NUCLEAR SCIENCE AND TECHNIQUES **26**, 040605 (2015)

An improvement in APOR process I-uranium/plutonium separation process[*]

XIAO Song-Tao (肖松涛),[†] LI Li (李丽), YE Guo-An (叶国安), LUO Fang-Xiang (罗方祥),
LIU Xie-Chun (刘协春), YANG He (杨贺), and LAN Tian (兰天)
China Institute of Atomic Energy, P. O. Box 275 (26), Beijing 102413, China

(Received June 27, 2014; accepted in revised form September 20, 2014; published online August 20, 2015)

The reduction stripping behavior of Pu(IV) from 30%TBP/OK with hydroxysemicarbazide (HSC) was investigated, and the separation efficiency of HSC and DMHAN-MMH for U/Pu partitioning in Purex process was compared. The results show that HSC can effectively realize the separation of Pu from U; using mixer-settlers to simulate U/Pu separation in 1B bank of PUREX, from 16-stage counter current extraction experiment (in which 6 stages for supplemental extraction, 10 stages for stripping) with flow rate ratio $(\mathrm{1BF} : \mathrm{1BX} : \mathrm{1BS}) = 4 : 1 : 1$ in 1B contactor, good result was achieved that the yields are both more than 99.99% for uranium and Pu, the separation factor of plutonium from uranium ($SF_{\mathrm{Pu/U}}$) is 2.8×10^4, and separation factor of uranium from plutonium ($SF_{\mathrm{U/Pu}}$) is 5.9×10^4. As a stripping reductant, HSC can effectively achieve the separation of Pu from U and the separation effect is nearly the same with DMHAN-MMH, which contributed to replace enough the latter with HSC in the U/Pu separation in Advanced Purex Process Based on Organic Reagent (APOR) process.

Keywords: APOR process, Hydroxysemicarbazide, Reduction stripping, Separation of Pu from U, Pu(IV)

DOI: 10.13538/j.1001-8042/nst.26.040605

I. INTRODUCTION

Purex (Plutonium and Uranium Recovery by Extraction) has been employed worldwide in spent fuel reprocessing. For the typical Purex process, it always includes three extraction cycles, namely U/Pu codecontamination and separation cycle, uranium purification cycles and plutonium purification cycles. The Purex process was first developed in USA in the 1949's, and thereafter it has been ameliorated by researchers in many countries, including France, UK, Russia, Japan, China, and so on [1].

In the U/Pu codecontamination and separation cycle of Purex process, the separation of Pu from U is always realized through stripping of Pu(IV) loaded in organic phase with reductant aqueous solution based on a reduction of Pu(IV) to Pu(III). Therefore, the improvement of reductants is one of the principal research content in the Purex process. From 1962's, U(IV)-N_2H_4 have been studied as reductants in 1B unit of Purex process and applied to the French Marcoule Nuclear fuel reprocessing plant [2], and now they have been successfully applied in the commercial reprocessing plants,such as UP-2, UP-3, THORP, etc.

One important trend in developing future reprocessing extraction processes is the use of high efficient salt-free reductants in U Pu separation and purification process, such as hydrazine [3] and hydroxylamine [4, 5] as well as their derivatives [6–9]. N,N-dimethylhydroxylamine (DMHAN) is one derivative of hydroxylamine in which two H atoms are both substituted by methyl group and it was studied as Pu(IV) reductant by Koltunov *et al.* [6] of Russia. The results showed that DMHAN can reduce Pu (IV) at a very rapid rate in nitric acid solution.

APOR process [10] developed by ZHANG Xian-Ye *et al.* of China Institute of Atomic Energy, using two organic reagents, namely, N,N-dimethylhydroxylamine (DMHAN) and monomethyl hydrazine (MMH), is a two cycle aqueous reprocessing process. Compared with the U(IV) and hydrazine, reductants of DMHAN and MMH have three self-evident superiorities: 1) elimination of U and Pu interference from Tc, hence the simplified first cycle and reduced hot region facilities (by 2/5); 2) 20 times increase of separation factors, with improved quality of the U and Pu products; and 3) removing the security risk of hydrazoic acid explosion. However, there are still some improvements to be done for the APOR process: 1) DMHAN and MMH, as modified liquid rocket propellants, is of high heat of combustion, hence a certain security hidden danger in its application in large nuclear fuel reprocessing plant [11]; 2) the preparation of DMHAN is not suitable in vicinity of the nuclear plant because of the vacuum pyrolysis process in synthesis [12]; 3) oxidation products and radiolysis products are complex [13].

For all these reasons, we developed the new organic reductant, namely, hydroxysemicarbazide (HSC) [14, 15], to substitute DMHAN and MMH as reductant reagents of U/Pu separation. Its usage allows the following benefits: 1) good hydrophilicity (molecular formula is $\mathrm{HONHCONHNH_2}$); 2) quick reduction of Pu(IV) to Pu(III) [14]; 3) the need of just small quantity of HNO_3 to neutralize its alkalescence of $\mathrm{pH} = 8.0$, hence the reduced effect of salting out on extraction distribution of Pu(III); and 4) decreased solid waste and reduced environmental impact.

In this paper, the application of HSC in U/Pu separation is studied and compared with DMHAN-MMH. The concentration effects of HSC, HNO_3 in stripping solution, NO_3^-, U in organic phase, phase-contacting time, phase ratio and temperature on reaction rate of Pu(IV) are investigated. To evaluate feasibility of applying HSC in the partitioning of uranium and plutonium in the Purex process, HSC is compared with DMHAN-MMH using U/Pu separation process carried out with 16-stage counter current extraction in 1B contactor.

---

[*] Supported by China National Nuclear Corporation fund (No. 61601090401)

[†] Corresponding author, xiao_songtao@126.com

II. EXPERIMENTAL

A. Reagents and apparatus

All chemicals used were of analytical-reagent grade. HSC and DMHAN were synthesized as describes in Ref. [12, 16] in purity of over 99%. MMH was provided by China Aerospace Science & Industry Corp., in purity of over 99%. Pu(IV) stock solutions in nitric acid were obtained in the following steps, reduction by Fe(II), and oxidation by HNO_2, and purification with 2606 anion-exchange resin named poly(4-vinyl-N-methylpyridinum nitrate).

The concentration of macro-scale uranium in aqueous phase was measured by redox titration method, while in organic phase the U(VI) was back-extracted by mixed acid before titration. The micro-scale uranium in aqueous phase was measured by TOPO extraction–Br-PADAP coloring–spectrophotometry (at wavelength of 575 nm), while in organic phase the U(VI) was back-extracted by saturated Na_2CO_3 before extracted by TOPO [17]. Plutonium concentration of stock solutions was measured by K-edge and alpha counter while in process by scintillation counter [18] (LS-6000L, Beckman Corp.). The nitric acid concentration was measured by titration with NaOH after complex of U(VI) and Pu(IV) by $(NH_4)_2C_2O_4$, while in organic phase the nitric acid was back-extracted by deionization before PH titration (PHS-3C, Shanghai Precision Instrument Co., Ltd., China).

DC-1020 low-temperature thermostatic water bath (Ningbo Xinzhi Biological Technology Co., Ltd., China) was used to keep constant temperature in the reduction stripping experiments, and the mixer-settler extractor was used in bench-scale experiments.

B. Procedures

The single-stage back-extraction test in the centrifugal extraction tube includes the following steps: keeping the solution at constant temperature for 0.5 h, introducing it into centrifugal extraction tubes according to design phase ratio, stirring it for a few minutes, and standing it for separation and analysis.

Multi-stage counter current tests were carried out using miniature mixer-settlers, the flowsheet was shown in Fig. 1. The flow streams to the mini-banks were specified to simulate U/Pu separation in 1B bank of PUREX. Note that the mini-banks with phase ratio of 1.5 : 1 have 16 stages, 10 stages for reduction stripping and the rest for supplemental extraction. The volumes of mixing chamber and settling chamber are 3 and 5 mL, respectively. Residence time of 60 s is maintained in each mixing chamber. Flow rate ratio of the feed solution (1BF), uranium supplement extraction stream (1BS) and aqueous stripping stream (1BX) (1BF : 1BS : 1BX) is 4 : 1 : 1.

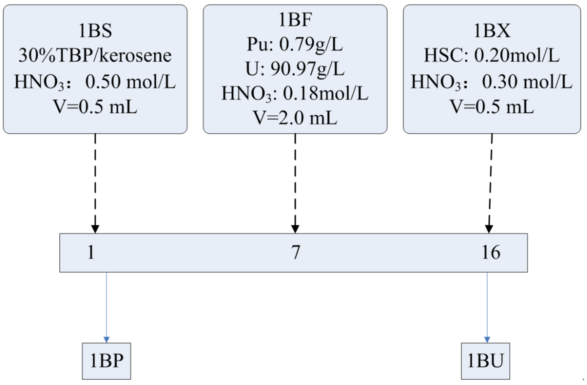

Fig. 1. (Color online) Flowsheet diagram of experiment for simulating 1B of Purex process.

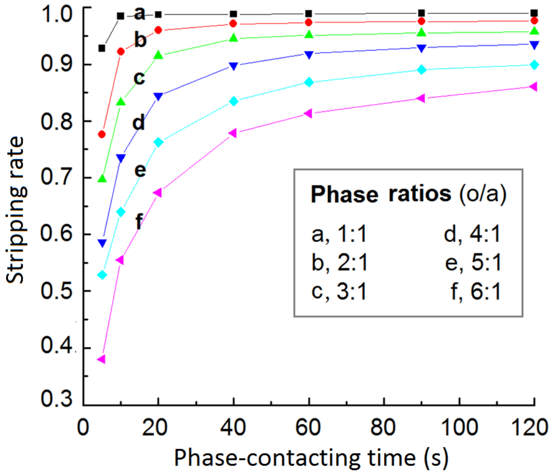

Fig. 2. (Color online) The stripping rate of Pu(IV) at different phase ratios as function of phase-contacting time. $c(\mathrm{Pu(IV)})_0 = 0.8\,\mathrm{g/L}$, $c(\mathrm{HSC}) = 0.05\,\mathrm{mol/L}$, $c(\mathrm{H^+}) = 0.4\,\mathrm{mol/L}$, $c(\mathrm{NO_3^-}) = 0.4\,\mathrm{mol/L}$, $T = 21\,^\circ\mathrm{C}$.

III. RESULTS AND DISCUSSION

A. The single-stage test of Pu(IV) stripping by HSC

Effects of phase-contacting time, phase ratio, concentrations of H^+, NO_3^-, HSC, U in organic phase and temperature on stripping rate of Pu(IV) were investigated.

1. *Influence of phase-contacting time*

At 21 °C, with phase ratio varying from 1 : 1 to 6 : 1, the stripping rates of Pu(IV) as function of phase-contacting time were obtained at the concentrations of HSC, H^+, NO_3^- and Pu(IV) in the organic phase being 0.05 mol/L, 0.4 mol/L, 0.4 mol/L and 0.8 g/L, respectively. The results are given in Fig. 2. The stripping rates of Pu(IV) at all phase ratios increased with phase-contacting time. At a same contact time, the stripping rate decreased with increasing phase ratio. At a phase ratio of $1:1$ and phase-contacting time of 5 s, the stripping rate was 92.8%; while for phase ratio of $4:1$, the stripping rate was 58.7% and 84.5% at phase-contacting time of 5 s and 20 s, respectively. It is worth noting that the reduction stripping of Pu(IV) could go basically balance at $4:1$ phase ratio and 30 s phase-contacting time, and the stripping rate was 91.9%. Therefore, at a large phase ratio, appropriate increase in contact time can improve the stripping rate of Pu(IV) significantly.

2. *Influence of H^+ concentration*

At 21 °C, phase-contacting time of 30 s and phase ratios of $1:1$ and $4:1$, the influence of H^+ concentration on the stripping rate was investigated at the concentrations of HSC, NO_3^- and Pu(IV) in organic phase being 0.05 mol/L, 0.4 mol/L and 0.8 g/L, respectively. The results are shown in solid signs in Fig. 3. The stripping rate decreased with increasing H^+ concentration. This is because that the reaction rate between HSC and Pu(IV) is inversely proportional, in a power of 0.43, to H^+ concentration [4]. An increase of H^+ concentration will decrease the amount of Pu(III) reduced by Pu(IV). Additionally, there is rapid extraction/back-extraction equilibrium of Pu(IV) between aqueous and organic (30% TBP/kerosene) phase, and Pu(III) is hardly extracted by 30% TBP/kerosene, hence the decrease of Pu(IV) stripping into aqueous phase. When the H^+ concentration increased from 0.3 to 1.0 mol/L, the Pu(IV) stripping rate decreased from 89.2% to 70.7%. So, lower H^+ concentration is preferred for back-extraction of Pu(IV) by HSC. Increasing the stripping time to 90 s and keeping the parameters of other conditions, the results are illustrated in blanket signs in Fig. 3. We suggest that extending the stripping time properly may increase the stripping rate if higher H^+ concentration is needed according to the process.

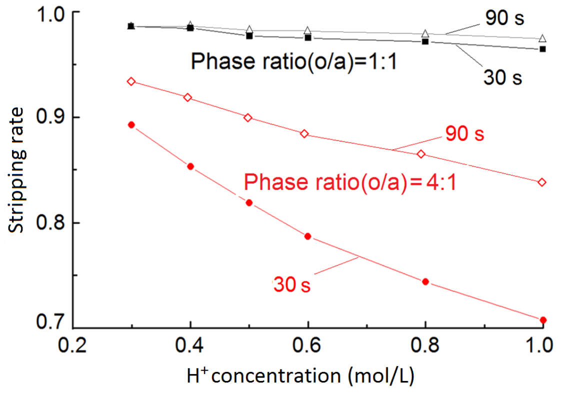

Fig. 3. (Color online) Effect of the H^+ concentration of on the stripping of Pu(IV) at different phase ratios and stripping time. $c(\mathrm{Pu(IV)})_0 = 0.8\,\mathrm{g/L}$, $c(\mathrm{HSC}) = 0.05\,\mathrm{mol/L}$, $c(\mathrm{NO_3^-}) = 0.4\,\mathrm{mol/L}$, $T = 21\,°\mathrm{C}$.

3. *Influence of NO_3^- concentration*

At 21 °C, phase-contacting time of 30 s and phase ratios of $1:1$ and $4:1$, the influence of NO_3^- concentration on the stripping rate was investigated when the concentration of HSC, H^+ and Pu(IV) in organic phase were 0.05 mol/L, 0.4 mol/L and 0.8 g/L, respectively. The result was given in Fig. 4. The stripping rate of Pu(IV) decreased with the increasing NO_3^- concentration. This is because that the reaction rate between HSC and Pu(IV) is inversely proportional, in a power of 0.58, to NO_3^- concentration, so an increase in NO_3^- concentration decreases, the amount of Pu(III) is reduced by Pu(IV), hence the decrease of Pu(IV) back-extraction to aqueous phase. Another reason is the salting out effect and synergistic extraction effect in the extraction process, so distribution ratio of Pu(IV) between TBP/kerosene and nitric acid aqueous solutions increase with the NO_3^- concentration, so the amount of Pu(IV) back-extraction to aqueous phase and the stripping rate of Pu(IV) decrease. From Fig. 4, at phase ratio $= 4:1$, when the NO_3^- concentration increased from 0.4 to 1.0 mol/L, the stripping rate of Pu(IV) decreased from 85.3% to 77.6%. Thus, lower NO_3^- concentration is helpful for the back-extraction of Pu(IV) by HSC.

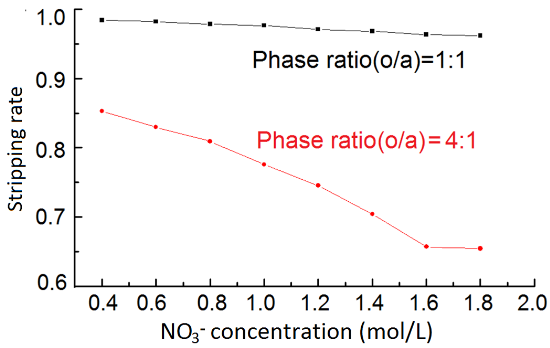

Fig. 4. (Color online) Effect of NO_3^- concentration on the stripping of Pu(IV) at different phase ratios. $c(\mathrm{Pu(IV)})_0 = 0.8\,\mathrm{g/L}$, $c(\mathrm{HSC}) = 0.05\,\mathrm{mol/L}$, $c(\mathrm{H^+}) = 0.4\,\mathrm{mol/L}$, $T = 21\,°\mathrm{C}$, stripping time $= 30\,\mathrm{s}$.

4. *Influence of HSC concentration*

At 21 °C, phase-contacting time of 30 s and phase ratios of $1:1$ and $4:1$, the influence of HSC concentration on the stripping rate was investigated at concentrations of NO_3^-, H^+ and Pu(IV) in organic phase being 0.4 mol/L, 0.4 mol/L and 0.8 g/L, respectively. As shown in Fig. 5, the stripping rate of Pu(IV) increases with HSC concentration. Below 0.05 mol/L, an increase in HSC concentration increases the stripping rate of Pu(IV) significantly, while over 0.05 mol/L, the HSC concentration affects just a little the stripping rate of Pu(IV). Therefore, in using HSC as a reductant for U/Pu separation, 0.01 mol/L is the best concentration.

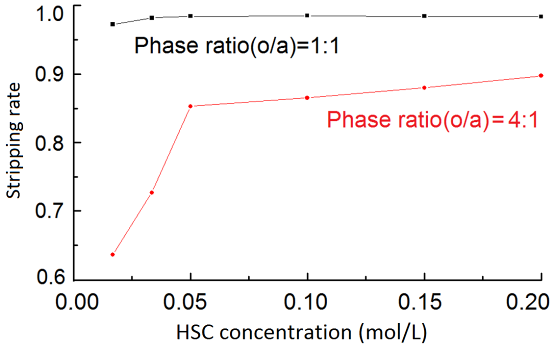

Fig. 5. (Color online) Effect of HSC concentration on the stripping of Pu(IV) at different phase ratios. $c(\mathrm{Pu(IV)})_0 = 0.8$ g/L, $c(\mathrm{H}^+) = 0.4$ mol/L, $c(\mathrm{NO_3^-}) = 0.4$ mol/L, $T = 21$ °C, stripping time = 30 s.

5. *Influence of U(VI) concentration*

At 21 °C, phase-contacting time of 30 s and phase ratios of 1:1 and 4:1, the influence of U(VI) concentration on the stripping rate was investigated at concentrations of $\mathrm{NO_3^-}$, HSC, $\mathrm{H^+}$ and Pu(IV) in organic phase being 0.4 mol/L, 0.05 mol/L, 0.4 mol/L and 0.8 g/L, respectively. As shown in Fig. 6, the stripping rate of Pu(IV) increases with the U(VI) concentration. For phase ratio $= 4:1$, the stripping rate increased by 7.5% at 5.0 g/L of U(VI) concentration.

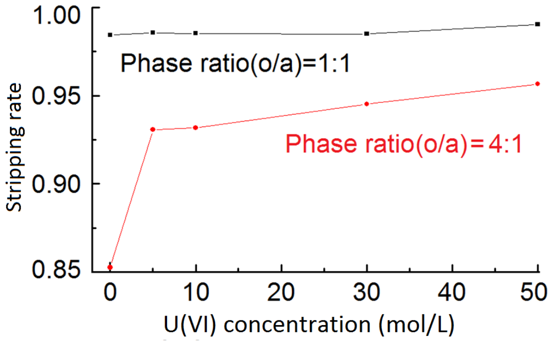

Fig. 6. (Color online) Effect of U(VI) concentration on the stripping of Pu(IV) at different phase ratios. $c(\mathrm{Pu(IV)})_0 = 0.8$ g/L, $c(\mathrm{HSC}) = 0.05$ mol/L, $c(\mathrm{H}^+) = 0.4$ mol/L, $c(\mathrm{NO_3^-}) = 0.4$ mol/L, $T = 21$ °C, stripping time = 30 s.

6. *Influence of temperature on the reaction*

At phase-contacting time of 30 s and phase ratios of $1:1$ and $4:1$, the influence of temperature on the stripping rate was investigated at concentrations of $\mathrm{NO_3^-}$, HSC, $\mathrm{H^+}$ and Pu(IV) in organic phase being 0.4 mol/L, 0.05 mol/L, 0.4 mol/L and 0.8 g/L, respectively. As shown in Fig. 7, the stripping rate of Pu(IV) increased with temperature. This is because the Pu(IV) distribution of decreases with increasing temperature, hence the increase of Pu concentration in aqueous phase; also, a temperature increase accelerates the reaction rate between HSC and Pu(IV): a 10 °C increase accelerates the reaction rate by about 10 times. So, an appropriately high temperature increases the stripping rate of Pu(IV) in the process of U/Pu separation.

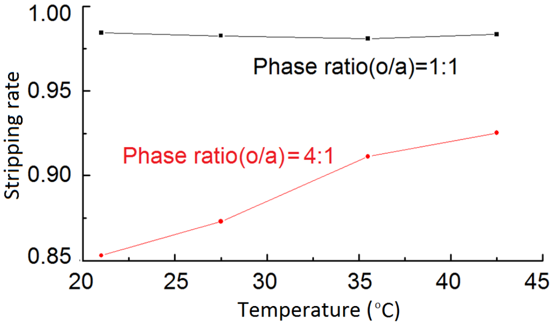

Fig. 7. (Color online) Effect of temperature on the stripping of Pu(IV) at different phase ratios. $c(\mathrm{Pu(IV)})_0 = 0.8$ g/L, $c(\mathrm{HSC}) = 0.05$ mol/L, $c(\mathrm{H}^+) = 0.4$ mol/L, $c(\mathrm{NO_3^-}) = 0.4$ mol/L, stripping tome = 30 s.

B. The multi-stage counter current tests of Pu(IV) stripping

Under the same experimental conditions, with DMHAN-MMH or HSC as a reductant of Pu(IV), and using the mixer-settler, comparative experiments were conducted to evaluate the feasibility of applying HSC in the partitioning of uranium and plutonium. The flow sheet is shown in Fig. 1. 1BF solution contains 90.97 g/L U, 0.79 g/L Pu and 0.18 mol/L $\mathrm{HNO_3}$-30% TBP/kerosene; 1BS is 30% TBP/kerosene with 0.50 mol/L $\mathrm{HNO_3}$, and 1BX is an aqueous solution with 0.10 mol/L DMHAN, 0.15 mol/L MMH and 0.30 mol/L $\mathrm{HNO_3}$ or with 0.30 mol/L HSC and 0.30 mol/L $\mathrm{HNO_3}$.

At the beginning, the mixer-settler was fed with solution containing no uranium. Two hours later, 1BF solution with uranium and plutonium was fed. After two hours, instantaneous samples were taken and analyzed every 30 min in 6h.

1. *DMHAN-MMH as the reductant of Pu(IV)*

The experiment, conducted according to the above conditions, achieved a steady state after 2.5 h. The concentration profiles of uranium, nitric acid and plutonium are shown in Fig. 8. At the stripping step, both the organic and aqueous phase had higher concentration of uranium, while the organic phase had a lower concentration of nitric acid (about 0.02 mol/L). At the supplement extraction step, the higher concentration of nitric acid in 1BS could help uranium extraction, thus a satisfactory result was achieved with the U concentration being only 5×10^{-3} g/L in the 1BP outlet. And the Pu concentration in 1BU was 25 µg/L. The calculation results show that the U and PU yields are 99.99% and 99.96%, respectively; the material balances of U and $\mathrm{HNO_3}$ are 99.9%

and 98.5%, respectively; and the separation factor of Pu from U ($SF_{\text{Pu/U}}$) is 2.5×10^4, and separation factor of U from Pu ($SF_{\text{U/Pu}}$) is 6.0×10^4.

In Fig. 8, the uranium concentration declined to 30 mg/L when the stages for supplemental extraction is 4, offering a higher uranium yield of 99.99%; a plutonium concentration peak appeared at Stage 7, indicating a cumulative process of plutonium in aqueous phase. We attribute this to the oxidation product of DMHAN, namely formaldehyde.

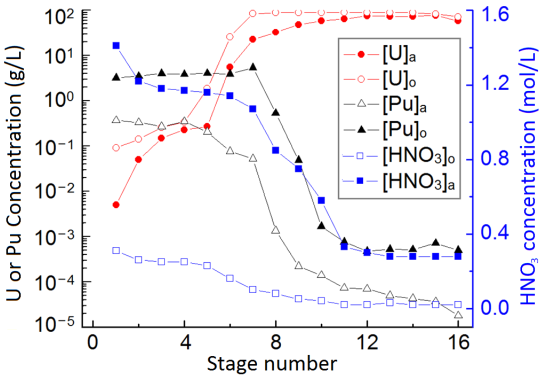

Fig. 8. (Color online) The concentration profile of U, Pu and HNO_3 in the experiment of DMHAN-MMH.

2. *HSC as the reductant of Pu(IV)*

The experiment, conducted at the same conditions, achieved a steady state after 2.0 h. The concentration profiles of uranium, nitric acid and plutonium are shown in Fig. 9. At the stripping step, both the organic and aqueous phase had a higher concentration of uranium, while the organic phase had a lower HNO_3 concentration of about 0.02 mol/L. At the supplement extraction step, the higher HNO_3 concentration in 1BS could help uranium extraction, thus a satisfactory result was achieved with uranium concentration being only 6.11×10^{-3} g/L in the 1BP outlet. In the experiment contained no plutonium, the uranium yield was 99.998%, and the U and HNO_3 material balances were 99.92% and 97.5%, respectively.

At supplement extraction step, uranium concentration in aqueous phase declined to 50 mg/L at stage 2, offering higher yield of more than 99.99%. At higher stage numbers, it decreased little; while the distribution ratio of plutonium was almost the same as those in stages 1–6, without plutonium peak, unlike the result using DMHAN-MMH as reductant. So it is considered that there are no uncertainties in U/Pu separation process. Plutonium concentration in aqueous phase and organic phase is about 3.2 g/L and 0.2 g/L, respectively, the concentration difference is small between each stage. The distribution of nitric acid in the supplement extraction step is similar to that of plutonium.

At the stripping step, uranium concentration did not change obviously and a lower concentration in organic phase outlet agrees with the theory calculation; while in aqueous phase, it increased significantly in stages 7–11. In stages 11–16, the concentration was almost the same. In organic phase, plutonium concentration decreased significantly in stages 7–11, but kept nearly the same in stages 12–16; while in aqueous phase, plutonium concentration showed a trend of orders of magnitude lower in stages 7–11, but kept almost the same in stages 11–16. The HNO_3 concentration in organic phase decreased significantly in stages 7–13, but kept almost the same in stages 14–16; while the HNO_3 concentration in aqueous phase decreased obviously in stages 7–12, but kept almost the same in stages 13–16.

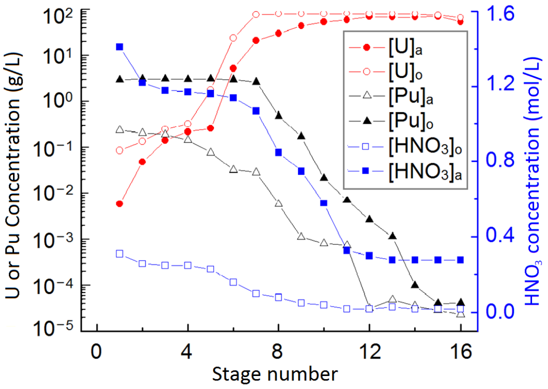

Fig. 9. (Color online) The concentration profile of U, Pu and HNO_3 in the experiment of HSC.

Calculation results show that the separation factor of plutonium from uranium ($SF_{\text{Pu/U}}$) is 2.8×10^4, and separation factor of uranium from plutonium ($SF_{\text{U/Pu}}$) is 5.9×10^4.

Compared the results of experiments using DMHAN-MMH or HSC as reductant for separating Pu from U, we can see that the separation factor of $SF_{\text{Pu/U}}$ and $SF_{\text{U/Pu}}$ are nearly the same. From the point of the view, HSC can replace DMHAN-MMH in separation Pu from U in APOR process.

IV. CONCLUSION

As a new organic reductant, HSC can effectively realize the separation of Pu from U, and excellent result is achieved, that is, the separation factor of plutonium from uranium ($\text{SF}_{\text{Pu/U}}$) is 2.8×10^4, and separation factor of uranium from plutonium ($SF_{\text{U/Pu}}$) is 5.9×10^4.

The separation effect using HSC as reductant is nearly the same as DMHAN-MMH, and further, no peak of plutonium exists compared to the result using DMHAN-MMH as reductant.

So HSC has good application prospects in Purex process, and is a more likely alternative reductant in separation of Pu from U in APOR process.

[1] Suzuki A. Handbook on process and chemistry of nuclear fuel reprocessing (Version 2). Japan Atomic Energy Agency, Tokyo, 2008 .

[2] Schlea C S, Caverly M R, Henry H E, *et al.* Uranium(IV) nitrate as a reducing agent for plutonium(IV) in the Purex process. Technical report, DP-808, E. I. du Pont de Nemours & Co., Savannah River Laboratory, Aiken, SC, 1963. DOI: 10.2172/4651247

[3] Koltunov V S and Baranov S M. Organic derivatives of hydrazine and hydroxylamine in future technology of spent nuclear fuel reprocessing. Global'95, Versailles, France, 1995, 577–583.

[4] Yarbro S L, Schreiber S B, Ortiz E M, *et al.* Reducing Pu(IV) to Pu(III) with hydroxylamine in nitric acid solutions. J Radioanal Nucl Ch, 1998, **235**: 21–25. DOI: 10.1007/bf02385931

[5] Scott Barney G. A kinetic study of the reaction of plutonium(IV) with hydroxylamine. J Inorg Nucl Chem, 1976, **38**: 1677–1681. DOI: 10.1016/0022-1902(76)80660-4

[6] Koltunov V S and Baranov S M. Organic derivative of hydrazine and hydroxylamine in future technology of spent fuel reprocessing. Radiokhimiya, 1993, **35**: 67–71.

[7] Yan T H, Zhen W F, Ye G A, *et al.* Synthesis of dihydroxyurea and its application to the U/Pu split in the PUREX process. J Radioanal Nucl Ch, 2009, **279**: 293–299. DOI: 10.1007/s10967-007-7289-1

[8] Zhang A Y, Hu J X, Zhang X Y, *et al.* Hydroxylamine derivative in Purex process. J Radioanal Nucl Ch, 2002, **252**, 565–571. DOI: 10.1023/A:1015867107219

[9] Li G L and He H. Study on mechanism for oxidation of N, N-dimethylhydroxylamine by nitrous acid. J Radioanal Nucl Ch, 2011, **287**, 673–678. DOI: 10.1007/s10967-010-0912-y

[10] He H, Ye G A, Tang H B, *et al.* An advanced Purex process based on salt-free reductants. Radiochim Acta, 2014, **102**: 127–133. DOI: 10.1515/ract-2014-2120

[11] Liu Z J and Guo B. Review of gas test technology of hydrazine rocket propellant. Low Temp Special Gases, 2007, **25**: 37–42. (in Chinese) DOI: 10.3969/j.issn.1007-7804.2007.02.009

[12] Li G L, He H, Hu J X, *et al.* The synthesis and characterization of N, N-dimethylhydroxylamine. Chemical Reagents. 2006, **28**: 202–204. (in Chinese) DOI: 10.3969/j.issn.0258-3283.2006.04.004

[13] Chen H, He H, Li G L, *et al.* Pulse radiolysis of aqueous N, N-Dimethylhydroxylamine solutions. J Radiat Res Radiat Process, 2010, **28**: 138–143. (in Chinese)

[14] Xiao S T, Ye G A, Liu X C, *et al.* Kinetics of reaction between Pu and Hydroxysemicarbazide in nitric acid solution. Atom Energy Sci Technol. 2011, **45**: 277–281. (in Chinese) DOI: 10.7538/yzk.2011.45.03.0277

[15] Xiao S T, Ye G A, Liu X C, *et al.* Study on kinetics of reaction between Hydroxysemicarbazide and HNO_2 in perchloric acid solution. Atom Energy Sci Technol. 2011, **45**: 403–407. (in Chinese) DOI: 10.7538/yzk.2011.45.04.0402

[16] Ren S J, Wang R B, Komatsu K, *et al.* Synthesis, biological evaluation, and quantitative structure activity relationship analysis of new schiff bases of hydroxysemicarbazide as potential antitumor agents. J Med Chem, 2002, **45**: 410–419. DOI: 10.1021/jm010252q

[17] Johnson D A and Florence T M. Spectrophotometric determination of uranium(VI) with 2-(5-bromo-2-pyridylazo)-5-diethyl- aminophenol. Anal Chim Acta, 1971, **53**: 73–79. DOI: 10.1016/S0003-2670(01)80072-6

[18] Yu E J, Huang H A, Zhou X Q, *et al.* Plutonium portioning in the third plutonium cycle of the Purex process with hydrazine stabilized hydroxylamine nitrate. Atom Energy Sci Technol. 1993, **27**: 80–88. (in Chinese)