Separation Science and Technology
Publication details, including instructions for authors and subscription information:
http://www.informaworld.com/smpp/title~content=t713708471

Recent R&D Studies Related to Coprocessing of Spent Nuclear Fuel Using *N,N*-Dihexyloctanamide

P. N. Pathak[a]; D. R. Prabhu[a]; A. S. Kanekar[a]; V. K. Manchanda[a]
[a] Radiochemistry Division, Bhabha Atomic Research Centre, Trombay, Mumbai, India

To cite this Article Pathak, P. N. , Prabhu, D. R. , Kanekar, A. S. and Manchanda, V. K.(2009) 'Recent R&D Studies Related to Coprocessing of Spent Nuclear Fuel Using *N,N*-Dihexyloctanamide', Separation Science and Technology, 44: 15, 3650 — 3663
To link to this Article: DOI: 10.1080/01496390903183147
URL: http://dx.doi.org/10.1080/01496390903183147

Separation Science and Technology, 44: 3650–3663, 2009

ISSN: 0149-6395 print/1520-5754 online
DOI: 10.1080/01496390903183147

Recent R&D Studies Related to Coprocessing of Spent Nuclear Fuel Using *N,N*-Dihexyloctanamide

P. N. Pathak, D. R. Prabhu, A. S. Kanekar, and V. K. Manchanda
Radiochemistry Division, Bhabha Atomic Research Centre, Trombay,
Mumbai, India

Abstract: The PUREX process has undergone several modifications to address the issues of high burn up, fewer solvent extraction cycles, and reduced waste arisings. Advanced fuel cycle scenarios have led to a renewed international interest in the development of separation schemes for co-recovering U/Pu from spent fuels. Completely incinerable *N,N*-dihexyloctanamide (DHOA) has been identified as a promising candidate for the reprocessing of spent fuels. Batch extraction studies were carried out to evaluate DHOA and TBP for the coprocessing (co-extraction and co-stripping) of U and Pu from spent fuel under varying concentrations of nitric acid and of uranium as well as under simulated pressurized heavy water reactor spent fuel feed conditions. At 50 g/L U in 4 M HNO_3, D_{Pu} values for 1.1 M DHOA and 1.1 M TBP solutions in *n*-dodecane were 7.9 and 3.8, respectively. In contrast, significantly lower D_{Pu} value at 0.5 M HNO_3 (4×10^{-3}) for DHOA as compared to TBP (4×10^{-2}) suggested that it was a better choice for coprocessing of spent nuclear fuel. This behavior was attributed to the change in stoichiometry of extracted species at lower acidity vis-a-vis the higher acidity. These studies suggest that plutonium fraction can be enriched with respect to uranium contamination in the product stream. DHOA displays better extraction behavior of plutonium and stripping behavior of uranium under simulated feed conditions. DHOA appears distinctly better than TBP with respect to fission product/structural material decontamination of U/Pu.

Keywords: Coprocessing, dihexyloctanamide, plutonium, PUREX, TBP, uranium

Received 26 December 2008; accepted 10 June 2009.

Address correspondence to V. K. Manchanda, Radiochemistry Division, Bhabha Atomic Research Centre, Trombay, Mumbai 400 085, India. Fax: (+91) 22 25505151. E-mail: vkm@barc.gov.in

INTRODUCTION

To achieve optimum peaceful utilization of fissile materials, separation scientists will have to ensure the reprocessing of spent fuels, and afford effective defenses against proliferation. The proliferation resistant fuel cycle has led to a renewed international interest in the development of new separation schemes for coprocessing of U/Pu present in dissolver solution (1–6). This has an additional advantage with respect to criticality problems. The coprocessing option appears particularly promising for the reprocessing of Pu based fast reactor spent fuels with lower uranium component. Therefore, efforts are being made worldwide on a whole range of processes that must be taken up for reprocessing–recycling goals. This includes the development of a reprocessing scheme for spent nuclear fuel to get mixed uranium-plutonium oxide as the product can be used directly in processes for manufacturing mixed oxide (MOX) nuclear fuels. At the Savannah River Laboratory, a coprocessing flowsheet was proposed for recovering and purifying light water reactor (LWR) fuels to achieve partial partitioning of uranium and plutonium to eliminate streams with pure plutonium. Hydroxylamine nitrate was the reductant in these tests. Plutonium was enriched with respect to uranium concentrated by factors of 6 to 27.4 (7). Another approach for recycling spent nuclear fuels with the aim of minimizing the number of stages and the risk of proliferation of nuclear materials was coextraction of uranium, plutonium, and a part of fission products (0.1 to 10%) (8). However, the plutonium product stream obtained after the partitioning step would contain a part of extracted uranium and most of the radioactive fission products extracted during the extraction step. This plutonium product stream was proposed to be processed by a sol-gel method in order to obtain plutonium-uranium-fission products mixed oxide, which would be subsequently used to manufacture fresh nuclear fuels. Although the presence of neutron absorbing fission products in fresh nuclear fuels does not pose problem in fast reactor operations, a substantial increase in fissile content is required for thermal light water reactors. This new type of fuel has to be tested in a reactor before its actual use. Also, the presence of radioactive fission products at all stages in spent fuel reprocessing and fresh fuel production chains requires additional radiation protection systems in the operating plants. This option appears to be expensive in view of the additional shielding requirements due to high dose and evaluation studies on recycled fuel.

In this context, Baron et al., proposed a new scheme for reprocessing spent nuclear fuel, which should allow effective decontamination of uranium and plutonium from the fission products. The final product is uranium-plutonium oxide that can be directly used for the manufacture of MOX nuclear fuels after suitable adjustment of Pu to U ratio using existing Pu stocks (8). Unlike PUREX process, the proposed scheme aims at

i. co-extraction of U(VI) and Pu(IV) from the feed solution,
ii. scrubbing of fission products from the loaded organic phase using nitric acid, and
iii. the partial separation of plutonium from uranium (during reductive stripping step) and to keep it together throughout for all subsequent steps including mixed oxide fuel fabrication.

The uranium present in the organic phase is stripped separately by dilute nitric acid and kept separately. The extractant may be a trialkyl phosphate, such as tri-*n*-butyl phosphate (TBP), triisobutyl phosphate (TiBP), or a triisoamyl phosphate (TiAP) in *n*-dodecane (9). However, the preferred extractant is 30% TBP/*n*-dodecane being used in the PUREX process. The salient features of this process are:

a. it never allows plutonium to be left without uranium minimizing the risk of plutonium being misappropriated for military purposes, and
b. it provides mixed uranium-plutonium oxide powder as the product that can be used directly for the manufacture of MOX nuclear fuels for fast reactors.

However, decades of experience in nuclear fuel reprocessing based on the PUREX process has identified few problems of TBP essentially arising due to the degradation products of TBP, which affect the extractive and stripping behavior of U/Pu and its hydrodynamic properties such as viscosity, density and phase disengagement time (10,11). These shortcomings are of serious concern for the reprocessing of short-cooled thermal reactor fuels or fast reactor spent fuels. In addition, TBP introduced for reprocessing leaves behind ~20 volume% of phosphoric acid which has to be neutralized and thereafter either bituminized or converted into phosphate glass (12). Evaluation of alternate extractants is therefore desirable, which can overcome these problems.

N,N-dialkyl amides have been identified as alternative extractants to TBP (13–16). GANEX (Group ActiNide Extraction) and ARTIST (Amide-based Radio-resources Treatment with Interim Storage of Transuranics) processes have been proposed at CEA, France and at JAERI, Japan, respectively (17,18). These separations are achieved by use of amidic extractants in accordance with the CHON principle. The GANEX process involves a step of preliminary uranium separation using a branched amide, di(2-ethylhexyl)isobutyramide, followed by the group extraction of transuranic elements, along with the residual uranium, from the raffinate. On the other hand, the ARTIST process proposes to recover and stock all actinides, uranium, and a mixture of transuranics separately, and to dispose only fission products. The branched-alkyl monoamide (BAMA) proposed to be used in ARTIST process is *N,N*-di-(2-ethylhexyl)butyramide (D2EHBA).

Extensive laboratory batch studies as well as mixer settler studies were performed at BARC, India, to evaluate *N,N*-dialkyl amides as extractants for the reprocessing of spent uranium and thorium based fuels (16). Straight chain *N,N*-dihexyloctanamide (DHOA) was identified as a promising alternative to TBP for the reprocessing of spent uranium based fuels. DHOA extracts Pu(IV) more efficiently than TBP, both at trace-level concentration as well as under uranium loading conditions. Mixer settler experiments on uranium extraction suggested that the behavior of DHOA is similar to that of TBP during the extraction cycle but is better than TBP during the stripping cycle. DHOA offers better fission product decontamination than that of TBP.

Third phase formation behavior of any extractant for a particular metal ion under a specified condition is expressed in terms of Limiting Organic Concentration (LOC) of the metal ion beyond which the organic phase splits into two phases. During the reprocessing of spent nuclear fuel by solvent extraction, the occurrence of the third phase in the extraction system is undesirable as apart from limiting the throughput of the plant, it presents criticality hazard. This is particularly relevant to fast reactor fuel reprocessing, where the plutonium concentration is fairly high. At 3.3 M HNO_3, the LOC (g/L) values of Pu(IV) and U(VI) in 1.1 M DHOA/*n*-dodecane system were $\sim$49 and $\sim$98, respectively. The corresponding Pu(IV)-LOC value for 1.1 M TBP has been reported to be $\sim$53 g/L (19). No third phase formation was observed in the case of uranium extraction using TBP as the extractant under identical conditions. Radiolytic degradation studies of DHOA showed the formation of caprylic acid, dihexylamine, and dihexylketone, which could be easily washed out. Unlike TBP, irradiated DHOA showed negligible retention of Pu, U and fission products in the organic phase after the stripping cycle (20).

The present paper compares the behavior of DHOA and TBP as extractants for the coprocessing of uranium based fuels under

a. varying concentrations of nitric acid and of uranium, and
b. simulated pressurized heavy water reactor (PHWR) spent fuel feed conditions.

Batch experiments have been done for the performance evaluation of TBP and DHOA in extraction, scrubbing and stripping cycles. Extraction behaviors of Am(III), fission products and structural materials were also investigated under simulated PHWR feed conditions using both the extractants.

EXPERIMENTAL

Materials

DHOA was synthesized in our laboratory as per the reported method (16). TBP and *n*-dodecane used in this study were of AR grade. However, TBP was washed with alkali prior to its use in the present work. ^{233}U tracer ($\sim 10^{-4}$ M) was purified by anion exchange to eliminate the daughter products of ^{232}U and was found by α spectrometry to be free from ^{228}Th and its daughter products (21). Pu (principally ^{239}Pu) was purified by solvent extraction procedure using HTTA (2-theonyltrifluoroacetone) as extractant and its radiochemical purity was ascertained by gamma spectrometry for the absence of ^{241}Am (22). Pu(IV) was extracted by 0.5 M HTTA in xylene at 1.0 M HNO_3 and stripped by 8.0 M HNO_3 and was used as stock for Pu(IV). Further, the valency of Pu in the aqueous phase was adjusted and maintained in the tetravalent state by the addition of 0.05 M $NaNO_2$ + 0.005 M NH_4VO_3 (holding oxidants). Other radionuclides such as (^{241}Am, ^{147}Nd, ^{137}Cs, ^{152,154}Eu, ^{95}Zr, ^{106}Ru/^{106}Rh, ^{85,89}Sr, ^{59}Fe, and ^{51}Cr) were checked for their purity by gamma spectrometry. A stock solution of natural uranium (oxide form) was prepared by dissolving in nitric acid, and used after suitable dilution as per the requirement in solvent extraction experiments.

Extraction Procedure

Pre-equilibrated 1.1 M solutions of DHOA and TBP in *n*-dodecane with respective nitric acid solutions were used for solvent extraction experiments under desired experimental conditions. Desired volumes of the pre-equilibrated organic phases (1.1 M TBP/DHOA) and the aqueous phases containing metal ions were kept for equilibration in the water bath for 30 minutes at 25°C. The two phases were then centrifuged and assayed by taking suitable aliquots (25–50 μL) from both the phases. ^{233}U and Pu in the organic and aqueous phases were estimated by liquid scintillation counting using dioxane based scintillator. The composition of the scintillator 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 liter of dioxane. The correction for counts in both the phases due to the decay products of natural uranium was done by performing experiments under identical conditions using natural uranium solutions under the desired conditions. The distribution ratio of the metal ions (D_M) was defined as the ratio of concentration of metal ion in the organic phase (expressed in terms of counts per unit volume per minute) to that in the aqueous phase. For radiometric assay, usually concentration and aliquot sizes were adjusted suitably to give a count rate of 10000 to 50000 counts/min. However, in the cases where the count rates were lower, a long period counting was performed to keep relative standard deviation values within ±5%. All the experiments were carried out at least in duplicate and the material balance was within error limits (±5%).

RESULTS AND DISCUSSION

Effect of Uranium Concentration

Extraction studies of U(VI) and Pu(IV) were carried out at varying acidities (0.5–4 M HNO_3) and uranium concentrations (50–300 g/L) using 1.1 M solutions of TBP as well as of DHOA in *n*-dodecane. Third phase was observed in the case of 1.1 M DHOA/*n*-dodecane for $[U]_{initial} > 100$ g/L ($V_O/V_A = 1$). V_O/V_A was maintained as 3 for $[U]_{initial}$ of 200 g/L and 3.5 for $[U]_{initial}$ of 300 g/L U. V_O and V_A refer to the volume of the organic and aqueous phases used in the solvent extraction experiments. The D_U and D_{Pu} values

i. increased with increased aqueous phase acidity at a fixed uranium concentration, and
ii. decreased with increased uranium concentration at a fixed acidity for both the extractants (Figs. 1–4).

Generally, D_U values were higher but D_{Pu} values were lower in the case of TBP than those of DHOA. It is also evident that unlike TBP, D_U does not decrease significantly in case of DHOA with uranium concentration in the range 100–300 g/L. DHOA displayed fascinating behavior with respect to D_{Pu} values. Typically at 50 g/L U (which is expected to be present in fast reactor spent fuel feed solution), the D_{Pu} values were: 4.0×10^{-2} (0.5 M HNO_3) and 3.8 (4 M HNO_3) for 1.1 M TBP and 4.0×10^{-3}(0.5 M HNO_3) and 7.9 (4 M HNO_3) for 1.1 M DHOA. Irrespective of the uranium concentration, D_{Pu} values were larger for DHOA between 3–4 M HNO_3 but lower between 0.5 – 1.0 M HNO_3 as compared to the corresponding values with TBP. This was attributed to the change

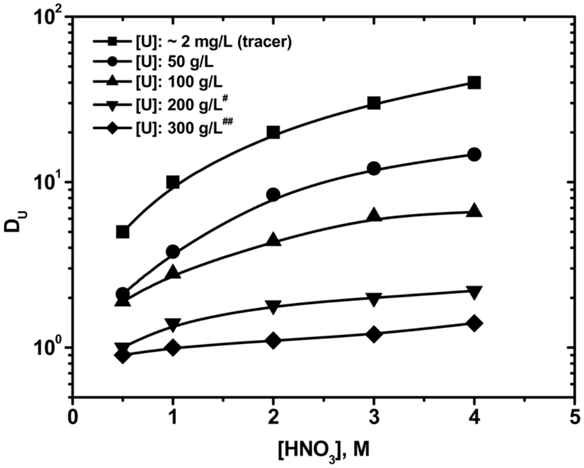

Figure 1. Variation of D_U with nitric acid concentration; Extractant: 1.1 M TBP/*n*-dodecane; Temperature: 25°C; #: $V_O/V_A = 3$; ##: $V_O/V_A = 3.5$.

in the nature of extractable species at 0.5 and 4.0 M HNO_3 using DHOA as the extractant. These special features of DHOA make it a promising candidate to coprocess uranium and plutonium present in the dissolver solution. This is particularly important for the coprocessing of fast reactor spent fuels

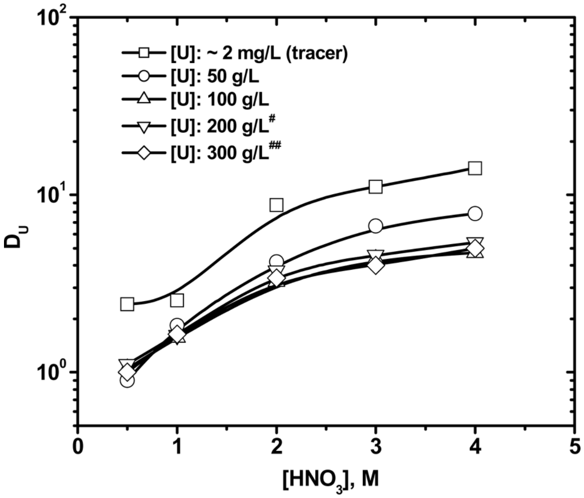

Figure 2. Variation of D_U with nitric acid concentration; Extractant: 1.1 M DHOA/*n*-dodecane; Temperature: 25°C; #: $V_O/V_A = 3$; ##: $V_O/V_A = 3.5$.

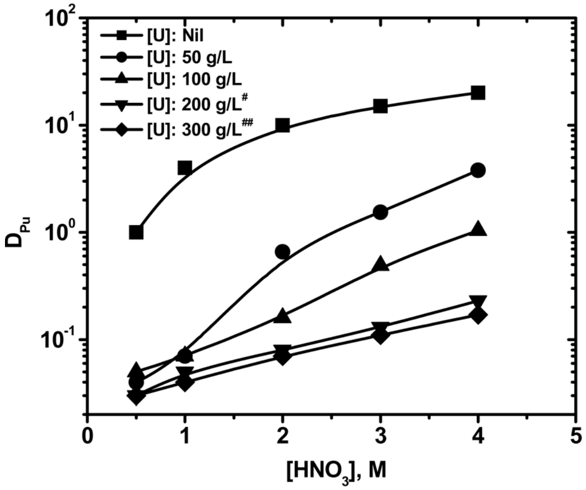

Figure 3. Variation of D_{Pu} with nitric acid concentration; Extractant: 1.1 M TBP/*n*-dodecane; Temperature: 25°C; #: $V_O/V_A = 3$; ##: $V_O/V_A = 3.5$.

containing larger quantities of plutonium. In comparison to TBP, Pu can be better extracted from 4 M HNO_3 and better stripped from the loaded organic phase at 0.5 M HNO_3 (without any reducing agent) using DHOA.

To ascertain the stoichiometry of the extracted species, the D_{Pu} values were obtained as a function of DHOA concentration

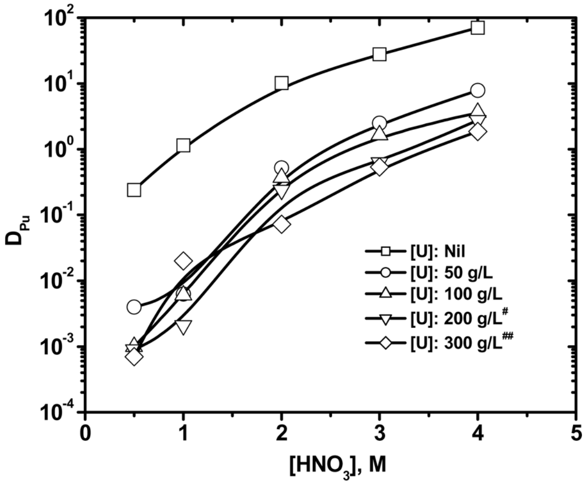

Figure 4. Variation of D_{Pu} with nitric acid concentration; Extractant: 1.1 M DHOA/*n*-dodecane; Temperature: 25°C; #: $V_O/V_A = 3$; ##: $V_O/V_A = 3.5$.

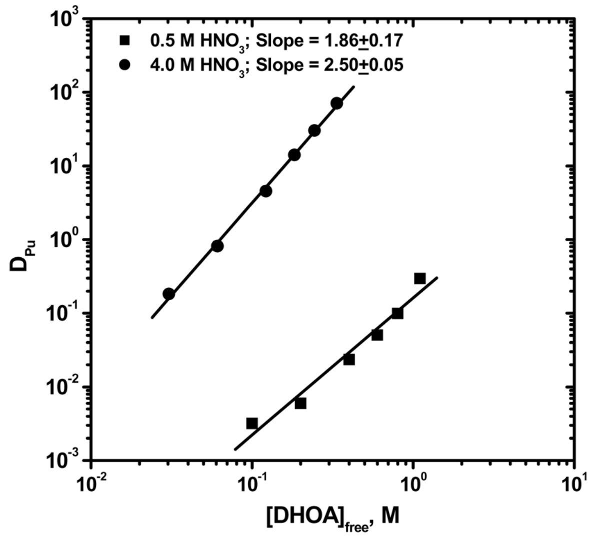

Figure 5. Variation of log D_{Pu} vs log $[DHOA]_{free}$ at 0.5 M and 4 M HNO_3.

(0.1–1.1 M) at 0.5 and 4.0 M HNO_3. Fig. 5 shows that the slopes of the log D_{Pu} vs log $[DHOA]_{free}$ concentration at these two acidities are different. It appears that a disolvated species is formed at lower acidity while more than two DHOA molecules are involved in the extracted species of Pu(IV) at higher acidity. The data indicate that DHOA appears better extractant than TBP for coprocessing of fast reactor fuels and a lesser number of stages are required for extraction and stripping of plutonium as compared to that of uranium. Figure 6 compares the separation factor (SF: D_U/D_{Pu}) values for 1.1 M TBP and 1.1 M DHOA solutions in *n*-dodecane in the presence of 50 g/L U in 0.5 and 4 M HNO_3. It is evident that the Pu/U ratio can be increased in the case of DHOA during the stripping step at 0.5 M HNO_3. This observation suggests that DHOA appears promising for the coprocessing of fast reactor spent fuels.

Extraction Studies under Simulated PHWR Spent Fuel Feed Conditions

The flow sheet developed at Savannah River Plant, USA and by the French group aimed at enriching plutonium stream (by reductive stripping) with respect to uranium (7,8). By contrast, Zabunoglu and Özdemir proposed a two-step flow sheet viz. co-decontamination (of U and Pu) and co-stripping, for coprocessing of spent light water

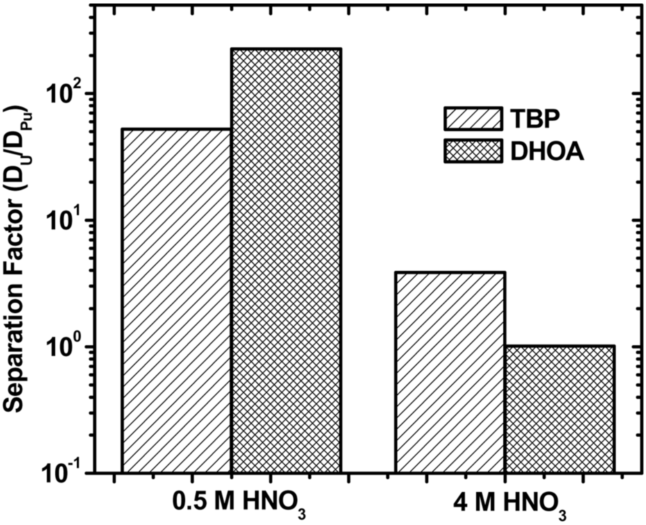

Figure 6. Separation factor of U and Pu at 0.5 M and 4 M HNO_3; Extractant(s): 1.1 M TBP and 1.1 M DHOA solutions in *n*-dodecane; [U] = 50 g/L; Temperature = 25°C.

reactor fuels similar to conventional PUREX process (3). Fission product decontamination was done by acid scrub (2.5 M HNO_3). No reducing agent was proposed for the stripping of plutonium (and uranium), instead dilute nitric acid solution (0.01 M) was used in flow sheet development studies. In this context, it was of interest to evaluate DHOA and TBP as extractant for coprocessing of uranium and plutonium under PHWR spent fuel feed conditions. Table 1 compares the extraction behavior of uranium and plutonium under simulated PHWR feed conditions containing 300 g/L U at 4 M HNO_3 maintaining $V_O/V_A = 5$. Whereas quantitative extraction (>99%) of Pu(IV) was achieved in two successive extraction stages employing 1.1 M DHOA as extractant; three stages were required for its extraction using 1.1 M TBP as extractant. Both DHOA and TBP required two extraction stages for quantitative extraction of uranium.

Scrubbing conditions were optimized using loaded 1.1 M TBP and 1.1 M DHOA solutions in *n*-dodecane (obtained after extraction cycle) as organic phases and 2–4 M HNO_3 solutions maintaining $V_O/V_A = 5$ (Table 2). Based on these studies, 4 M HNO_3 was found suitable as scrubbing solution [TBP: 3.9 (D_{Pu}), 10 (D_U); DHOA: 6.6 (D_{Pu}), 5.4 (D_U)]. DHOA offered higher D_{Pu} values both under extraction and scrubbing stages as compared to those of TBP.

Table 1. Extraction data of U and Pu under simulated PHWR feed conditions; Extractant(s): 1.1 M TBP and 1.1 M DHOA solutions in *n*-dodecane; [HNO_3]: 4 M; V_O/V_A: 5; Temperature: 25°C

	1.1 M TBP		1.1 M DHOA	
Stage	E_U (%)	E_{Pu} (%)	E_U (%)	E_{Pu} (%)
I	97.3	83.3	95.6	93.7
II	99.9	97.2	99.8	*99.9*
III	–	*99.5*	–	–

One of the interests in the coprocessing of U/Pu from the spent fuels is to avoid the stringent/critical chemical conditions for the partitioning of Pu from uranium, which is achieved by the use of reducing agents like U(IV) and hydrazine. By contrast, only dilute nitric acid is proposed to be used for the stripping of uranium and plutonium from the loaded organic phase. The purpose of co-stripping was to wash the loaded organic phases (TBP/DHOA) with dilute nitric acid solution with a recovery as high as possible. Therefore, stripping studies were carried out on the loaded TBP and DHOA phases using 0.1 M HNO_3 without any reducing agent. The choice of 0.1 M HNO_3 was as the strippant was based on the minimum aqueous phase acidity requirement to avoid hydrolysis/polymerization of plutonium. It should be noted that acidity build up in the aqueous phase was also expected during the stripping cycle.

Table 3 shows that quantitative stripping of uranium can be achieved in four contacts in the case of DHOA as extractant. By contrast, a minimum of six stages were needed for its stripping from loaded TBP phase. However, three stages were sufficient for the quantitative stripping of Pu from loaded TBP/DHOA phases. These studies suggest that

Table 2. Scrubbing studies on loaded organic phases containing U and Pu under simulated PHWR feed conditions; Extractant(s): Loaded 1.1 M TBP and 1.1 M DHOA solutions in *n*-dodecane (during extraction cycle); [HNO_3]: 4 M; V_O/V_A: 5; Temperature: 25°C

	1.1 M TBP		1.1 M DHOA	
[HNO_3], M	D_U	D_{Pu}	D_U	D_{Pu}
2	6.5	1.4	4.0	2.3
3	9.2	2.6	4.6	5.4
4	10.0	3.9	*5.4*	*6.6*

Table 3. Stripping behavior of U and Pu from loaded organic phases under simulated PHWR feed conditions; [HNO_3]: 0.1 M; Temperature: 25°C

		Pu Stripping, %		U Stripping, %	
Stages	V_O/V_A	1.1 M TBP	1.1 M DHOA	1.1 M TBP	1.1 M DHOA
I	2	78	78	20	26
II	2	89	90	47	60
III	1	>99	>99	70	90
IV	1	–	–	85	>99

co-processing of PHWR spent fuel can be achieved in lesser number of stages using DHOA as extractant (extraction: 2; stripping: 4) as compared to that of TBP (extraction: 3; stripping: 6).

Extraction Behavior of Americium, Fission Products, and Structural Materials

It was of interest to investigate the behavior of Am(III), fission products (viz. ^{147}Nd, ^{137}Cs, ^{152,154}Eu, ^{95}Zr, ^{106}Ru/^{106}Rh, ^{85,89}Sr) and structural materials (viz. ^{59}Fe and ^{51}Cr) under the simulated feed conditions using 1.1 M TBP and 1.1 M DHOA solutions in *n*-dodecane as the extractants. Table 4 clearly indicates that DHOA offers better decontamination from Am(III), fission products, and structural materials as compared to those of TBP.

Table 4. Fission products/structural materials distribution behavior under simulated PHWR feed conditions; [HNO_3]: 4 M; V_O/V_A: 5

	D_M	
Metal ion	1.1 M TBP	1.1 M DHOA
Am	4.3×10^{-2}	6.6×10^{-3}
Cs	3.1×10^{-2}	$<10^{-4}$
Eu	4.1×10^{-2}	2.3×10^{-3}
Zr	0.2	5×10^{-2}
Ru	5×10^{-2}	1×10^{-2}
Fe	5.5×10^{-2}	3.8×10^{-3}
Nd	3.3×10^{-2}	$<10^{-4}$
Cr	$<10^{-4}$	$<10^{-4}$

CONCLUSIONS

Batch extraction studies suggest that DHOA is a better choice for coprocessing of spent nuclear fuel than TBP. It offers better extraction of Pu under feed conditions (4 M HNO_3) and its easy stripping at 0.5 M HNO_3 without using any reducing agent. This observation has been attributed to the formation of disolvated species at 0.5 M HNO_3; while more than two DHOA molecules are involved in the extracted species at 4 M HNO_3. DHOA displays better extraction behavior of plutonium and stripping behavior of uranium under simulated feed conditions. Quantitative extraction of Pu can be achieved in two contacts using 1.1 M DHOA as the extractant; while more than three stages are required for 1.1 M TBP as the extractant. Four contacts of 0.1 M HNO_3 is sufficient for quantitative uranium stripping in the case of DHOA. On the other hand, a minimum of six stages were needed for its stripping from loaded TBP phase. DHOA appears distinctly better than TBP with respect to Am(III), fission products and structural materials decontamination of U/Pu.

REFERENCES

1. Knighton, J.B.; Baldwin, C.E. (1979) Pyrochemical coprocessing of uranium dioxide-plutonium dioxide LMFBR fuel by the salt transport method. Rocky Flats Report RFP-2887, CONF-790415-29.
2. Zabunoğlu, O.H.; Özdemir, L. (2005) Purex co-processing of spent LWR fuels: Comparative fuel cost analyses. *Ann. Nucl. Energy*, *32*: 137.
3. Zabunoğlu, O.H.; Özdemir, L. (2005) Purex co-processing of spent LWR fuels: Flow sheet. *Ann. Nucl. Energy*, *32*: 151.
4. Pereira, C.; Leite, E.M. (1998) Non-proliferating reprocessed nuclear fuels in pressurized water reactors: Fuel cycle options. *Ann. Nucl. Energy*, *25*: 937.
5. Pobereskin, M.; Kok, K.D.; Madia, W.J. (1978) Coprocessing – an evaluation of chemical reprocessing without plutonium separation. *Nucl. Technol.*, *41*: 149.
6. Sunanta, P.; Reiner, P. (1980) Recycling of actinides in light water reactors. *Nucl. Technol.*, *51*: 7.
7. Statton, M.A.; Thompson, M.C. (1979) Flowsheet for Coprocessing Uranium and Plutonium, USDOE Report, DP-1505.
8. Baron, P.; Dinh, B.; Masson, M.; Drain, F.; Emin, J. Process for reprocessing a spent nuclear fuel and of preparing a mixed uranium-plutonium oxide. World International Patent Organization, Patent No. WO/2007/135178, November 29, 2007.
9. Vasudeva Rao, P.R. (1998) Extraction of actinides by trialkyl phosphates. *Min. Pro. Ext. Met. Rev.*, *18*: 309.
10. McKay, H.A.C.; Miles, J.H.; Swanson, J.L. (1990) The Purex Process, "Science and Technology of Tributyl Phosphate," v.3. In: *Applications of*

Tributyl Phosphate in Nuclear Fuel Reprocessing; Schulz, W.W.; Burger, L.L.; Navratil, J.D.; Bender, K.P., eds.; CRC Press Inc.: Boca Raton, Florida.

11. Siddall, T.H.; Wallace, R.M. (1960) Effect of solvent degradation on the PUREX process. USDOE-Report DP-286.
12. Deshingkar, D.S.; Ramaswamy, M.; Kartha, P.K.S.; Kutty, P.V.E.; Ramanujam, A. (1989) Treatment of tributyl phosphate wastes by extraction cum pyrolysis process. Report BARC – 1480, Bhabha Atomic Research Centre, Mumbai, India.
13. Siddall, T.H. (1961) Effects of structure of N,N-disubstituted amides on their extraction of actinide and zirconium nitrates and of nitric acid. *J. Phys. Chem.*, *64*: 1863.
14. Musikas, C. (1987) Solvent extraction for the chemical separations of the 5 f elements. *Inorg. Chim. Acta*, *140*: 197.
15. Gasparini, G.M.; Grossi, G. (1986) Long chain disubstituted aliphatic amides as extracting agents in industrial applications of solvent extraction. *Solv. Extr. Ion Exch.*, *4*: 1233.
16. Manchanda, V.K.; Pathak, P.N. (2004) Amides and diamides as promising extractants in the back end of the nuclear fuel cycle: An overview. *Sep. & Pur. Technol.*, *35*: 85.
17. Miguirditchian, M.; Sorel, C.; Camès, B.; Bisel, I.; Baron, P. (2008) Extraction of uranium(VI) by N,N-di-ethyl(2-hexyl)isobutyramide (DEHIBA): From the batch experimental data to counter-current process. ISEC-2008, Tucson, Arizona.
18. Sasaki, Y.; Suzuki, S.; Tachimori, S.; Kimura, T. (2003) An innovative chemical separation process (ARTIST) for treatment of spent nuclear fuel. Atoms for Prosperity: Updating Eisenhowers Global Vision for Nuclear Energy, Global 2003, p. 1266.
19. Vasudeva Rao, P.R.; Kolarik, Z. (1996) A review of third phase formation in extraction of actinides by neutral organophosphorus extractants. *Solv. Extr. Ion Exch.*, *14*: 955.
20. Parikh, K.J.; Pathak, P.N.; Misra, S.K.; Tripathi, S.C.; Dakshinamoorthy, A.; Manchanda, V.K. (2009) Radiolytic degradation studies on *N,N*-dihexyloctanamide (DHOA) under PUREX process conditions. *Solv. Extr. Ion Exch.*, *27*: 244.
21. Srinivasan, N.; Nadkarni, M.N.; Balasubramanian, G.R.; Chitnis, R.T.; Siddiqui, H.R. (1972) Pilot plant for the separation of U-233 at Trombay. BARC Report-643, Bhabha Atomic Research Centre, Mumbai, India.
22. Sajun, M.S.; Ramakrishna, V.V.; Patil, S.K. (1981) The effect of temperature on the extraction of plutonium(IV) from nitric acid by tri-*n*-butyl phosphate. *Thermochim. Acta*, *47*: 277.