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Selective separation of uranium from sulfuric acid media using a polymer inclusion membrane containing alamine336

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Abstract

This study reports the extraction of U(VI) using poly(vinyl chloride)-based polymer inclusion membrane (PIM), possessing the extractant alamine336 and the plasticizer Polyoxyethylene alkyl ether (POE). The effects of transport parameters such as alamine336 and POE concentration in the PIM composition, uranium and sulfuric acid concentration in the feed phase, and the receiving phase type and concentration were discussed. Under the optimum conditions, $91.02 \pm 2\%$ of uranium was transported through the PIM. The prepared PIM showed excellent selectivity for U(VI) from the sulfuric acid leach solution. The stability of the PIM was found to be acceptable for 5 days.

Keywords Polymer inclusion membrane · Uranium · Alamine336 · Sulfuric acid leach solution · Selectivity · Stability

Introduction

Liquid membrane (LM) technique is an efficient and simple separation method that can compete with conventional solvent extraction techniques (Aguilar and Cortina 2008). Some advantages of this method compared with the solvent extraction are simultaneous extraction and receiving at one stage, low consumption of extractant, possible complete transfer of solute because of non-equilibrium mass transfer. In comparison with the solvent extraction method, formation of the third phase, phase entrainment, and flooding can be avoided in liquid membrane technique. However, the significant disadvantage of the liquid membrane is the low stability, which should be considered before any large-scale applications (Kislik 2010; Mahanty et al. 2015).

Polymer inclusion membranes (PIMs) are similar to the supported liquid membranes (SLMs) where an extractant is entrapped within a polymer matrix, usually with adding a plasticizer to modify the chemical species solubility within the membrane (Almeida et al. 2012; St John et al. 2012). In the PIM technique, the loosing of the carrier from the base polymer into the aqueous phases is very low and can

Parisa Zaheri pzaheri@aeoi.org.ir alleviate the stability issues of LMs (Mahanty et al. 2016). So, in this investigation, the separation of uranium from sulfuric acid media was studied using the PIM system.

Liquid membrane techniques have been used for the separation of uranium by many investigators (Mohapatra et al. 2006; El Sayed 2003; Shamsipur et al. 2010; Davarkhah et al. 2013; Biswas et al. 2013; Elsayed et al. 2013). However, there are not many studies on the separation of uranium using PIMs. Matsuoka et al. (1980) reported the transport of uranium using a PIM containing TBP (three butyl phosphate) as an extractant. However, because of the high water solubility of TBP, the PIM has not enough stability for practical application. Bayou et al. used cellulose triacetate (CTA) PIM, including trioctylphosphine oxide (TOPO) as an extractant and 2-nitrophenyl octyl ether (NPOE) as a plasticizer for investigating the transport of uranium and molybdenum ions. They compared the transport properties of this PIM with an SLM and showed that the uranium and molybdenum transport efficiency was high using the PIM system. However, they were not able to extract uranium completely (Bayou et al. 2010). St John et al. used some commercial extractants for uranium separation from sulfate media using a PIM system. They showed that the PIM with compositions 40% (m/m) di-(2-ethylhexyl)phosphoric acid (D2EHPA) and 60% (m/m) poly(vinyl chloride) (PVC) had superior ability to separate U(VI) (St John et al. 2010). In another work, they showed that uranium transport across a PIM containing 35% D2EHPA, 10% NPOE and 55% PVC in comparison

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with a PIM consisting of 45% D2EHPA and 55% PVC, was only a little faster. So, the use of PIM without an expensive plasticizer was preferred (St John et al. 2012). Mahanty et al. (2016) demonstrated actinides separation from acidic feed solutions using a (CTA)-based PIM composed of C-pivot tripodal diglycolamide (T-DGA) as a carrier and NPOE as a plasticizer. They also used a branched diglycolamide as an extractant for this purpose (Mahanty et al. 2015).

So far, many high-molecular-weight amines have been used for extraction of uranium from the ore leached with sulfuric acid (Boirie 1958; Sato 1963; Kumar et al. 2010). Also, the price and extraction efficiency of these amines have been assessed. On the other hand, an amine extractant breaks down to nitrogen and oxygen; so, its disposition is easier in comparison with organophosphate extractants (Ohashi et al. 2014). However, the use of these amines as the carrier in the membrane systems is restricted (Babcock et al. 1980). One of the most efficient commercial amines for the extraction of uranium from the leaching solution is alamine336. However, despite the extensive use of alamine336 in the field of solvent extraction (Kumar et al. 2010; Nasab 2014; Quinn et al. 2013; Kim et al. 2012; Ramadevi et al. 2012) and liquid membrane (Wongkaew et al. 2014; He et al. 2000), the use of alamine336 in the PIM system for the extraction of uranium has not been reported yet in the literature. The only unsuccessful report in this regard relates to St John et al. (2010) research group that could not prepare PIMs containing alamine336 with sufficient transparency and flexibility, and also reasonably resistant to mechanical stress to extract uranium from 0.1-M H₂SO₄. Therefore, in this study, the PIM containing alamine336 as an extractant was prepared and the transport of uranium through that was investigated for the first time. Various parameters such as extractant and plasticizer concentration in the membrane, sulfuric acid concentration in the feed phase, receiving agent type and concentration, and uranium concentration in the feed phase have been investigated. The selectivity and stability of the prepared PIM were also checked.

Experimental

Reagents

Tri-octyl/decyl amine (alamine336) (NetSun Co., China), poly(vinyl chloride) with high molecular weight (Merck), and tetrahydrofuran (THF) (HPLC grade, Merck) were all used as received. Polyoxyethylene alkyl ether (POE) with three polar oxyethylene groups and an alkyl chain length of 12 was prepared from Kimyagaran Emrooz Co. (Iran). Uranyl acetate dehydrate, H_2SO_4 , (NH₄)₂CO₃, Na₂CO₃, HNO₃, H_2SO_4 , HCl, NaNO₃, NaF, NH₄Cl, and NaHCO₃ were purchased from Merck.

Apparatus

The pH value of aqueous solutions was measured using a pH meter (Sartorius).

The concentrations of U(VI) ions in aqueous phases were measured using Inductivity Coupled Plasma-Atomic Emission Spectrometry (ICP-AES, Optima 7300 DV, America).

An INSIZE digital micrometer was used for measuring the PIM thickness.

The characterization of the PIM was studied using the FT-IR spectrometer (Brukere Model).

Membrane preparation

In a glass beaker, specified amounts of PVC, POE, and alamine336 were mixed with 10 mL of THF. Throughout this paper, the amount of PIM compositions was reported in weight percent. The glass beaker was put on a magnetic stirrer for 2 h to have a uniform solution of all ingredients. Afterward, the mixture was transferred into a flat bottom petri dish to evaporate the THF and forming the PIM film. To reduce the evaporation rate of THF and have a uniform PIM film, the petri dish was covered with a perforated aluminum sheet. After 24 h, the content of the petri dish was immersed in cold water to remove the resulting polymer film. The prepared PIM had an oily surface that shows there is a little bonding capacity between the PVC and alamine336. So, to increase the hydrogen bonding capacity of alamine336, it was protonated with 0.1 mol/L sulfuric acid. To determine the thickness of the prepared PIM, the thicknesses of several points of that were averaged. A circular segment of the PIM with a diameter of 57 mm was cut and placed in a permeation cell. The effective diameter of the PIM was 47 mm. All experiments were repeated three times.

Transport studies

A glass cell comprised of two compartments for the feed and receiving solutions was used to study the transport properties of the prepared PIM. For this purpose, the prepared PIM was clamped between these two compartments. Both compartments were stirred with a magnetic stirrer at 600 rpm. The extraction and back-extraction rate and the average backextraction flux (J_{av}) were revealed by monitoring U(VI) concentration in the aqueous phases by Eqs. (1)–(3).

Etraction% =
$$\frac{C_{\text{U},0} - C_{\text{U},\text{f}}}{C_{\text{U},0}} \times 100,$$
 (1)

Back - Etraction% =
$$\frac{C_{\text{U,r}}}{C_{\text{U,0}}} \times 100,$$
 (2)

$$J_{\rm av} = \left(\frac{C_{\rm U,r}}{t}\right) \left(\frac{V}{A}\right),\tag{3}$$

where $C_{U^{\dagger}0}$ is the primary U(VI) concentration in the feed solution. $C_{U^{\dagger}f_{,}}$ and $C_{U^{\dagger}r_{,}}$ are the concentrations of U(VI) in the feed and receiving solutions at time *t*, respectively. *V* is the receiving phase volume, and *A* is the PIM effective surface area, respectively.

The separation factor (S_F) was calculated as in Eq. 4:

$$S_{\rm F(U/X)} = \frac{(C_{\rm U}/C_{\rm X})_{\rm r}}{(C_{\rm U}/C_{\rm X})_{\rm f}},$$
(4)

where $C_{\rm U}$ is the U(VI) concentration, and $C_{\rm X}$ is the other ions concentration in the sulfuric acid leach solution. Subscripts r and f represent the receiving phase and the feed phase, respectively. All experiments were conducted at ambient temperature (25 °C).

Results and discussion

Fig. 1 Prepared PIM contain-

(0.1 mol/L)

ing **a** alamine336, **b** protonated alamine336 with sulfuric acid

Influence of alamine336 content in the PIM

As mentioned above, to increase the hydrogen bonding capacity of alamine336, it was protonated with 0.1 mol/L



sulfuric acid before use for preparing a PIM. The results in Fig. 1 show that the PIM containing protonated alamine336 is transparent and without oily surface. To see the effect of alamine336 content on the uranium transport, various amounts of that were used in the PIM composition according to Table 1. As revealed in Fig. 2, U(VI) was not transported in the absence of alamine336. By adding alamine336 to the PIM composition, the flux of U(VI) grew considerably as the alamine336 content was enhanced to 40 wt%. In these conditions, the presence of numerous extracted species at the interface of the feed and membrane rises the transport of U(VI) across the PIM (St John et al. 2012). The mechanistic description of U(VI) transport with alamine336 was explained in Sect. 3.6. Further increase in the amount of alamine336 led to a PIM with an oily surface and weak mechanical resistance, and hence it was difficult to handle the membrane for transport experiments. So, the high content of the alamine336 (above 40 wt%) was not investigated.

Influence of plasticizer content in the PIM

The use of plasticizers in the PIM structure often modifies the membrane plasticity and softness that provides conditions for easy transport of metal ions through that (Mahanty et al. 2016). The influence of plasticizer content was studied using PIMs, including the different amounts

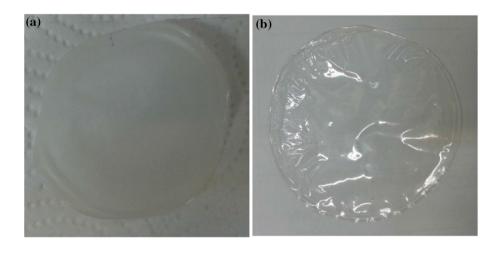


Table 1PIMs containingvarious content of alamine336

PIM composition			Alamine336 wt%	PIM thickness	Appearance	
PVC (mg)	POE (mg)	Alamine336 (mg)		(µm)		
215	65	0	0	25±3	Flexible, transparent	
173	65	42	15	25 ± 3	Flexible, transparent	
150	65	65	23.21	25 ± 3	Flexible, transparent	
103	65	112	40	25 ± 3	Flexible, transparent	
47	65	168	60	25 ± 3	Oily surface, poor mechanical resistance	

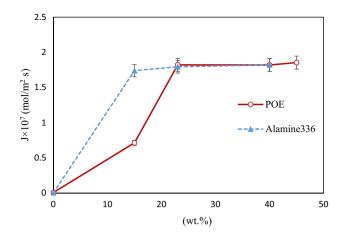


Fig.2 Transport of U(VI) across alamine336/PVC PIMs with different content of alamine336 or POE (feed phase: 1.0×10^{-4} mol/L U(VI) in 0.1 mol/L H₂SO₄; receiving phase: 0.5 mol/L (NH₄)₂CO₃)

of POE (Table 2). The obtained results in Fig. 2 show there is no transport of U(VI) through the PIM without any plasticizer. After that, increasing the plasticizer content up to 23.21 wt% increases PIM permeability significantly. Because, the presence of POE in the PIM structure improves the movements of extractant and extracted species (Yildiz et al. 2016). After that, there is no change in transport efficiency with increasing plasticizer amount. So, 23.21 wt% POE was chosen as the optimal content in subsequent experiments.

Influence of sulfuric acid concentration in the feed phase

The effect of sulfuric acid concentration in the feed phase on U(VI) flux was investigated in the range of 0.01-1.0 mol/L. As shown in Fig. 3, the maximum flux was obtained for 0.1 mol/L sulfuric acid. Kumar et al. (2010) observed this trend for the solvent extraction of uranium using alamine336. Based on the results obtained from solvent extraction studies in the literature (TRÉMILLON 1965), the maximum flux in the presence of 0.1 mol/L sulfuric acid can be attributed to the conversion of alamine336 (R₃N) in the membrane phase into

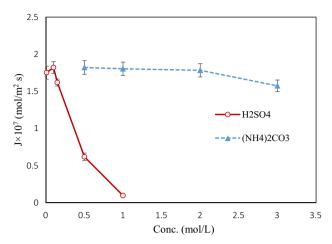


Fig. 3 Transport of U(VI) across (40 wt% Alamine336/23.21 wt% POE/36.78 wt% PVC) PIMs with different concentrations of sulfuric acid in the feed phase (feed phase: 1.0×10^{-4} mol/L U(VI); receiving phase: 0.5 mol/L (NH₄)₂CO₃) and with different concentrations of ammonium carbonate in the receiving phase (feed phase: 1.0×10^{-4} mol/L U(VI) in 0.1 mol/L H₂SO₄)

favorable amine sulfate species according to the following reaction,

$$\left(\mathrm{H}^{+} + \mathrm{HSO}_{4}^{-}\right) + 2\mathrm{R}_{3}\mathrm{N} \rightleftharpoons (\mathrm{R}_{3}\mathrm{NH})_{2}\mathrm{SO}_{4},\tag{5}$$

and subsequently, the extraction of uranium U(VI) according to the following reaction,

$$UO_2SO_4 + 2(R_3NH)_2SO_4 \rightleftharpoons (R_3NH)_4UO_2(SO_4)_3.$$
(6)

The drastic decline in the flux of U(VI) for concentrations higher than 0.1 mol/L sulfuric acid can also be described by conversion of alamine336 (R_3N) in the membrane phase into unfavorable amine bisulfate species as follows:

$$\left(\mathrm{H}^{+} + \mathrm{HSO}_{4}^{-}\right) + \mathrm{R}_{3}\mathrm{N} \rightleftharpoons \mathrm{R}_{3}\mathrm{NH}^{+}\mathrm{HSO}_{4}^{-},\tag{7}$$

which leads to undesirable extraction of U(VI) into the PIM as Eq. 8 shows,

$$UO_2SO_4 + 4R_3NH^+HSO_4^- \rightleftharpoons (R_3NH)_4UO_2(SO_4)_3 + 2(H^+ + HSO_4^-).$$
(8)

Table 2	PIMs containing
various	content of POE

PIM composition			POE wt%	PIM thick-	Appearance
PVC (mg)	POE (mg)	Alamine336 (mg)		ness (µm)	
168	0	112	0	25±3	Low flexibility, transparent
126	42	112	15	25 ± 3	Flexible, transparent
103	65	112	23.21	25 ± 3	Flexible, transparent
56	112	112	40	25 ± 3	Oily surface, non-transparent

Influence of receiving agent type and concentration

Experiments were also made to study the effect of the receiving agent type in the internal phase. For this purpose, ten different receiving agents, NaF, H₂O, NaHCO₃, Na₂CO₃, NH₄Cl, (NH₄)₂CO₃, H₂SO₄, HNO₃, HCl and NaNO₃ with the same concentration of 0.5 mol/L were used. Figure 4 shows that, under the same experimental conditions, the flux of uranyl ions into the receiving phase decreases in the order $(NH_4)_2CO_3 > NaHCO_3 > NaF > Na_2CO_3 > NH_4Cl > H_2SO_4$ > NaNO₃> HCl> H₂O> HNO₃. Thus, ammonium carbonate is the most efficient agent in receiving uranium from PIM containing alamine336. The results also showed that the basic agents play their roles as a receiving phase better than the acidic and neutral agents for the back-extraction of uranium. Thus, to the best of our knowledge, the most efficient mechanism for the recovery of uranium from the membrane is to neutralize the protonated amine in the structure of ion-associated complex at the interface of membrane and receiving phase, thereby the uranyl ion is abandoned into the receiving phase. Obviously, this mechanism depends on the pH and the alkaline solutions as a stripper facilitate its occurrence.

The effect of the variation of ammonium carbonate concentration in the receiving phase was also studied; the results in Fig. 3 show that with an increase in ammonium carbonate concentration higher than 0.5 mol/L, the flux of U(VI) diminishes. Thus, a solution of 0.5 mol/L ammonium carbonate could be used as the most suitable receiving phase.

Influence of U(VI) concentration in the feed phase

The variation of flux with the U(VI) concentration in the feed solution was studied in the range of 1.0×10^{-4} to 8.33×10^{-4} mol/L. The obtained results in Fig. 5 show that the U(VI) concentration has a significant effect on the transport of U(VI) through a (40 wt% alamine336/23.21 wt%)

POE/36.78 wt% PVC) PIM. According to Eq. 6, the stoichiometric ratio of U(VI) to alamine336 is 1:4. For the studied range of uranium concentration, the mole ratio of U(VI) to alamine336 is between 1:5 and 1:40. As the mole ratio of U(VI) to alamine336 is always more than the stoichiometric ratio, increasing the U(VI) concentration in the feed solution leads to increasing the U(VI) flux (St John et al. 2012). In other words, in this region, the transport is mainly determined by the uranyl ions diffusion through the boundary layer in the feed phase and the reaction of uranyl ions and alamine336 molecules (Wijers et al. 1988).

Extraction and receiving mechanism of U(VI) at the PIM interfaces

There are various species of dissolved uranyl ions in the sulfuric acid medium such as UO_2^{2+} , UO_2SO_4 , $UO_2(SO_4)_2^{2-}$ and

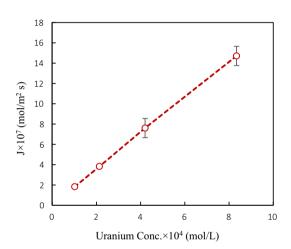


Fig. 5 Transport of U(VI) across (40 wt% Alamine336/23.21 wt% POE/36.78 wt% PVC) PIMs with different initial concentrations of U(VI) in the feed phase (feed phase: U(VI) in 0.1 mol/L H_2SO_4 ; receiving phase: 0.5 mol/L (NH₄)₂CO₃)

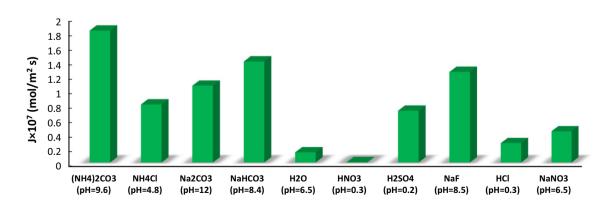


Fig.4 Transport of U(VI) across (40 wt% Alamine336/23.21 wt% POE/36.78 wt% PVC) PIMs with different receiving agents (feed phase: 1.0×10^{-4} mol/L U(VI) in 0.1 mol/L H₂SO₄)

 $UO_2(SO_4)_3^{4-}$ whose concentration is a function of the free sulfate concentration in the solution. In 0.1 mol/L sulfuric acid solution, the concentration of free sulfate is almost 0.032 mol/L (Brubaker Jr 1957). Considering the latter concentration of the free sulfate, and according to the calculations of Boirie (1958), the dominant species of uranyl ion in the solution is UO_2SO_4 . On the other hand, based on the solvent extraction studies in 0.1 mol/L sulfuric acid medium, alamine336 (R₃N) at the feed side interface forms the neutral species of (R₃NH)₂SO₄ by accepting protons. Thus, at the mentioned level of sulfuric acid concentration, uranyl ions are essentially extracted as the ion-associated complex of (R₃NH)₄UO₂(SO₄)₃ (Boirie 1958). Therefore, at the feed–membrane interface, the main reaction can be written as follows:

$$UO_2SO_4 + 2(R_3NH)_2SO_4 \rightleftharpoons (R_3NH)_4UO_2(SO_4)_3.$$
(9)

The reaction at the membrane–receiving phase interface and in the presence of 0.5 mol/L ammonium carbonate is as follows:

$$(R_3NH)_4UO_2(SO_4)_3 + 7(NH_4)_2CO_3 \rightleftharpoons 4R_3N + UO_2(CO_3)_3^{-4} + 4HCO_3^{-} + 3SO_4^{-2} + 14NH_4^{+}.$$
(10)

The transport of uranyl ions is coupled with the co-transport flow of the constituent ions of sulfuric acid, i.e., sulfate and proton ions. This transport mechanism of the uranyl sulfate as a predominant species of uranium in the feed phase across a PIM is schematically described in Fig. 6.

Membrane surface characterization

The surface characterization of the prepared PIM was studied using the FT-IR technique. The FT-IR spectrum of the PVC powder, POE, the PIM including PVC + POE and the PIM including PVC + POE + alamine336, is shown in Fig. 7. Figure 7a shows the characteristic vibrational C–H, C–Cl, and CH₂ bands of the PVC at 1431, 2916, and 619/cm, respectively. As shown in Fig. 7c, two peaks of C–O and O–H bands appear at 1118 and 3442, respectively, because of adding POE plasticizer to PVC. However, in comparison to the FT-IR spectrum of the pure POE (Fig. 7b), the O–H band has low peak intensity. This demonstrated that the POE plasticizer and PVC were banded together by O–H groups (Zaheri and Ghassabzadeh 2017).

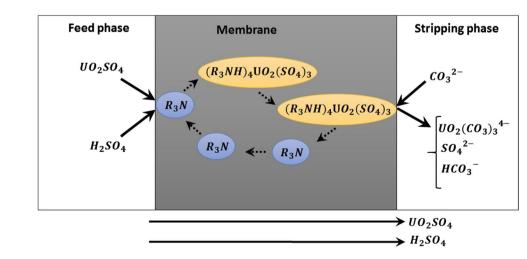
The FT-IR spectrum of the PIM containing PVC+POE+ alamine336 in Fig. 7d reveals the additional bands at 1253 and 1454/cm that relate to the C–N and CH_2 groups of alamine336 extractants. In this case, it also can be seen that the O–H band of POE has low peak intensity. This indicated that alamine336, PVC, and POE were bonded via O–H groups and this connection is strong. This is evidence of a significant increase in the uranium flux through the membrane in the presence of alamin336.

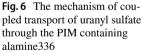
Selectivity of PIM

To study the selectivity of the synthesized membrane, in the course of a competitive transport experiment from the feed phase containing sulfuric acid leach solution, the percentage of the metal ions U(VI), Al^{3+} , Ca^{2+} , Mg^{2+} , Mn^{2+} , Fe^{3+} released in the receiving phase under optimum conditions was determined. The achieved results (Table 3) show a high selectivity of the PIM including alamine336 towards uranium ions and also a high efficiency for uranium recovery in this competitive experiment.

The kinetic of uranium transport across the PIM

Under the achieved optimum conditions, the concentration variation of uranium in the feed, membrane, and receiving phases versus time is shown in Fig. 8a. The kinetic of the transport of uranium across the PIM was described as a first-order reaction,





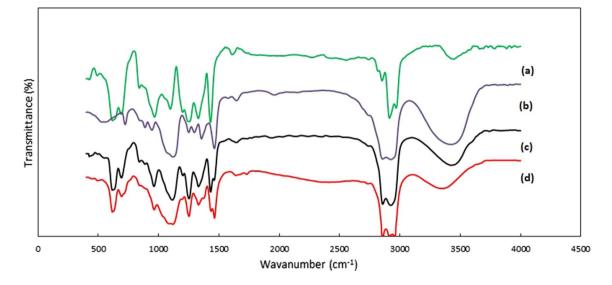


Fig. 7 FT-IR spectra of PVC (a), POE (b), PIM including PVC + POE (c) and PIM including PVC + POE + alamine336 (d)

competitive transport experiment			
Cation	Initial conc. (mg/L)	Conc. in the receiving phase after 12 h (mg/L)	Back-extraction (%)
U(VI)	92	83.73	$91.02 (\pm 2)^{a}$
Ca ²⁺	373	1.31	0.00
Fe ³⁺	768	b	0.00
Mn ²⁺	663	b	0.00
Mg ²⁺	2488	2.37	0.00
Al ³⁺	481	b	0.00

Table 3 Results of the transport of uranium and other metal ions in a

Feed phase: sulfuric acid leach solution containing 0.1 M H_2SO_4 , U(VI), Ca²⁺, Fe³⁺, Mn²⁺, Mg²⁺, Al³⁺; receiving phase: 0.5 M (NH₄)₂CO₃; transport time 12 h, Temperature 25 °C

^aThe value mentioned in the parenthesis is the standard deviation (σ_{N-1}) for three independent experiments

^bInferior than the detection limit

$$\ln\left(\frac{C_{\rm U,f}}{C_{\rm U,0}}\right) = -kt,\tag{11}$$

where k and t are the rate constant and time of transport, respectively.

To determine k value, the variation of $\ln(C_{U,f'}C_{U,0})$ vs. time is plotted in Fig. 8b. There was a linear relationship between $\ln(C_{U,f'}C_{U,0})$ and time, which was confirmed by the high value of R². The k value was calculated to be 6×10^{-5} /s. Jin et al. (1989), Kozlowski and Walkowiak (2002) also concluded the first-order reaction for metal ions.

The performance of the PIM system containing (40 wt% alamine336, 23.21 wt% POE and 36.78 wt% PVC) was compared in terms of permeability, stability, and U(VI)/Fe³⁺ separation factor with other PIM systems in Table 4. Since the experimental conditions of the used PIM systems by other investigators are different, instead of flux, the permeability was used to evaluate their transport properties by the following equation,

Fig. 8 a Concentration profile of uranium in the feed, receiving and membrane phases, **b** kinetic plot of uranium transport through the PIM

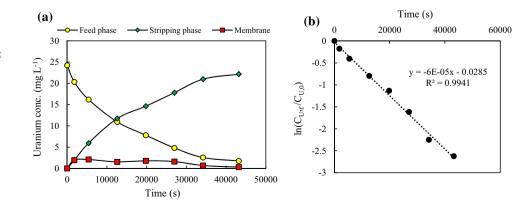


 Table 4
 Comparison of PIM systems used for transport of uranium

PIM system	10 ⁶ P (m/s)	Stability	$S_{\rm F}({\rm U/Fe^{3+}})$	References
45 wt% D2EHPA + 55 wt% PVC	6.96	> 10 days	582	St John et al. (2012)
75 wt% TBP+25 wt% PVC	2.40	1.2 days	650	Bloch et al. (1967)
62 wt% TBP+38 wt% CTA	4.76	<1 h	-	Matsuoka et al. (1980)
18 wt% TOPO + 42 wt% NPOE + 40 wt% CTA	2.15	>16 days	-	Bayou et al. (2010)
40 wt% Alamine336+23.21 wt% POE+36.78 wt% PVC	6.74	>5 days	7307	This study

Table 5 The extraction and back-extraction of U(VI) for five consecutive cycles of experiments (membrane: (40% Alamine336/23.21% POE/36.78% PVC); feed phase: 1.0×10^{-4} mol/L U(VI) in 0.1 mol/L H₂SO₄; receiving phase: 0.5 mol/L (NH₄)₂CO₃)

	Extraction (%)	Back-extraction (%)
Cycle 1	95.52	93.66
Cycle 2	95.11	93.53
Cycle 3	95.84	94.08
Cycle 4	94.06	92.65
Cycle 5	93.74	90.85

$$P = \left(\frac{V}{A}\right)k.$$
(12)

The results show that the prepared PIM in this study has exclusive transport properties and long-term stability. It is also evident that the prepared PIM has the highest separation factor towards uranium ions and Fe^{3+} ions.

Stability of PIM

The stability of the selected PIM was studied by conducting five consecutive cycles of extraction and back-extraction under the same experimental conditions. Each cycle of these five experiments was run with new feed and receiving solutions. The results in Table 5 show that the extraction and back-extraction efficiency was over 90.00% in the five cycles of experiments. The slight decline in the extraction efficiency may be related to the transporting the extractant into the aqueous solutions (Kaya et al. 2013). However, the PIM technique, due to enfolding an extractant within a polymer structure, is very persistent to the loss of the extractant in comparison with liquid membrane and solvent extraction methods.

Conclusion

The separation of U(VI) from a sulfuric solution was performed using a PVC-based PIM, including protonated alamine336 extractant and POE plasticizer. The obtained results revealed that the transport of U(VI) through the prepared PIM is affected by the amounts of the extractant and plasticizer in the PIM composition, the sulfuric acid concentration in the feed phase, and the receiving phase concentration. Under the assessed optimum conditions (PIM consisting of (40 wt% alamine336, 23.21 wt% POE and 36.78 wt% PVC), 0.1 mol/L sulfuric acid in the feed solution and 0.5 mol/L ammonium carbonate as a receiving solution), the acceptable performance with average flux value as 1.82×10^{-7} mol/m² s and very high selectivity value as 7307 for U(VI)/Fe³⁺ was achieved for the prepared PIM.

Investigating the effect of receiving agent type on uranium transport through PIM showed that the basic agents act as a better receiving phase than the acidic and neutral agents for the back extraction of uranium.

The prepared PIM showed long-term stability for five consecutive extractions of uranium.

According to high selectivity, stability, and a few use of alamine336 in the prepared PIM, it is an eco-friendly and affordable process for selective extraction of uranium from the sulfuric acid leach solution.

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