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Solvent extraction studies of protactinium for its recovery from short-cooled spent fuel and high-level waste solutions in thorium fuel cycle using diisobutyl carbinol (DIBC) as extractant

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ABSTRACT

Conditions have been optimized for selective removal of protactinium by solvent extraction from short-cooled spent fuel dissolver solution and from high-level waste solution under simulated conditions employing diisobutyl carbinol (DIBC) as extractant. These studies include: effect of acidity, extractant and thorium concentration on the extraction behavior of protactinium. Selectivity of the extractant for protactinium has been investigated with respect to U, Pu, Th, and the fission/activation products. These studies suggest that 40% DIBC (~2.02 M) solution in *n*-dodecane can be employed for the recovery of protactinium from short-cooled spent fuel dissolver solution and from high-level waste solutions. Extraction of Pa increased with aqueous phase acidity due to the formation of protonated species of DIBC, $[H(DIBC)_{1-2}]^+$ and anionic $Pa(OH)_2(NO_3)_4]^-$ species. There was an enhancement in Pa extraction at lower acidities ($\leq$4 M HNO_3) with increased Th concentration (up to 200 g/l) beyond which a decrease was observed. At 200 g/l Th in 4 M HNO_3, the separation factor (SF) values of Pa over U and Th are ~100, >10^5, respectively. DIBC displays good fission/activation product decontamination under the conditions of these studies.

Keywords: Protactinium; Thorium; AHWR; Reprocessing; High-Level waste; Diisobutyl carbinol

1. Introduction

Thorium is 3 to 4 times more abundant than uranium and is an easily exploitable resource for nuclear power in many countries. Unlike natural uranium, which contains ~0.7% fissile ^{235}U isotope, natural thorium does not contain any 'fissile' material and is made up of the 'fertile' ^{232}Th which can be converted to 'fissile' ^{233}U, thereby enlarging the fissile material resources. Thorium fuels and fuel cycles are particularly relevant to countries like India having large thorium deposits (approx 600,000 tons) but very limited uranium reserves (approx 80,000 tons) to sustain its long-term nuclear power programme [1]. Several thermal reactor systems have been proposed for thorium utilization in India's nuclear power program. Amongst them, the development of advanced heavy water reactor (AHWR) is relatively on an advanced stage. Other important options for thorium utilization program are: (a) Molten Salt Breeder Reactor (MSBR), (b) Aqueous Suspension Reactor (ASR) [2–5]. These reactors are expected to work on circulating fluid fuel (molten fluoride/slurry of thorium

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oxide in water) and provide an option of on-line fuel reprocessing, which removes fission products and minimizes the losses of neutrons. In thorium breeding scenarios, the intermediate product ^{233}Pa would be removed from the reactor and allowed to decay into highly pure ^{233}U. The reprocessing of MSBR is rather complex and the effectiveness of this reprocessing rests mainly on the extraction and storage of the protactinium away from the neutron flux so as to avoid the production of ^{234}U by neutron capture. The half-life of ^{233}Pa is 27 d and its extraction has to be markedly faster if it is to be efficient.

Currently, AHWR appears most promising nuclear power reactor system for the utilization of ^{233}U as the fissionable material and thorium as a fertile material. Many processes have been developed in the past for the separation of ^{233}U from Th, ^{233}Pa and the fission products and from each other [6–11]. However, THOREX process employing 5% (v/v) tri-*n*-butyl phosphate (TBP) dissolved in aliphatic diluent has been evaluated in a greater detail form Indian perspective [11]. Recently, a modified flow sheet has been proposed for the recovery of ^{233}U, Pu from AHWR spent fuel using 5% TBP/*n*-dodecane as the solvent [12]. However, Pa behavior under process conditions is not well understood. Therefore, there is a need to develop processes for selective extraction of ^{233}Pa from spent fuels in these reactors. AHWR spent fuels are expected to initially reach a burn up of 15,000 MWd/T which will be enhanced up to 24,000 MWd/T under equilibrium conditions. It would be necessary to reprocess some of the fuel bundles after short-cooling every year to achieve a burn up of ~40,000 MWd/Te. This again necessitates the knowledge of extraction behavior of protactinium under process conditions [2]. A process to recover ^{233}Pa prior to the processing of short-decayed fuel would minimize inventory charges of stored fissionable material. The separated ^{233}Pa, which will decay to isotopically pure ^{233}U, can reduce the volume of radioactive product to be stored. In addition, this would lead to significant decrease in the activity of the solutions in the thorium-uranium recovery procedure. The production of ^{233}Pa and ^{231}Pa in nuclear reactor can be shown by the following reactions:

1. $^{232}\mathrm{Th}(\mathrm{n},\ 2\mathrm{n})\ ^{231}\mathrm{Th} \overset{\bar{\beta}}{\rightarrow}\ ^{231}\mathrm{Pa}(\mathrm{n},\ \gamma)\ ^{232}\mathrm{Pa} \overset{\bar{\beta}}{\rightarrow}\ ^{232}\mathrm{U}$

2. $^{232}\mathrm{Th}(\mathrm{n},\gamma)\ ^{233}\mathrm{Th} \overset{\bar{\beta}}{\rightarrow}\ ^{233}\mathrm{Pa} \overset{\bar{\beta}}{\rightarrow}\ ^{233}\mathrm{U}(\mathrm{n},\gamma)\ ^{234}\mathrm{U}$
$\overset{\alpha}{\rightarrow}\ ^{230}\mathrm{Th}(\mathrm{n},\gamma)\ ^{231}\mathrm{Th} \overset{\bar{\beta}}{\rightarrow}\ ^{231}\mathrm{Pa}(\mathrm{n},\gamma)\ ^{232}\mathrm{Pa} \overset{\bar{\beta}}{\rightarrow}\ ^{232}\mathrm{U}$

The (*n*, 2*n*) reactions encountered during the irradiation of thorium lead to the formation of long-lived ^{231}Pa and relatively short-lived ^{232}U (~70 y) with its hard beta, gamma emitting daughter products. Thus, ^{233}U produced in nuclear reactor is contaminated with ^{232}U and the level of contamination depends on the isotopic composition of initial thorium fuel, the burn-up and the neutron spectrum encountered in the reactor. The presence of long-lived alpha emitter ^{231}Pa (3.3×10^4 y) and significant amount of aluminium along with fluoride adds new dimensions in high-level waste (HLW) management of AHWR spent fuel. Pentavalent ^{231}Pa (PaO^{3+}) is of special concern in AHWR-HLW as it is capable to migrate much more in water/soil compared to other ions. Therefore, there is need to establish a suitable process employing an appropriate solvent for this specific task. Even though production of minor actinide in Th-^{233}U fuel is orders (~10^2–10^6 time) times less compared to those in U-^{235}U fuel, production of minor actinides in AHWR is expected to be higher than that in PHWR due to the use of ^{239}Pu in AHWR fuel which leads to higher generation of Am and Cm isotopes [13,14]. Typically, the amount of ^{231}Pa produced in (Th-^{233}U)O_2 MOX fuel is expected to be 3 g/Te at 20,000 MWd/Te of burn-up and its concentration in AHWR-HLW is expected to be ~1 mg/l [13]. In addition to above process requirements, reliable high precision measurements of ^{231}Pa have widespread implications in the study of earth sciences. ^{231}Pa along with its parent nuclide ^{235}U, is suitable for U/Pa dating of continental or marine carbonates up to ages of approximately 150 ky and ^{231}Pa is an important tracer in Paleoceanography [15].

The behavior of protactinium ions in solution is very complex because of the tendency of the element to hydrolyze and polymerize and large number of ionic species being formed. The simultaneous presence of Pa ions with different charges, polymerization etc. interferes with the choice of a suitable method for the determination of Pa and its separation from other elements. Solvent extraction has been successfully utilized for the separation of Pa in several studies. Recovery of Pa from aqueous solutions has been carried out using several extractants such as trilauryl amine (TLA), TBP, methyl isobutyl ketone (MIBK), diisopropyl ketone/carbinol (DIPK/DIPC), and thenoyltrifluoroacetone (HTTA) [16–19]. Sill proposed procedures for the preparation of ^{233}Pa tracer mainly using diisobutyl carbinol (DIBC: 2,6 dimethyl 4-heptanol) [20]. In our laboratory, the distribution behavior of Pa was investigated in the presence of varying amounts of Th (up to 250 g/l) and U (20 g/l) from aqueous nitric acid solutions (1–6 M) employing a 1 M solution of di-2-ethylhexyl-isobutylamide (D2EHIBA) in *n*-dodecane [21]. D2EHIBA was identified as promising alternative of TBP for selective recovery of ^{233}U from irradiated thorium. Other extractants used to study the extraction behavior of Pa(V) from various aqueous solutions are diethylhexyl phosphoric acid (HDEHP), and tricaprylylmethyl ammonium chloride (Aliquat 336) in toluene [22]. However, DIBC deserves special mention in view of its selective nature towards Pa from acid solutions [23].

A brief survey of the literature of protactinium recovery and consideration of possible application of the findings to the development of AHWR reprocessing scheme indicated that further study was needed for the extraction of protactinium with as a head-end process. This paper deals with the extraction behavior of protactinium, thorium and uranium employing DIBC as an extractant. Conditions for extracting protactinium from solutions of irradiated thorium by DIBC have been arrived at in laboratory tracer experiments. The extraction data have been used to optimize the conditions for selective extraction of protactinium over U, Th, Pu, and fission/activation products.

2. Experimental

DIBC (Aldrich), *n*-dodecane (Lancaster) were used as received. ^{233}U tracer (~10^{-4}M) was purified by anion exchange procedure [24]. The purity was checked alpha and gamma ray spectrometry. ^{233}Pa was separated from irradiated ^{232}Th (in APSARA reactor) by a radiochemical technique using DIBC as the extractant [25]. Its purity was checked by gamma ray spectrometry (Characteristic peak: 311.9 keV). Thorium (nitrate from) obtained from Indian Rare Earths Limited, Mumbai, was used wherever necessary. The inner sides of the borosilicate glass tubes (used for solvent extraction studies) were coated using 5% solution of dimethyl dichlorosilane (DMDCS) in toluene prior to Pa to avoid its loses on the walls [26]. The solution was then drained out at least after 3 h and dried before their use in solvent extraction studies.

Equal volumes (0.5 or 1.0 ml) of the pre-equilibrated organic phases and the aqueous phases under desired conditions were kept for equilibration in a water bath for 30 min at 25°C. Acid uptake in the organic phase was carried out by alkalimetry in neutralized water-ethanol medium using standard alkali and phenolphthalein as indicator. Gamma spectrometry was carried out for measuring ^{233}Pa activity. ^{233}U was estimated by liquid scintillation counting using suitable aliquots (generally 50 µl). The composition of the scintillation medium was: 0.7% (w/v) 2,5-diphenyloxazole (PPO), 0.03% (w/v) 1,4-di-[2-(5-phenyloxazoyl)]-benzene (POPOP), 1% (w/v) trioctyl phosphine oxide (TOPO), and 10% (w/v) naphthalene dissolved in one litre of dioxane. Thorium estimation was carried out by ethylenediamine tetraacetic acid (EDTA) complexometric titration employing xylenol orange as indicator, and by spectrophotometry using Thoron (2-(2-hydroxy-3,6-disulpho-1-napthylazo) benzene arsenic acid) as chromophore and V 53 UV–vis spectrophotometer (from JASCO, Japan) [27]. The distribution ratio of the metal ions (D_M) was defined as the ratio of (i) concentrations for thorium, and (ii) activities in a given volume for Pa, U, fission/activation products in the organic phase to that in the aqueous phase.

$$D_M = [M]_{\mathrm{org}}/[M]_{\mathrm{aq}} \tag{1}$$

Material balance and the reproducibility of the distribution data was within error limits (±5–10%).

3. Results and discussion

3.1. Acid uptake behavior of DIBC

The understanding of acid uptake behavior of an extractant is important in devising process flow sheet. In this context, nitric acid extraction studies were carried out in two different conditions viz. (a) at different acidities (1–6 M HNO_3) using 100% DIBC (5.1 M), and (b) at different DIBC concentrations (10–100%; 0.51–5.1 M) in *n*-dodecane at 4 M HNO_3. The acid uptake behavior by DIBC (A) can be represented as:

$$\mathrm{H}^+_{\mathrm{aq}} + \mathrm{NO}^-_{3\,\mathrm{aq}} + n\mathrm{A}_{\mathrm{org}} \xrightleftharpoons{K_H} \mathrm{HNO}_3 \cdot n\mathrm{A}_{\mathrm{org}} \tag{2}$$

$$K_H = \frac{[\mathrm{HNO}_3 \cdot n\mathrm{A}]_{\mathrm{org}}}{[\mathrm{H}^+]_{\mathrm{aq}}[\mathrm{NO}_3^-]_{\mathrm{aq}}[\mathrm{A}]^n_{\mathrm{free,org}}} \tag{3}$$

where $[\mathrm{H}^+]_{\mathrm{org}} = [\mathrm{HNO}_3{\cdot}nA]_{\mathrm{org}}$ and $[\mathrm{H}^+]_{\mathrm{aq}} = [\mathrm{NO}_3^-]_{\mathrm{aq}}$

$$K_H = \frac{[\mathrm{H}^+]_{\mathrm{org}}}{[H^+]^2_{\mathrm{aq}}[\mathrm{A}]^n_{\mathrm{free,\,org}}} \tag{4}$$

Taking logarithm on both sides of Eq. (4) and rearranging

$$\log\left[\mathrm{H}^+\right]_{\mathrm{org}} - 2\log\left[\mathrm{H}^+\right]_{\mathrm{aq}} = n\log\left[\mathrm{A}\right]_{\mathrm{free}} + \log K_H \tag{5}$$

$[\mathrm{H}^+]_{\mathrm{aq}}$ was calculated from the titre value (T) and dissociation constant of HNO_3 (K_a).

$$K_a = \frac{[\mathrm{H}^+]_{\mathrm{aq}}[\mathrm{NO}_3^-]_{\mathrm{aq}}}{[\mathrm{HNO}_3]_{\mathrm{aq}}} = \frac{[\mathrm{H}^+]^2_{\mathrm{aq}}}{T - [\mathrm{H}^+]_{\mathrm{aq}}} \tag{6}$$

where $T = ([\mathrm{H}^+]_{\mathrm{aq}} + [\mathrm{HNO}_3]_{\mathrm{aq}})$ in aqueous phase. Therefore

$$\begin{gathered}\left[\mathrm{H}^+\right]_{\mathrm{aq}}^{\;2} + K_a \cdot \left[\mathrm{H}^+\right]_{\mathrm{aq}} - K_a \cdot T = 0 \\ \left(K_a = 23.5,\ \text{literature value}\right)\end{gathered} \tag{7}$$

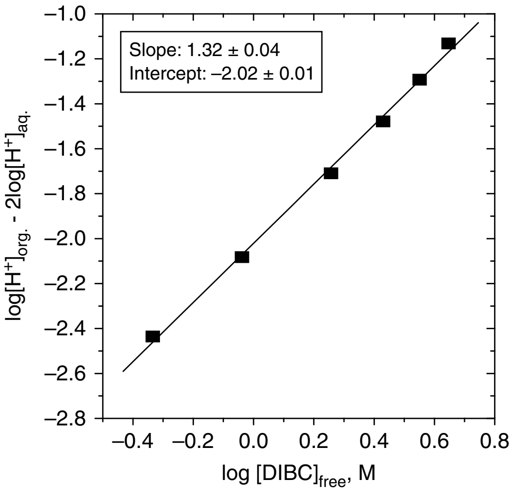

Fig. 1. K_H determination of DIBC; $[HNO_3]$: 4 M; Diluent: *n*-dodecane; *T*: 25°C.

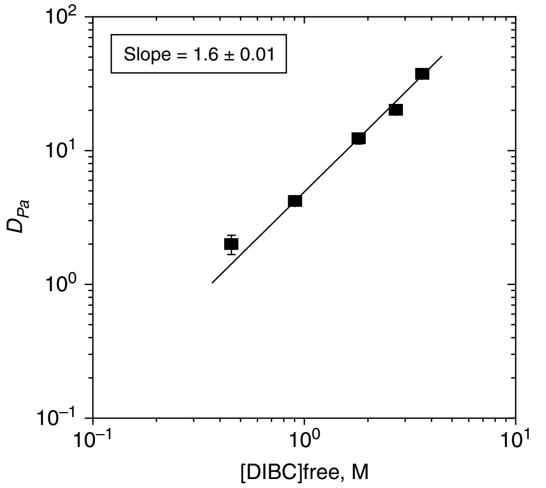

Fig. 2. Variation of D_{Pa} with DIBC conentration; Diluent : *n*-dodecane; $[HNO_3]$: 4 M; *T*: 25°C.

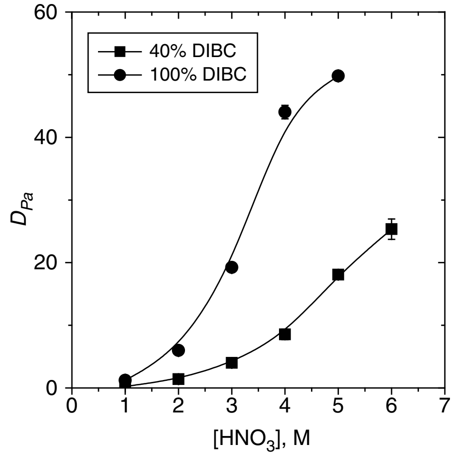

Fig. 3. D_{Pa} as a function of nitric acid concentration; Diluent: *n*-dodecane; *T*: 25°C.

$[H^+]_{aq}$ was obtained by solving the above quadratic equation, and $[DIBC]_{free} = [DIBC]_{initial} - [H^+]_{org}$ (assuming one HNO_3 was bound to one DIBC molecule). From Eq. (5), it can be seen that a plot of $\{\log[H^+]_{org} - 2\log[H^+]_{aq}\}$ against $\log[DIBC]_{free}$ should give a straight line with slope n i.e. the number of DIBC molecules bound to each HNO_3 in the adduct formed and K_H value can be found from the intercept.

For 100% DIBC, the acid uptake in the organic phase increased gradually from 0.05 M to 1.5 M HNO_3 with increased initial aqueous phase acidity from 1 to 6 M HNO_3. On the other hand, DIBC concentration variation experiments for Pa extraction at 4 M HNO_3 indicated towards the formation of predominantly 1:1 species (slope: 1.32) (Fig. 1). Similar observations have been reported during the extraction of HCl by DIBC [28]. The acid uptake constant, K_H, was calculated from the intercept value as 0.01.

3.2. Effect of DIBC concentration

Fig. 2 suggests the involvement of ~1–2 DIBC molecules during Pa extraction process through the formation of ion-paired complex [28]. Similar solvation has been observed during nitric acid extraction. Therefore, the nitric acid is extracted as an ion pair of the type $[H(DIBC)_{1-2}]^+ \cdot [NO_3^-]$ [28]. The activity of this species increases with increased nitric acid concentration and that of free DIBC decreases. Therefore, it is logical to conclude that Pa forms complex with protonated DIBC rather than free DIBC. Also, equal solvation numbers of nitric acid and Pa-species indicate that they interact in 1:1 ratio. Thus, protactinium is extracted as monobasic complex of general formula $HPa(OH)_m(NO_3)_n \cdot$ 1–2 DIBC, with $m + n = 6$, and $n \geq 1$. Hardy et al. have reported that the hydrolyzed cationic, neutral and anionic species $[Pa(OH)_2(NO_3)_4]^-$ of Pa in nitric acid are in equilibrium and depending on the aqueous phase acidity, the interchange between them is fast [18]. Fig. 3 shows the variation of D_{Pa} values with aqueous phase acidity for pure DIBC (100%; 5.05 M) and 40% (2.02 M) DIBC/*n*-dodecane. As expected, D_{Pa} values are significantly higher for 100% DIBC in the entire range of acidity. However, 40% DIBC/*n*-dodecane solution was chosen for Pa recovery experiments in view of the relatively lower concentration of ^{233}Pa (in dissolver solution) and ^{231}Pa (in HLW solution), and sufficiently high D_{Pa} value (8.55 ± 0.83).

3.3. Effect of thorium

Fig. 4 shows the variation of D_{Pa} with increasing concentrations of Th (up to 200 g/l) at different acidities. Generally, there is an increase in D_{Pa} values with increased

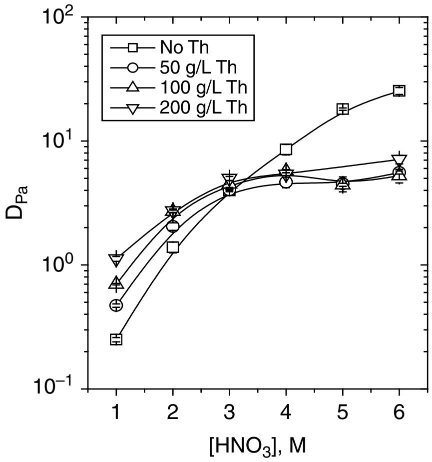

Fig. 4. Effect of Th on D_{Pa} as a function of acidity; Extractant: 40% DIBC/*n*-dodecane ; *T*: 25°C.

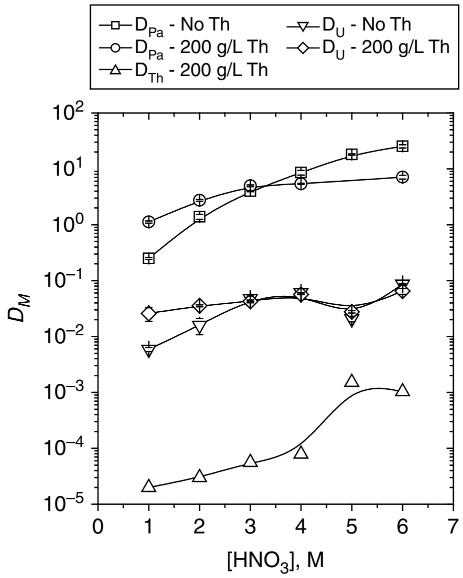

Fig. 5. Effect of Th on D_{Pa}, D_U and D_{Th} at different acidities; Extractant: 40% DIBC/*n*-dodecane ; *T*: 25°C.

aqueous phase acidity which can be attributed to the formation of protonated species of DIBC $[H(DIBC)_{1-2}]^+$ and anionic $Pa(OH)_2(NO_3)_4]^-$ species. This observation is further supported by enhanced extraction of Pa at lower acidities (up to 4 M) with increased Th concentration (up to 200 g/l). However, there is a decrease in D_{Pa} values beyond 4 M HNO_3 indicating a decrease in the activity of $[H(DIBC)_{1-2}]^+$ species due to partial loading of organic phase with thorium even though the distribution ratio values are too low (Fig. 5). Typically, in the presence of 200 g/l at 4 M HNO_3, the separation factor (SF) values of Pa over U (D_{Pa}/D_U) and Th (D_{Pa}/D_{Th}) are ~100, >10^5, respectively. The relatively lower SF value of Pa over U was attributed to latter's tendency to form anionic species which is more favored at higher nitrate concentrations.

3.4. Fission/activation products extraction behavior

Extraction studies were also performed to check the selectivity of DIBC over the fission products during the recovery of protactinium from short-cooled AHWR spent fuel dissolver solution or from high-level waste solution generated after the reprocessing step. These studies were carried out using 100 and 200 g/l Th solutions at 4 M HNO_3 spiked with an aliquot of a HLW sample generated after the reprocessing of pressurized heavy water reactor (PHWR) spent fuel with burn up of ~6500 MWD/T employing the PUREX process as the aqueous phase and 40% DIBC/*n*-dodecane solution as solvent. After equilibration, the phases are separated and assayed for individual fission/activation product (M: ^{137}Cs, ^{144}Ce, ^{106}Ru, ^{60}Co etc.) activity using gamma spectrometry. The distribution ratio (D_M) values for these radionuclides were of the order of ~10^{-4} suggesting better decontamination of Pa from fission/activation products in the presence of macro concentrations of thorium in the feed/waste solutions.

4. Conclusions

Solvent extraction studies have been carried out to optimize the conditions for Pa extraction from short-cooled dissolver and HLW solutions using DIBC as the extractant. The study suggests that protactinium is extracted as monobasic complex of general formula $HPa(OH)_m(NO_3)_n \cdot 1\text{–}2$ DIBC, with $m + n = 6$, and $n \geq 1$. Based on ligand variation studies under pure tracer conditions, 40% DIBC/*n*-dodecane (2.02 M) solution was chosen for Pa recovery (D_{Pa}: 12, tracer condition) from dissolver/HLW solutions. Extraction of Pa increases with aqueous phase acidity due to the formation of protonated species of DIBC, $[H(DIBC)_{1-2}]^+$ and anionic $Pa(OH)_2(NO_3)_4]^-$ species. There was an enhancement in Pa extraction at lower acidities (up to 4 M) with increased Th concentration (up to 200 g/l); the decrease in D_{Pa} values beyond 4 M HNO_3 was attributed decreased activity of $[H(DIBC)_{1-2}]^+$ species and to partial loading of organic phase with thorium. At 200g/l Th in 4 M HNO_3, the separation factor (SF) values of Pa over U and Th are ~100, >10^5, respectively. DIBC displays good fission/activation product decontamination under the conditions of these studies. These studies, therefore, suggest that 40% DIBC (2.02 M) solution *n*-dodecane can be used for Pa recovery from dissolver solution of short-cooled spent fuel as well as from HLW solution. However, radiolytic stability of the solvent under process conditions also needs to be examined.

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