Facilitated transport of uranium(VI) across supported liquid membranes (SLM) containing T2EHDGA: Part II. Nature of feed, pore size and temperature on pertraction rates

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ABSTRACT

Facilitated transport of uranyl ion was investigated across PTFE supported liquid membranes using T2EHDGA in *n*-dodecane as the carrier extractant containing 30% *iso*-decanol as the phase modifier. The feed was usually 3 M HNO_3 while the strip phase constituted pH 2 solution. About 11 h were required for quantitative transport of U(VI) from a feed of 3 M HNO_3 using 0.2 M T2EHDGA in *n*-dodecane containing 30% *iso*-decanol as the carrier extractant. Transport studies carried out using various mineral acid as feed indicated the transport trend as $HNO_3 \gg HCl \sim HClO_4$ which did not change significantly when the respective sodium salts were used in the feed. Effect of various parameters such as membrane pore size, U concentration in the feed and temperature, etc. on the transport rate was also investigated. Comparisons were made with the transport behaviour of plutonyl ion and TODGA system.

Keywords: Uranium; High level waste; Separation; Uranium; Supported liquid membrane; Diglycolamides; T2EHDGA

1. Introduction

'Actinide partitioning' is emerging as the key step in the 'Partitioning & Transmutation' strategy for the safe management of high level waste (HLW) emanating from the reprocessing of spent nuclear fuel material [1]. Out of the various reagents tested for the extraction of minor actinides from HLW feed solutions, the diglycolamides have shown promise in terms of better extraction as well as stripping efficiency, radiation stability, reusability, etc. [2–4]. Moreover, the diglycolamides are considered 'green' alternatives to reagents such as CMPO (carbamoyl methyl phosphine oxide) [5], TRPO (tri-*n*-alkyl phosphine oxide) [6] and DIDPA (di-*iso*-decyl phosphoric acid) [7]. Though TODGA (*N,N,N',N'*-tetraoctyl diglycolamide), a substituted diglycolamide, has been extensively studied by various research groups, its branched homolog namely *N,N,N',N'*,-tetra(2-ethyl hexyl) diglycolamide (T2EHDGA) has not been well studied. Due to its tendency to form third phase at higher acidities, *iso*-decanol has been used as a phase modifier in T2EHDGA extraction systems. Systematic studies on the role of *iso*-decanol and third phase formation in T2EHDGA system have been carried out [8]. Manohar et al. have reported the solvent extraction studies using T2EHDGA for its possible application in 'actinide partitioning' [9].

The high level waste invariably contains some amounts of U from the losses from the PUREX (Plutonium Uranium Reduction Extraction) cycle. A typical HLW composition is given in Table 1. The advantages of

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Table 1
Composition of a typical high level waste (SHLW) for pressurized heavy water reactor (PHWR); acidity: ~3 M HNO$_3$

Constitutent	Concentration (mg/l)	Constitutent	Concentration (mg/l)
Sr	186.3	Sb	4.7
Rb	74.5	Se	12.3
Zr	771.3	Dy	2.0
Ag	18.6	Sn	15.6
Ba	308.8	Te	102.8
Cd	16.3	Sm	163.8
Ce	532.5	Tb	5.0
Cs	543.8	Gd	165
Fe	500	Eu	22.6
Cr	100	Pr	243.8
Co	127.5	Nd	862.5
Na	3000	La	263.8
Ni	100	Pd	267.5
U	18,325	Ru	463.8
Y	99	–	–
Mo	731.3	–	–

T2EHDGA based 'actinide partitioning' process include no prior separation of U [10] unlike the CMPO-based TRUEX (Trans Uranium Extraction) process where the presence of macro concentration of U could lead to third phase formation [11]. In view of this, the understanding of the extraction behaviour of U(VI)-T2EHDGA extraction system is of great relevance. We have recently reported the solvent extraction and supported liquid membrane (SLM) transport behaviour of U using T2EHDGA as the extractant [12]. The major advantage of SLM based separation methods is extremely low solvent inventory and easy scale up option available [13–15]. Moreover, in SLM system, the extraction and stripping takes place simultaneously making it an efficient separation method. Finally, some of the major disadvantages of solvent extraction system such as third phase formation and phase entrainment can be alleviated in the SLM based separation method [16].

Our earlier SLM study with the U(VI)-T2EHDGA system has indicated that the feed acidity is of great significance. As the extracted complex depends largely on the complexing ability of the anion, the nature of mineral acid may play a significant role on the transport of uranyl ion. Ansari et al. have reported the aggregation behaviour of the diglycolamides is primarily dependent on the mineral acid type as well as concentration [17]. In view of these, the role of the nature of the mineral acid on the transport of UO$_2^{2+}$ was

investigated. Comparison of the transport data of UO$_2^{2+}$ was also made with those obtained with PuO$_2^{2+}$. It was also pertinent to investigate the role of membrane pore size and temperature effect on the transport behaviour of UO$_2^{2+}$. Finally, in view of the presence of large concentration U in the HLW, it was required to understand the effect of U carrier concentration on the transport efficiency.

2. Experimental

2.1. Materials

Tetra(2-ethylhexyl)diglycolamide (T2EHDGA) was procured from Thermax Ltd, Pune, India and was characterized by GC-MS and elemental analyzer and the purity was checked by HPLC (Dionex, Germany) and distribution studies. PTFE membranes used in the present study were procured from Sartorius, Germany. ^{233}U tracer was purified from the associated radiotracer impurities by ion-exchange methods reported earlier [18]. Stock solutions of the radiotracers were made in dilute nitric acid after checking the purity by alpha spectrometry. All the other reagents were of AR grade and were used without further purification. Studies with macro-concentration of U were carried out using nitrate salts of natural U and spiking with the radiotracer, ^{233}U. Assaying of ^{233}U was done by alpha-liquid scintillation counting using a LSC system (Hidex, Finland).

2.2. Transport studies

The SLM studies were carried out using 16 ml glass transport cells with feed/strip solutions stirred at 200 rpm which was optimum as reported by us earlier [19]. The membrane thickness was measured by a Mitutoyo Digital micrometer while porosities of the membranes were measured by an Electroscan 2020 environmental scanning electron microscope (ESEM) [19]. The thickness of the membranes was found to be 55 μm each for 0.2 and 0.45 μm membranes while it was 75 μm each for 1.2 and 5.0 μm membranes. The active area for transport was measured and found to be 7.06 cm^2. The porosities of the membranes with pore sizes 0.2, 0.45, 1.2 and 5.0 μm were 51%, 64%, 74% and 84% while the effective area of the membrane flat sheets was determined as 3.60, 4.52, 5.22 and 5.93 cm^2, respectively. The micro porous membrane filters were soaked in the carrier solution (usually 0.2 M T2EHDGA in *n*-dodecane, also containing 30% *iso*-decanol as the phase modifier) prior to use. The submerged membrane was removed from the solution and wiped carefully with a tissue paper to remove the excess fluid on the support. Usually, the feed compartment contained 3.0 M HNO$_3$ while the receiver phase solution was

pH 2.0 solution. Aliquots were removed from the feed as well as the receiver compartments in regular intervals and assay of [233]U was done as mentioned above. The transport studies were carried out at ambient temperatures (24°C ± 1°C). Temperature variation studies were carried out using a thermostated bath at the specified temperature with an error of 0.5°C. The material balance in these studies was found to be within ± 5%.

2.3. Transport equations

Transport of a metal ion in supported liquid membranes containing organophilic carrier extractant generally involves five distinct steps, viz., (1) diffusion of the metal ion from the bulk feed phase to the feed–membrane interface, (2) extraction of the metal ion by the carrier molecules at the feed–membrane interface, (3) diffusion of the metal–carrier complex inside the membrane phase, (4) stripping of the metal ion at the membrane–receiver interface, and (5) diffusion of the stripped metal ion from the membrane–receiver interface. The transport experiments are carried out under the conditions that the distribution coefficient (symbolized as D) is much larger at the feed–membrane interface as compared to that at the membrane–receiver interface. The mass transfer across the supported liquid membrane can be presented by the permeability coefficient (P) which is obtained from the equation:

$$\ln\left(C_{f,0}/C_{f,t}\right) = QPt/V_f \tag{1}$$

where V_f, $C_{f,0}$ and $C_{f,t}$ represent the volume of the feed and concentrations of feed at starting time and after time 't', respectively. Q is expressed as the product of the geometrical surface area (A) and the porosity (ε). The percent metal ion transport (% T) at a given time is determined by the following equation,

$$\%T = 100\left(C_{r,t}\right)/C_{f,0} \tag{2}$$

where $C_{r,t}$ is the concentration of the metal ion in the receiver phase at a given time 't'. All the experiments were repeated and the accepted data were within the error limits of $\pm 5\%$.

3. Results and discussion

The transport studies were carried out using 0.2 M T2EHDGA in *n*-dodecane containing 30% *iso*-decanol as the phase modifier. The transport profile for a feed and receiver containing 3 M HNO$_3$ and pH 2.0 solutions, respectively, indicated >80% transport after 5 h (Fig. 1). which subsequently resulted in quantitative transport

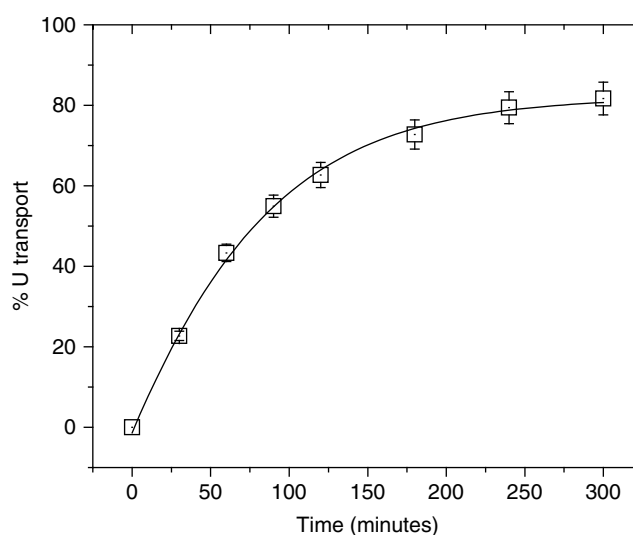Fig. 1. Transport profile of U(VI) with 0.45 μm PTFE flat sheet membranes. Feed: 3 M HNO$_3$; receiver: pH 2.0 solution; carrier: 0.2 M T2EHDGA in *n*-dodecane.

after 11 h The transport studies were repeated several times and reasonable reproducibility was obtained as indicated in the figure. The transport data were comparable with those obtained in the UO$_2^{2+}$ – TODGA transport system using comparable extractant concentration (both cases about 80% transport was noticed after 5 h).

3.1. Nature of feed solution

We have reported earlier that the transport of U increased with increasing feed acidity [11]. This was a consequence of the extraction equilibrium

$$UO_2^{2+} + 2\left(NO_3^-\right) + nT2EHDGA_{(o)} \leftrightarrow$$
$$UO_2\left(NO_3\right)_2 nT2EHDGA_{(o)} \tag{3}$$
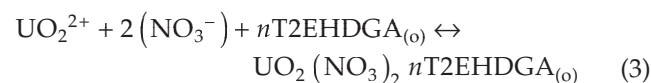
which moves to the right with increasing nitrate ion concentration (directly proportional to nitric acid concentration). As the HLW contains about 3 M HNO$_3$, it was required to investigate the transport of U at 3.0 M HNO$_3$. It was also required to investigate the effect of various mineral acid types such as HCl and HClO$_4$ on the U transport rates. Fig. 2 gives the transport profiles under these feed conditions at 3.0 M acidity. The transport rates followed the trend: HNO >> (HCl ~ HClO$_4$. This was in sharp contrast to the extraction of Am^{3+} reported with these acids which was explained on the basis of the reverse micelle formation ability of these acids as indicated by the metal ion extraction ability which followed the trend HClO$_4$ > HNO$_3$ > HCl. Increasing the acid concentration from 3.0 to 6.0 M indicated spectacular increase in the transport rate with HCl while
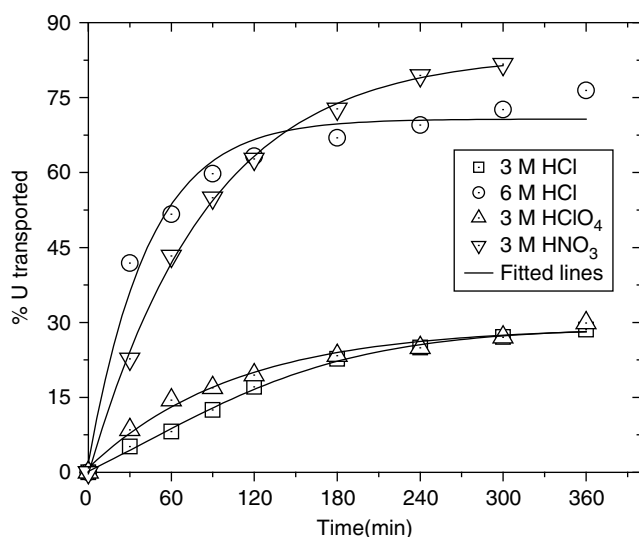
Fig. 2. Transport profile of U(VI) using different mineral acid types in the feed. Receiver: pH 2.0 solution; carrier: 0.2 M T2EHDGA in *n*-dodecane. Pore size: 0.45 µm.
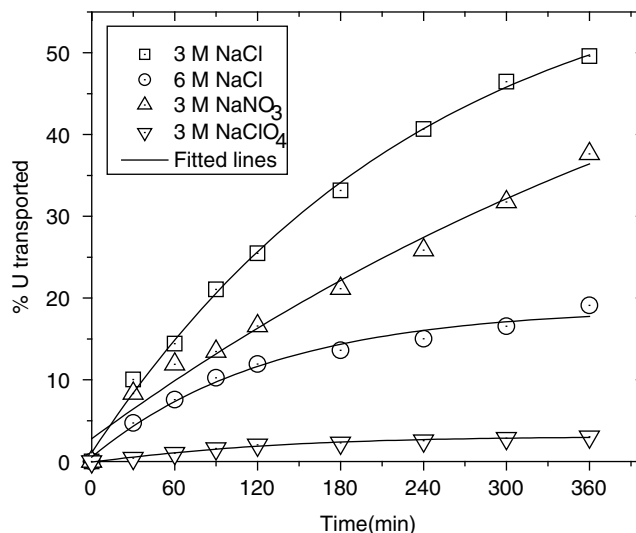

Fig. 3. Transport profile of U(VI) using different sodium salts of different mineral acids as the feed. Receiver: pH 2.0 solution; carrier: 0.2 M T2EHDGA in *n*-dodecane. Pore size: 0.45 µm.

there was no significant change in case of HNO_3. This is, of course, in line with the extraction behaviour of the metal ions observed with HCl and HNO_3 [17]. The permeability coefficients were calculated using Eq. (1) and are listed in Table 2. It is not surprising that the P values followed the same trend as the transport profiles.

We have reported that the extraction of U increased significantly when the feed contained $NaNO_3$ in stead of HNO_3 [12]. In order to discount the effect of co-transport of hydrogen ion which adversely affected the transport of U, several experiments were carried out using the respective sodium salts in the feed. The results are presented in Fig. 3 while the permeability coefficients are listed in Table 2. The permeability data indicated that while the permeability coefficient for uranyl ion decreased significantly when the feed condition was changed from 3 M HNO_3 to 3 M $NaNO_3$ an entirely opposite effect was observed with 3 M HCl and 3 M NaCl. This is in sharp

contrast to the SLM system containing CMPO as the carrier extractant, where significant increase in the metal ion transport was observed with $NaNO_3$ as compared to HNO_3 in the feed [20]. On the other hand, the transport rate became negligible when 3 M $NaClO_4$ was used as the feed. This behaviour is rather intriguing and need a more detailed investigation. The significant increase in the P value, while changing the feed from 3 M $NaNO_3$ to 6 M $NaNO_3$, was ascribed to the increased tendency to form the extractable species with increased nitrate ion. On the other hand, a sharp decline in the transport rate and P value was seen when the feed concentration of NaCl was increased from 3 to 6 M which may be attributed to the formation of non-extractable anionic species such as $UO_2Cl_3^-$ and $UO_2Cl_4^{2-}$.

3.2. Comparison with PuO_2^{2+} ion transport

HLW usually contains small amount of Pu due to the losses from the PUREX cycle. In view of the large number of metal ions present in the HLW and the oxidizing conditions prevailing, it was of interest to compare the transport behaviour of Pu in the +6 oxidation state. The transport profile of PuO_2^{2+} is presented in Fig. 4 along with that for UO_2^{2+} for comparison purpose. The transport rate for the PuO_2^{2+} ion was significantly lower as compared to that for UO_2^{2+} ion. This may be due to either the higher complex formation and hence extraction constants for the uranyl ion compared to the plutonyl ion or due to formation of extracted species with different stoichiometry than that for the uranyl ion [21]. Similar observation was made in the TODGA system

Table 2
Permeability data of UO_2^{2+} under varying feed conditions using 0.2 M T2EHDGA in *n*-dodecane + 30% *iso*-decanol as the carrier solvent

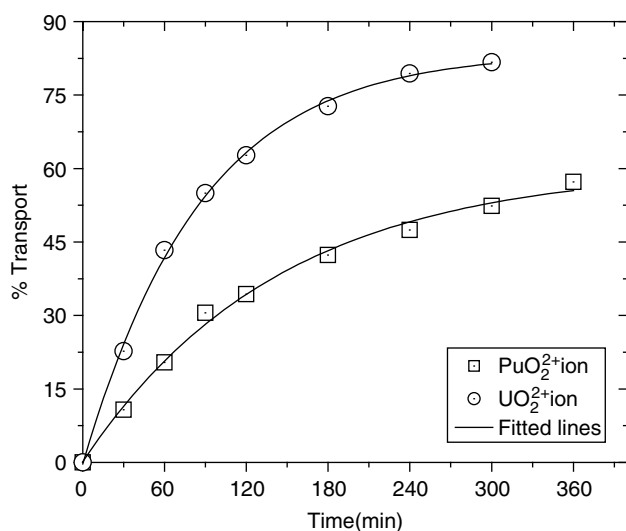| Feed	P (cm/s) × 10⁴	Feed	P (cm/s) × 10⁴
3.0 M HNO_3	3.37 ± 0.29	3.0 M $NaNO_3$	0.67 ± 0.03
3.0 M HCl	0.86 ± 0.12	3.0 M NaCl	1.12 ± 0.03
3.0 M $HClO_4$	0.73 ± 0.04	3.0 M $NaClO_4$	$(4.22 \pm 0.23) \times 10^{-2}$
6.0 M HNO_3	3.86 ± 0.23[a]	6.0 M $NaNO_3$	2.70 ± 0.30[a]
6.0 M HCl	2.27 ± 0.11	6.0 M NaCl	0.28 ± 0.02

[a]Data taken from Ref. [12].

Fig. 4. Transport profile of UO_2^{2+} and PuO_2^{2+} ions with 0.45 µm PTFE flat sheet membranes. Feed: 3M HNO_3; receiver: pH 2.0 solution; carrier: 0.2 M T2EHDGA in *n*-dodecane.

reported earlier [22]. A comparison of the permeability coefficients of UO_2^{2+} and PuO_2^{2+} ions is made in Table 3. TODGA is found to be a far superior extractant as compared to T2EHDGA as the *P* values for both UO_2^{2+} and PuO_2^{2+} are significantly higher even with 0.1 M TODGA as compared that of 0.2 M T2EHDGA as the carrier extractant (Table 3).

3.3. Effect of membrane pore size

Membrane pore size plays an important role in the transport of metal ions across an SLM. This has been shown by various workers for various metal ions and various extractants [23,24]. In diffusion controlled transport, as in the present case, the permeability coefficient of a diffusing species across a polymeric matrix can be given by Eq. [25]:

$$P = \frac{\varepsilon R^2}{\tau 2d_o} \tag{4}$$

Table 3
Permeability data of UO_2^{2+} and PuO_2^{2+} ions using TODGA (0.1 M) as the carrier extractant

Metal ion	Extractant	P (cm/s) × 10⁴	Ref.
UO_2^{2+}	T2EHDGA (0.2 M)	3.37 ± 0.29	Present work
PuO_2^{2+}	T2EHDGA (0.2 M)	1.29 ± 0.10	Present work
UO_2^{2+}	TODGA (0.1 M)	9.76 ± 0.15	[23]
PuO_2^{2+}	TODGA (0.1 M)	2.05 ± 0.06	[23]

where *R* is the membrane pore size, τ is the tortuosity factor, ε is the membrane porosity and d_o is the membrane thickness. This equation clearly indicates that with increasing pore size, the resistance felt by the diffusing complex decreases and as a result, the transport rate increases. But at the same time, from the Laplace's equation.

$$p = (2g/r_p)\cos\theta \tag{5}$$

it is quite clear that the trans-membrane pressure (*p*) required to displace the carrier from the membrane pore is inversely proportional to the pore size (r_p) of the membrane. So when the pore size increases, the trans-membrane pressure also decreases and hence the stability of the membrane due to release of the organic carrier molecule from the pores also decreases. Both these factors play an opposing role on the transport of any metal ion across an SLM of varying pore size. Keeping these factors in consideration, the transport studies of U were carried out from 3 M HNO_3 feed using PTFE membranes of varying pore sizes, viz. 0.2, 0.45, 1.20 and 5.0 µm. As reported earlier, for different metal ions the variation in transport rate is affected differently with membrane pore size variation [24–26]. It is clear from Fig. 5 that for U-T2EHDGA system, transport rate increases up to 0.45 µm and decreased there after similar to the Am-T2EHDGA system reported earlier [27]. This signifies that up to 0.45 µm the increase in transport due to less hindrance of U-T2EHDGA complex and thereafter the loss of carrier molecule due to lower trans-membrane pressure becomes dominant causing in decrease in transport rate. Due to this reason, 0.45 µm PTFE have been used as membrane filters throughout the present study.
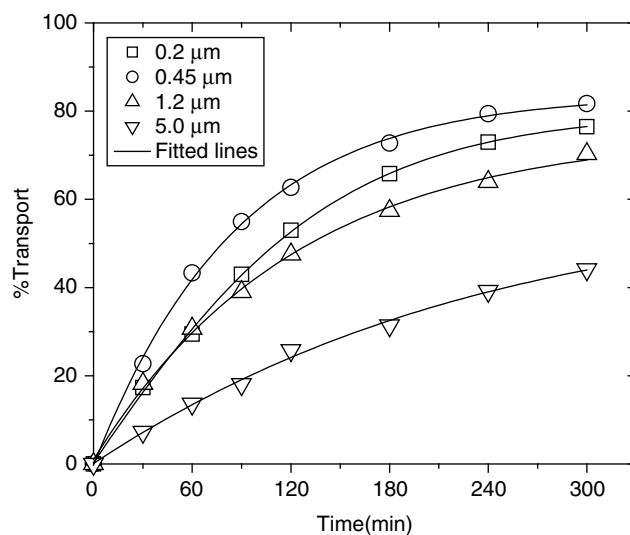

Fig. 5. Transport of U(VI) using 0.2 M T2EHDGA/*n*-dodecane from 3 M HNO_3 with varying membrane pore sizes.

3.4. Thermodynamics study

As mentioned above, the transport process is a combination of extraction, diffusion and stripping steps. As diffusion is the rate determining step, the transport rates are decided by the diffusion coefficients or the permeability coefficients. It was decided to carry out transport studies as a function of temperature and the thermodynamic parameters were determined from the Van't Hoff equation and Gibbs–Helmholtz equation as follows:

$$\Delta G^o = -2.303RT \log P \tag{6}$$

and

$$\Delta G^o = \Delta H^o - T\Delta S^o \tag{7}$$

From Eqs. (6) and (7),

$$\mathrm{Log} P = -(\Delta H^o/2.303R)(1,000/T) + (\Delta S^o/2.303R) \tag{8}$$

The plot of log P vs. 1,000/T is illustrated in Fig. 6 and from the slope and intercept the ΔH^o and ΔS^o values were calculated as 10.24 ± 0.79 kJ/mol and −31.82 ± 2.55 J/K/mol, respectively. Endothermicity of the transport process suggests that with increasing temperature the diffusion is facilitated by the decreasing viscosity of the membrane phase which favours the mobility of the complex and T2EHDGA molecules alike. At the same time, the complexed molecules indicate a decrease in their entropy values resulting in a reasonably high negative entropy term. Similar data treatment has been done by Lee and Yang for the Eu(III) transport studies in SLM system containing PC-88A in kerosene as the carrier extractant [28].
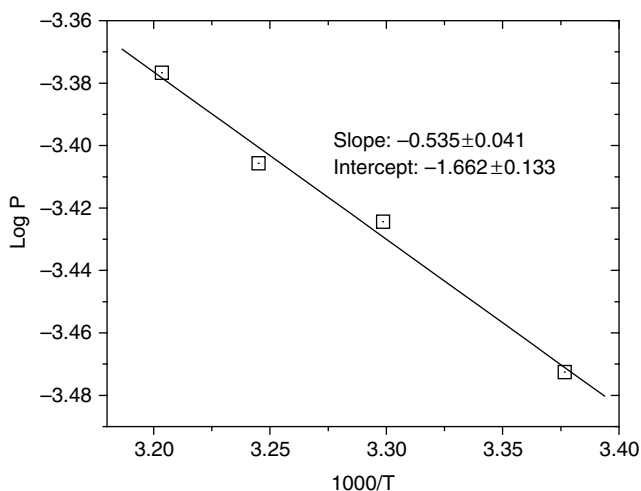
3.5. Effect of U concentration

Concentration of U in the feed plays an important role in the transport rate of U(VI) ion. It is clear from Eq. (3) that the complexation and the transport rate increases with increasing U concentration in the feed. However, due to limitation in the carrier concentration in the membrane phase, the U transport rate may actually appear to decrease though the flux values increase. Transport studies were carried out using U concentration from 0.1 to 20 g/l and the results are shown in Fig. 7. As expected, the transport rates decreased with increasing U concentration. The permeability coefficients also decreased with increasing U concentration (Table 4). On the other hand, analysis of the flux vs. U concentration data indicates an initial increase was observed with a plateau approaching at higher U concentrations (Table 4). When the U concentration was 0.1 g/l the flux value
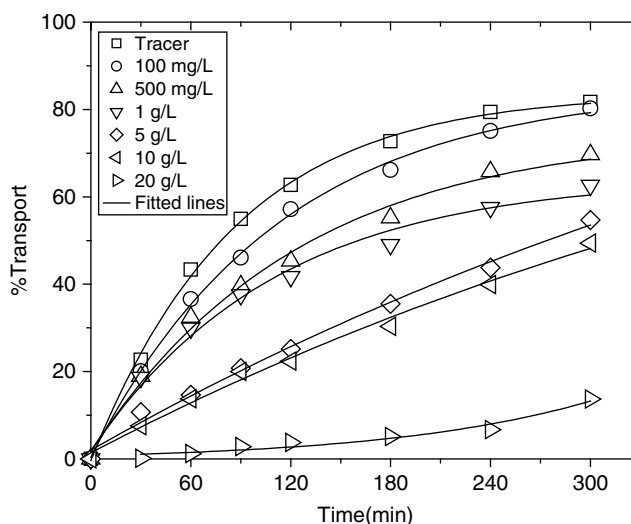
Fig. 7. Transport of U(VI) using 0.2 M TEHDGA against different concentration of U from 3 M HNO$_3$. Feed: 3 M HNO$_3$; receiver: pH 2.0 solution; carrier: 0.2 M T2EHDGA in *n*-dodecane.

Table 4
Permeability coefficients and flux values as a function of the U concentration in the feed

U Concentration (g/l)	P (cm/s) × 10^4	Flux (mol/cm^2 s) × 10^9
0.1	3.27 ± 0.19	0.14 ± 0.01
0.5	2.21 ± 0.17	0.47 ± 0.03
1	1.88 ± 0.08	0.79 ± 0.02
5	1.61 ± 0.12	3.38 ± 0.21
10	1.45 ± 0.06	6.01 ± 0.23
20	0.69 ± 0.05	5.81 ± 0.03

Fig. 6. Temperature dependence of permeability coefficient of U(VI) transport from 3 M HNO$_3$ using 0.2 M TEHDGA as the carrier extractant.

was 1.4×10^{-8} mol/cm^2 s which increased to 58.1×10^{-7} mol/cm^2 s for 20 g/l of U concentration. In view of the low flux obtained with the flat sheet SLM system, it was required to opt for hollow fiber supported liquid membrane system, which can result in spectacular increase in the mass transfer rates [29].

4. Conclusions

From the present studies, the following conclusions can be drawn. Nature of the mineral acid plays an extremely important role in the transport of the metal ion which includes the nature of complexed species in the feed phase. Plutonyl ion transport is much lower as compared to that of the uranyl ion and TODGA is found to be a more efficient extractant as compared to T2EHDGA. The studies with the varying membrane pore size suggested 0.45 μm as the optimum value. The transport rates of uranyl ion obtained in the present system with T2EHDGA as the carrier extractant matched very well with those reported earlier with the TODGA system and hold promise for application in the removal of U from acidic wastes including the PUREX raffinate and high level waste.

Acknowledgements

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