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Carrier-Mediated Transport of Molybdenum(VI) through Supported Liquid Membranes

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ABSTRACT

The liquid membrane transport of Mo(VI) from an acidic solution into an alkaline stripping solution was carried out by using a mobile carrier, 5,8-diethyl-7-hydroxydodecan-6-one oxime (LIX 63). Further transport of MoO_4^{2-} from the alkaline solution was performed by using another carrier, trioctylmethylammonium chloride (TOMAC). Molybdenum(VI) was effectively transported through double membranes composed of LIX 63/dilute NaOH solution/TOMAC from a feed solution (10^{-3} M HNO_3) into a product solution (1 M NaOH). Molybdenum(VI) was concentrated with high recovery into a small volume of product solution. The separation of Mo(VI) from several ions was performed by means of the double membranes.

INTRODUCTION

Molybdenum has extensive industrial applications and is regarded as one of the vital strategic metals in modern technology. Alkali or acidic leaching of molybdenum resources is usually accompanied by a variety of elements (1). Metal separation by solvent extraction has been established as a convenient technique and has been proposed for molybdenum refining (2–4). In our preceding paper, LIX 63 (5,8-diethyl-7-hydroxydodecan-6-one oxime) was verified to be an effective reagent for the extraction separation of Mo(VI) from impurities (5).

On the other hand, supported liquid membranes (SLM) have been developed as a promising technique for the separation and concentration of metal ions. LIX 63 has been successfully employed as a relevant carrier in SLM transport (6) and can be used for Mo(VI) transport.

A combination of two SLMs containing selective carriers of different chemical properties has been proposed as an advanced procedure for metal separation (7–10). The stripping solution of the first membrane is chosen to promote the extraction of a metal ion into the second membrane. Molybdenum(VI) can be extracted with LIX 63 as MoO_2^{2+} from acidic solutions and stripped into alkaline solutions as MoO_4^{2-}, which is extractable with a liquid anion exchanger such as trioctylmethylammonium chloride (TOMAC) (11, 12). A common dilute alkaline solution for stripping from LIX 63 and for extraction with TOMAC would be a convenient practice for the transport of Mo(VI) through double membranes. This appears to be advantageous over transport across a single membrane in regard to recovery and separation of Mo(VI), as the double membrane system improves the selectivity.

In the present study the transport behavior of Mo(VI) across a membrane impregnated with LIX 63 in kerosene was investigated. This system was combined with an alternate membrane containing TOMAC–10% 1-octanol in kerosene, and the selective transport of Mo(VI) was investigated through the double membranes.

EXPERIMENTAL

Materials

5,8-Diethyl-7-hydroxydodecan-6-one oxime (LIX 63; Henkel Corporation) was diluted with kerosene, and trioctylmethylammonium chloride (TOMAC, Koei Chemical Ind. Co.) in kerosene was modified with 10% (v/v) 1-octanol. The addition of 1-octanol served to enhance the solubilities of TOMAC and its complex by depressing emulsion formation, and hence stabilize the liquid membrane. An aqueous solution of molybdenum(VI) was prepared by dissolving $Na_2MoO_4 \cdot 2H_2O$ (Wako Pure Chemical Ind.) in water. The pH value was controlled with 0.01 M $(H,Na)CH_3COO$. A polytetrafluoroethylene (PTFE) membrane (Fluoropore FP-045, Sumitomo Electric Ind.) of 80 μm thickness, 74% porosity, and 0.45 μm pore size was used as an inert support. Other reagents used were of guaranteed grade.

Extraction and Analytical Procedures

An aqueous solution containing 5×10^{-4} M ($M = mol \cdot dm^{-3}$) Mo(VI) was shaken at 245 strokes per min (SPM) with an equal volume of a kerosene solution of LIX 63 at 25°C. The two phases were separated by centrifugation at 3000 rpm for 1 minute. The concentration of Mo(VI) in the aqueous phase was determined by inductively coupled plasma-atomic emission spectrometry (ICP-AES) (model SEIKO SPS 1200 A) at 201.030 nm. The analysis was performed using standard and blank solutions having background compositions similar to the sample solution to be tested. The metal concentration in the organic phase was calculated from a mass balance before and after extraction.

Membrane Permeation

An apparatus for single SLM transport was comprised of inner and outer polypropylene compartments (6). The PTFE membrane (26 cm^2) was attached to the bottom of the inner compartment and soaked with organic solution containing the carrier, and the excess solution was removed by wiping with a filter paper. A feed solution (100 cm^3) containing the metal ion (5×10^{-4} M) was placed in the outer compartment, and an equal volume of product solution was placed in the inner compartment. The apparatus was shaken at 120 SPM at 25°C, and aliquots of the feed and product solutions were periodically withdrawn for metal analysis by ICP-AES.

For transport through double membranes, an apparatus was composed of three compartments corresponding to the feed, intermediate, and product solutions. The first membrane (SLM1, 45 cm^2) and the second membrane (SLM2, 26 cm^2) were attached to the bottoms of the intermediate and product compartments and impregnated with the LIX 63 and TOMAC solutions, respectively. The feed, intermediate, and product solutions were poured into their corresponding compartments.

The support PTFE membrane was used several times after washing with acetone and finally with ethanol to remove the organic solution, and drying in air. The repeated use of the membrane support was found to have little effect on the transport behavior of Mo(VI) species.

RESULTS AND DISCUSSION

Solvent Extraction of Molybdenum with LIX 63

Molybdenum(VI) forms various species depending on conditions in the aqueous solutions. A mono cationic form, MoO_2^{2+}, is present in high acidity (13), and its complex species with some anions exist at higher acid concentrations. A series of polymolybdate species is predominant in weakly acidic solutions; however, Mo(VI) exists as a mono anionic form, MoO_4^{2-}, in alkaline solutions (14).

Molybdenum(VI) is extractable with LIX 63 in an acidic region according to the following equilibrium (5):

$$MoO_2^{2+} + 2HL_{(o)} \rightleftharpoons MoO_2L_{2(o)} + 2H^+ \qquad (1)$$

where HL represents LIX 63 and the subscript (o) refers to the organic phase.

The extraction equilibrium and the rate of extraction were investigated at various aqueous conditions prior to investigation of Mo(VI) transport across the SLM. Figure 1 shows the extraction of Mo(VI) with 10^{-2} M LIX 63 in kerosene from different acidic media: HNO_3, $HClO_4$, H_2SO_4, and HCl. Quantitative extraction of Mo(VI) was accomplished from acids below 5 M. The extraction of Mo(VI) decreased with a further increase in acid concentration. A sharp drop in extraction from HCl solution is due to the formation of unextractable anionic chloro complexes (15). The complex formation of Mo(VI) with anions such as NO_3^-, ClO_4^-, and SO_4^{2-} is not clearly identified. In subsequent experiments nitric acid was employed due to high extraction in a wide concentration region.

The effect of contact time on the extraction of Mo(VI) was investigated at different HNO_3 concentrations in the range from 10^{-3} to 3 M. As Fig. 2 shows, the extraction was fairly fast at lower HNO_3 concentrations and slowed down with an increase in concentration. Almost quantitative

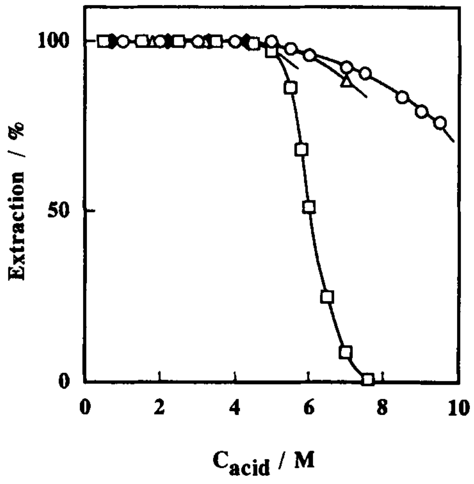

FIG. 1 Extraction of Mo(VI) with 0.01 M LIX 63 in kerosene at 3 hours. (○) HNO_3, (△) $HClO_4$, (◆) H_2SO_4, (□) HCl.

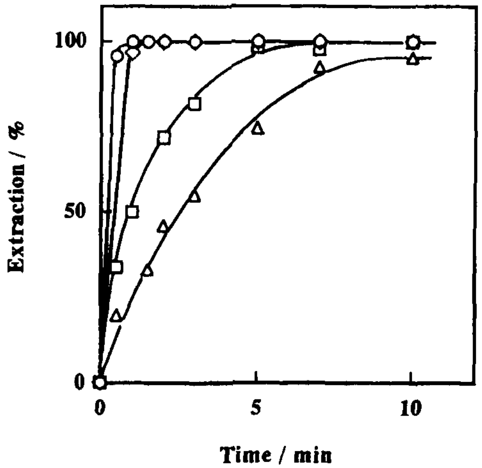

FIG. 2 Effect of contact time on the extraction of Mo(VI) with 0.01 M LIX 63 in kerosene from different concentrations of HNO_3. C_{HNO_3}: (○) 10^{-3} M, (◇) 0.1 M, (□) 1 M, (△) 3 M.

extraction was accomplished in a short time at a low HNO_3 concentration (below 0.1 M). As is evident from Eq. (1), the extraction of Mo(VI) is governed by the hydrogen ion concentration.

The extraction rate of Mo(VI) was further examined by varying the initial pH value from 3 to 6, and the results obtained are presented in Fig. 3. Fast extraction was observed at pH 3, and the extraction rate decreased with increasing pH. The cationic Mo(VI) species was readily extracted from lower pH solutions, forming the MoO_2L_2 complex. The existence of polymetallic species at higher pH reduced the rate of extraction (16). Extraction was rather depressed at higher pH levels due to the predominance of the unextractable MoO_4^{2-} form. The deficiency of H^+ restricts the conversion of MoO_4^{2-} to the MoO_2^{2+} form. Thus, a slightly acidic solution is suitable for the extraction of Mo(VI).

Application to Transport across LIX 63-SLM

LIX 63 is an effective extractant for Mo(VI), and it would be useful as a mobile carrier in SLM transport. A membrane impregnated with LIX 63 in kerosene was contacted with a feed solution initially containing the metal ion and a stripping product solution. The driving force for the transport of a metal ion across an SLM is usually provided by the concentration gradient of a chemical species between the two sides of the membrane.

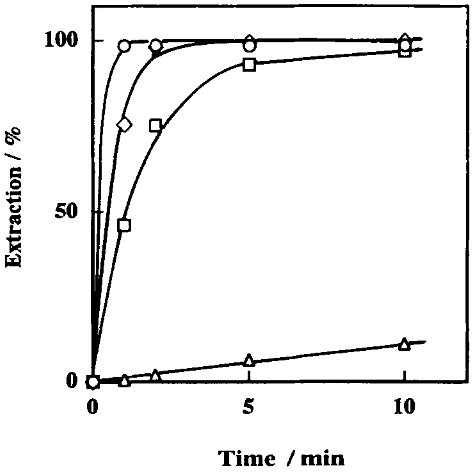

FIG. 3 Effect of contact time on the extraction of Mo(VI) with 0.01 M LIX 63 in kerosene at different pH values. pH: (○) 3 (◇) 4.5, (□) 5, (△) 6.

Molybdenum(VI) is considerably extractable from acidic solution with LIX 63, but it is poorly extractable from alkaline medium; hence, hydrogen ion concentrations in the feed and product sides are important factors affecting the transport of Mo(VI).

The transport of Mo(VI) from 0.1 M HNO_3 into 0.1 M NaOH was investigated through an SLM containing LIX 63 of different concentrations. As shown in Fig. 4, the Mo(VI) fraction in the feed side decreased with shaking time due to extraction into the membrane phase. Only a small fraction of Mo(VI) was extracted with 10^{-3} M LIX 63, whereas almost complete extraction was accomplished after 5 hours with 1 M LIX 63. The fraction of Mo(VI) in the product side increased with a time lag, compared with the decreasing fraction in the feed side, because of insufficient stripping into the product side; in this case a considerable portion of metal ion is retained in the SLM. The fraction of Mo(VI) moved into the product side varies in a similar manner in the range of LIX 63 concentration from 0.01 to 0.1 M. At the high LIX 63 concentration of 1 M, the stripping was rather depressed due to the high stability of the metal complex in SLM.

Figure 5 shows the transport of Mo(VI) from different HNO_3 concentrations. The rate of transport markedly decreased with increasing acid concentration. Complete extraction from the feed side of 10^{-3} M HNO_3 was

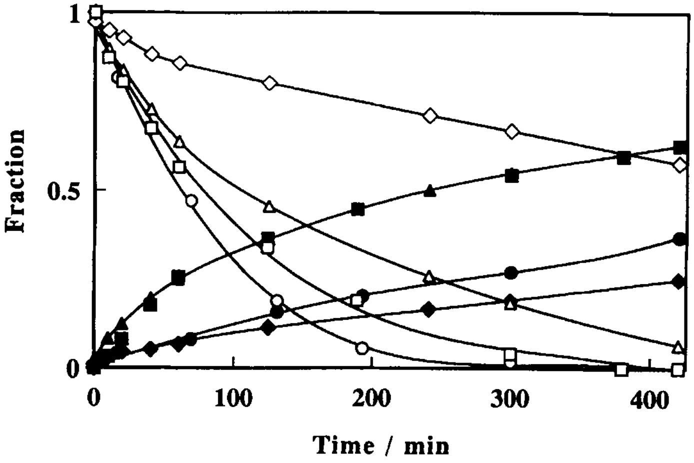

FIG. 4 Effect of LIX 63 concentration on the transport of Mo(VI); feed: 0.1 M HNO_3; product: 0.1 M NaOH. $C_{LIX\,63}$: (◇, ◆) 10^{-3} M, (△, ▲) 10^{-2} M, (□, ■) 0.1 M, (○, ●) 1 M; open symbols: feed; filled symbols: product.

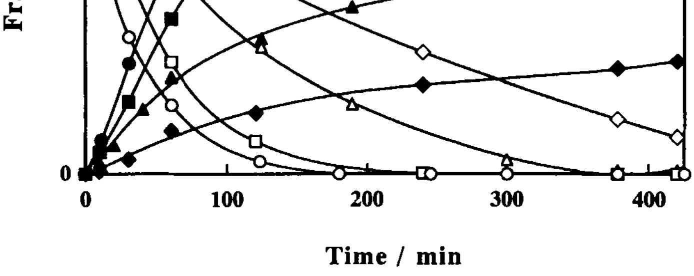

FIG. 5 Effect of HNO_3 concentration in the feed solution on the transport of Mo(VI); SLM: 0.1 M LIX 63 in kerosene; product: 0.1 M NaOH. C_{HNO_3}: (○, ●) 10^{-3} M, (□, ■) 10^{-2} M, (△, ▲) 0.1 M, (◇, ◆) 1 M; open symbols: feed; filled symbols: product.

attained after 3 hours, while more than 80% of Mo(VI) was transported into the product side.

The transport of Mo(VI) at different pH values of the feed solution is shown in Fig. 6. The extraction of Mo(VI) into the SLM was fast at pH 4.5 and slowed down at higher pH. The transport behavior in Figs. 5 and 6 corresponds to the pattern observed in liquid–liquid extraction experiments.

The stripping of Mo(VI) was performed by shifting the acidity to an alkaline region where the MoO_2^{2+} species is converted to the poorly extractable MoO_4^{2-} species. This conversion governs the stripping process. The transport of Mo(VI) was investigated by adopting different NaOH concentrations in the product side. As shown in Table 1, the recovery increased with increasing NaOH concentration up to 0.01 M and became unaltered at about 85% at higher concentrations.

Double Membrane Transport

Although complete stripping of the metal species is essential for quantitative transport process, a portion of Mo(VI) species was found to be retained in the LIX 63 SLM. Incomplete stripping may cause substantial problems during continuous operation due to accumulation of the metal

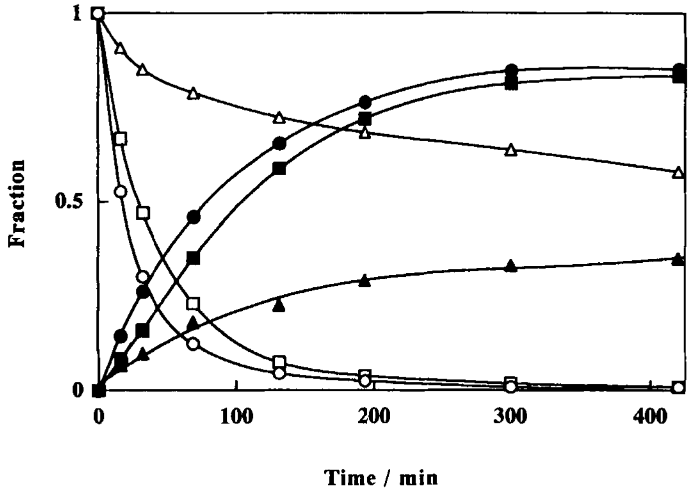

FIG. 6 Effect of pH of feed solution on the transport of Mo(VI); SLM: 0.1 M LIX 63 in kerosene; product: 0.1 M NaOH. pH of feed: (○, ●) 4.5, (□, ■) 5, (△, ▲) 6; open symbols: feed; filled symbols: product.

TABLE 1
Effect of NaOH Concentration on the Transport of Mo(VI)[a]

NaOH (M)	$[Mo]_{f,t}/[Mo]_{f,0}$, 7 hours	Recovery (%), 7 hours
1×10^{-3}	0.065	75.2
5×10^{-3}	0.028	80.1
1×10^{-2}	0.008	83.2
5×10^{-2}	0.001	84.8
1×10^{-1}	0.001	85.0
5×10^{-1}	0.001	85.1

[a] Feed: 10^{-3} M HNO_3; SLM: 0.1 M LIX 63 in kerosene.

species in SLM, resulting in reduction of the overall efficiency of the process. Therefore, the stripping conditions on LIX 63 SLM should be improved. Removing of the metal ion from the product side enhances the liberation of the retained metal species from the SLM. Further transport of the stripped metal ion through a second membrane is a promising procedure to improve the stripping of Mo(VI) from the LIX 63 first membrane. Trioctylmethylammonium chloride (TOMAC) is expected to act as an appropriate carrier in the second membrane, based on the transport behavior of Mo(VI) (11, 12).

TOMAC has anion-exchange characteristics and showed high extractability for anionic forms of Mo(VI). MoO_4^{2-} was highly extractable with TOMAC from a dilute NaOH solution, but the extraction was poor from a concentrate NaOH solution. The uptake of Mo(VI) with TOMAC from an alkaline solution proceeds through the following anionic-exchange equilibria (12):

$$R_3R'NCl_{(o)} + OH^- \rightleftharpoons R_3R'NOH_{(o)} + Cl^- \tag{2}$$

$$2R_3R'NOH_{(o)} + MoO_4^{2-} \rightleftharpoons (R_3R'N)_2MoO_{4(o)} + 2OH^- \tag{3}$$

where $R_3R'NCl$ represents TOMAC.

Additional transport of MoO_4^{2-} across the TOMAC-SLM from the stripping solution of the LIX 63-SLM appears to be suitable for establishing a double membrane system. A proposed transport scheme is illustrated in Fig. 7. Here, SLM1 and SLM2 refer to the membranes containing LIX 63 and TOMAC, respectively, and the intermediate solution refers to the stripping solution of SLM1.

On the basis of the transport behavior of Mo(VI) through individual membranes, LIX 63-SLM, and TOMAC-SLM (11, 12), a double mem-

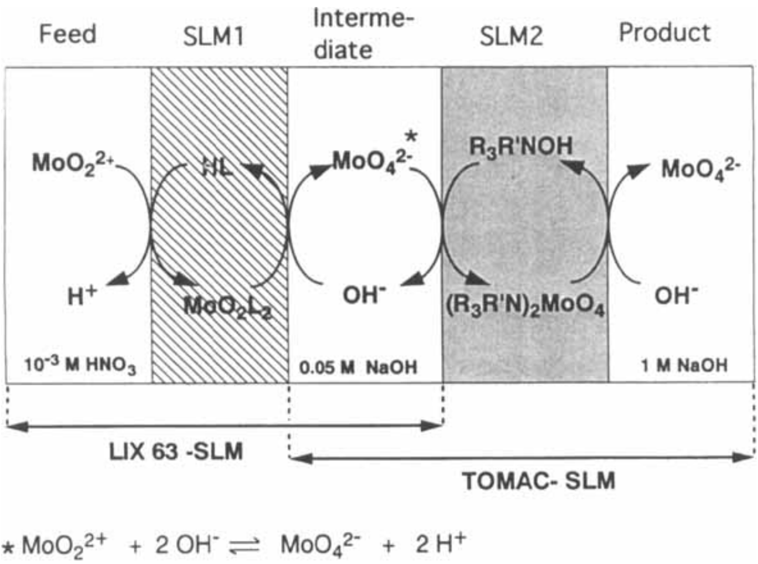

FIG. 7 Scheme of the transport of Mo(VI) through double membranes.

brane experiment was carried out from the feed solution of 10^{-3} M HNO_3 via the intermediate solution of 5×10^{-2} M NaOH into the product solution of 1 M NaOH. The transport profile through the LIX 63-SLM and TOMAC-SLM is illustrated in Fig. 8. The metal ion was rapidly extracted by the LIX 63-SLM from the feed solution. The fractions of Mo(VI) in the intermediate solution initially increased with time due to successive stripping and then decreased owing to extraction into the TOMAC-SLM. On the other hand, the fraction of Mo(VI) in the product solution slowly increased, indicating limited transport across the second SLM. Molybdenum(VI) in an acidic feed solution exists as MoO_2^{2+} which is readily extracted to the LIX 63-SLM, and the formed complex (MoO_2L_2) is diffused through the membrane and dissociated in the alkaline intermediate solution. The MoO_2^{2+} is converted to MoO_4^{2-}, and the latter is extracted with TOMAC-SLM. The complex $(R_3R'N)_2MoO_4$ is diffused and the MoO_4^{2-} is released into the product solution of 1 M NaOH.

For a feasible transport process, sufficient transport of Mo(VI) from the intermediate solution into the product side is essential. The rate of extraction of Mo(VI) with an SLM is inversely proportional to the volume of feed solution (11):

$$k_{f,obs} = (J/[Mo]_{f,0}) \cdot S/V_f$$

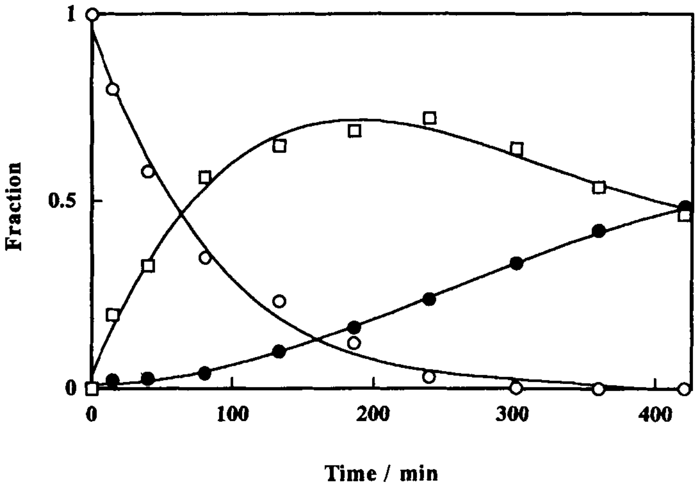

FIG. 8 Transport of Mo(VI) through double membranes; feed: 10^{-3} M HNO_3 (100 cm^3); SLM1: 0.1 M LIX 63 in kerosene; intermediate: 5×10^{-2} M NaOH (100 cm^3); SLM2: 1 M TOMAC–10% 1-octanol in kerosene; product: 1 M NaOH (100 cm^3); (○) feed, (☐) intermediate, (●) product.

where $k_{f,\,obs}$ (s^{-1}) is the apparent transport rate constant from the feed side, J is the flux ($mol{\cdot}cm^{-2}{\cdot}s^{-1}$), $[Mo]_{f,0}$ is the concentration of Mo(VI) in the feed solution at time zero, S is the geometric area of the SLM, and V_f is the volume of feed solution. The transport rate can be accelerated by reducing the feed volume and increasing the membrane area. Here, the intermediate solution is simultaneously acting as a product solution for SLM1 and as a feed solution for SLM2, and a decrease in its volume is favorable for promoting the transport rate across SLM2. Transport experiments were carried out by varying the volume of intermediate solution (V_i) at constant volume (100 cm^3) of feed and product solutions. As shown in Fig. 9, Mo(VI) was slowly transported in the product side at a large V_i of 100 cm^3 due to slow extraction with SLM2. The transport into the product solution was markedly enhanced at a small V_i of 20 cm^3 owing to fast extraction into SLM2. The recovery in the product solution was about 75% after 7 hours of transport, and the Mo(VI) in the intermediate solution was about 3%. The unrecovered Mo(VI) is considered to be retained as complexes with reagents in SLMs.

The transport was performed by varying LIX 63 concentrations from 10^{-1} to 10^{-3} M in SLM1 at a V_i of 20 cm^3. As shown in Fig. 10, the release of Mo(VI) into the product side was improved at a lower LIX 63

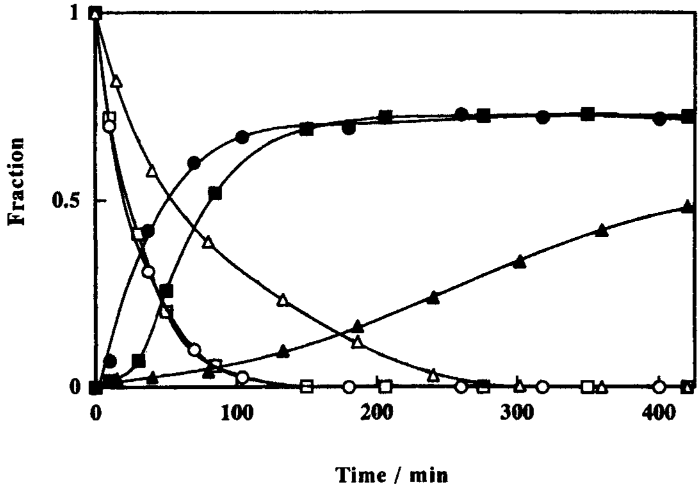

FIG. 9 Transport of Mo(VI) through double membranes at different volumes of intermediate solution; (○, ●) 20 cm^3, (□, ■) 50 cm^3, (△, ▲) 100 cm^3; open symbols: feed; filled symbols: product. Other conditions are the same as those in Fig. 8.

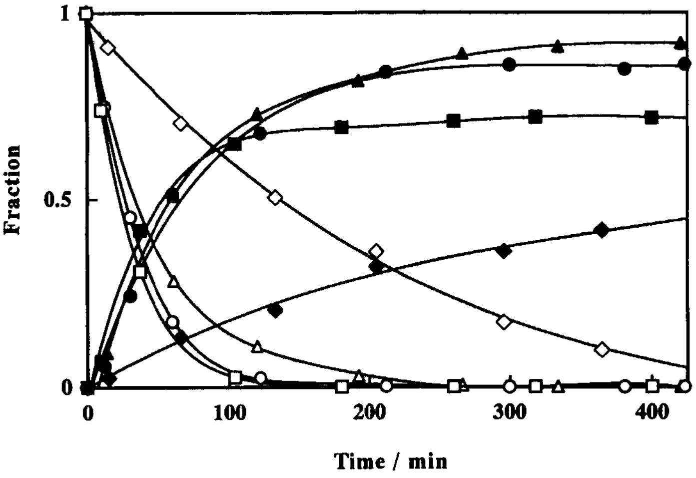

FIG. 10 Transport of Mo(VI) through double membranes at different LIX 63 concentrations in SLM1; intermediate volume: 20 cm^3. $C_{\text{LIX 63}}$: (◇, ◆) 10^{-4} M, (△, ▲) 10^{-3} M, (○, ●) 10^{-2} M, (□, ■) 0.1 M; open symbols: feed; filled symbols: product. Other conditions are the same as those in Fig. 8.

concentration. About 95% of Mo(VI) was recovered at 10^{-3} M LIX 63, indicating little retention of Mo(VI) in SLM1. The continuous removal of the metal ion from the intermediate solution enhanced the liberation of the retained metal ion from SLM1 at low LIX 63 concentration. The rate of metal extraction from the feed side significantly decreased with a further decrease in LIX 63 concentration. On the other hand, high TOMAC concentration in SLM2 is important for fast extraction of Mo(VI) from the intermediate solution.

In order to concentrate the metal species in the product side, the transport was tested by reducing the volume of product solution at the feed volume to 100 cm^3. The results of recovery and enrichment factor ($[\mathrm{Mo}]_{\mathrm{p,t}}/[\mathrm{Mo}]_{\mathrm{f,0}}$) related with the volume ratio of the feed to product solutions, V_f/V_p, are summarized in Table 2. Molybdenum(VI) was concentrated in the product solution with high recovery; however, the recovery decreased with further increases of the V_f/V_p value. The fraction of Mo(VI) in the intermediate solution was found to be small with a few exceptions for high V_f/V_p cases.

Transport of Respective Ions

Hydrometallurgical processing of molybdenum resources involves bringing many associated elements into solutions. Separation of molybdenum from these impurities is of practical importance. LIX 63 has been shown to be a selective extractant for Mo(VI) over several ions owing to the high complexing ability to cationic Mo(VI) species from acidic solutions. On the other hand, TOMAC can extract anionic Mo(VI) species together with a variety of anions. The connection of LIX 63 and TOMAC

TABLE 2
Effect of the Volume Ratio of Feed to Product Solutions on the Transport of Mo(VI) through Double Membranes[a]

V_p (cm^3)	V_f/V_p	$[\mathrm{Mo}]_{\mathrm{p,t}}/[\mathrm{Mo}]_{\mathrm{f,0}}$, 7 hours	Recovery (%), 7 hours
100	1	0.95	95.0
50	2	1.9	94.9
25	4	3.7	93.6
10	10	7.8	77.6

[a] Feed: 10^{-3} M HNO_3; intermediate: 5×10^{-2} M NaOH; product: 1 M NaOH; SLM1: 10^{-3} M LIX 63 in kerosene; SLM2: 1 M TOMAC–10% 1-octanol in kerosene.

TABLE 3
Transport of Respective Ions through Double Membranes[a]

Element	Fraction, 7 hours		
	Feed	Intermediate	Product
Ca(II)	0.98	0.01	0.00
Cu(II)	0.98	ND[b]	ND
Fe(III)	0.96	ND	ND
Zn(II)	0.99	0.01	0.00
P(V)	0.98	0.00	0.00
Si(IV)	0.95	0.01	0.01
Al(III)	0.99	ND	ND
Co(II)	0.94	0.01	0.00
Ni(II)	0.96	0.01	0.00
V(IV)	0.00	0.14	0.60

[a] Feed: 10^{-3} M HNO_3; intermediate: 5×10^{-2} M NaOH; product: 1 M NaOH; SLM1: 10^{-3} M LIX 63 in kerosene; SLM2: 1 M TOMAC–10% 1-octanol in kerosene.

[b] Not determined.

in double membranes provides for the selective transport of Mo(VI). The transport of several ions in a mixture was examined by the use of the present double membranes, and the fractions in the feed, intermediate, and product solutions after 7 hours are summarized in Table 3.

Molybdenum concentrates usually contain some impurities such as Ca(II), Cu(II), Fe(III), Zn(II), P(V), and Si(IV) which are partially or totally dissolved during leaching processes (1). As shown in Table 3, these ions showed little transport through the double membranes, with a large portion of them remaining in the feed solution.

Metal ions such as Al(III), Co(II), Ni(II), and V(IV) are present together with Mo(VI) in spent catalysts (17), and their separation is necessary for metal recycling. Al(III), Co(II), and Ni(II) can be easily isolated to remain in the feed solution. Only vanadium(IV) was partially transported from the feed side into the product solution because its permeation properties are similar to those of Mo(VI).

CONCLUSION

Molybdenum(VI) was transported across a LIX 63 liquid membrane with a recovery of more than 80%. Almost complete and selective transport of Mo(VI) were accomplished by double membranes containing LIX

63 and TOMAC carriers. The high complexing ability of LIX 63 with Mo(VI) in acidic solution provides for its separation from most varieties of accompanying ions.

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