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Short communication

Innovative approach to enhance uranium ion flux by consecutive extraction via hollow fiber supported liquid membrane

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ABSTRACT

The enhancement of liquid membrane performance in this study, by consecutive extraction using a synergistic extractant, is the first to demonstrate that the synergistic extractant, normally employed in the solvent extraction process, can also be extended to extraction applications via a liquid membrane. The study focuses on increasing the flux of metal ions using a consecutive extraction reaction which occurs inside the liquid membrane phase. Uranium ions in trisodium phosphate solution, a by-product of monazite processing, are the specie of interest in this experiment. The feed solution is trisodium phosphate containing uranium ions, whereas the stripping solution is nitric acid. The extractant is Aliquat336 mixed with TBP in kerosene. The results demonstrate that when Aliquat336 is mixed with TBP, the flux of uranium ions is synergistically increased because of a consecutive extracting reaction in the liquid membrane phase. The highest flux is 3600 mg m^{-2} s^{-1} with 0.06 M TBP and 0.6 M Aliquat336. Synergistic extraction is also obtained because the flux is much higher than that obtained using a single extractant.

1. Introduction

Monazite ore is a natural phosphate of the rare earths, mainly the cerium and lanthanum metals, and usually contains some uranium and thorium. The hydroxide compounds of uranium, thorium and other rare earths, and the by-product trisodium phosphate, were separated by filtration. Trisodium phosphate was sent to the crystallization unit, whereas the hydroxide compounds were then dissolved with HCl to remove the gangue and undigested components. Uranium and thorium in chloride solution were selectively precipitated with NaOH solution, and the remaining mixed rare earths were all precipitated into a mixed hydroxide cake. Cerium in the mixed hydroxide cake was later separated by leaching with HNO_3 [1]. Unfortunately, uranium has high solubility in water; therefore filtration is not a suitable technique to separate uranium from trisodium phosphate solution. The contaminated uranium is traditionally separated from trisodium phosphate solution by either ion exchange or chemical precipitation. In general, uranium is a radioactive element which is useful at the front and back ends of the nuclear fuel cycle. However uranium is known to cause serious environmental damage and acute toxicological effects in mammals, and its compounds are potential carcinogens [1].

Efficient extraction and purification of metal ions from an aqueous solution using a liquid membrane have gone through many years of study. This liquid membrane combines solvent extraction and stripping processes in a single step and does not require phase-mixing for good mass transfer. Instead, the membrane will allow two phases to remain in contact with each other through a rigid, thin and porous membrane, stabilizing the interface between the two phases.

This liquid membrane is well recognized as an excellent process treatment with dilute solutions for recovery of many kinds of metal: for example, heavy metals [2,3], precious metals [4], rare earth metals [5,6], toxic metals [7,8] and radioactive metals [9]. Generally it offers greater benefits over other processes, especially for separation and recovery from industrial wastes, pollutant removal and bio-separation [10,11]. Hollow fibers are one of the most popular materials built to support a liquid membrane. They are widely used in industry because of several beneficial features: for example, less extractant required compared to conventional solvent extraction; larger surface area per volume; lower energy; higher selectivity [5].

A hollow-fiber module is composed of an outer shell, which is a single nonporous material through which the materials inside cannot be transported. Inside that shell are multiple thin fibers

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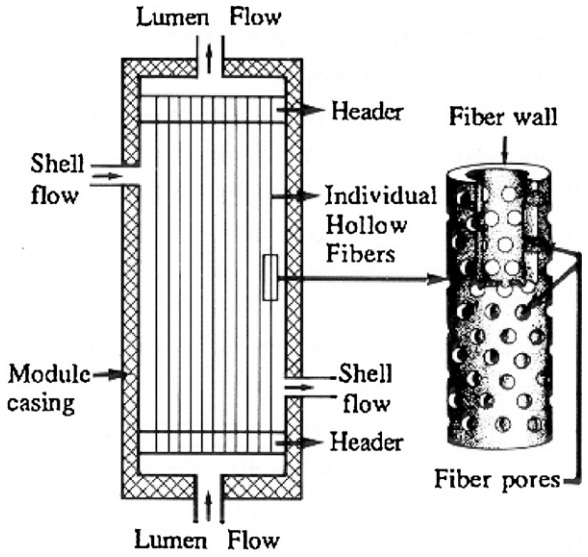

Fig. 1. Hollow fiber module which is used to support liquid membrane and a close up of the cross section of a single hollow fiber.

running the length of the shell, all in nice, neat rows. What occurs is that the source phase is piped through the system from top to bottom, and the pores in the fibers themselves are filled with the organic phase. The carriers in that phase then transport the source across to the stripping phase; then the stripping phase is forced out through the sides of the shell. Fig. 1 represents a hollow-fiber module which is used to support a liquid membrane; also shown is a close-up of the cross section of a single hollow fiber.

A liquid membrane as an extractant carrier was immobilized in the pores of a hydrophobic microporous supporter; this membrane binds one of the components very selectively from the feed solution, and separates the aqueous feed solution from the strip solution. The species are accumulated in the strip at concentrations generally greater than those in the feed solution. The permeation of the species is due to a chemical potential gradient (the driving force of the process) existing between two opposite sides of the liquid membrane [1,12,13].

However, some disadvantages of a liquid membrane are also apparent. An increase in extractant concentration can yield higher extractability only up to a point; beyond that, little or no benefit in extractability will be gained, as can be seen by the viscosity of the liquid membrane [14,15]. This work aimed at overcoming the disadvantages of a liquid membrane. Consecutive extraction was implemented via hollow-fiber supported liquid membrane (HFSLM). Aliquit336 and TBP were selected as extractants to separate uranium (a radioactive element from monazite processing) ions from trisodium phosphate.

A conventional liquid membrane reaction, and the transportation of metal ions (uranium ions) with Aliquat336 as an extractant, are described in Eqs. (1) and (2) [16,17]:

$$[UO_2(CO_3)_3]^{4-} + 4R_4NNO_3 \leftrightarrow (R_4N)_4UO_2(CO_3)_3 + 4NO_3^- \quad (1)$$

$$(R_4N)_4UO_2(CO_3)_3 + 4NO_3^- \leftrightarrow [UO_2(CO_3)_3]^{4-} + 4R_4NNO_3 \quad (2)$$

The uranium ions react with Aliquat336 to produce a complexing agent at the interface between the feed and the liquid membrane. The complex formed then diffuses through the membrane phase to the interface membrane/strip-phase where the decomplexation of the metal ions occurs. The scheme of metal transport is shown in Fig. 2a [18].

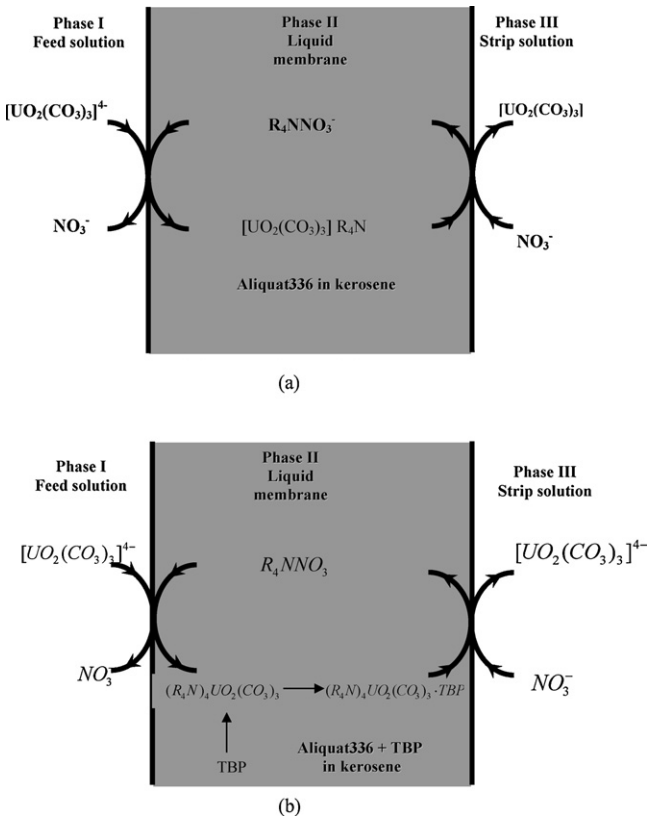

Fig. 2. (a) Schematic diagram of conventional transport mechanism. (b) Schematic diagram of novel transport mechanism.

Unfortunately, the extraction from a conventional mechanism appears unsatisfactorily low, due to the disadvantages described earlier. To overcome this problem, TBP was added to the liquid membrane phase and mixed with Aliquat336. The mechanism begins with uranium ions reacting with Aliquat336, thus producing a complexing agent in the membrane phase similar to conventional extraction in Eq. (3). Subsequently, in the liquid membrane phase, TBP reacts with the complexing agent, producing a TBP complex according to Eq. (4) and diffusing through the liquid membrane to the stripping phase. Finally, in the stripping process in Eq. (5), nitrate ions react with the TBP complexing agent, producing uranium ions in the stripping solution. The mechanism presents with two extraction reactions and is known to be a novel transport mechanism, simply illustrated in Fig. 2b.

$$[UO_2(CO_3)_3]^{4-} + 4R_4NNO_3 \leftrightarrow (R_4N)_4UO_2(CO_3)_3 + 4NO_3^- \quad (3)$$

$$(R_4N)_4UO_2(CO_3)_3 + TBP \leftrightarrow (R_4N)_4UO_2(CO_3)_3 \cdot TBP \quad (4)$$

$$(R_4N)_4UO_2(CO_3)_3 \cdot TBP + 4NO_3 \leftrightarrow [UO_2(CO_3)_3]^{4-} + TBP + 4R_4NNPO_3 \quad (5)$$

As such, study of synergistic extraction is a challenge work to overcome the liquid membrane disadvantages.

2. Experimental

2.1. Feed and chemicals

Trisodium phosphate from monazite processing was obtained from the Rare Earth Research and Development Center, Office of Atoms for Peace, Thailand. The feed concentration was 45 ppm uranium ions in trisodium phosphate solution without further