行政院國家科學委員會專題研究計劃成果報告

交流型平板式薄膜萃取器效率之研究(2/3)
——回流對交流型平板式薄膜萃取器效率之影響

計 劃 類 別： ■個別型計劃 □整合型計劃

計 劃 編 號： NSC 89-2214-E032-010

執 行 時 間： 89 年 8 月 1 日至 90 年 10 月 31 日

個別型計劃： 計劃主持人： 葉和明
共同主持人：

整合型計劃： 總計劃主持人：
子計劃主持人：

注： 整合型計劃總報告與子計劃成果報告請分開編印各成一冊，彙整一起繳交國科會

處 理 方 式： ■可立即對外提供參考(已發表於 90 年 12 月 7 日舉行的第 23 屆輸送現象與其應用研討會)
□一年後可對外提供參考
□二年後可對外提供參考
(必要時，本會得展延發表時限)

執 行 單 位： 淡江大學化工研究所

中華民國九十年十二月十日

中文摘要

薄膜溶劑萃取裝置主要藉一微孔薄膜將兩液相隔離，而溶質自萃餘相通過薄膜而傳入萃取相。本研究就回流效應對交流式平板型薄膜萃取器效率之影響，作理論與實驗之探討。質量傳送之理論分析，乃類比熱交換器中之熱傳送。實驗係藉微孔聚丙烯所製成之薄膜，作為可通過之隔網，並利用甲基異丙酮為溶劑，在各種溶液濃度與流率以及回流比下，萃取水中之醋酸。理論推測值與實驗值甚為吻合。本研究並證實回流操作，確能顯著提高萃取效率。

當液相(萃餘相或萃取相)之分佈係數較大及溶液濃度較高時，質量傳送之阻力主要受此相所控制，此時薄膜萃取器若加設回流裝置，可增強流體速度而減小其質量傳送阻力，進而提高萃取效率。

薄膜萃取操作優於傳統萃取操作，蓋因前者可以克服後者的這些缺點：氾濫、混合、兩相密度差、液相流速之互相牽制…等。因此薄膜萃取之研究，乃目前分離程序中相當熱門之一環，而本研究所推介之可提高萃取效率的回流效應，可促使薄膜萃取操作之應用更為廣泛。

Proceedings 2001 Symposium on Transport Phenomena and Its Applications page 497-500

A Study on The Performance of Crossflow Flat-Plate Membrane Extractions

Ho –Ming Yeh, Hsian –Hau Cheng, and Yi –Heng Tzeng.

葉和明 鄭翔豪 曾一恆

Department of Chemical Engineering, Tamkang University, Tamsui 251, Taiwan, R.O.C

ABSTRACT

The effects of recycle on membrane extraction through a cross-flow rectangular module have been studied both theoretically and experimentally. Theoretical analysis of mass transfer in cross-flow device with and without recycle was analogous to heat transfer in cross-flow heat exchangers. Experiments were carried out with the use of a membrane sheet made of microporous polypropylene as a permeable barrier to extract acetic acid from aqueous solution by methyl isobutyl ketone. Theoretical predictions are in agreement with the experimental results. Contrast a device without recycle, improvement in mass transfer is achievable if cross-flow membrane extraction is operated in a device of same size with recycle which provides the increase of fluid velocity, resulting in reduction of mass-transfer resistance.

Keywords: Solvent extraction; Microporous membrane; Recycle effect; Cross flow

Introduction

The performance of membrane solvent extraction through rectangular mass exchangers has been discussed in the previous work (Yeh and Hsu, 1999; Yeh and Chen, 2000), under cocurrent-flow, countercurrent-flow and cross-flow operations. Though many research works concerning microporous membrane ex-traction have been reported (Kiani et al., 1984; Basu & Sirkar, 1991; Ding & Cussler, 1991) none of them have considered the effects of recycle of the fluids at the ends. Recently, the effect of recycle on the performance in parallel-flow devices was first investigated by Teramoto et al. (1994), and was later reported by the present author (Yeh, et al., 1998). It was found in previous work that recycle can enhance the mass transfer in parallel-flow rectangular membrane extractors, especially for operations with higher inlet volume rate or shorter conduit length. It is the purpose of present work to investigate theoretically and experimentally the effects of recycle on mass transfer through cross-flow rectangular membrane extractors.

2.Theory

Fig. 1 shows the membrane extraction system, in which an impermeable plate with negligible thickness is placed in vertical to the upper plate and the membrane sheet at the center line of channel a (phase a), to divide the channel into two subchannels (subchannels a_1 and a_2) of same size, and a pump is installed for recycle. Before entering the subchannels a_1, for double-pass system with a recycle at the inlet, the raffinate phase (phase a) with inlet volume rate Q_a and concentration $C_{a,i}$ will mix the fluid exiting from subchannel a_2 with volume rate RQ_a, which is controlled by means of a conventional pump situated at the beginning of subchannel a_1. The extraction phase (phase b) with inlet volume rate Q_b flows steadily through channel b. The total mass transfer area of membrane surface, $S=B\times L=2(L\times B/2)$, are the same for both devices with and without recycle, as shown in the figure.

2.1. Governing equations

Fig. 1 illustrates the flows in a cross-flow membrane extractor with recycle. The expressions for mass transfer rates are

$$W=Q_a(C_{a,i}-C_{a,e})=(1+R)Q_a(C^0_{a,i}-C_{a,e}) \quad (1)$$

$$=Q_b(C_{b,e}-C_{b,i}) \quad (2)$$

Since the impermeable plate divides the raffinate phase into two flow regions, mass balances will be also taken for each flow region. By taking the mass balances through a differential area dxdy in flow region 1, one obtains

$$-[(1+R)Q_a/(B/2)]dydC_{a,1}$$

$$=Kdxdy(H_{ac}C_{a,1}-H_{bc}C_{b,1}) \quad (3)$$

$$=[(Q_b/L)dx]dC_{b,1} \quad (4)$$

Similarly, for flow region 2

$$[(1+R)Q_a/(B/2)]dydC_{a,2}$$

$$=Kdxdy(H_{ac}C_{a,2}-H_{bc}C_{b,2}) \quad (5)$$

$$=[(Q_b/L)dx]dC_{b,2} \quad (6)$$

The boundary conditions for so-lving $C_{a,1}$, $C_{a,2}$, $C_{b,1}$ and $C_{b,2}$ from Eqs.(3)-(6) are:

at x=0, $C_{a,1} = C^0_{a,i}$, $C_{a,2} = C_{a,e}$ (7),(8)

at x=L, $C_{a,1} = C_{a,2} = C_{a,1e}$ (9),(10)

at y=0, $C_{b,1} = C_{b,i}$ (11)

at y=B/2, $C_{b,1} = C_{b,1e}$ (12)

at y=B, $C_{b,2} = C_{b,e}$ (13)

Inspection of Eqs.(7)-(13) shows that the outlet concentrations, $C_{a,1e}$, $C_{a,e}$, $C_{b,1e}$, as well as the mixed inlet concentration, $C^0_{a,i}$, are not specified a priori. Mathematically, more relations for mass-transfer rates in regions 1 and 2, as well as for the mixing effect at the inlet, are needed for determination of these values. They are

$$(1+R)Q_a(C^0_{a,i}-C_{a,1e})=Q_b(C_{b,1e}-C_{b,i}) \quad (14)$$

$$(1+R)Q_a(C_{a,1e}-C_{a,e})=Q_b(C_{b,e}-C_{b,1e}) \quad (15)$$

$$C_{a,i}+RC_{a,e}=(1+R)C^0_{a,i} \quad (16)$$

Solving above equations one obtains the equation for mass transfer rate as

$$W=(1+R)AQ_a\left[C_{a,i}-(H_{bc}/H_{ac})C_{b,i}\right] \quad (17)$$

where

$$A=\frac{1-\left(\frac{2(1+R)}{n}+\ell(1+R)+\frac{1}{1-e^{n\ell/2(1+R)}}-\frac{\ell(1+R)}{1-e^{-n\ell/2}}\right)B}{(1+R)-\frac{2(1+R)}{n}-\frac{1}{1-e^{n\ell/2(1+R)}}+\frac{\ell(1+R)}{1-e^{-n\ell/2}}} \quad (18)$$

$$B=\left[\frac{\ell(1+R)}{1-e^{-\frac{n\ell}{2}}}+\frac{1}{1-e^{-\frac{n}{2(1+R)}}}-\frac{2(1+R)}{n}\right]^{-1} \quad (19)$$

$$\ell=\frac{Q_aH_{bc}}{Q_bH_{ac}} \quad (20)$$

$$n=\frac{KSH_{ac}}{Q_a} \quad (21)$$

3. Experiment

3.1. Experimental performance

The chemical, materials, dimensions of apparatus and experimental procedure are exactly the same as those employed and performed in previous work (Yeh and Chen, 2000), except that in present study, a two-pass membrane extractor with recycle (see Fig.1), instead of a single-pass membrane extractor without recycle, was employed. Experiments were carried out with the use of a membrane sheet (L=B=16.5 cm) made of microporous polypropylene (Gelman sciences, average pore size of 0.2 μm, porosity of 70% and thickness of 178 μm) as a permeable barrier to extract acetic acid (reagent ACS grade, Fisher) from aqueous solution by methyl isobutyl ketone (MIBK, reagent grade, Fisher). The membrane sheet was inserted between two horizontally parallel plates of stainless steel, with same distance from them to divide the conduit into two channels (channels a and b, or phases a and b) of same height (h=0.19 cm). For membrane extraction with recycle, an impermeable plate was placed additionally in vertical to the upper plate and the membrane sheet at the centerline of channel a to divide channel a into two subchannels (subchannels a and b) of height h and width (B/2) for recycle, as shown in Fig. 1.

3.2 Comparison of theoretical predictions with experimental results

The experimental conditions are as follows. Aqueous solution: Q_a=0.184~1.426 cm^3/s; $C_{a,i}=4.96\times10^{-4}$, 2.02×10^{-3} mole/cm^3. Organic solution: Q_b=0.125 cm^3/s; $C_{b,i}$=0. Since acetic-acid aqueous solution (phase a) and MIBK organic solution (phase b) employed in present experimental work are immisible, it does not need to fill another fluid (phase c) in the pores of the membrane. Further, microporous poly-propylene used in present experiment is hydrophobic membrane, the organic solution wets the membrane, and thus H_{bc}=1 and H_{ac}=0.524 at 25°C (Yeh and Huang, 1995). Some experimental results of $C_{a,e}$ were obtained and the experimental values of mass transfer rate, W, were calculated from Eq. (1). The results for W are plotted in Figs. 2 and 3.

The theoretical predictions were calculated from Eq. (17) with the use of the following correlation equations for overall mass transfer coefficients (Yeh and Chen, 2000):

$K\times10^4$(cm/s)=$7.256V_a^{0.104}$, for $C_{a,i}=4.96\times10^{-4}$ mole/cm^3 ; (22)

$K\times10^4$ (cm/s)=$4.734V_a^{0.124}$, for $C_{a,i}=2.02\times10^{-3}$ mole/cm^3 ; (23)

, as well as the relation of V_a to Q_a (cm^3/s)

$$V_a = \frac{Q_a(1+R)}{h(B/2)} = \frac{Q_a(1+R)}{(0.19)(165/2)}, \text{ cm/s} \qquad (24)$$

The results of theoretical prediction are also plotted in Figs. 3 and 4 for comparison. It is seen from these figures that the theoretical predictions are in agreement with the experimental results.

4.Conclusion

The improvement of performance in membrane extraction by operating a device with recycle is best illustrated by calculating the percentage increase in mass-transfer rate based on the operation in a device of same size without recycle

$$I = \frac{W - W_0}{W_0} \qquad (25)$$

The experimental values of W_0 and W were used to calculate I by Eq. (25), and the results are presented in Table 1. From these results we see that operation with recycle of higher reflux ratio substantially improves the mass transfer rate. The enhancement increases with increasing reflux ratio, especially for larger inlet concentration $C_{a,i}$ in raffinate phase.

Nomenclature

B	membrane width (cm)
$C_{a,1}$, $C_{a,2}$	solute concentration distribution in channel a_1, in channel a_2, of raffinate phase (mole/cm^3)
$C_{b,1}$, $C_{b,2}$	solute concentration distribution within $0 \le y \le B/2$, within $B/2 \le y \le B$, of extract phase (mole.cm^3)
$C_{a,i}$, $C_{b,i}$	inlet solute concentration in phase a, in phase b (mole/cm^3)
$C_{a,e}$, $C_{b,e}$	outlet solute concentration in phase a, in phase b (mole/cm^3)
$C_{a,1e}$, $C_{b,1e}$	solute concentration of phase a at x = L, of phase b at y = B/2 (mole/cm^3)
$C^0_{a,i}$	mixed inlet concentration in raffinate phase (mole/cm^3)
H_{ac}, H_{bc}	distribution coefficient between phase a and phase c, between phase b and phase c
h	height of flow channel (cm)
I	improvement of mass-transfer rate defined by Eq. (24)
K	overall mass-transfer coefficient (cm/s)
L	length of membrane sheet. (cm)
ℓ ,n	dimensionless group de-fined by Eq.(20), by Eq.(21)
Q_a, Q_b	volume flow rate in phase a, in phase b (cm^3/s)
R	reflux ratio
S	total mass-transfer area in a membrane sheet, BL (cm^2)
W	total mass-transfer rate in a membrane extractor(mole/s)
W_0	W obtained in a device of same size without recycle (mole/s)
x, y	rectangular coordinates(cm)
V_a, V_b	fluid velocity in phase a, in phase b (cm/s)

Acknowledgements

The authors wish to express their thanks to the National Science Council of R.O.C for financial aid with Grant No. NSC 89-2214-E-032-010

References

Basu,R. and K. K. Sirkar, "Hollow Fiber Contained Liquid Membrane Separation of Citric Acid", *AIChE J.*, **37**, 383 (1991).

Ding H. B. and E. L. Cussler, "Fractional Extraction with Hollow Fibers with Hydrogel-Filled Walls", *AIChE J.*, **37**, 855.

Kiani, A., R. R. Bhave and K. K. Sirkar, "Solvent Extraction with Immobilized Interfaces in Microporous Membrane", *J. of Membr. Sci.*, **20**, 125 (1984).

Yeh, H. M. and Y. K. Chen, (2000). "Membrane Extraction through Cross-Flow Rectangular Modules", *J. of Membr. Sci.*, **170**, 235.

Yeh, H. M. and Y. S. Hsu,. "Analysis of Membrane Extraction through Rectangular Mass Exchangers", *Chem. Eng. Sci.*, **54**, 897 (1999).

Yeh, H. M. and C. M. Huang, "Solvent Extraction in Multipass Parallel-Flow Mass Exchangers of Microporous Hollow-Fiber Modules", *J. of Membr. Sci.*, **103**, 135 (1995).

Yeh, H. M., Y. Y. Pen and Y. K. Chen, "Solvent Extraction through a Double-Pass Parallel-Plate Membrane Channel with Recycle", *J. of Membr. Sci.*, **163**, 177 (1999).

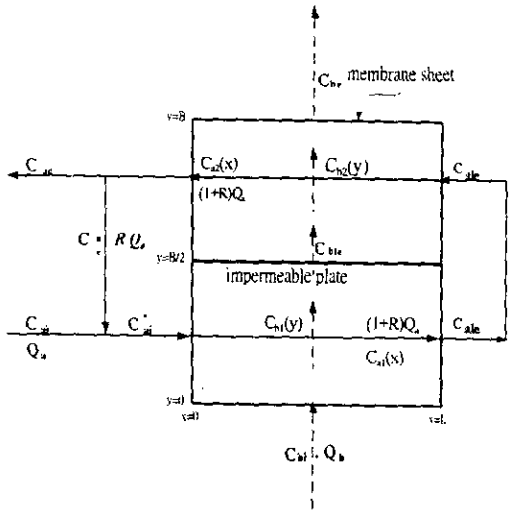

Fig.1. Double-pass cross-flow flat-plate membrane extractor with recycle

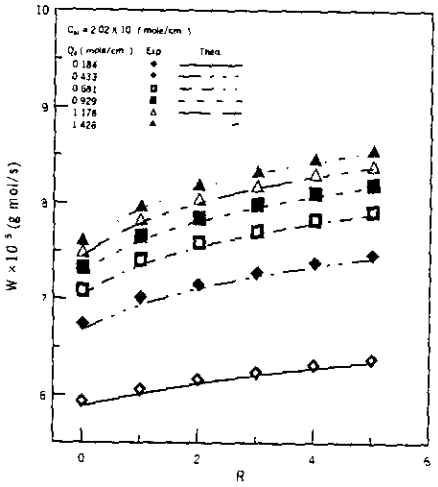

Fig.3. W vs. R: $C_{a,i}$=2.02×10^{-3} mole/cm^3; Q_b=0.125cm^3/s

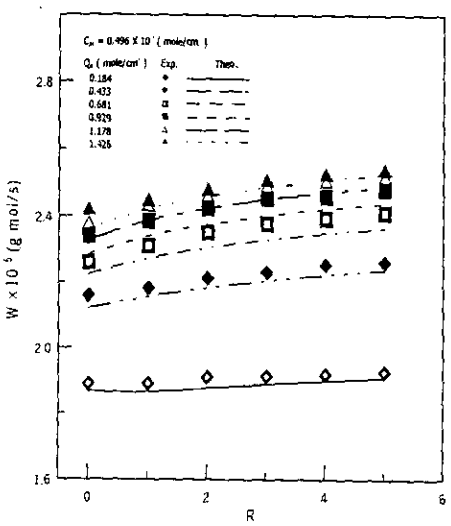

Fig.2. W vs. R: $C_{a,i}$=0.496×10^{-3} mole/cm^3; Q_b=0.125cm^3/s

Table 1

Experimental values of W_n (Yeh & Chen, 2000) and I for water-acetic acid-MIBK system obtained with Q_b=0.125 cm^3/s and $C_{b,i}$=0

$C_{a,i}\times10^3$ (mole/cm^3)	Q_a (cm^3/s)	$W_n\times10^5$ (mole/s)	I(%)		
			R=1	R=3	R=5
0.496	0.184	1.9670	-3.84	-1.33	-1.91
	0.433	2.1044	3.72	6.05	7.49
	0.681	2.2064	4.72	7.84	9.32
	0.929	2.2575	5.68	8.72	9.96
	1.178	2.3560	3.30	5.85	7.05
	1.426	2.4527	-1.15	2.38	3.66
2.02	0.184	6.1401	-1.24	1.70	3.92
	0.433	6.5037	7.93	11.90	14.78
	0.681	6.9326	6.92	11.30	14.26
	0.929	7.3205	4.62	9.20	12.06
	1.178	7.5274	4.13	8.85	11.61
	1.426	7.6719	4.01	8.74	11.65