Transfer of uranium(VI) through emulsion liquid membrane with N,N-dibutyloctanamide as a carrier*

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Abstract The transfer behavior of uranium(VI) through an emulsion liquid membrane using N,N-dibutyloctanamide(DBOA) as a mobile carrier has been investigated. The result shows that the liquid membrane process can effectively and rapidly concentrate uranium(VI) from nitric acid medium, and the optimum conditions for the transfer of uranium(VI) were obtained

Keywords Uranium(VI), N,N-dibutyloctanamide(DBOA), Emulsion liquid membrane

1 Introduction

The liquid membranes, both emulsion liquid membranes(ELM) and supported liquid membranes(SLM), are widely used in various fields: the elimination of radioactive metals from the nuclear waste[1]; the concentration of metal ions in hydrometallurgy[2,3] and some medical techniques.[4] The liquid membrane process involves the dispersion of globules of oil and water emulsion into a third phase containing the material to be concentrated. This technique exhibits the advantage of a very high transfer surface and a huge concentrating capacity.

2 Experimental

DBOA was synthesized and purified as described previously[5]; kerosene was purchased from Nanking Chemical Plant and purified by distillation. All other chemicals used were of the A R grade without further purification.

W/O emulsion was prepared by mixing 10.0ml of the oil phase and 10.0 ml of the internal aqueous phase in a flask at 1500±50 r/m. for 15 min. The oil phase was composed of DBOA as a carrier, 4%(w/v) span80 as an emulsifier and kerosene as solvent. An aqueous sulfuric acid was used as the internal aqueous phase for stripping, uranyl nitrate (10^{-3}mol/L) aqueous solution containing nitric acid at required concentration as external phase.

ELM transfer operation was performed by pouring 4.0ml of the W/O emulsion into 20.0ml external phase stirred at 250±20 r/m. Samples were taken from external phase at proper time intervals. Experiments were carried out at room temperature(20±1°C) except marked.

Uranium(VI) ion in external phase was analyzed by the Arsenazo-Ⅲ spectrophotometry.

3 Results and discussion

The extraction of U(VI) by DBOA from nitric acid media has been established in our previous works. The extracting and stripping (by sulfuric acid) process can be expressed as follows:

$$UO_2^{2+} + 2NO_3^- + 2DBOA_{(o)} = UO_2(NO_3)_2 \cdot 2DBOA_{(o)} \quad (1)$$

$$UO_2(NO_3)_2 \cdot 2DBOA_{(o)} + 2H^+ = UO_2^{2+} + 2DBOA \cdot HNO_{3(o)} \quad (2)$$

$$3UO_2^{2+} + 6SO_4^{2-} \longrightarrow UO_2SO_4 + UO_2(SO_4)_2^{2-} + UO_2(SO_4)_3^{4-} \quad (3)$$

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3.1 Transfer of uranium(VI) through ELM

The emulsion liquid membrane used in this work is shown schematically in Ref.[2]. Uranium(VI) is transported from the external phase to the internal phase via kerosene membrane. Movement of the charged species through the organic membrane is accomplished by the presence of the carrier(DBOA). After complexation of uranyl ion with DBOA and nitrate anion at the interface between external phase and membrane, the complex diffuse down with its concentration gradient. At the other interface , the metal ion is released into the internal phase via complex $UO_2(NO_3)_2 \cdot 2DBOA_{(o)}$. At this stage the carrier diffuses back across the liquid membrane. The net result is the transfer of uranium(VI) from external phase to the internal phase across the bulk of organic phase.

3.2 Effect of nitric acid in the external phase

The transfer of U(VI) through ELM containing 0.10 mol/L DBOA was carried out at different concentrations of nitric acid in the external phase. Fig.1 shows the time-dependent fractions (C_t/C_o) of U(VI) in external solution, where C_t and C_o are the concentrations of U(VI) at time t and the initial, respectively.

It can be seen from Fig.1 that it has advantage of transfer for U(VI) to increase the concentration of nitric acid in external solutions since the fraction vs time curves fall from smoothly to suddenly. If C_{HNO_3} >5.0 mol/L the divergence of fraction vs time curves is not evidently. So the proper condition for the transport of U(VI) is that the concentration of nitric acid in external phase should be within the range of 3~5 mol/L.

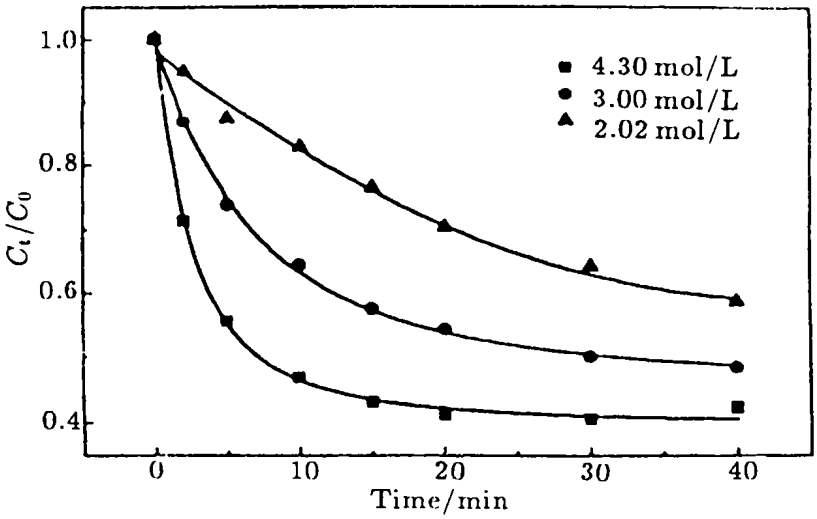

Fig.1 Effect of HNO_3 concentration in external phase on the transfer of uranium (VI)
ELM: 0.1 mol/L DBOA and 4% (W/V) span 80 in kerosens; Internal phase: 3.5 mol/L H_2SO_4; External phase: 0.001 mol/L uranyl nitrate in nitric acid solution; Stirring speed: 250±20 r/m; 293 K

3.3 Effect of DBOA concentration in ELM

The transfer of U(VI) through the ELM containing DBOA with different concentration is presented in Fig.2. The experimental results indicate that the transfer of U(VI) speeds up with increasing the concentration of DBOA in ELM. However, as $C_{DBOA(O)}$ exceeds 0.30 mol/L, the ELM becomes thick and its stability decreases. The proper concentration of DBOA in ELM is about 0.30 mol/L.

3.4 The influence of temperature on the transfer of uranium(VI)

Fig.3 shows the influence of temperature on the transfer of U(VI). The lower the temperature is the faster the uranium(VI) transfer. Moreover, the stability of ELM decreases greatly if the temperature is over 25°C. Therefore, it is necessary to control the temperature under 25°C.

It was concluded that the transfer of U(VI) was accomplished through a ELM containing

DBOA as the mobile carrier from nitric acid solution to the sulfuric acid solution. Uranium(VI) was concentrated effectively in the internal sulfuric acid solutions under the proper conditions. In this way, radioactive waste can be treated with this ELM to eliminate the radioactive contamination.

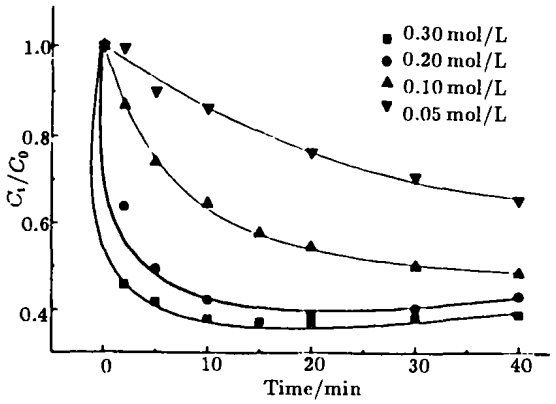

Fig.2 Transfer of U(VI) with different concentrations of DBOA in ELM

ELM: DBOA and 4%(W/V) span 80 in kerosene; Internal phase: 3.5 mol/L H_2SO_4; External phase: 0.001 mol/L uranyl nitrate and 3.0 mol/L HNO_3; Stirring speed: 250±20 r/m; 293 K

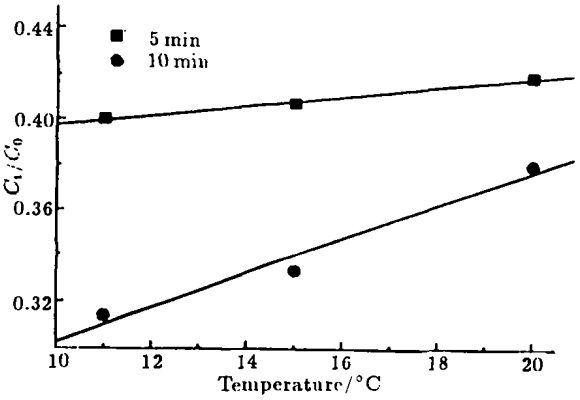

Fig.3 Effect of temperature on the transfer of uranium(VI)

ELM: 0.30 mol/L DBOA and 4% (W/V) span 80 in kerosene; Internal phase: 3.5 mol/L H_2SO_4; External phase: 0.001 mol/L uranyl nitrate in 1.0 mol/L HNO_3; Stirring speed: 250±20 r/m

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