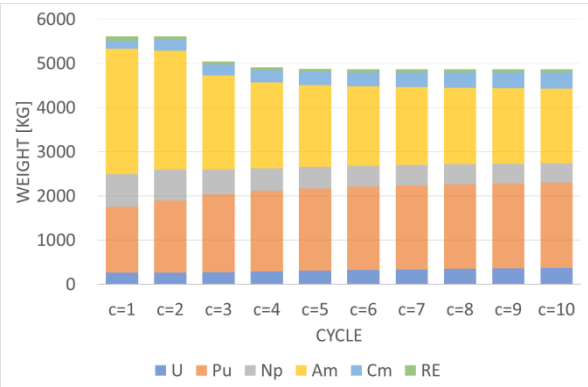

FIG. 5. Amount of each element at BOC (CASE1).

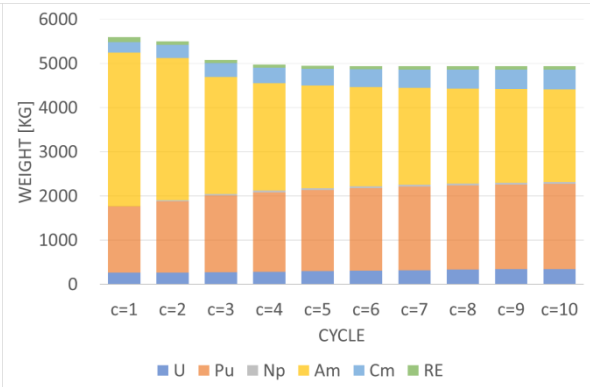

FIG. 6. Amount of each element at BOC (CASE2).

4. CONCLUSION

This study investigates the new ADS fuel composition based on the SELECT process. Among the raffinates and fractions produced by the SELECT process, two (U-Pu fraction and MA fraction) or three (U-Pu fraction, Raffinate 3, and MA fraction) of them can be used for the ADS. Although the new fuel composition includes U and RE as the impurity, it is confirmed that the accompaniment of 15at.% U and 4at.%RE is no problem for the ADS neutronics design. The k_{eff} value, proton beam current and RLPD are appropriate and there is no deterioration for the transmutation amount. These results also indicate that the DF values for RE in the reprocessing process are also adequate.

As the future work, it is necessary to discuss how to treat or discard Raffinate 4 (and Raffinate 3 if it is not used for the ADS fuel). Especially, Raffinate 4 includes about 10%Cm, so its treatment will be difficult. Another step to separate Cm from Raffinate 4 may be required. For the ADS neutronics design, new subcriticality should be investigated with the use of the new fuel composition.

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