



## 中文摘要

本文以理論分析與實驗操作並進，探討交流型平板式薄膜萃取器中之質量傳送效率。理論分析乃類比交流型平板式熱交換器中之熱質傳送理論，並假設通道截面積上溶質之濃度為均勻。實驗則藉微孔聚丙烯薄膜，以甲基異丁酮萃取水中之醋酸。實驗結果印證了理論之預測值。

# A Study on the Performance of Cross-Flow Rectangular Membrane Extrators

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**Abstract** - The mass transfer for membrane extraction through a cross-flow parallel-plate module has been studied both theoretically and experimentally. Theoretical analysis of mass transfer in cross-flow membrane extractors was analogous to heat transfer in cross-flow heat exchangers with the assumption that the concentration variation in the cross sections of flow channel was negligible. Experiments were carried out with use of membrane sheet made of microporous polypropylene coated with polytetrafluoroethylene as a permeable barrier to extract acetic acid from aqueous solution by methyl isobutyl ketone. Theoretical predictions are in agreement with the experimental results. The simpler but still precise equation for predicting the total mass-transfer rate might be more powerful than the exact solution obtained in the previous work to overcome the mathematical difficulties in device design.

*Key words* : Solvent extraction ; Microporous membrane ; Cross flow

## INTRODUCTION

When solvent extraction is carried out in a microporous membrane device, the membrane is generally in contact with two kinds of fluid at two sides (phase a and phase b) and if fluids a and b are miscible, then in the pores of the membrane are filled with another fluid (phase c) which is immiscible with these two fluids. The solute is extracted from phase a to phase c and then to phase b, or vice versa (Lo and Baird, 1980; Kiani et al., 1984). Thus, membrane solvent extraction is a dispersion-free separation process which can overcome the application limitations of conventional solvent extraction, such as flooding, intimate mixing, limitations on independent phase flow rate variations, requirement of density difference and inability to handle particulates (Prasad et al., 1986).

Like heat exchangers, mass exchangers of membrane solvent extraction may be extremely different in design and construction, but in principle the only differences are those of relative direction of the two fluids, phases a and b. Accordingly, distinction is made between parallel flow and cross flow. The direction of two fluids in cross flow may generally be different by 90 angular degrees. A theoretical study on membrane extraction through cross-flow rectangular modules was done by the present

authors (Yeh and Hsu, 1999). The equation they derived is though an exact solution for estimating the mass-transfer rate, but is an integration form and its applications are cumbersome. The purpose of present work is to derive a simpler but still precise equation for mass transfer by neglecting the concentration variation in the cross sections of flow channel. Both the exact and approximate solutions will then be confirmed with the experimental result.

## THEORY

The schematic diagram in Fig. 1 may serve to explain the nomenclature to be employed for cross flow. This system consists of two channels, for the fluids a and b, respectively, which are separated by a microporous membrane sheet through which solute is extracted and transferred perpendicularly to its exposed surfaces. The assumptions made in this analysis are: steady state, no chemical reaction, uniform velocities over the cross section of flow, constant rates of flows, constant mass-transfer coefficients and constant distribution coefficients. Further, here we only consider the simplest type of cross-flow systems in which the flow directions of the two fluids are perpendicular, instead of being oblique, to each other. By taking the mass balances for membrane solvent extraction through a

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differential area  $dx dy$  in a cross-flow mass exchanger, two partial differential equations for solute concentrations are obtained

$$(KB/Q_a)(H_{ac}C_a - H_{bc}C_b) = -(\partial C_a/\partial x) \quad (1)$$

$$(KB/Q_b)(H_{ac}C_a - H_{bc}C_b) = (\partial C_b/\partial y) \quad (2)$$

where  $H_{ac}$  and  $H_{bc}$  are the distribution coefficients of solute between two different phases, while  $K$  denotes the overall mass-transfer coefficient.

In addition to all the assumptions made previously, here for simplicity, we will further assume uniform concentrations in the cross sections of flow channel. This is valid when  $C_a$  and  $C_b$  do not vary sensitively with  $y$  and  $x$ , respectively. Therefore,  $C_a(x, y) \approx C_a(x)$  and  $C_b(x, y) \approx C_b(y)$ , and Eqs. (1) and (2) become

$$-Q_a dC_a = KB(H_{ac}C_a - H_{bc}C_b)dx \quad (3)$$

$$Q_b dC_b = KL(H_{ac}C_a - H_{bc}C_b)dx \quad (4)$$

The outlet concentrations,  $C_{a,e}$  and  $C_{b,e}$ , were determined from Eqs. (3) and (4), by following the procedure of Bowman et al. (1940) with the use of the following boundary conditions:

$$C_a = C_{a,i} \text{ at } x = 0 \quad (5)$$

$$C_a = C_{a,e} \text{ at } x = L \quad (6)$$

$$C_b = C_{b,i} \text{ at } y = 0 \quad (7)$$

$$C_b = C_{b,e} \text{ at } y = B \quad (8)$$

Finally the rate of mass transfer was obtained as

$$W = Q_a(C_{a,i} - C_{a,e}) \quad (9)$$

$$= Q_b(C_{b,e} - C_{b,i}) \quad (10)$$

$$= \frac{(Q_a H_{ac})(H_{ac}C_{a,i} - H_{bc}C_{b,i})}{\left[ \frac{1}{1-e^{-n}} \right] + \left[ \frac{\ell}{1-e^{-n\ell}} \right] - \left( \frac{1}{n} \right)} \quad (11)$$

where

$$\ell = Q_a H_{bc} / Q_b H_{ac} \quad (8)$$

$$n = (KSH_{ac})/Q_a \quad (9)$$

and  $S (=BL)$  is the mass transfer area.

## EXPERIMENTAL

### Chemicals and materials

The following chemicals were used in this study: (1) glacial acetic acid (reagent ACS grade, Fisher); (2) methyl isobutyl ketone (MIBK; reagent grade, Fisher); (3) sodium hydroxide (certified ACS grade, Fisher); (4) phenolphthalein (loose crystals, Coleman and Bell).

The permeable barrier is a membrane sheet made of microporous polypropylene coated with polytetrafluoroethylene (Gelman Science, Inc.). This membrane has average pore size of  $0.2 \mu\text{m}$ , porosity of 70 % and thickness of  $178 \mu\text{m}$ . Since microporous polypropylene is hydrophobic membrane, the organic solution (solute: acetic acid; solvent: MIBK) wets the membrane, and thus  $H_{bc} = 1$  and  $H_{ac} = 0.524$  at  $25^\circ\text{C}$  (Yeh and Huang, 1995).

### Apparatus and procedure

A parallel conduit of stainless steel with working dimensions,  $16.5 \text{ cm (L)} \times 16.5 \text{ cm (B)} \times 0.38 \text{ cm (= } 0.3978\text{-}0.0178) \text{ cm (2h)}$ , was constructed for cross-flow membrane extraction. A membrane sheet having total surface area of  $272.25 \text{ cm}^2$  ( $S = BL$ ) with the properties mentioned earlier, was inserted as a permeable barrier between two parallel plates, with same distance from them to divide the conduit into two channels (phase a and phase b) of height  $h$ .

The apparatus used for membrane extraction is the same as that in the previous works (Yeh and Huang, 1995; Yeh et al., 1999), except that the experiment was run in cross flow, instead of running in parallel flow. The procedure was the same as that done by earlier investigators (Prasad and Sirka, 1988; D'Elia et al., 1986). Since the membrane used in this study is hydrophobic, the aqueous pressures were maintained at higher pressures than the organic pressures to prevent solvent mixing between phases. Under specific operation conditions, samples of outlet streams were analyzed at 10 min intervals until steady state was reached as indicated by no change in

acetic acid concentration ( $C_{a,e}$  and  $C_{b,e}$ ) by titration with sodium hydroxide over a period of one hour. The time for steady state was about 30 min.

The operating conditions are as follows. Aqueous solution:  $Q_a = 0.184 \sim 1.426 \text{ cm}^3/\text{s}$ ;  $C_{a,i} = 4.96 \times 10^{-4}, 2.02 \times 10^{-3} \text{ mole/cm}^3$ . Organic solution:  $Q_b = 0.125 \sim 1.5 \text{ cm}^3/\text{s}$ ;  $C_{b,i} = 0$ .

### *Comparison of theoretical predictions with experimental results*

The experimental values of  $C_{a,e}$  were obtained. These experimental results were used to determine overall mass-transfer coefficients with the use of the calculating equations derived by the exact solution (Yeh and Hsu, 1999).

It was found that all overall mass-transfer coefficients  $K$  increase slightly with fluid velocities, especially with  $v_b$ . Therefore, we may assume that  $K$  is independent of the velocity in the organic phase. Accordingly, the following correlation equations were reached with  $L = 16.5 \text{ cm}$ ,  $B = 16.5 \text{ cm}$  and  $h = 0.19 \text{ cm}$  (Yeh et al., 2000)

$$K \times 10^4 (\text{cm/s}) = 7.256 v_a^{0.104}, \quad \text{for } C_{a,i} = 4.96 \times 10^{-4} \text{ mole/cm}^3 \quad (14)$$

$$K \times 10^4 (\text{cm/s}) = 4.734 v_a^{0.124}, \quad \text{for } C_{a,i} = 2.02 \times 10^{-3} \text{ mole/cm}^3 \quad (15)$$

Figures 2 and 3 show the experimental values of overall mass-transfer rates  $W$ . The theoretical predictions were calculated from Eq. (11) of the approximate solution, with the use of the experimental values of  $K$  calculated from the correlation equations, Eqs. (14) and (15), and the results are also plotted in Figs. 2 and 3 for comparison. It is seen from these figures that the theoretical predictions are in agreement with the experimental results.

## DISCUSSION AND CONCLUSION

The exact solution for membrane extraction in cross-flow rectangular devices was given in the previous work with the consideration of nonuniform concentrations in the cross sections of flow channel (Yeh and Hus, 1999). A simpler but still precise solution has been obtained in present work with the assumption of uniform

cross-section concentrations, analogy to the work done by Bowan et al. (1940) for heat exchangers. The experimental work was carried out with a parallel conduit of stainless steel inserted with a membrane sheet made of microporous polypropylene coated with polytetrafluoroethylene to extract acetic acid from aqueous solution by methyl isobutyl ketone. Since the organic solution wetted the membrane during operation, the membrane used in this study is hydrophobic. The overall mass-transfer coefficients were determined from the experimental data and two correlation equations for overall mass-transfer coefficients related to feed concentrations and fluid velocity in phase a were derived. Theoretical predictions obtained from both the exact and approximate solutions are in agreement with the experimental results. It is showed that the deviation of the approximate solution from the exact solution is less than 4% in the present experimental ranges (Yeh and Chen, 2000).

## NOMENCLATURE

$B$	Membrane width (m)
$C_a, C_b$	Solute concentration distribution in phase a, in phase b (kg mole/m <sup>3</sup> )
$C_{a,i}, C_{b,i}$	Outlet solute concentration in phase a, in phase b (kg mole/cm <sup>3</sup> )
$H_{ac}, H_{bc}$	Distribution coefficient between phase a and phase c, between phase b and phase c
$h$	Height of flow channel (cm)
$K$	Overall mass-transfer coefficient (m/s)
$L$	Effective length of a mass exchanger (m)
$\ell, n$	Dimensionless group defined by Eq. (12), (13)
$Q_a, Q_b$	Volume flow rate in phase a, in phase b (m <sup>3</sup> /s)
$S$	Overall mass-transfer area of a microporous membrane sheet, BL (m <sup>2</sup> )
$W$	Total mass-transfer rate in a mass exchanger (kg mole/s)
$x, y$	Rectangular coordinates (m)
$v_a, v_b$	Fluid velocity in phase a, in phase b (cm/s)

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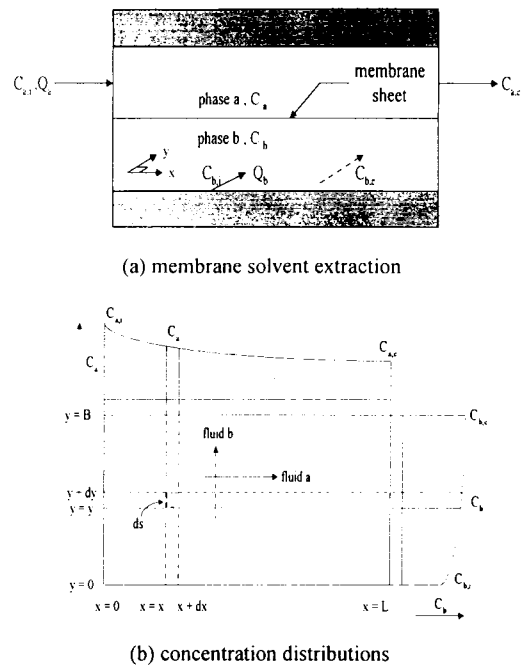


Fig. 1. Schematic diagram of membrane solvent extraction in rectangular mass exchanger of cross flow

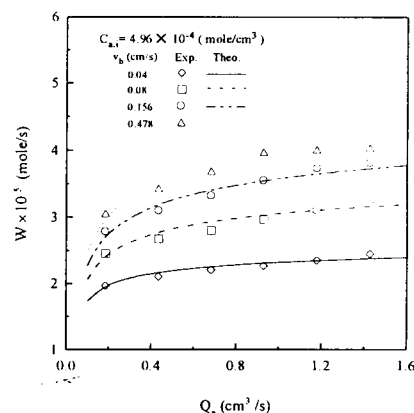


Fig. 2 Total mass-transfer rate vs.  $Q_a : C_{a1} = 0.496 \times 10^{-3} \text{ mole/cm}^3$

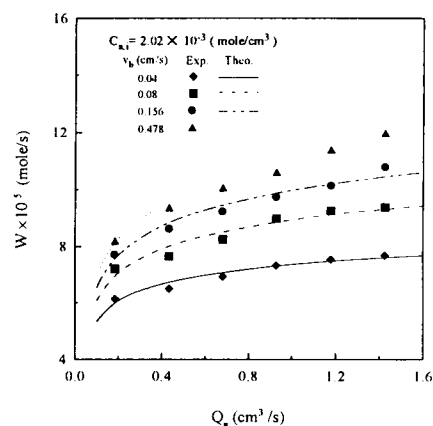


Fig. 3 Total mass-transfer rate vs.  $Q_a : C_{a1} = 2.02 \times 10^{-3} \text{ mole/cm}^3$