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## Effect of Recycling of Feed Solution on the Efficiency of Supported Liquid Membrane Module

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### ABSTRACT

The effect of recycling of a feed solution from the outlet to the inlet of the feed channel of a plate-and-frame type supported liquid membrane module on the recovery of zinc was investigated under the condition that the feed side mass transfer resistance is predominant. Di(2-ethylhexyl)phosphoric acid was used as the carrier. It was found that at a given feed flow rate, recycling of the feed solution is effective for improving the recovery of zinc since recycling brings about an increase of the flow rate in the feed channel and also the mass transfer coefficient in the feed side. The experimental data could be satisfactorily simulated by a proposed theoretical model.

### INTRODUCTION

In the separation by supported liquid membranes (SLMs), mass transfer resistance in a feed phase can be predominant if the distribution ratio of a permeant species at the feed–liquid membrane interface is very large (1). In such cases the permeation rate can be increased by increasing  $u_F$ , the linear velocity of the feed solution in the feed channel of an SLM module, since the mass transfer coefficient in the feed solution,  $k_F$ , increases with an increase in  $u_F$  (2).

This suggests that for increasing the recovery of a permeant species for a given feed flow rate, recycling of a part of the feed solution is favorable

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since the recycling brings about an increase in  $u_F$  and also  $k_F$ . The mode of the recycling operation is schematically shown in Fig. 1.

In this study the effect of recycling of the feed solution on the recovery of zinc by a plate-and-frame type supported liquid membrane module was experimentally and theoretically investigated. As the carrier of zinc, di(2-ethylhexyl)phosphoric acid (D2EHPA) was used.

## EXPERIMENTAL

### Reagents

D2EHPA was a generous gift from Daihachi Chemical Industry Co. Ltd., Japan. Liquid membrane solutions were prepared by dissolving D2EHPA in *n*-dodecane. Aqueous zinc solutions were prepared by dissolving zinc nitrate in deionized water. The pH of the solution was adjusted to 3 by the addition of nitric acid. Aqueous hydrochloric acid solution (2 mol/dm<sup>3</sup>) was used as the stripping solution.

### Apparatus and Procedure

The plate-and-frame type supported liquid membrane module (SLM module) used in the experiment, which is similar to that used elsewhere (3), is schematically shown in Fig. 2. The length  $L$ , the width  $W$ , and the depth  $d$  of both the feed and the strip channels were 98, 1.35, and 0.1 cm, respectively. In each channel a mesh spacer (opening, 0.89 cm; thickness, 0.1 cm) was inserted. Fluoropore FP-010 made of polytetrafluoroethylene (Sumitomo Electric Co., Ltd.) was used as the support membrane of the SLM.

The schematic diagram of the experimental apparatus is shown in Fig. 3. An aqueous zinc solution was supplied to the feed channel of the SLM

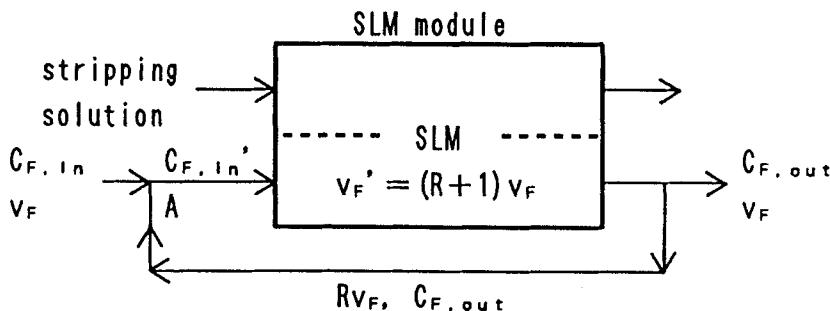


FIG. 1 Operation of supported liquid membrane module with recycling of feed solution.

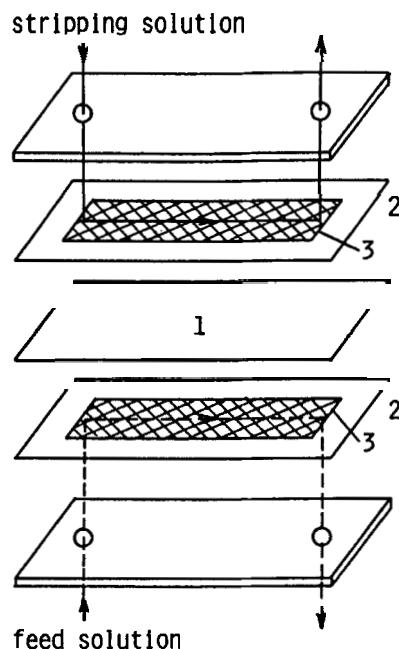


FIG. 2 Schematic diagram of plate-and-frame type supported liquid membrane module. 1: Supported liquid membrane. 2: Gasket. 3: Mesh spacer.

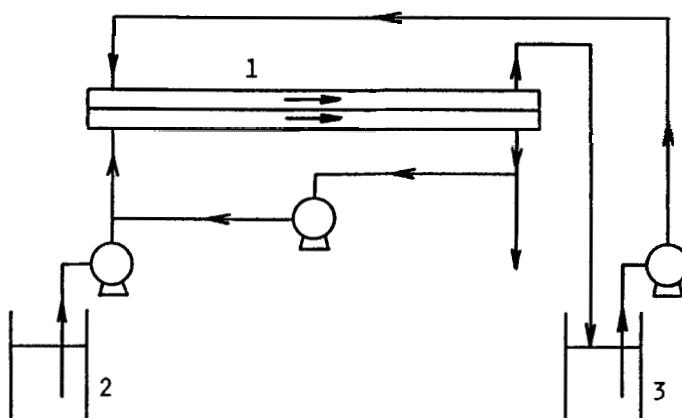


FIG. 3 Schematic diagram of experimental apparatus. 1: Supported liquid membrane module. 2: Reservoir of feed solution. 3: Reservoir of stripping solution.

module by a peristaltic pump at a prescribed flow rate,  $v_F$ , and a part of the solution leaving the feed channel was recycled to the inlet of the feed channel by a roller pump at a flow rate of  $Rv_F$ , where  $R$  is the recycling ratio. The module was operated in a once-through mode with recycling for the feed solution and a circulation mode for the stripping solution. To prevent the degradation of the SLM due to the dissolution of the membrane solution, a small amount of organic membrane solution was added to the stripping solution (3). The zinc concentration in the aqueous solutions was determined by atomic absorption spectrophotometry. All experiments were conducted at 25°C.

## RESULTS AND DISCUSSION

### Determination of Mass Transfer Coefficient in the Feed Solution

As described earlier, when the distribution ratio of a permeant species at the liquid membrane-feed solution interface is sufficiently high, its permeation rate is limited by the mass transfer resistance in the feed solution. In order to determine the relationship between the mass transfer coefficient in the feed solution  $k_F$  and the linear velocity of the feed solution  $u_F$ , a series of experiments were performed under the condition of low hydrogen ion concentration, low zinc concentration in the feed, and high carrier concentration, i.e., under the condition of  $\text{pH}_{F,\text{in}} = 3$ ,  $C_{F,\text{in}} = 23 \text{ ppm}$ , and  $[(\text{HL})_2] = 0.698 \text{ mol/dm}^3$ . In this case the distribution ratio of zinc, which is defined as the ratio of the concentration in the organic phase to that in the aqueous phase, was estimated as high as  $1.5 \times 10^4$  (4). The SLM module was operated in a once-through mode for the feed solution without recycling. The value of  $k_F$  was calculated by the following equation (2):

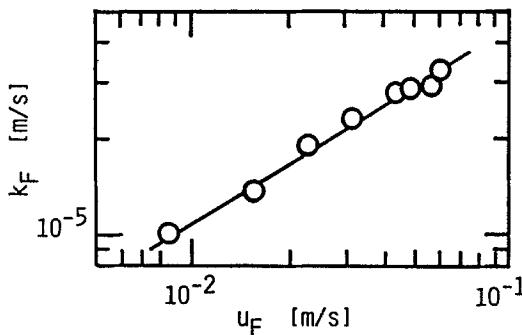
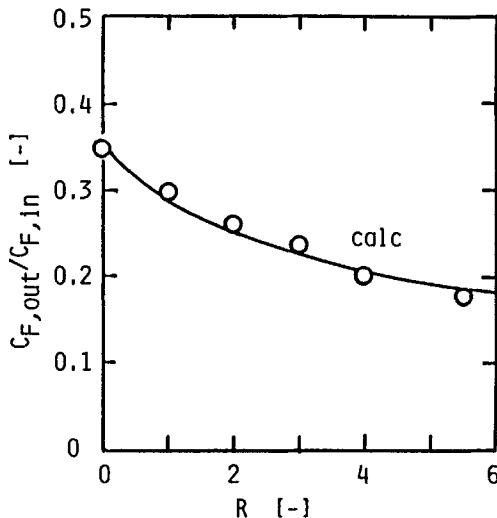
$$k_F = (v_F/S) \ln(C_{F,\text{in}}/C_{F,\text{out}}) = (u_F d/L) \ln(C_{F,\text{in}}/C_{F,\text{out}}) \quad (1)$$

The result is shown in Fig. 4. The data were correlated by the following equation:

$$k_F = 1.82 \times 10^{-4} u_F^{0.615} \quad (2)$$

### Effect of Recycling Ratio on the Recovery of Zinc

Figure 5 shows the effect of the recycling ratio  $R$  on the dimensionless outlet concentration of zinc,  $y$  ( $= C_{F,\text{out}}/C_{F,\text{in}}$ ). It is clearly seen that as  $R$  increases, the outlet concentration decreases, indicating that recycling is effective for obtaining high recovery.

FIG. 4 Effect of linear velocity  $u_F$  on mass transfer coefficient  $k_F$ .FIG. 5 Effect of recycling ratio  $R$  on dimensionless outlet concentration  $y$ .  $C_{F,in} = 23$  ppm,  $pH_{F,in} = 3.0$ ,  $[(HL)_2] = 0.698$  mol/dm<sup>3</sup>,  $v_F = 8$  cm<sup>3</sup>/min.

### Simulation of Experimental Data

The material balance equation at Point A in Fig. 1 is expressed as follows:

$$v_F C_{F,in} + R v_F C_{F,out} = (1 + R) v_F C'_{F,in} \quad (3)$$

The relation between  $C'_{F,in}$  and  $C_{F,out}$  is given as follows (2):

$$C_{F,out} = C'_{F,in} \exp(-k_F S/v_F) = C'_{F,in} \exp[-k_F S/(1 + R)v_F] \quad (4)$$

From Eqs. (3) and (4), the following equation is obtained:

$$y = \frac{C_{F,out}}{C_{F,in}} = \frac{\exp\{-k_F S/(1 + R)v_F\}}{1 + R[1 - \exp\{-k_F S/(1 + R)v_F\}]} \quad (5)$$

The linear velocity in the feed channel  $u_F$  is given by  $(1 + R)v_F/(Wd)$ . The solid line in Fig. 5 is the result calculated by Eq. (5) in which  $k_F$  was estimated by Eq. (2). It can be seen that the experimental data are in good agreement with the theoretical predictions.

Since the enhancement of the recovery of zinc by the recycling is due to the increase of  $k_F$  with increasing  $u_F$ , the effect of recycling is considered to be more effective when the value of  $n$  in the relation  $k_F = \text{constant} \times u_F^n$  is larger. Therefore, it should be noted that the recycling of feed solution is not effective for a hollow fiber module with the feed solution flowing in the lumen side of the hollow fibers since the value of  $n$  for this case is as low as  $1/3$  (5).

## SYMBOLS

$C$	concentration of zinc (mol/m <sup>3</sup> )
$d$	channel depth of SLM module (m)
$k_F$	feed side mass transfer coefficient (m/s)
$L$	channel length of SLM module (m)
$R$	recycling ratio
$S$	membrane area (m <sup>2</sup> )
$u_F$	linear velocity in feed channel (m/s)
$v_F$	volumetric feed rate (m <sup>3</sup> /s)
$v'_F$	volumetric rate in feed channel (m <sup>3</sup> /s)
$W$	channel width of SLM module (m)
$y$	$C_{F,out}/C_{F,in}$
$[(HL)_2]$	concentration of dimer of D2EHPA (mol/dm <sup>3</sup> )

## Subscripts

$F$	feed solution
in	inlet
out	outlet

## ACKNOWLEDGMENT

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