

# Enhanced Separation of Yttrium(III) from Iron(III) in the Presence of Complexing Agents through Supported Liquid Membrane

A. Tsuyoshi, T. Abiko, H. Hoshi and K. Akiba

*Institute for Advanced Materials Processing, Tohoku University,  
Katahira-2, Aoba-ku, Sendai, 980-8577*

The selective transport of yttrium(III) in the presence of iron(III) through a supported liquid membrane (SLM) has been performed by employing di(2-ethylhexyl)phosphoric acid (DEHPA) as a mobile carrier. Yttrium was preferentially transported across the SLM from the feed solution into the product solution of 1 M H<sub>2</sub>SO<sub>4</sub>. The transport of iron(III) was suppressed in the presence of complexing agents in the feed solution, and the separation of yttrium(III) from iron(III) was sufficiently enhanced by the addition of phosphoric acid or EDTA.

## § 1. Introduction

The separation of rare earth (RE) elements from iron(III) is an important subject in material processing, and solvent extraction with acidic organophosphorus compounds has been widely used for the extraction separation of RE elements [1,2]. The preferential extraction of yttrium(III) with DEHPA in the presence of iron(III) was previously performed based on the fast kinetics for yttrium extraction[3]. The extraction system is applied to the transport of metal species across a liquid membrane employing an extractant as a mobile carrier. The previous studies have demonstrated the SLM transport system, and the selective transport of yttrium(III) in the presence of iron(III) was accomplished by employing a mobile carrier of acidic organophosphorus compounds [4,5].

The present study has been performed for the improvement in the separation of yttrium(III) from iron(III) through an SLM with the aid of complexing agents for the suppression of the transport of iron(III).

## § 2. Experimental

### 2.1 Materials

Di(2-ethylhexyl)phosphoric acid (DEHPA) (Daihachi Chem. Ind. Co., Ltd.) was diluted with kerosene. A radioisotope, <sup>88</sup>Y, was produced from the <sup>89</sup>Y( $\gamma$ , n)<sup>88</sup>Y reaction by the bremsstrahlung from a linear electron accelerator of Tohoku University, and <sup>59</sup>Fe (DuPont/NEN Research Products) was obtained as a hydrochloric acid solution. A PTFE membrane (Fluoropore FP045, Sumitomo Electric Ind.) used as an inert support has an average pore size of 0.45  $\mu$ m, a porosity of 74% and a thickness of 80  $\mu$ m.

### 2.2 Liquid membrane transport

An equipment for membrane transport comprised of an inner and outer compartments separated

by a supported liquid membrane (SLM) of  $26 \text{ cm}^2$  impregnated with  $0.02 \text{ M}$  ( $\text{M} = \text{mol dm}^{-3}$ )  $(\text{DEHPA})_2$  (as a dimer) in kerosene. A feed solution ( $100 \text{ cm}^3$ ) containing  $10^{-4} \text{ M}$  each yttrium(III) and iron(III) respectively labeled with  $^{88}\text{Y}$  and  $^{59}\text{Fe}$  was put in the outer compartment. A stripping solution ( $10 \text{ cm}^3$ ) of  $1 \text{ M}$  sulfuric acid was placed in the inner compartment. These transport compartments were horizontally shaken at  $100 \text{ strokes/min}$  at  $25^\circ\text{C}$ . Samples were withdrawn from the feed and product solutions, and their radioactivities were measured with a well-type NaI (Tl) scintillation detector.

### § 3. Results and Discussion

#### 3.1 Liquid-membrane transport

The transport of yttrium(III) and iron(III) was carried out through the liquid membrane impregnated with  $0.02 \text{ M}$   $(\text{DEHPA})_2$  in kerosene. Figure 1 shows the time-dependent fractions of metal species in the feed solution of  $0.5 \text{ M}$   $(\text{H,Na})\text{Cl}_2\text{CHCOO}$  (pH 2.0) and the stripping product solution of  $1 \text{ M}$   $\text{H}_2\text{SO}_4$ . The fraction of yttrium(III) in the feed solution decreases with time and that in the product solution increases and approaches to the fraction of 1 after 300 min. The transport profiles for both sides are closely symmetrical, indicating that metal species retained in SLM are negligibly small. As for iron(III), the transport is rather low, and the fraction of iron(III) transported after 300 min was about 0.7. For the sufficient separation of yttrium(III) from iron(III), it is important to suppress the iron(III) transport.

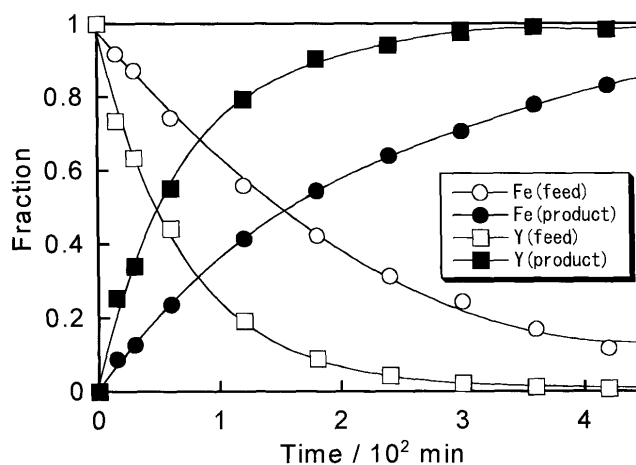


Fig.1. Transport of Y(III) and Fe(III) through SLM. SLM :  $0.02 \text{ M}$   $(\text{DEHPA})_2$  in kerosene. Feed :  $0.5 \text{ M}$   $(\text{H,Na}) \text{Cl}_2\text{CHCOO}$ , pH 2.0,  $10^{-4} \text{ M}$  Y(III) and Fe(III) ; product :  $1 \text{ M}$   $\text{H}_2\text{SO}_4$ ;  $25^\circ\text{C}$ .

#### 3.2 Effect of phosphoric acid in the feed solution

Conditions on the feed side were examined by the addition of complexing agents in order to enhance the mutual separation of yttrium(III) from iron(III). Figure 2 shows the metal transport across the SLM in the presence of  $0.5 \text{ M}$  phosphate buffer (pH 2.0). The transport profiles for yttrium(III) are similar to those in the absence of phosphate, whereas the transport of iron(III) is extremely depressed due to the formation of phosphate complexes.

Figure 3 shows plots of  $\ln ([M]_{f,t}/[M]_{f,0})$  against time  $t$ , where  $[M]_{f,0}$  and  $[M]_{f,t}$  are the concentrations of metal ion in the feed solution at time zero and a given time,  $t$ (s), respectively. As Fig.3 shows, a linear

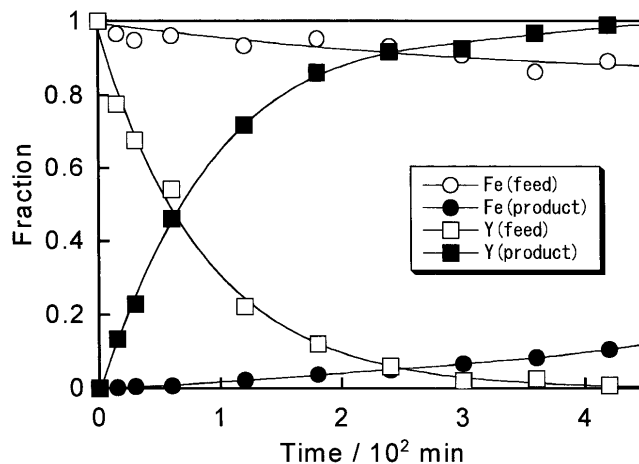


Fig.2. Transport of Y(III) and Fe(III) through SLM. SLM: 0.02 M (DEHPA)<sub>2</sub> in kerosene. Feed: 0.5 M (H,Na) H<sub>2</sub>PO<sub>4</sub>, pH 2.0; product: 1 M H<sub>2</sub>SO<sub>4</sub>.

correlation was obtained as :

$$\ln ([M]_{f,t} / [M]_{f,0}) = -k_{obs} t \quad (1)$$

The apparent rate constant for metal transport,  $k_{obs}$ , can be evaluated from the slope of the linear line. Variations in  $k_{obs}$  values against phosphate buffer concentration are shown in Fig.4. The values for yttrium(III) are large and only slightly decrease with increasing concentration of phosphate buffer, while those for iron(III) more sharply decrease.

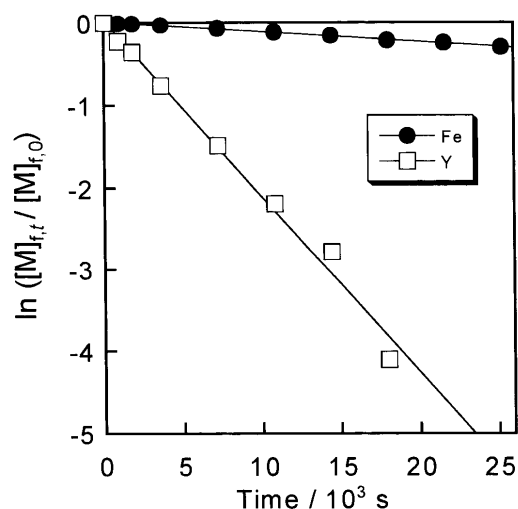


Fig.3. Decreases in the fractions of metal species on the feed side. SLM: 0.02 M (DEHPA)<sub>2</sub> in kerosene. Feed: 0.25 M (H, Na)H<sub>2</sub>PO<sub>4</sub>, pH 2.0; product: 1 M H<sub>2</sub>SO<sub>4</sub>.

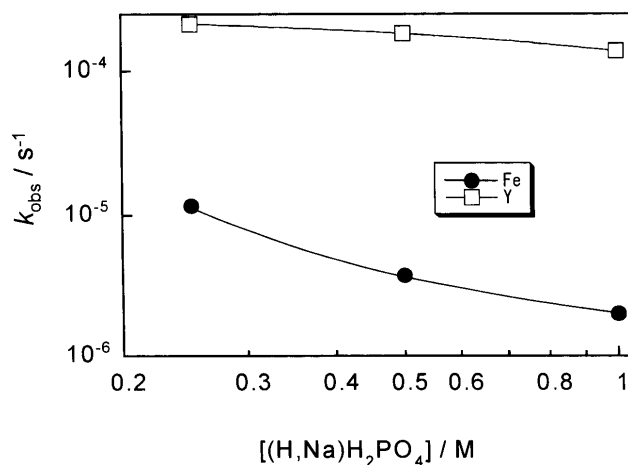


Fig.4. Effect of the concentration of phosphate buffer in the feed solution on  $k_{obs}$ . The other conditions are the same as those in Fig.2.

The separation factor in liquid membrane transport at time,  $\alpha_T$ , can be expressed by the ratio of the  $k_{obs,1}$  and  $k_{obs,2}$  values for desired metals,  $M_1$  and  $M_2$ , respectively,

$$\alpha_T = k_{obs,2} / k_{obs,1} \quad (2)$$

The separation factor for yttrium(III) over iron(III) rises with increasing concentration of phosphate buffer, and reached to 70 in the presence of 1 M (H,Na)H<sub>2</sub>PO<sub>4</sub>.

Transport experiment was carried out to examine extraction conditions by varying pH at constant

0.5 M  $(\text{H,Na})\text{H}_2\text{PO}_4$  in the feed solution. Figure 5 shows plots of  $k_{\text{obs}}$  against pH in the feed solution. The values for yttrium(III) remain practically constant irrespective of pH, while those for iron(III) increase with increase in pH. The separation factors are reduced from above  $10^3$  around pH 1.5 to 20 around pH 2.5.

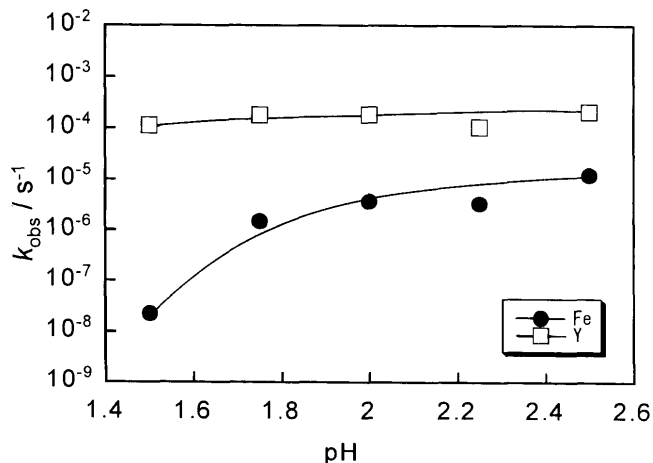


Fig.5. Effect of pH of the feed solution on  $k_{\text{obs}}$ . The other conditions are the same as those in Fig.2.

### 3.3 Effect of EDTA in the feed solution

Transport of iron(III) species across the membrane would be probably depressed in the presence of a complexing agent, ethylenediamine-N,N,N',N'- tetraacetic acid (EDTA). The metal transport was performed in the presence of  $5 \times 10^{-4}$  M EDTA. As Fig.6 shows, the transport of yttrium is similar to that in the absence of EDTA, while almost no transport of iron(III) was observed due to the complex formation of iron(III) with EDTA.

The values of  $k_{\text{obs}}$  for both yttrium(III) and iron(III) are illustrated as a function of pH in Fig.7. The  $k_{\text{obs}}$  value for yttrium(III) slightly decreases with increasing pH, while that for iron(III) remains very low level of  $10^{-7}$ , in which transport of iron(III) is negligibly small. The  $\alpha_T$  values exceed  $10^3$  at pH 1-

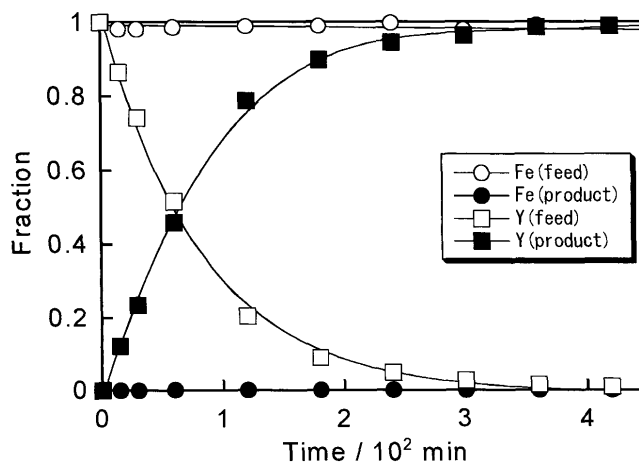


Fig.6. Transport of Y(III) and Fe(III) through SLM. SLM : 0.02 M  $(\text{DEHPA})_2$  in kerosene. Feed : 0.5 M  $(\text{H,Na})\text{Cl}_2\text{CHCOO}$ , pH 1.5,  $5 \times 10^{-4}$  M EDTA ; product : 1 M  $\text{H}_2\text{SO}_4$ .

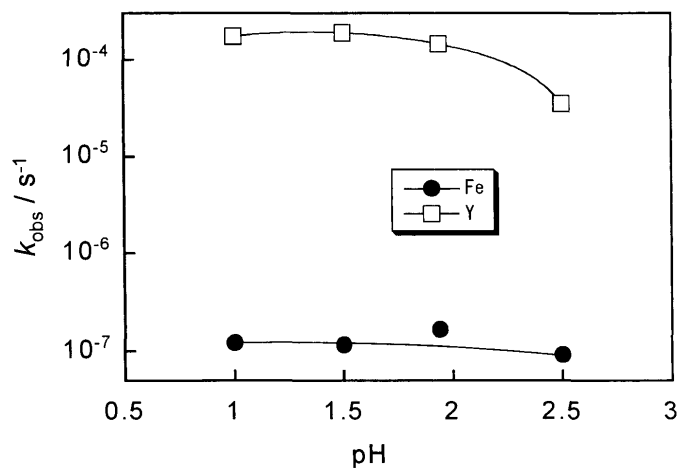


Fig.7. Effect of EDTA concentration on  $k_{\text{obs}}$ . The other conditions are the same as those in Fig. 6.

1.5, and decrease with rising pH above 1.5. The transport of iron(III) was thoroughly suppressed with EDTA, and quantitative separation of yttrium was accomplished. The separation of yttrium from iron(III) was thus enhanced by controlling pH in the presence of complexing agents.

### Acknowledgement

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