

Homogeneous liquid–liquid extraction of europium from aqueous solution with ionic liquids

Ying Dai^{1,2} · Bin Cao^{1,2} · Shimin Zhong² · Guanbao Xie² · Youqun Wang^{1,2} · Yuhui Liu^{1,2} · Zhibin Zhang^{1,2} · **Yunhai Liu1,2 · Xiaohong Cao1,2**

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Abstract

Comparing with the traditionally immiscible two-phase extraction, the homogeneous liquid–liquid extraction technique shows potential in industrial separation engineering due to nearly infnite contact interface. In this work the ionic liquid (IL) compounds such as *N*-(carboxymethyl)-*N*,*N*-dimethylethanaminium bis-trifluoromethane-sulfonimide ([DHbet][Tf₂N]) and *N*-(carboxyethyl)-trimethylammonium bistrifluoromethane-sulfonimide ([THbet][Tf₂N]) were synthesized. The homogeneous extraction behaviors of europium with two ILs were studied as functions of solution pH, ionic strength, contact time, and initial europium concentration. The results indicated that both homogeneous extractions were dependent on pH and independent on ionic strength. The extraction capacities for $[DHbet][Tf_2N]$ and $[THbet][Tf_2N]$ were 3.29 mmol/L and 3.16 mmol/L, respectively. ILs could be recovered using 1.0 M hydrochloric acid. The mole-ratio method indicated the formation of a mononuclear complex between the europium ion and IL. Total europium extraction efficiencies of more than 91% for [DHbet][Tf₂N] and more than 90% for [THbet][Tf₂N] were obtained by quadruple-stage countercurrent extraction. The result proves the feasibility of the homogeneous liquid–liquid extraction technique as an alternative option for europium separation from aquatic solution.

Keywords Homogeneous extraction · Europium · Complex formation · Ionic liquid

Introduction

The nuclear electricity power station generates high density energy with nearly greenhouse gas zero emission $[1-3]$ $[1-3]$. Nevertheless the resultant spent fuel contains more than 300 kinds of radioactive nuclides including fission products, activation elements, ultra uranium nuclides and unutilized uranium [[4](#page-5-2)], which in most cases is processed to recover uranium and plutonium with the well-known PUREX extraction process [\[5,](#page-5-3) [6](#page-5-4)]. The residual wastewater as a result of the process retains major radioactivity and a large proportion of nuclides. The treatment and ultimate disposal of the

 \boxtimes Xiaohong Cao xhcao321@163.com

¹ State Key Laboratory of Nuclear Resources and Environment, East China University of Technology, Nanchang 330013, Jiangxi, China

wastewater is thus a challenging problem. The radioactive metal americium-241 (241 Am) is significant on account of high radioactivity (specifc activity of 3.43 Ci/g), heavy metal toxicity and a long half-life of 432.2 years [\[7](#page-6-0)]. Exposure to 241 Am would cause the lesion of tissues and cancer [\[8](#page-6-1)]. With the aim of diminishing the adverse effect, the separation of 241 Am from aquatic waste is being attempted with adsorption $[9, 10]$ $[9, 10]$ $[9, 10]$ $[9, 10]$, solvent extraction $[11–13]$ $[11–13]$ $[11–13]$, and chemical precipitation $[14]$ $[14]$. It is worth noting that the solvent extraction technique shows promising with advantages of large handling capacity, continuous operation and high enrich-ment factor based on the multi-stage operation [\[15–](#page-6-7)[21](#page-6-8)]. Although being powerful, efficient and reliable, the traditionally immiscible two-phase extraction technique deploys too much not friendly organic solvent of volatilization, hazard and toxicity [\[13](#page-6-5), [16](#page-6-9), [22](#page-6-10)]. Replacing solvent and altering extraction way would promote the applicability of solvent extraction.

Ionic liquids (ILs) refer to fuid salts made of ions and possess unique physicochemical properties such as lowvolatilization, low melting point, high cohesive energy and

School of Chemistry, Biological and Materials Sciences, East China University of Technology, Nanchang 330013, Jiangxi, China

non-combustibility compared with molecular liquids [\[23](#page-6-11)]. Over the past decades ILs has been extensively studied in the fields of catalysis $[24]$ $[24]$, synthesis $[25]$ $[25]$, separation $[26]$ $[26]$ $[26]$, transformation and utilization of biomass [[27\]](#page-6-15). The traditionally immiscible two-phase extraction processes utilize ILs as diluents, which do not participate coordination reaction. However the viscosity of ILs restricts the mass transfer area, increases the equilibrium time, and limits the separation performance [[25](#page-6-13)]. Recently the hydro-soluble ILs at a certain temperature shows attractivity. It means changing the temperature to interconvert two-phases and homogeneous phase. The phase interconversion phenomenon based on temperature is thermomorphic behavior, based on which the homogeneous liquid–liquid extraction (HLLE) technique is established. Several prominent advantages associated with the technique include infnite contact interface and fast kinetics. For example 1-hexyl-3-methylimidazolium tetrafluoroborate $[C_6mim][BF_4]$ could separate silver metal from 4,4-bis-(dimethylamino)-thiobenzophenone solution by HLLE, in which the critical thermomorphic temperature was at 323 K [\[28](#page-6-16)]. Onghena and Binnemans [[29\]](#page-6-17) proved the binary phase of betainium bis(trifuoromethylsulfonyl)imide with water could form a homogeneous phase at \geq 328 K to extract metals. Based on the reported references, it is convinced that the HLLE technique based on a specifc IL can be applied in the domain of Am extraction.

In the present work, we synthesized two novel ionic liquids named *N*-(carboxymethyl)-*N*,*N*-dimethylethanaminium bistrifluoromethanesulfonimide ([DHbet][Tf₂N]) and *N*-(carboxyethyl)-trimethylammonium bistrifuoromethane-sulfonimide ([THbet][Tf₂N]) (shown in Fig. [1](#page-1-0)), which could form homogeneous phase with water at 358.15 K, 343.15 K, respectively. Efects of pH, contact time and ionic strength on the homogeneous extraction of europium were studied, in which europium worked as a nonradioactive surrogate for Am due to same oxidation state, similar ionic radius and coordination chemistry [[30\]](#page-6-18). The complex formations and dissociation constants were discussed using the moleratio method. The stripping experiment was conducted to study the recovery performance. Multistage counter-current extraction was simulated.

Fig. 1 The chemical structures of **a** [DHbet][Tf₂N] and **b** [THbet] $^{1+00}$, $^{1+10}$, $^{19-0}$, $^{19-0}$, $^{19-0}$, $^{19-0}$, $^{10-0}$, $^{11/2}$, $^{10-0}$, $^{11/2}$, $^{10-0}$, $^{11/2}$, $^{10-0}$, $^{11/2}$, $^{10-0}$, $[Tf_2N]$

Reagents and instruments

Reagents

All chemical reagents used in the study were of analytical grade and directly used without purifcation unless otherwise specifed. Europium nitrate hexahydrate, *N*,*N*-Dimethylethylamine, trimethylammonium and bistrifuoromethanesulfonimide lithium salt were purchased from Sinopharm Chemical Reagent Co., Ltd. (Shanghai, China). Arsenazo III purchased from the Aladdin Chemistry Co., Ltd. (Shanghai, China) was used to quantitatively analyze the concentration of the europium ion.

Instruments

FT-IR spectrums were measured in ATR mode using a Ge crystal plate on a Vector Model (Bruker, Switzerland) spectrometer. ${}^{13}C$ NMR and ${}^{1}H$ NMR spectrums were recorded in an Avance DMX500 (Bruker, Switzerland) spectrometer. The metal ion concentration was determined by using a 721E model UV–Vis spectrophotometer (Shanghai Spectrum, China).

Preparation of [DHbet][Tf₂N] and [THbet][Tf₂N]

 $[DHbet][Tf_2N]$ was synthesized according to the following the two-step method. A 500 mL three-necked flask with a refux condenser was flled with chloroacetic acid $(0.317 \text{ mol}, 30 \text{ g})$ and DMF (100 mL) . The system was stirred to form a homogeneous phase, to which *N*,*N*-dimethylethylamine (0.547 mol, 40 g) was batch-wise added. The mixture was heated to a constant temperature of 60 °C for 6 h followed by suction fltration, vacuum drying. After that, a white powder solid as the crude product was obtained. The further purifcation was operated by the recrystallization method. The pure product, *N*-(carboxymethyl)-*N*,*N*dimethylethanaminium hydrochloride, was separated out from 150 mL ethyl alcohol.

The ionic liquid, [DHbet][Tf₂N], was obtained by the replacement reaction. The method in detail was described as the following. *N*-(carboxymethyl)-*N*,*N*-dimethylethanaminium (0.0871 mol, 14.45 g) and bistrifuoromethanesulfonimide lithium (0.0871 mol, 25 g) were added into distilled water (20 mL), followed by stirring for 1 h at room temperature. Then the mixture was centrifuged to separate the sublayer ionic liquid, which was washed five times to remove the reaction byproduct, LiCl. Afterward, the pure ionic liquid, [DHbet][Tf₂N], was obtained. FT-IR (cm⁻¹): 1740, 1468, 1415, 1340, 1340, 1129, 1045, 1179, 735, 653, 607; 3.51 (s, 2H, CH₂), 3.22 (s, 6H, $2 \times$ CH₃), 1.34 (s, 3H, CH₃); ¹³C-NMR (75 MHz, DMSO, δ/ppm): 167.28 (COO), 120.93, 118.10 (CF₃), 60.38 (CH₂), 50.93 (CH₃), 40.53 (CH₂).

According to the above path [THbet][Tf₂N] was synthesized from the initial raw material of trimethylammonium. FT-IR (cm−1): 1731, 1480, 1335, 1130, 1046, 1177, 739, 785, 613. 1 H-NMR (300 MHz, DMSO, δ/ppm): 3.56 (s, 2H, CH₂), 2.77 (s, 2H, CH₂), 3.06 (s, 9H, $3 \times CH_3$). ¹³C-NMR (75.47 MHz, DMSO, δ/ppm): 172.55 (COO), 121.29, 118.91 $(2 \times CF_3)$, 61.58 (CH₂), 44.16 (CH₂–CH₂), 54.02 (CH₃).

Homogeneous solvent extraction study

Prior to a homogeneous extraction operation, the ionic liquid ([DHbet][Tf₂N] or [THbet][Tf₂N]) was preequilibrated to a certain pH by once contacting with an identical volume of metal-free solution of specifc pH. The pH of aqueous phase was adjusted with 0.5 mol/L nitric acid and NaOH solution. Therefore, equal volume of IL and the aqueous phase containing europium of a certain concentration were set into a conical fask and heated to the critical thermomorphic temperature to form a homogeneous system. After a certain period, the solution was cooled down to form two phases. The residual europium concentration in the aqueous solution was detected with the arsenazo III method with the UV–Vis spectrophotometer. The distribution ratio (*D*) was calculated by taking the ratio of equilibrium concentration of metal ion in the organic phase and in the aqueous phase (Eq. [1\)](#page-2-0).

$$
D_{\rm Eu} = \frac{[C]_{\rm org}}{[C]_{\rm aq}} = \frac{C_{\rm i} - C_{\rm f}}{C_{\rm f}} \times \frac{V_{\rm aq}}{V_{\rm org}} \tag{1}
$$

where $[C]_{\text{org}}$ and $[C]_{\text{aq}}$ represent europium content in the ionic liquid phase and the aqueous phase (mol/L), respectively; C_i and C_f are the initial and residual concentration of europium (mol/L), respectively; V_{aq} and V_{org} are volumes of the aqueous phase and ionic liquid (mL). All extraction experiments were duplicated at least to make reliable and repeatable results.

Results and discussion

Efect of pH on the extraction

The solution pH plays essential role in the metal species distribution and the extraction extent [[31](#page-6-19), [32](#page-6-20)]. Considering europium hydrolysis beyond pH 7.0 [\[33,](#page-6-21) [34](#page-6-22)], the effect of pH in the range of 1.0–7.0 on the extraction was investigated. The result is shown in Fig. [2a](#page-2-1). One could see that as pH increased from 1.0 to 3.0, the distribution ratio (*D*) increased from 1.39 to 1.91 for [DHbet][Tf_2N] and from 1.11 to 1.73 for [THbet][Tf₂N], further slightly decreased to 1.85 for [DHbet][Tf₂N] and to 1.67 for [THbet][Tf₂N] as pH increased to 7.0. The explanation for the plot was the following. The contributing coordination group when extracting europium was the carboxyl in the chemical structure of two ILs [[35\]](#page-6-23). As pH increased from 1.0 to 3.0, the less protonated carboxyl group coordinated europium efectively. Once pH beyond 3.0 the slight protonation exerted weak infuence on the extraction and the value of *D* kept constant. Further experiments were conducted at $pH = 3.0$.

As shown in Fig. [2](#page-2-1)b, the extraction using [DHbet] [Tf₂N] and [THbet][Tf₂N] at $pH = 3.0$ weakly dependent on ionic strength was accordance with the chemical process in nature [[36](#page-6-24)–[38\]](#page-6-25).

Fig. 2 Effects of **a** pH and **b** ionic strength on europium extraction with [DHbet][Tf₂N] and [THbet][Tf₂N] ($C_{Eu} = 5$ mmol/L, $V_{aq}/V_{org} = 1$, *t*=20 min and *T*=358.15 K)

Fig. 3 Efects of contact time on europium extraction with [DHbet] [Tf₂N] and [THbet][Tf₂N] ($C_{Eu} = 5$ mmol/L, pH = 3.0, $V_{a0}/V_{org} = 1$ and $T = 358.15$ K)

Contact time

The extraction at diferent contact times was researched and the result was shown in Fig. [3.](#page-3-0) It could be seen that in the first 5 min *D* reached 1.62 for [THbet][Tf₂N] and 1.70 for [DHbet][Tf₂N]. After 10 min the values were 1.69 for [THbet][Tf₂N] and 1.90 for [DHbet][Tf₂N] and kept constant further, illustrating the accomplishment of equilibrium. The results proved that the extractions were fast processes. The traditionally immiscible two-phase extraction is based on two immiscible solvents, in which the limited contact interface depresses the substance transmission, leading a relatively long equilibrium time. For example, Tan et al. [\[39\]](#page-6-26) found that $2,6$ -bis(5,6-diethyl-1,2,4-triazin-3-yl)pyridine dissolved in room temperature ionic liquids exhibited slow kinetics over 24 h in a two-phase extraction toward europium. The extraction herein utilized the ability of the specifc ionic liquid to form a homogeneous phase with aqueous solution at a certain temperature. The homogeneous system made the contact interface infnite, resulting suffciently reaction between the ILs and target in a short time.

Efect of initial concentration of europium on the extraction

The experiments varying the initial europium concentration at 0.5, 1.0, 3.0, 5.0, 10.0, 15.0, 40.0 mmol/L were conducted to investigate the maximum extraction capacity of [DHbet] [Tf₂N] and [THbet][Tf₂N]. As shown in Fig. [4](#page-3-1), the europium concentration in ionic liquid phase increased rapidly from 0.3 to 3.29 mM for [DHbet][Tf₂N] and from 0.3 to 3.16 mM for $[THbet][Tf_2N]$ as the initial europium concentration varied from 0.5 to 5.0 mM. This was attributed to the greater

Fig. 4 Effects of initial europium concentrations on [DHbet] [Tf₂N] and [THbet][Tf₂N] (pH=3.0, $V_{aq}/V_{org}=1$, $t=20$ min and *T*=358.15 K)

gradient caused by the higher europium concentration. On the other hand, the concentration of europium extracted kept at constant once the initial europium concentration beyond 5.0 mM. This was because the ionic liquid phases reached saturation. The maximum europium extraction capacity were determined as 3.29 mmol/L for [DHbet][Tf₂N] and 3.16 mmol/L for [THbet][Tf₂N].

Stripping performance

The stripping research was carried out using hydrochloric acid as stripping agent. Solutions of ionic liquids saturated with europium were prepared in advanced. The stripping experiment was conducted by mixing the stripping

Fig. 5 Effect of hydrochloric acid concentration on the stripping performance

hydrochloric acid solution of 0.2, 0.4, 0.6, 0.8, or 1.0 mol/L with the equal volume of the europium saturated ionic liquids. The europium recovery efficiency corresponding to the hydrochloric acid concentration is shown in Fig. [5](#page-3-2). It could be seen that as the acid concentration increased, the recovery percentage increased. The hydrochloric acid of 1.0 mol/L could recovery 94% europium from Eu-[DHbet][Tf₂N] and 93% europium from Eu-[THbet][Tf₂N], respectively.

Complex formation and dissociation constants of Eu-[DHbet][Tf₂N] and Eu-[THbet][Tf₂N]

In the mole-ratio method, a series of aqueous samples were made in which the concentration of ionic liquid ([DHbet] [Tf₂N] or [THbet][Tf₂N]) kept constant while that of europium varies. The maximum absorbance–wavelength of the complex were qualitatively measured as 281 nm for [DHbet] [Tf₂N] and as 289 nm for [THbet][Tf₂N] firstly which were located in the ultraviolet spectral region due to the weak conjugation efect. Then the absorbance of each aqueous sample was surveyed and plotted versus the mole ratio between europium and the ionic liquid. It is a reasonable prediction that the absorbance would increase as the mole ratio increased and reach a constant after the complex concentration come up to the maximum. In the curve, a break appears at the mole ratio of the complex composition. As shown in Fig. 6 , two tangents intersecting at $(1.0, 0.79)$ for [DHbet][Tf₂N] and (1.0, 0.74) for [THbet][Tf₂N] indicated that Eu and the ionic liquids reacted in 1:1 ratio that were mononuclear complex.

For a typical complex dissociation reaction:

 $Eu_mIL_n \Leftrightarrow mEu + nIL$

The dissociation constant of the complex, *K*, can be expressed as the following equation (Eq. [2\)](#page-4-1).

$$
K = \frac{[C_{\rm Eu}]^m [C_{\rm IL}]^n}{[C_{\rm Eu_mIL_n}]} = \frac{[m\alpha C]^m [n\alpha C]^n}{(1-\alpha)C} = \frac{m^m n^n \alpha^{m+n} C^{m+n-1}}{1-\alpha}
$$
(2)

where α is the dissociation degree, equal to the difference value of 1.0 and the ratio for a defned coordination value (A_d) to the theoretical maximum absorbance number for coordination (A_0) , namely 1.0- A_d/A_0 , and *C* is the complex concentration at the break point.

Since both *m* and *n* values had been determined as 1.0 and *α* value of Eu-[DHbet][Tf₂N] was calculated as 0.113, the value for Eu-[THbet][Tf₂N] was 0.108, $C = 1.0$ mmol/L, the dissociation constant for Eu-[DHbet][Tf₂N] was 0.0143 and for Eu-[THbet][Tf₂N] was 0.0131 .

Multistage countercurrent extraction process

Multistage countercurrent extraction process is an important separation technique and is often simulated using the cascade extraction in lab [[40](#page-6-27)]. This study used a four-stage cascade extraction (shown in Fig. [7\)](#page-5-5), in which the europium concentration of the feed solution = 5.0 mmol/L, $V_{\text{a}q}/V_{\text{org}}=1$, contact time=20 min, and $T=358.15$ K. The europium concentrations in every round rafnate for [DHbet] [Tf₂N] were $C(R_1) = 15.31$ mg/L, $C(R_2) = 14.03$ mg/L, $C(R_3) = 16.98$ mg/L, $C(R_4) = 21.29$ mg/L and for [THbet] [Tf₂N] were $C(R_1) = 15.31$ mg/L, $C(R_2) = 14.03$ mg/L, $C(R_3) = 16.98 \text{ mg/L}, C(R_4) = 21.29 \text{ mg/L},$ respectively. Theoretically $C(R_x)$ increased as the cascade number increased. Generally $C(R_1)$ should be less than $C(R_2)$ due to the more fresh ionic liquid was used in every vessel. However the value of $C(R_1)$ was greater than $C(R_2)$ in this study. This was attributed to unsteady state in the frst two rounds. Thus one could see that $C(R_3)$ and $C(R_4)$ values were greater than $C(R_1)$ and $C(R_2)$ values, and $C(R_3) < C(R_4)$. The total

Fig. 6 Formation curves of \mathbf{a} Eu-[DHbet][Tf₂N] and \mathbf{b} Eu-[THbet][Tf₂N] complex system

Fig. 7 Extraction of europium in four-stage cascade extraction process using [DHbet] [Tf_2N] or [$THbet$][Tf_2N]. $(C_{Eu} = 5.0 \text{ mmol/L}, V_{\text{ao}}/V_{\text{org}} = 1,$ $t = 20$ min and $T = 358.15$ K)

extraction efficiencies were calculated as 91.12% europium for [DHbet][Tf₂N] and as 90.55% europium for [THbet] $[Tf_2N]$ by using Eq. [3](#page-5-6). It was convincible that the extraction efficiency could be further improved by more than 4-stage extraction.

$$
Ex(Eu)\% = \left(1 - \frac{\sum_{i=1}^{4} C_{\text{R}i} \times V_{\text{R}i}}{C_{\text{F}} \times V_{\text{F}}}\right) \times 100\%
$$
 (3)

Conclusion

In consideration of drawbacks including limited contact interface and relatively long equilibrium time of the traditionally immiscible two-phase extraction, a homogeneous solvent extraction technique toward europium was initially developed herein based on ionic liquid $[DHbet][Tf_2N]$ and [THbet][Tf₂N]. Effects of pH, contact time and ionic strength on the europium extraction were studied. It was demonstrated that the europium extraction was dependent on pH. The optimal value was 3.0. Almost infnite contact interface created by the homogeneous phase state possessed fast mass transfer kinetics, made the extraction reach equilibrium within 10 min. The maximum europium extraction capacity were determined as 3.29 mmol/L for [DHbet][Tf_2N] and 3.16 mmol/L for [THbet][Tf₂N]. The extracted europium was efectively stripped by hydrochloric acid. Once contact with hydrochloric acid (1.0 mol/L) could strip ca. 94% of the extracted europium. The mole-ratio method demonstrated europium and ILs reacted in 1:1 ratio that is mononuclear complex. Simulating four-stage counter-current operation extracted more than 91% europium for [DHbet][Tf₂N] and more than 90% europium for [THbet][Tf₂N]. The study shows that the homogeneous solvent extraction process based on [DHbet][Tf₂N] and [THbet][Tf₂N] are effective for europium separation.

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