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Plutonium and other actinides behaviour in NEXT process

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Abstract

The behaviour of Pu and other actinides in some characteristic processes of the NEXT process, i.e. the crystallization process and the U–Pu–Np co-recovery process, was investigated with referring to the experimental results obtained in our experimental facility, CPF. In the crystallization process, the existence of Pu(VI) in the dissolver solution brought the co-crystallization of Pu(VI) with U, and low DF_{Pu}. Such co-crystallization could be prevented by adjusting the Pu valence to Pu(IV). In the crystallization with the dissolver solution containing Pu(IV), meanwhile, DF_{Cs} showed significant low values, which might be caused by the formation and crystallization of some kinds of Cs compounds with Pu(IV). In the U–Pu–Np co-recovery process which had high [HNO_3] feed solution obtained from the crystallization process, it was confirmed experimentally that almost all the Np could be extracted with U and Pu in the extraction section. Some considerable parameters, e.g. temperature, acidity of stripping solution, were pointed out for preventing the leakage of U and Pu to the solvent and for avoiding the Pu polymer formation in the stripping section using only diluted HNO_3 as the stripping solution. Under the appropriate stripping condition taking account of these parameters, the calculation and experimental results showed that these elements could be stripped efficiently.

Keywords: NEXT process; Crystallization; Extraction; TBP; Actinides

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1. Introduction

Since 1997, Japan Atomic Energy Agency (JAEA) has been keeping on the feasibility study on the commercialized fast reactor cycle systems, which aims for ensuring safety, economical competitiveness, efficient utilization of resources, proliferation resistance, and decreasing environmental burden. For meeting these requirements, the investigation and the evaluation of several conceptual reactors, reprocessing and fuel fabrication for the future fast reactor cycle system have been carried out in the feasibility study. On this investigation and evaluation, the advanced aqueous reprocessing system named New Extraction system for TRU (Np, Pu, Am and Cm) recovery (NEXT) process is selected as one of the promising processes for a spent nuclear fuel reprocessing [1]. The NEXT process basically consists of three characteristic processes as shown in Fig. 1; the crystallization process for recovering a part of U from dissolver solution, the U–Pu–Np co-recovery process with single cycle flowsheet using TBP as an extractant and the Am–Cm recovery process with extraction chromatography. In this study, we will discuss the behaviour of Pu and other actinides in the crystallization process and the U–Pu–Np co-recovery process with referring to the experimental results obtained in our experimental facility, Chemical Processing Facility (CPF).

2. Crystallization process

In the crystallization process, over 70% of U in the dissolver solution is recovered as uranyl nitrate hexahydrate (UNH) crystal by cooling the dissolver solution, which is based on the temperature dependence of the solubility of UNH crystal into HNO_3. Although U is the major element in the spent fuel of fast reactor, the dissolver solution also contains a significant amount of Pu, minor actinides (MA) and fission products (FP). Therefore, it is important for the selective U recovery from the dissolver solution to investigate (and control, if needed) the behaviour of these elements in the crystallization process.

2.1. Crystallization experiments

Table 1 summarizes the experimental conditions of several crystallization experiments carried out in CPF [2–4]. The effect of the Pu valence on its behaviour in the crystallization process

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Table 1
Condition of the crystallization experiments

	Feed solution					Cooling condition	Washing solution
	$[H^+]$ (M)	[U] (g/L)	[Pu] (g/L)	$[^{137}Cs]$ (Bq/mL)	$[^{155}Eu]$ (Bq/mL)		
Run1 (U–Pu(IV)–HNO_3)	2.7	540	51	–	–	50–40 °C → 15–10 °C	180 g U/L ($UO_2(NO_3)_2$)–4 M HNO_3
Run2 (U–Pu(VI)–HNO_3)	5.6	610	47	–	–		
Run3 (U–Pu(VI, IV)–HNO_3)	5.6	550	40	–	–		
Run4 (dissolver solution[a])	6.0	410	200	5.6×10^{8}[b]	2.6×10^{7}		8 M HNO_3
Run5 (dissolver solution[a])	3.9	536	68	4.8×10^{8}[b]	5.6×10^{6}		5 M HNO_3
Run6 (U–Pu(IV)–Cs–HNO_3)	8.1	409	42	2.1×10^{3}[b]	–		

[a] Dissolver solution of the MOX fuel irradiated in "JOYO" (burn-up: 32 ∼ 64GWd/t) (valence of Pu was adjusted to Pu(IV)).
[b] Estimated [Cs] (g/L) are 0.71 (Run4), 2.1 (Run5) and 4.6 (Run6), respectively.

was investigated mainly in the experiments using U–Pu–HNO_3 solution as the feed solution. The structure of the crystal obtained in some experiments was determined by X-ray diffraction (XRD) analysis using RIGAKU RINT2100. In the experiments using the dissolver solution of the MOX fuel irradiated in the fast reactor "JOYO", the decontamination factors (DF) of FP as well as Pu to U crystal were measured for estimating the behaviours of those elements in the crystallization process. In Run5 and Run6, an appropriate amount of $CsNO_3$ was added into the feed solution for investigating the effect of the Cs concentration on its behaviour. The crystals obtained in the experiments were washed by appropriate HNO_3 solution for removing the mother solution on their surface. The experimental procedures have been reported in detail by our previous reports [2–4].

Table 2
Decontamination factors of Pu to U in the crystal [3]

		DF_{Pu}[a]
Run1	Before washing	4.9
	After washing	29
Run2	Before washing	1.3
	After washing	1.5
Run3	Before washing	3.6
	After washing	4.9

[a] DF_{Pu} was calculated by $DF_{Pu} = (C_{f,Pu}/C_{f,U})/(C_{c,Pu}/C_{c,U})$; $C_{x,y}$: concentration of element y in x; x; f: feed solution, c: crystal; y; U: uranium, Pu: plutonium.

2.2. Plutonium behaviour in the crystallization process

Table 2 summarizes the DF of Pu to the U crystal (DF_{Pu}) in some crystallization experiments using U–Pu–HNO_3 solution as the feed solution. These results show clearly that Pu has the different behaviour in the crystallization process according to its valence in the dissolver solution. Under the condition that Pu was adjusted to Pu(IV) in the feed solution, comparatively high DF_{Pu} could be achieved, especially by washing the U crystal, which suggested that little Pu was accompanied with the U crystal, and the only surface of the U crystal was contaminated with mother solution containing Pu. On the other hand, if Pu(VI) existed in the feed solution, the values of the DF_{Pu} were considerably low even after washing the U crystal, which implied that some kind of Pu compound was precipitated and contained in the U crystal. These differences of the Pu behaviours, which was caused by the Pu valence in the feed solution, were also reflected in the color of the crystal after washing; lemon yellow in Pu(IV) feed, while orange in Pu(VI) feed. Our previous study showed that Pu(VI) is not crystallized in Pu(VI)–HNO_3 solution under these crystallization conditions [5]. It is, therefore, considered that Pu(VI) would be co-crystallized with U in U–Pu(VI)–HNO_3 solution. Table 3 shows the lattice constants of the U crystal obtained in

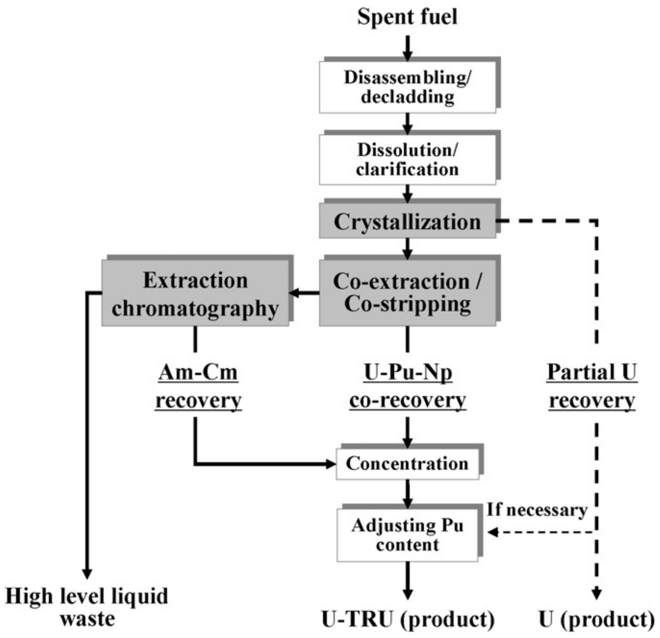

Fig. 1. Schematic flow of the NEXT process.

Table 3
Lattice constants of the crystal crystallized in U–Pu(VI)–HNO_3 system

	a	b	c	Pu contents (%)
U crystal obtained in U–Pu(VI)–HNO_3[a]	13.18	8.02	11.47	6.4
UNH crystal [5]	13.14	8.01	11.43	–

[a] Feed solution: $[H^+]$: 6.7 M, [U]: 344 g/L, [Pu]: 37 g/L (Pu(VI)).

Table 4
Decontamination factors of Pu and FPs to U in the crystal

	DF[a]		
	Pu	^{137}Cs	^{155}Eu
Run4			
Before washing	5.6	1.2	4.2
After washing	25	0.8	27
Run5			
Before washing	5.9	1.4	11.7
After washing	22.9	2.1	>92.8
Run6			
Before washing	4.5	2.0	–
After washing	4.7	0.4	–

[a] DF was calculated by $DF_{Pu} = (C_{f,M}/C_{f,U})/(C_{c,M}/C_{c,U})$; $C_{x,y}$: concentration of element *y* in *x*; *x*; f: feed solution, c: crystal; *y*; U: uranium, M: Pu, ^{137}Cs, ^{155}Eu.

the experiment with U–Pu(VI)–HNO_3 feed solution. The lattice constants of pure UNH crystal, which has been reported by Moseley et al. [6], are also listed in this table. The positions of the XRD peaks obtained on the analysis of the U crystal gave good agreement with those assigned to the UNH crystal, and lattice constants estimated from these peaks showed the almost same values as those of the UNH crystal. These suggest that parts of the U atoms (U(VI)) in the UNH crystals are replaced by the Pu atoms (Pu(VI)) during the crystallization with U–Pu(VI)–HNO_3 feed solution. These experimental results shows the importance of the adjustment of the Pu valence to Pu(IV) for preventing Pu co-crystallization and keeping high DF_{Pu}.

Table 4 summarizes the DF of Pu and some FP to the U crystal in the crystallization experiments using the dissolver solution of the MOX fuel irradiated in the fast reactor "JOYO" as the feed solution (Run4 and Run5). In these experiments, the Pu valence in the dissolver solution was adjusted to be Pu(IV) by NO_x bubbling, which brought the reasonable DF_{Pu} after washing the U crystal. The DF of FP showed that their behaviours in the crystallization were complicated. The DF of lanthanides (Eu) indicated that, on the assumption that the behaviour of Eu could be extrapolated to the other lanthanides, most of these elements existed in the mother solution on the surface of the U crystal and could be removed effectively by washing the U crystal. On the other hand, the behaviour of Cs was supposed to be quite different from those of Pu and lanthanides, because the DF_{Cs} showed low value and it was not improved even after washing the U crystal. This suggests that the U crystal accompanied not only mother solution containing Cs on its surface but some kind of Cs compound which had low solubility into washing solution, HNO_3, in these experimental conditions. Fig. 2 shows that the appearance of the U crystal obtained in the crystallization experiment using U–Pu–Cs–HNO_3 solution as the feed solution (Run6). This experiment was carried out for confirming the Cs behaviour without any FP under the crystallization condition where the Pu valence in the feed solution was adjusted to be Pu(IV). The values of DF_{Pu} and DF_{Cs} after washing U crystal were estimated to be 4.7 and 0.4, respectively, in this experiment (7.7 mg Pu and 5.8 mg Cs in 1 g crystal). Although these values were comparatively lower than those in the above experiments using the dissolver solution as the feed solution, which might be caused by the difference of several experimental conditions (higher Cs and HNO_3 concentration) as mentioned in the following paragraph, it was confirmed that Cs could be hardly separated from the U crystal even by washing the U crystal. In this experiment, a small amount of green crystal was observed in the U crystal after washing the U crystal (see Fig. 2). Cesium can crystallize as $CsNO_3$ or $Cs_2UO_2(NO_3)_4$ in $CsNO_3$–$UO_2(NO_3)_2$–H_2O system, but the Cs concentration in this experiment (and other crystallization experiments) was much lower than the value which gave the crystals of these compounds [7]. On the other hand, it has been also reported that whitish green crystal of $Cs_2Pu(NO_3)_6$ is formed by mixing $CsNO_3$ and $Pu(NO_3)_4$ in high nitric acid solution and the solubility of Pu and Cs in this system is estimated to be 3–4 g/L (28 °C) [8]. This suggests that Cs might form double nitrate with Pu, and crystallize as $Cs_2Pu(NO_3)_6$ under the condition of our crystallization experiments, which was observed as a small amount of green crystal in the U crystal on Run6.

Fig. 2. U crystal in the crystallization experiment (Run6) (after washing).

Veirs et al. have reported that Pu(IV) takes three major species expressed as $[Pu(NO_3)_n]^{4-n}$ ($n = 2, 4, 6$) in HNO_3 and the ratio of $[Pu(NO_3)_6]^{2-}$ increases with the concentration of nitrate ion ($[NO_3^-]$) [9]. Higher ratio of $[Pu(NO_3)_6]^{2-}$ will bring more formation and crystallization of $Cs_2Pu(NO_3)_6$; higher $[NO_3^-]$ will bring lower solubility of Cs (and Pu(IV)) and lower DF_{Cs} (and DF_{Pu}). This would be one of the reasons which caused the difference of DF_{Cs} and DF_{Pu} in our crystallization experiments shown in Tables 2 and 3. The identification of the green crystal obtain in the U crystal and detailed investigation of this compound are now in progress, which is indispensable for clarifying the process condition without the formation of such undesirable crystal. These studies will bring several effective methods for achieving high DF_{Cs}, e.g. decreasing $[NO_3^-]$ in the feed solution, and be also useful, if further required, for developing some purification method for the U crystal contaminated with this green crystal.

3. Uranium–plutonium–neptunium co-recovery process

After the crystallization process, U, Pu and Np are co-recovered in the U–Pu–Np co-recovery process with single cycle flowsheet using TBP as an extractant (see Fig. 3). This process has several characteristics compared with the conventional PUREX process;

- co-extraction of U, Pu and Np by TBP;
- elimination of the Pu partitioning section;
- co-stripping of U, Pu and Np by diluted HNO_3 without any reductants nor complexants.

Among the actinide elements, Np has the complex behaviour in the extraction section because of its variable valences in HNO_3; extractable Np(IV) and Np(VI), and inextractable Np(V). It is, therefore, important for the effective co-extraction of Np with U and Pu to investigate and control the Np behaviour in the extraction section.

With the elimination of the Pu partitioning section, no Pu reduction to Pu(III) occurs in the process, and Pu is co-stripped with U (and Np) just by diluted HNO_3 solution without any reductants nor complexants. The stripping reaction of Pu (Pu(IV)) and U (U(VI)) are exothermic and endothermic, respectively [10], which means that the careful temperature control is one of the important factors in the stripping section for the effective (and complete) co-stripping of U and Pu (see Fig. 4). In addition, it is required for preventing the formation of Pu polymer under low acidity to adjust $[HNO_3]$ to be suitable value in the stripping section.

Based on these backgrounds, this study mentions the results of U–Pu–Np co-recovery experiments carried out in CPF, especially the Np behaviour in the extraction section, and U and Pu behaviour in the stripping section.

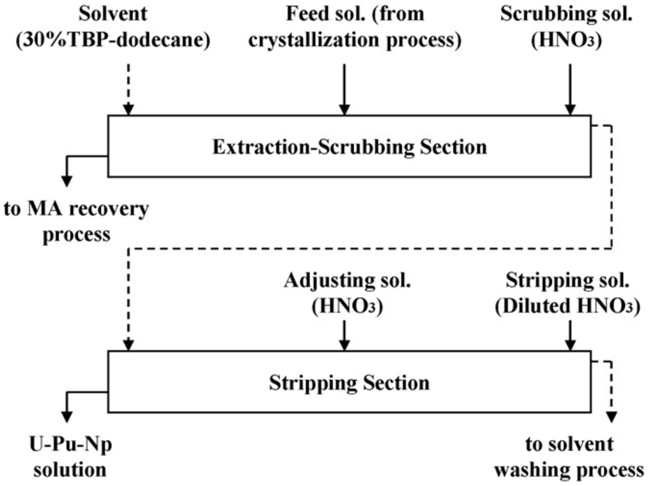

Fig. 3. Typical flowsheet of the U–Pu–Np co-recovery process.

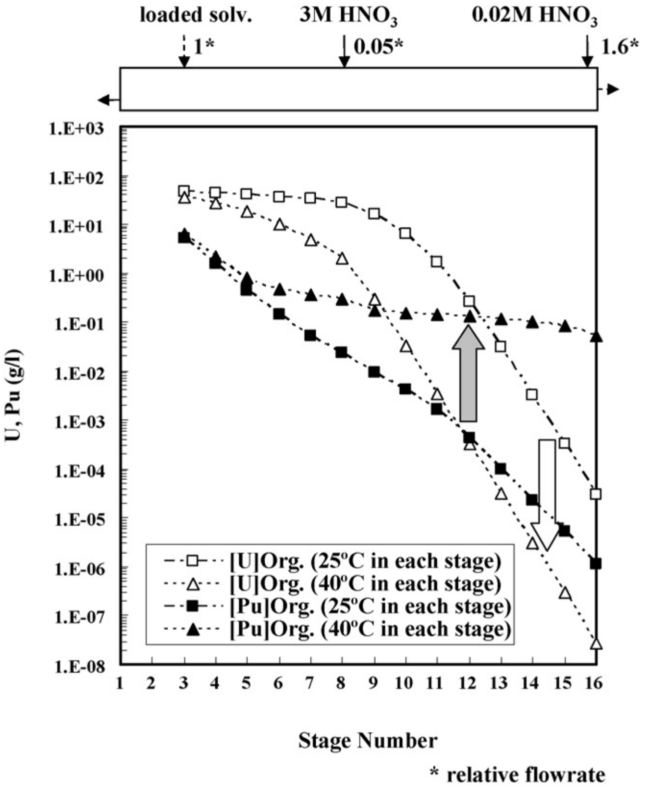

Fig. 4. Temperature dependence of U and Pu profiles in the stripping section (calculation results based the flowsheet of Run2 in Table 5).

3.1. Uranium–plutonium–neptunium co-recovery experiments

Table 5 summarizes the experimental conditions of the counter-current experiments for U–Pu–Np co-recovery. All these experiments used the centrifugal contactors which had about 10 mL of mixing zone and about 15 mL of settling zone in each stage. In these experiments, the dissolver solution of the MOX fuel irradiated in the fast reactor “JOYO” was supplied as the feed solution. The acidity of the feed solution was adjusted to be comparatively higher value than that in the conventional PUREX process, which was based on the acidity of the dissolver solution after the crystallization process. The valence of Pu in the feed solution was confirmed as Pu(IV) by absorbance spectra in UV–vis region before these experiments. The experimental procedures have been given in detail by our previous reports [11,12].

For evaluating the behaviour of U, Pu and Np in these experiments, the simulation of the extraction and stripping of these elements was also carried out with the simulation code MIXSET-X [13].

3.2. Neptunium behaviour in the extraction section

Table 6 shows the ratios of U, Pu and Np leaked to the raffinate in the U–Pu–Np co-recovery experiments. The leakage of U and Pu to the raffinate could be kept under the lower analytical limit and that of Np was also suppressed under 2% in all experiments.

Table 5
Condition of the U–Pu–Np co-recovery experiments

Fuel	Fast reactor JOYO core fuel		
	Run1	Run2	Run3
[U], [Pu], [Np] in feed sol. (g/L)	80, 32, 0.15	138, 37, 0.86	150, 13, 0.05
$[HNO_3]$ in feed sol. (M)	6.2	5.2	5.2
Pu valence in feed sol. (additional reductant)	Pu(IV)	Pu(IV) (+$NaNO_2$)	Pu(IV)(+NOX)
$[HNO_3]$ in scrubbing sol. (M)	2	2	5
$[HNO_3]$ in adjusting sol. (M)	10	3	3
Temp. of the reagents (°C)			
Stripping sol.	25	45	45
The others	25	25	25
Solv./feed (flowrate ratio)	2.0	2.3	2.1
Stripping/solv. (flowrate ratio)	1.2	1.6	1.5
Number of stripping stages	12	14	14

The oxidation of inextractable Np(V) by HNO_3 is described by the following equilibrium reaction.

$$2NpO_2{}^{+} + NO_3{}^{-} + 3H^{+} \leftrightarrow 2NpO_2{}^{2+} + HNO_2 + H_2O \qquad (1)$$

This equation shows that higher $[HNO_3]$ brings more oxidation of inextractable Np(V) to extractable Np(VI). In addition, the distribution ratio of Np(VI) by TBP in the extraction section increases with $[HNO_3]$. These indicate that the U–Pu–Np co-recovery process with high $[HNO_3]$ feed solution obtained from the crystallization process is favourable to the effective Np(V) oxidation to Np(VI) and its extraction. Under the same experimental condition except for $[HNO_3]$ in the scrubbing section, the leakage of Np to the raffinate seemed to decrease with increasing of $[HNO_3]$ in the scrubbing solution. It is, therefore, expected that high $[HNO_3]$ scrubbing solution also gives the similar good effect on the Np oxidation and extraction.

The increase of $[HNO_2]$ shifts the equilibrium (1) to the left hand, which means the suppression of Np(V) oxidation, while high $[HNO_2]$ increases the reaction rate of Np(V) by HNO_3 (reaction (1)) as given by the following equation [14].

$$-\frac{d[Np(V)]}{dt} = \frac{1.8 \times 10^{-3}[H^{+}][NO_3{}^{-}][HNO_2][Np(V)]}{[HNO_2] + [Np(V)]} \qquad (2)$$

Although the analysis of $[HNO_2]$ was not carried out in our experiments, the experimental results showed the almost all the Np could be extracted under a certain extraction condition as stated above. Our recent simulation study with MIXSET-X showed that the effect of HNO_2 on the Np extraction was remarkable in low $[HNO_3]$ feed solution, but under high $[HNO_3]$ condition almost all the Np could be extracted regardless of $[HNO_2]$ [15]. These experimental and calculation results implies that $[HNO_2]$ might not have so large influence on the Np oxidation and extraction in the U–Pu–Np co-recovery process with high $[HNO_3]$ feed solution although further quantitative

Table 6
Leakage of U, Pu and Np to the raffinate in the extraction section

	Raffinate	Loaded solvent
Run1		
U (%)	<0.1[a]	>99.9
Pu (%)	<1.2[b]	>98.8
Np (%)	0.9	99.1
Run2		
U (%)	<0.1[a]	>99.9
Pu (%)	<0.7[b]	>99.3
Np (%)	1.5	98.5
Run3		
U (%)	<0.1[a]	>99.9
Pu (%)	0.03	99.97
Np (%)	1.0	99.0

[a] Under the analytical lower limit ([U] in samples <0.03 g/L).
[b] Under the analytical lower limit ([Pu] in samples <0.2 g/L).

Table 7
Leakage of U, Pu and Np to the spent solvent in the stripping section

	Product	Spent solvent
Run1		
U (%)	70.5	29.5
Pu (%)	>96.4	<3.6[a]
Run2		
U (%)	>99.7	<0.3[b]
Pu (%)	>96.5	<3.5[a]
Run3		
U (%)	>99.9	<0.1[b]
Pu (%)	99.9	0.1
Np (%)	>98.9	<1.1[c]

[a] Under the analytical lower limit ([Pu] in samples <0.2 g/L).
[b] Under the analytical lower limit ([U] in samples <0.03 g/L).
[c] Under the analytical lower limit ([^{237}Np] in samples <7.4 Bq/mL).

Table 8
Effect of the stripping flowrate and the temperature on the U and Pu leakage to the spent solvent in the stripping section

Stripping/solv. (flowrate ratio)		Temp. of stripping sol. (°C)	
		25	45
1.2	U (%)	27.9	8.3
	Pu (%)	<0.1	0.2
1.4	U (%)	8.7	<0.1
	Pu (%)	<0.1	<0.1

Calculation results based on the flowsheet of Run1.

Table 9A
The value of $t_{\text{Pu polymer}}$ in the stripping section without adjusting solution

	Stage no.				
	4	6	8	10	12
$[HNO_3]$ (M)	0.15	3.0×10^{-2}	2.1×10^{-2}	2.0×10^{-2}	2.0×10^{-2}
[Pu] (g/L)	9.9	3.2	0.15	1.4×10^{-2}	1.2×10^{-5}
T (°C)	35	36.5	38.1	39.6	41.4
$t_{\text{Pu polymer}}$ (min)	2.2×10^{2}	0.71	15	1.9×10^{4}	2.9×10^{7}

Table 9B
The value of $t_{\text{Pu polymer}}$ in the stripping section with adjusting solution

	Stage no.				
	4	6	8	10	12
$[HNO_3]$ (M)	0.25	0.15	0.14	2.1×10^{-2}	2.0×10^{-2}
[Pu] (g/L)	9.6	3.3	0.28	1.5×10^{-2}	4.4×10^{-5}
T (°C)	35	36.5	38.1	39.6	41.4
$t_{\text{Pu polymer}}$ (min)	2.9×10^{3}	1.1×10^{3}	4.2×10^{4}	5.8×10^{2}	4.0×10^{6}

Calculation results based on the flowsheet of Run2.

investigation with analyzing $[HNO_2]$ should be required experimentally.

3.3. Uranium and plutonium behaviour in the stripping section

Table 7 shows the ratios of U and Pu leaked to the solvent in the U–Pu–Np co-recovery experiments. In the experiment using the stripping solution at R.T., the stripping of U was insufficient and about 30% of U leaked to the solvent. For preventing U leakage to the solvent, several methods can be considered, e.g. (1) increasing the stripping flowrate (increasing the ratio of the stripping flowrate to the solvent flowrate), (2) increasing the temperature in the stripping section and (3) decreasing $[HNO_3]$ in the stripping section. Every method, however, has its own problem; the method (1) increases the volume of stripping reagent (liquid waste), the method (2) increases the Pu (Pu(IV)) leakage to the solvent, and the method (3) increases the possibility of the Pu polymer formation in the stripping section. It is, therefore, desirable for preventing U leakage to combine these methods effectively. Table 8 shows the effect of the stripping flowrate (the ratio of the stripping flowrate to the solvent flowrate) and the temperature in the stripping section on the U and Pu leakage to the solvent, which was calculated by MIXSET-X. It is recognized that high temperature prevents the U leakage whereas that brings the considerable leakage of Pu. This Pu leakage, however, can be suppressed by slightly increasing the stripping flowrate as shown in the calculation results.

The Pu polymerization rate in HNO_3 has been reported by Scoazec et al., as the following experimental equation which estimates the time for changing 2% of Pu(IV) in the solution to the Pu polymer ($t_{\text{Pu polymer}}$) [16].

$$t_{\text{Pu polymer}}\,(\text{h}) = 7.66 \times 10^{-6}[\text{Pu}]^{-1.6}[\text{HNO}_3]^{4.6} \exp\frac{12300}{T\,(\text{K})} \quad (3)$$

Table 9A summarizes $t_{\text{Pu polymer}}$ estimated for the each stage in the stripping section using only 0.02 M HNO_3 stripping solution (without any addition of HNO_3 for adjusting the acidity). Under this stripping condition, the calculation results indicated the precaution against the possibility of the Pu polymer formation in some stages although the values of $t_{\text{Pu polymer}}$ were a little longer than the residence time (~tens second) in these stages. In the U–Pu–Np co-recovery flowsheet, therefore, additional HNO_3 is supplied into an appropriate stage for adjusting (increasing) the acidy, which brings the sufficient $t_{\text{Pu polymer}}$ for avoiding the Pu polymer formation (see Table 9B).

No formation of the Pu polymer in the stripping section was also confirmed by the U–Pu–Np co-recovery experiments using 0.02 M HNO_3 stripping solution with additional HNO_3 (adjusting solution).

As shown by these experimental and calculation results in the U–Pu–Np co-recovery process, it can be concluded that U and Pu can be stripped efficiently by choosing the appropriate stripping condition for preventing the U and Pu leakage to the solvent and avoiding the Pu polymer formation.

4. Conclusions

The behaviour of Pu and other actinides in some characteristic processes of the NEXT process, i.e. the crystallization process and the U–Pu–Np co-recovery process, was investigated with referring to the experimental results obtained in our experimental facility, CPF.

Plutonium showed the different behaviour in the crystallization process according to its valence in the dissolver solution. The existence of Pu(VI) in the dissolver solution brought the co-crystallization of Pu(VI) with U, and low DF_{Pu}. Such co-crystallization could be prevented by adjusting the Pu valence to Pu(IV). In the crystallization with the dissolver solution containing Pu(IV), meanwhile, DF_{Cs} showed significant low values, which might be caused by the formation and crystallization of some kinds of Cs compounds with Pu(IV). It should be further required for high DF_{Cs} to clarify the process condition without the formation of such Cs–Pu compounds, and/or to develop some purification methods for the U crystal contaminated with these compounds.

In the U–Pu–Np co-recovery process with high [HNO_3] feed solution obtained from the crystallization process, it was confirmed experimentally that almost all the Np could be extracted with U and Pu in the extraction section, which was also supported by the calculation with the simulation code MIXSET-X taking account of the Np redox reaction and its reaction rate. In the stripping section using diluted HNO_3 as the stripping solution without any reductant nor complexant, some considerable parameters, i.e. temperature, stripping flowrate, and acidity of stripping solution, were pointed out for preventing the leakage of U and Pu to the solvent and avoiding the Pu polymer formation. Under the appropriate stripping condition taking account of these parameters, the calculation and experimental results showed that these elements could be stripped efficiently.

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