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Effect of External Recycle and Double-Pass Barrier Location on Membrane Extraction in Cross-Flow Modules

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Abstract: The dispersion free membrane extraction through a cross-flow rectangular membrane module with arbitrary location of an impermeable plate placed for double-pass and external-recycle operations in raffinate phase, has been investigated. Theoretical treatment was analogous to the heat transfer in heat exchangers for the mass transfer in mass exchangers. An experiment was carried out in a cross-flow conduit of stainless steel inserted with a membrane sheet made of microporous polypropylene to extract acetic acid from aqueous solution by methyl iso-butyl ketone. Theoretical predictions are in agreement with the experimental results. Considerable improvement in mass-transfer performance is obtainable if the width of the first-pass subchannel, ΔB decreases, as well as the width of the second-pass subchannel, (1−Δ)B increases. The mass-transfer rate increases also when the reflux ratio increases. Based on the economic point of view, the suitable value of Δ (B is the width of a membrane sheet) may be 0.2.

Keywords: Cross flow, double-pass, external reflux, free barrier-location, membrane extraction

INTRODUCTION

The dispersion-free membrane extraction can overcome the application limitations of conventional solvent extraction, such as flooding, intimate

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mixing, limitations on independent phase flow rate variations, requirement of density difference, and inability to handle the particulate (1–3). The mass-transfer performance of solvent extraction through rectangular membrane modules has been analyzed under parallel-flow (cocurrent and countercurrent flows) and cross-flow operations (4–5). Under comparable conditions, most of the solute is extracted in the countercurrent-flow arrangement and the least in cocurrent flow. It was reported, however, that from secondary effects and high aspect ratio operations, cross flow may extract more solute than countercurrent flow (6). Recently, mass transfer for solvent extraction in hollow-fiber modules was investigated both theoretically and experimentally (7–9).

The reflux indeed has much influence in the heat and mass transfer, (10–19) which in turn plays a significant role in the design, calculation, and operation of the equipment. The effects of recycle on membrane extraction through a double-pass rectangular module have been studied both theoretically and experimentally (20–22). It is the purpose of the present study to investigate the performance in cross-flow modules with external recycle and double pass in the raffinate phase as well as with the arbitrary location of impermeable barrier for double pass.

THEORY

Unlike the computation of mass transfer in parallel-flow systems (either cocurrent or countercurrent flow), the theory of mass exchanger in the cross-flow system is rather complicated because the flow directions of the two fluids cross each other and the concentrations over the cross-section of the flow are nonuniform. 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. The schematic diagrams in Figs. 1 and 2 may serve to explain the nomenclature to be employed for a cross-flow module with double pass and external reflux in the raffinate phase where the impermeable barrier for double pass is located arbitrarily. This system consists of two channels, for the fluids a and b, respectively, which may be immiscible and separated by a microporous membrane sheet through which the solute is extracted and transferred perpendicularly to its exposed surfaces. In the case when fluids a and b are miscible, membrane pores are filled with another fluid (phase c) which is immiscible with original two fluids. The solute is extracted from phase a to phase c and then to phase b, or vice versa.

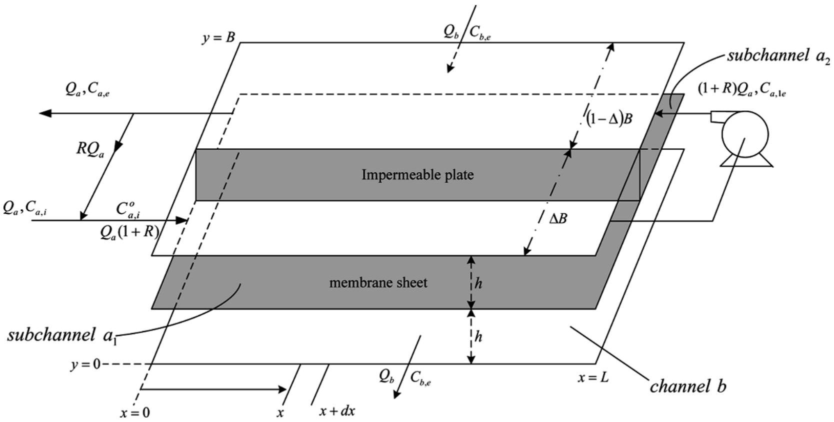

Figure 1. Schematic diagram of cross-flow rectangular membrane extractor with double pass and external recycle in the raffinate phase.

The overall mass balance is

$$\begin{aligned} Q_a(C_{a,i} - C_{a,e}) &= Q_a(1+R)(C^0_{a,i} - C_{a,e}) \\ &= Q_b(C_{b,e} - C_{b,i}) \end{aligned} \tag{1}$$

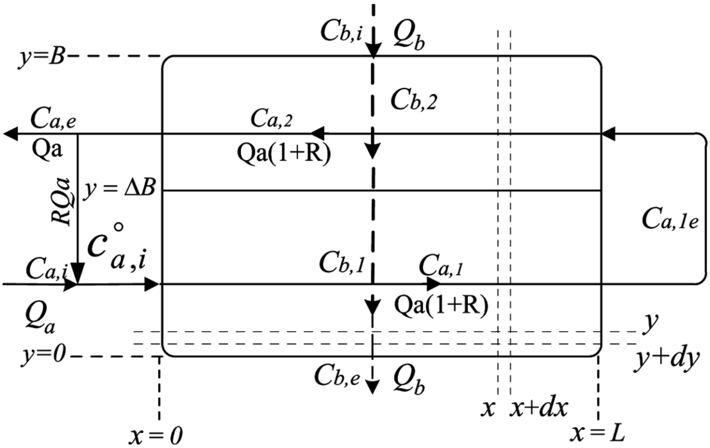

Figure 2. Flow sheet of cross-flow rectangular membrane extractor with double pass and external recycle in the raffinate phase.

Governing equations

As shown in Fig. 1, an impermeable plate with negligible thickness is placed in vertical to and between the upper plate and the membrane sheet, at a certain line in channel a (phase a) to divide the raffinate phase into two subchannels (subchannels a_1 and a_2) of widths ΔB and $(1 - \Delta)B$, respectively, for double-pass flow, and that a pump is installed for the external reflux. Thus, in the raffinate phase (phase a), the inlet fluid of volume rate Q_a mixed with the outlet reflux of volume rate RQ_a, flows steadily through subchannels a_1 and a_2. The extract phase (phase b) with inlet volume rate Q_b flows steadily through channel b and first across subchannel a_2 and then subchannel a_1.

The impermeable plate divides the raffinate phase into two flow regions. By taking the mass balances through a differential area dxdy in flow region 1, one obtains

$$K_1 dxdy(H_{ac}C_{a,1} - H_{bc}C_{b,1}) = -[\{Q_a(1+R)/\Delta B\}dy]dC_{a,1} \qquad (2)$$

$$K_1 dxdy(H_{ac}C_{a,1} - H_{bc}C_{b,1}) = -[(Q_b/L)dx]dC_{b,1} \qquad (3)$$

Similarly, for region 2

$$K_2 dxdy(H_{ac}C_{a,2} - H_{bc}C_{b,2}) = [\{Q_a(1+R)((1-\Delta)B)\}dy]dC_{a,2} \qquad (4)$$

$$K_2 dxdy(H_{ac}C_{a,2} - H_{bc}C_{b,2}) = -[(Q_b/L)dx]dC_{b,2} \qquad (5)$$

where K is the overall mass-transfer coefficient, while H_{ac} and H_{bc} are the distribution coefficients between two different phase, as defined by

$$H_{ac} = \frac{\text{solute concentration in phase c}}{\text{solute concentration in phase a}} \qquad (6)$$

The boundary conditions for solving $C_{a,1}$, $C_{a,2}$, $C_{b,1}$, and $C_{b,2}$ from Eqs. 2–5 are:

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

$$\text{at } x = L, \qquad C_{a,1} = C^0_{a,i}, \qquad C_{a,2} = C_{a,ie} \qquad (9,10)$$

$$\text{at } y = 0, \qquad C_{b,1} = C_{b,2} \qquad (11)$$

$$\text{at } y = \Delta B, \qquad C_{b,1} = C_{b,2} = C_{b,2e} \qquad (12)$$

$$\text{at } y = B, \qquad C_{b,2} = C_{b,i} \qquad (13)$$

Inspection of Eqs. (7)–(13) shows that the outlet concentrations, $C_{a,e}$, $C_{a,1e}$, $C_{b,2e}$, $C_{b,e}$, as well as the mixed inlet concentration, $C^0_{a,i}$, are not specified a priori. Mathematically, more relations for mass-transfer rates, W_1 and W_2, in regions 1 and 2, as well as for the mixing effect at the inlet, are needed for determination of these values. With the definitions of mean concentration differences, $(\Delta C)_{1,m})$ and $(\Delta C)_{2,m})$, in regions 1 and 2, respectively, we have

$$W_1 = Q_a(1+R)(C^0_{a,i} - C_{a,1e}) = Q_b(C_{b,e} - C_{b,2e}) = K_1 L(\Delta B)(\Delta C)_{1,m} \tag{14}$$

$$W_2 = Q_a(1+R)(C_{a,1e} - C_{a,e}) = Q_b(C_{b,2e} - C_{b,i}) = K_2 L((1-\Delta)B)(\Delta C)_{2,m} \tag{15}$$

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

Mass-Transfer Rate

The mathematical treatments for simultaneously solving these equations are rather cumbersome, and will not be presented here but are presented in the Appendix instead. The outlet concentration from phase a, thus obtained, is

$$C_{a,e} = \frac{(1+R)(H_{bc}/H_{ac})C_{b,i}\xi_{a,e} - C_{a,i}(1+\xi_{a,e})}{R\xi_{a,e} - 1} \tag{17}$$

where

$$\xi_{a,e} = \frac{\beta + \gamma}{\gamma(\beta - \alpha)} \tag{18}$$

$$\alpha = l(1+R) \tag{19}$$

$$\beta = \frac{1+R}{n_1\Delta} + \frac{l(1+R)}{1-e^{n_1 l\Delta}} - \frac{1}{1-e^{-n_1\Delta/(1+R)}} \tag{20}$$

$$\gamma = \frac{1+R}{n_2(1-\Delta)} + \frac{l(1+R)e^{n_2 l(1-\Delta)}}{1-e^{n_2 l(1-\Delta)}} - \frac{1}{1-e^{n_2(1-\Delta)/(1+R)}} \tag{21}$$

Finally, the total mass-transfer rate is readily obtained from Eq. (1), as well as from Eqs.(14) and (15), as

$$\mathrm{W} = W_1 + \mathrm{W}_2 = \mathrm{Q_a}(C_{a,i} - C_{a,e}) = \frac{\mathrm{Q}_a(1+\mathrm{R})[C_{a,i} - (\mathrm{H}_{bc}/\mathrm{H}_{ac})C_{b,i}]\xi_{a,e}}{\mathrm{R}\xi_{a,e} - 1} \tag{22}$$

EXPERIMENT

Apparatus and Method

Experiments were carried out with the use of a membrane sheet ($\mathrm{L} = \mathrm{B} = 16.5\,\mathrm{cm}$) made of microporous polypropylene (Gelman Sciences, average pore size $= 0.2\,\mu m$, porosity $= 70\%$ and thickness $\mathrm{t} = 178\,\mu m$) to extract acetic acid (reagent ACS grade, Fisher) from aqueous solution by methyl isobutyl ketone (MIBK, reagent grade, Fisher). The membrane sheet was inserted in parallel between two 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 ($\mathrm{h} = 0.19\,\mathrm{cm}$). Since microporous polypropylene is hydropholic membranes, the organic solution (solute: acetic; solvent: MIBK) wets the membrane, and thus $H_{bc} = 1$ and $H_{ac} = 0.524$ at 25□ (23). Accordingly, the aqueous pressures were maintained at higher pressures than the organic pressures to prevent solvent mixing between phases. The impermeable plate of negligible thickness was placed at arbitrary location and perpendicular to the upper plate and the membrane sheet to divide channel a into two subchannels (subchannels a_1 and a_2) of height h and variable widths (i.e., $\Delta = 0.25$, 0.5 and 0.75) for double-pass operation with external recycle, as shown in Fig. 1.

Comparison of Correlation Predictions with Experimental Results

Many experiment data of outlet solute concentration in phase a, $\mathrm{C_{a,e}}$, for various operating conditions were obtained and the corresponding values of mass-transfer rate, W, were then calculated from Eq.(1). Some of them are plotted in Figs. 3–5.

The following are the correlation equations (5) for estimating the values of the overall mass-transfer coefficient (24):

For $C_{a,\,i} = 0.5 \times 10^{-3}$ $(mole/cm^3)$,

$$\mathrm{K_i} \times 10^4 \ (cm/s) = 7.256\,\nu_{a,i}^{0.140}\,\nu_b^{0.022}, \quad \mathrm{i} = 1,2 \tag{23}$$

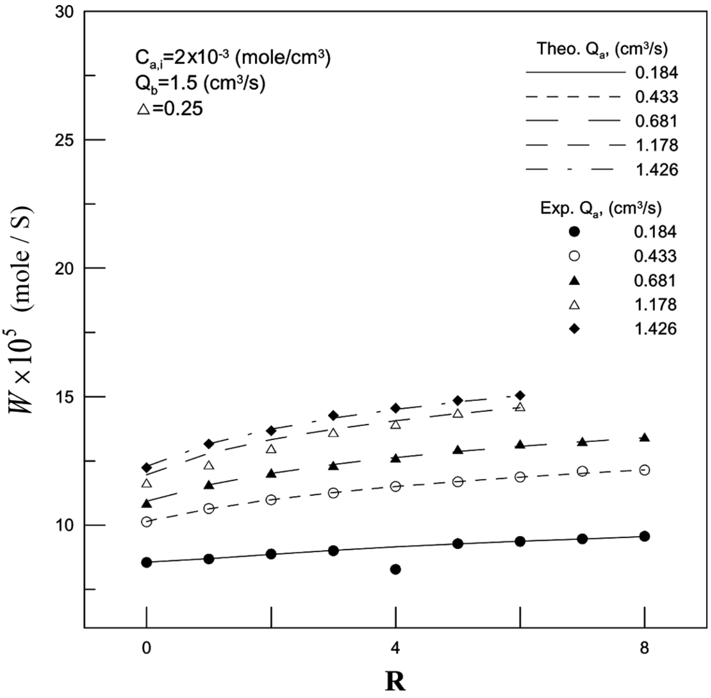

Figure 3. Comparison of theoretical predictions with experimental results, $C_{a,\,i} = 2 \times 10^{-3}$ *mole/cm*^{3}, $Q_b = 1.5$ *cm*^{3}*/s* and $\Delta = 0.25$.

while for $C_{a,\,i} = 2 \times 10^{-3}$ $(mole/cm^3)$,

$$K_i \times 10^4\,(cm/s) = 4.734\,\nu_{a,i}^{0.140}\,\nu_b^{0.022}, \quad \mathrm{i} = 1,2 \tag{24}$$

in which the fluid velocities, $\nu_{a,1}$, $\nu_{a,2}$ and ν_b (cm/s), are

$$\nu_{a,1} = \frac{(1+R)Q_a}{h\Delta B} \tag{25}$$

$$\nu_{a,2} = \frac{(1+R)Q_a}{h(1-\Delta)B} \tag{26}$$

$$\nu_b = \frac{Q_b}{hL} \tag{27}$$

The theoretical predictions of the mass-transfer rate were calculated from Eq. (22), and some of them are plotted in Figs. 3–5 for comparison with the experimental data. It is seen from these figures that the theoretical predictions are in agreement with the experimental results.

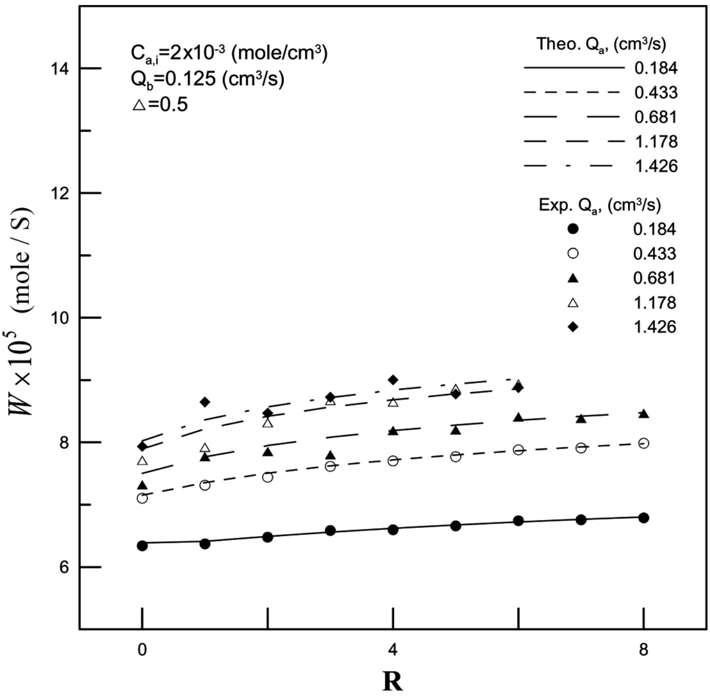

Figure 4. Comparison of theoretical predictions with experimental results, $C_{a,i} = 2 \times 10^{-3}$ *mole/cm*^{3}, $Q_b = 1.125$ *cm*^{3}*/s* and $\Delta = 0.5$.

RESULTS AND DISCUSSION

More predicted values of W are provided in Tables 1 and 2 for discussing the effect of R and Δ on mass-transfer rate. As indicated in Eqs.(23) and (24), mass-transfer coefficients, K_i, increase with the fluid velocities, $v_{a,i}$ and/or v_b, as well as with the flow rates, Q_a and/or Q_b. Accordingly, the total mass-transfer rate increases with Q_a and/or Q_b. This fact was confirmed experimentally, as shown in Figs. 3–5 and Tables 1 and 2.

Effect of Δon Performance

It is shown in Tables 1 and 2 that the mass-transfer rate W increases as the width of subchannel a_1, ΔB, decreases (or the width of subchannel a_2 increases). However, the increment of W turns down as Δ reaches 0.25, while the hydraulic dissipated loss thought to be still small but may increase rapidly. From the economic point of view, therefore, the

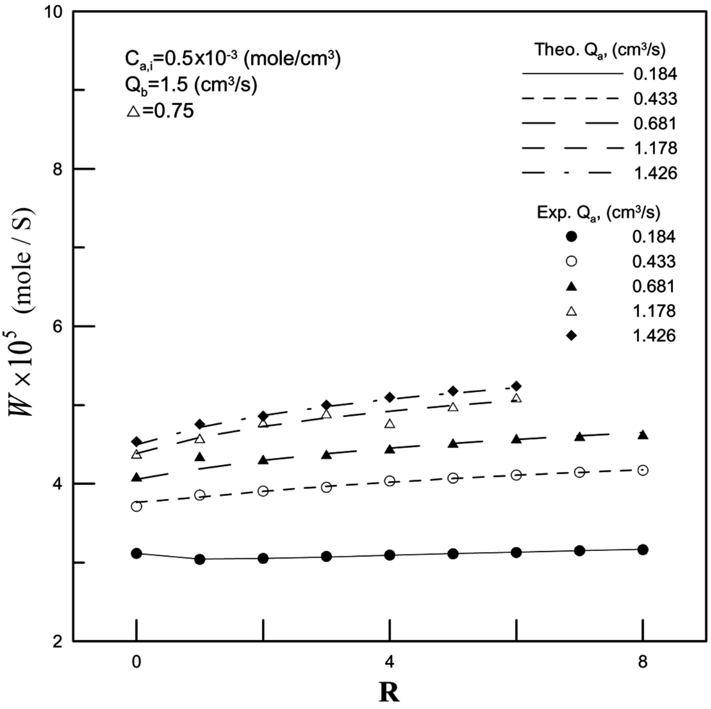

Figure 5. Comparison of theoretical predictions with experimental results, $C_{a,i} = 2\times10^{-3}$ *mole/cm*^{3}, $Q_b = 1.125$ *cm*^{3}*/s* and $\Delta = 0.75$.

suitably limited value of the small width fraction Δ of subchannel a_1, as well as the suitable barrier location for double-pass operation, ΔB, should be properly selected. Considerable improvement in performance is achievable by varying the barrier location from the centerline to decrease the width of subchannel a_1, especially for lower flow-rate operations. For the case of $Q_a = 0.5$ (cm^3/s) and R = 0 shown in Table 2, $W \times 10^5 = 13.1075$ mole/s for $\Delta = 0.1$, and 8.13538 mole/s for $\Delta = 0.5$, and thus the improvement for $\Delta = 0.1$ based on $\Delta = 0.5$ is: (13.1075 − 8.13538)/8.13538 = 63.62%.

Effect of R on Performance

Recycling operation also improves the mass-transfer rate W, except for the case of smaller inlet volume rate Q_a and the operation with reflux ratio R which is not large enough, as shown in Tables 1 and 2 for the case of $Q_a = 0.5$ (cm^3/s), $\Delta < 0.25$ and $R < 1.0$. This is because that in this case,

Table 1. Predicting results with $C_{a,i} = 0.5 \times 10^{-3}$ $mole/cm^3$, $Q_b = 0.5$ cm^3/s and $C_{b,i} = 0$

		$W \times 10^5 (mole/s)$					$E \times 10^8 (hp)$		
Q_a (cm^3/s)	R	$\Delta = 0.1$	$\Delta = 0.25$	$\Delta = 0.5$	$\Delta = 0.75$	$\Delta = 0.9$	$\Delta = 0.1$ (or 0.9)	$\Delta = 0.25$ (or 0.75)	$\Delta = 0.5$
0.5	0	3.277	2.831	2.034	1.142	0.525	0.685	0.347	0.269
0.5	0.5	3.233	2.819	2.060	1.178	0.550	1.500	0.738	0.562
0.5	1.0	3.240	2.839	2.092	1.210	0.569	2.640	1.285	0.972
0.5	2.0	3.285	2.894	2.154	1.260	0.597	5.897	2.848	2.145
0.5	4.0	3.382	2.994	2.250	1.331	0.637	16.320	7.851	5.897
1.0	0	3.464	3.010	2.184	1.240	0.575	2.640	1.285	0.972
1.0	0.5	3.516	3.071	2.251	1.292	0.604	5.897	2.848	2.145
1.0	1.0	3.573	3.131	2.308	1.334	0.627	10.457	5.037	3.786
1.0	2.0	3.677	3.233	2.398	1.397	0.660	23.486	11.291	8.477
1.0	4.0	3.833	3.384	2.579	1.484	0.705	65.179	21.304	23.486
1.5	0	3.600	3.135	2.285	1.364	0.607	5.897	2.848	2.145
1.5	0.5	3.689	3.227	2.370	1.364	0.639	13.226	6.366	4.783
1.5	1.0	3.769	3.304	2.437	1.410	0.663	23.486	11.291	8.477
1.5	2.0	3.898	3.427	2.542	1.480	0.699	52.802	25.362	19.030
1.5	4.0	4.083	3.601	2.687	1.575	0.747	146.610	70.391	52.802

the desirable effect of increasing the fluid velocity by applying the recycle cannot compensate for the situation that the driving force of mass transfer (concentration difference) in the extractor also decreases due to the remixing effect. In general, W increases with the recycle ratio, especially for higher flow rate. For the case of $Q_a = 1.5$ (cm^3/s) and $\Delta = 0.5$ shown in Table 2, $W \times 10^5 = 10.7486$ mole/s for R = 4, and 9.14037 mole/s for R = 0, and thus the improvement by recycle = (10.7468–9.14037)/9.14037 = 17.58%.

Effect of Δ on Hydraulic Dissipated Loss

The hydraulic dissipated loss in the parallel-plate channel may be estimated by

$$\underline{E = (\text{volume flow rate}) \times \Delta \mathrm{P}} \tag{28}$$

If laminar flow in the subchannels a_1 and a_2 is assumed, the pressure drop through the flow channels is (25)

$$\Delta P = \frac{12\mu L(\text{volume flow rate})}{\mathrm{h}^2 \times (\text{cross} - \text{section area of channel})} \tag{29}$$

Table 2. Predicting results with $C_{a,i}=2.0\times10^{-3}$ $mole/cm^3$, $Q_b=0.5$ cm^3/s and $C_{b,i}=0$

		$W\times10^5(mole/s)$				
Q_a (cm^3/s)	R	$\Delta=0.1$	$\Delta=0.25$	$\Delta=0.5$	$\Delta=0.75$	$\Delta=0.9$
0.5	0	13.108	11.325	8.135	4.566	2.102
0.5	0.5	12.932	11.277	8.238	4.714	2.199
0.5	1.0	12.959	11.356	8.370	4.839	2.274
0.5	2.0	13.139	11.574	8.615	5.039	2.389
0.5	4.0	13.528	11.977	9.001	5.326	2.546
1.0	0	13.857	12.039	8.735	4.959	2.300
1.0	0.5	14.063	12.285	9.003	5.170	2.418
1.0	1.0	14.292	12.525	9.231	5.335	2.507
1.0	2.0	14.705	12.932	9.594	5.588	2.640
1.0	4.0	15.334	13.534	10.110	5.936	2.821
1.5	0	14.400	12.542	9.140	5.215	2.427
1.5	0.5	14.758	12.907	9.479	5.455	2.555
1.5	1.0	15.075	13.217	9.750	5.641	2.652
1.5	2.0	15.591	13.709	10.168	5.920	2.796
1.5	4.0	16.331	14.405	10.747	6.300	2.990

Since the total hydraulic dissipated loss includes those in subchannels a_1 and a_2, and in channel b, we have

$$\begin{aligned} \mathrm{E} &= \mathrm{E}_{a,1}+\mathrm{E}_{a,2}+\mathrm{E}_{b} \\ &= \left[Q_a(1+R)\frac{12\mu_a \mathrm{L}\mathrm{Q}_a(1+R)}{h^3\Delta B}\right]+\left[Q_a(1+R)\frac{12\mu_a \mathrm{L}\mathrm{Q}_a(1+R)}{h^3(1-\Delta)B}\right]+\left[Q_b\frac{12\mu_b \mathrm{L}\mathrm{Q}_b}{h^3 B}\right] \\ &= \frac{12\mathrm{L}}{\mathrm{h}^3 B}\left[\frac{\mu_a\{Q_a(1+R)\}^2}{\Delta(1-\Delta)}+\mu_b Q_b^2\right] \end{aligned} \tag{30}$$

The total hydraulic dissipated losses E for various operating conditions were calculated by Eq.(30) with $L=B=16.5\,cm$, $\mathrm{h}=0.19\,\mathrm{cm}$, $\mu_a=1\times10^{-2}\,\mathrm{g}/cm\cdot s$ and $\mu_b=0.58\times10^{-2}\,\mathrm{g}/cm\cdot s$. Some of the results of E are also listed in Table 1. It is seen in this table that E increases when Δ goes far from 0.5 as well as when $\mathrm{Q_a}$ or R increases. However, the hydraulic dissipated loss is very small even for the critical case: $\mathrm{E}=1.466\times10^{-6}$ hp when $\Delta=0.1$ (or 0.9), $\mathrm{Q_a}=1.5\,cm^3/s$ and $\mathrm{R}=4$. Therefore, the operating costs in the device of present interest may be ignored.

Finally, the assumption of laminar flow should be checked. The Reynolds number in the subchannel a_1 may be defined with its equivalent diameter $\mathrm{D_{eq}}$ as

$$\begin{aligned} Re &= \frac{D_{eq} v_a \rho_a}{\mu_a} \\ &= [4(\Delta B)h/2(\Delta B + h)][Q_a(1+R)/(\Delta Bh)](\rho_a/\mu_a) \\ &= \frac{2Q_a(1+R)\rho_a}{(\Delta B + h)\mu_a} \end{aligned} \tag{31}$$

For the typical system ($\Delta = 0.1$, $R = 4$,, $Q_a = 1.5\,cm^3 s$) mentioned above, we have Re = 1305. Therefore, the assumption of laminar flow is acceptable.

Comparison of the Effects of Δ and R on Performances

It was found in the previous section that either increasing R or decreasing Δ may increase the mass-transfer rate in the ignorance of hydraulic dissipated loss. Further, varying Δ is much more effective than increasing R. As shown in Tables 1 and 2, the performance for $\Delta = 0.1$ without recycle ($R = 0$) are much higher than those for $\Delta = 0.5$ and 0.25 with $R = 4$. Therefore, the present system with the location of the impermeable plate moved away from the centerline, should be recommended to be employed, instead of using the one with the barrier located at the centerline ($\Delta = 0.5$), introduced in the previous work (22).

Model Accuracy

The model accuracy may be estimated using the definition

$$E = \frac{1}{N} \sum_{i=1}^{N} \frac{|W_{\exp} - W_{theo}|}{W_{\exp}} \tag{32}$$

where W_{theo} denotes the predicted value of W, while $W_{\exp}$ and N are the experimental data and the number of experimental measurements. The result of error analysis for 256 experimental measurements is about 2%.

CONCLUSION

The predicting equations for the mass-transfer rate in cross-flow membrane extractors with double-pass and external reflux in the raffinate phase, have been derived by mass balances. In the raffinate phase, an impermeable plate is placed at the arbitrary location for double-pass operations. Experimental works were carried out in a stainless steel parallel conduit inserted with a membrane sheet made of microporous polypropylene to extract acetic acid from aqueous solution by methyl isobutyl ketone. Theoretical predictions are in agreement with the experiment results.

Considerable improvement in mass transfer can be achieved by applying the recycling operation, and the mass-transfer rate increases with the reflux ratio R. Further improvement in mass-transfer performance is also obtainable if the location of the impermeable barrier for double pass is adjusted in the manner that the width of subchannel a_1 (ΔB, first-pass conduit for the raffinate phase) decreases, or the width of subchannel a_2 (second-pass conduit) increases. It was found that decreasing Δ is much more effective than increasing R. However, as seen in Tables 1 and 2, the increment of mass-transfer rate turns down as Δ reaches 0.25, while the hydraulic dissipated loss increases rapidly. Therefore, the suitable value of Δ may be 0.2.

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NOMENCLATURE

B = width of membrane sheet (cm)

C_a, C_b = bulk solute concentrations in the raffinate phase (phase a) and in extract phase (phase b) ($mole/cm^3$)

$C_{a,e}$, $C_{b,e}$ = outlet solute concentrations in phase a and in phase b ($mole/cm^3$)

$C_{a,1e}$ = outlet solute concentration in subchannel a_1 ($mole/cm^3$)

$C_{a,i}$, $C_{b,i}$ = inlet solute concentration in phase a, in phase b ($mole/cm^3$)

$C^0_{a,i}$ = mixed inlet concentration in phase a ($mole/cm^3$)

$C_{a1} C_{a2}$ = bulk solute concentration in subchannel a_1, in subchannel a_2 of phase a ($mole/cm^3$)

$C_{a,1m}$, $C_{a,2m}$ = average solute concentrations in phase *a* defined by Eqs. (A.1) and (A.2), respectively ($mole/cm^3$)

$C_{b,1m}$, $C_{b,2m}$ = average solute concentrations in phase *b* defined by Eqs. (A.3) and (A.4), respectively ($mole/cm^3$)

E = hydraulic dissipated loss (hp)
$n_{1,}$ n_2 = constant defined by Eq. (A.6), by Eq.(A.7)
l = constant defined by Eq. (A.5)
H_{ij} = distribution coefficient brtween phase i and phase j
h = half height of parallel channel, or the distance between the flat plate and membrane sheet (cm)
K_i = average overall mass-transfer coefficient (cm/s), $i = 1$ and 2 for regions 1 and 2, respectively (cm/s)
L = the length of membrane sheet (cm)
N = number of experimental measurements
ΔP = pressure drop through the flow channel (Pa)
R = reflux ratio, reverse volume rate RQ_a divided by inlet volume rate Q_a
S = overall mass-transfer area of a flat-plate membrane module LB, (cm/s)
$v_{a,1}$, $v_{a,2}$ = fluid velocity in subchannel a_1, in subchannel a_2 (cm/s)
v_b = fluid velocity in phase b (cm/s)
W = total mass-transfer rate ($mole/s$)
W$_1$, W$_2$ = mass-transfer rate in region 1, in region 2 ($mole/s$)
x = axis along the flow direction of phase a (cm)
y = axis along the flow direction of phase b (cm)
Δ = width fraction of subchannel a_1, $\Delta w/w$.
$\xi_{a,e}$, $\xi_{a,\ 1e}$ = constant defined by Eqs. (A.8) and (A.9), respectively.
α, β, γ = constant defined by Eq. (19), by Eq. (20), by Eq. (21).

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APPENDIX

A. Mean Concentration Difference

Define the following average concentrations and dimensionless groups:

$$\mathrm{C}_{a,1m} = \frac{1}{L}\int_{0}^{L} C_{a,1}dx \tag{A.1}$$

$$C_{a,2m} = \frac{1}{L}\int_{0}^{L} C_{a,2}dx \tag{A.2}$$

$$C_{b,1m} = \frac{1}{\Delta B}\int_{0}^{\Delta B} C_{b,1}dy \tag{A.3}$$

$$C_{b,2m} = \frac{1}{(1-\Delta)B}\int_{\Delta B}^{B} C_{b,2}dy \tag{A.4}$$

$$\ell = \frac{H_{bc}Q_a}{H_{ac}Q_b} \tag{A.5}$$

$$n_1 = \frac{K_1 BLH_{ac}}{Q_a} \tag{A.6}$$

$$n_2 = \frac{K_2 BLH_{ac}}{Q_a} \tag{A.7}$$

$$-\xi_{a,e} = \frac{H_{ac}(C^0_{a,i} - C_{a,e})}{H_{ac}C^0_{a,i} - H_{bc}C_{b,i}} \tag{A.8}$$

$$-\xi_{a,1e} = \frac{H_{ac}(C^0_{a,i} - C_{a,1e})}{H_{ac}C^0_{a,i} - H_{bc}C_{b,i}} \tag{A.9}$$

Accordingly,

$$\frac{\mathrm{H}_{bc}(C_{b,2e} - C_{b,e})}{H_{ac}C^0_{a,i} - H_{bc}C_{b,i}} = \ell(1+R)\xi_{a,1e} \tag{A.10}$$

$$\frac{H_{bc}(C_{b,i} - C_{b,2e})}{H_{ac}C^0_{a,i} - H_{bc}C_{b,i}} = -\ell(1 + R)(\xi_{a,1e} - \xi_{a,e}) \qquad \text{(A.11)}$$

$$\frac{\mathrm{H}_{bc}C_{b,i} - H_{ac}C_{b,e}}{H_{ac}C^0_{a,i} - H_{bc}C_{b,i}} = \ell(1 + R)\xi_{a,e} \qquad \text{(A.12)}$$

Integrating Eqs. (2) and (4) from $y = 0$ to $y = \Delta B$ and from $y = \Delta B$ to B, respectively, one obtains

$$-(Q_a(1 + R)/\Delta B)dC_{a,1} = K_1(H_{ac}C_{a,1} - H_{bc}C_{b,1m})dx \qquad \text{(A.13)}$$

$$(\mathrm{Q}_a(1 + R)/(1 - \Delta)B)dC_{a,2} = K_2(H_{ac}C_{a,2} - H_{bc}C_{b,2m})dx \qquad \text{(A.14)}$$

Similarly, integrating Eqs. (3) and (5) from $x = 0$ to L, we have

$$-(Q_b/L)dC_{b,1} = K_1(H_{ac}C_{a,1m} - H_{bc}C_{b,1})dy \qquad \text{(A.15)}$$

$$-(Q_b/L)dC_{b,2} = K_2(H_{ac}C_{a,2m} - H_{bc}C_{b,2})dy \qquad \text{(A.16)}$$

Integration of Eqs. (A.13) and (A.14) from $x = 0$ to L and rearrangement yields, respectively

$$\mathrm{H}_{bc}C_{b,1m} = \frac{H_{ac}(C_{a,1e} - C^0_{a,i})}{1 - \exp[-n_1\Delta/(1 + R)]} + H_{ac}C^0_{a,i} \qquad \text{(A.17)}$$

$$H_{bc}C_{b,2m} = \frac{H_{ac}(C_{a,1e} - C_{a,e})}{1 - \exp[(1 - \Delta)n_2/(1 + R)]} + H_{ac}C_{a,e} \qquad \text{(A.18)}$$

Integrating Eqs. (A.15) and (A.16) from $y = 0$ to $y = \Delta B$ and from $y = \Delta B$ to B, Respectively, one obtains

$$\mathrm{H_{ac}C_{a,1m}} = \frac{\mathrm{H_{bc}(C_{b,2e} - C_{b,e})}}{1 - E\mathrm{xp}(-\mathrm{n}_1\ell\Delta)} + \mathrm{H_{bc}C_{b,e}} \qquad \text{(A.19)}$$

$$\mathrm{H_{ac}C_{a,2m}} = \frac{\mathrm{H_{bc}(C_{b,i} - C_{b,2e})}}{1 - E\mathrm{xp}(\mathrm{n}_2\ell\Delta)} + \mathrm{H_{bc}C_{b,2e}} \qquad \text{(A.20)}$$

Integration of Eq. (A.13) from $\mathrm{x} = 0$ to x results in

$$H_{ac}C_{a,1} - H_{bc}C_{b,1m} = (H_{ac}C^0_{a,i}) - H_{bc}C_{b,1m}\exp\left[\frac{-n_1\Delta x}{L(1 + R)}\right] \qquad \text{(A.21)}$$

Integration of Eq. (A.21) from x = 0 to x = L and rearrangement results in

$$H_{ac}C_{a,1m} - H_{bc}C_{b,1m} = \frac{(1+R)}{n_1\Delta}\left(H_{ac}C_{a,i}^0 - H_{bc}C_{b,1m}\right)\left[1 - \exp\left(\frac{-n_1\Delta}{(1+R)}\right)\right] \tag{A.22}$$

Combination of Eqs. (A.17) and (A.22) to eliminate $(H_{ac}C_{a,i}^0 - H_{bc}C_{b,1m}) \times \left[1 - \exp\left(\frac{-n_1\Delta}{(1+R)}\right)\right]$ yields

$$H_{ac}C_{a,1m} - H_{bc}C_{b,1m} = \frac{(1+R)}{n_1\Delta}H_{ac}(C_{a,i}^0 - C_{a,1e}) \tag{A.23}$$

Consequently, the mean concentration difference in region 1 defined in Eq. (14) may be determined with the use of Eq. (A.23), i.e.

$$(\Delta C)_{1,m} = H_{ac}C_{a,1m} - H_{bc}C_{b,1m} \tag{A.24}$$

Similarly, the mean concentration difference in region 2 defined in Eq. (15) is

$$(\Delta C)_{2m} = H_{ac}C_{a,2m} - H_{bc}C_{b,2m} \tag{A.25}$$

$$= \frac{(1+R)}{n_2(1-\Delta)}H_{ac}\left(C_{a,1e} - C_{a,e}\right) \tag{A.26}$$

B Outlet Concentration

Substitution of Eqs. (A.17) and (A.19) into Eq. (A.23) to eliminate $C_{a,1m}$ and $C_{b,1m}$ coupled with the use of Eqs. (A.8)—(A.11) yields

$$\alpha\xi_{a,e} - \beta\xi_{a,1e} = -1 \tag{B.1}$$

By substituting Eq. (A.18) and (A.20) into Eq.(A.26) to eliminate $C_{a,2m}$ and $C_{b,2m}$ coupled with the with use of Eqs. (A.8)—(A.11), we have

$$\xi_{a,e} - \xi_{a,1e} = 1/\gamma \tag{B.2}$$

Solving Eqs. (B.1) and (B.2), we have Eq. (18) of the dimensionless outlet concentration of phase a

$$\xi_{a,e} = \frac{\beta+\gamma}{\gamma(\beta-\alpha)} \tag{18}$$

From Eq. (B.8) one obtains,

$$C_{a,i}^{0} = \frac{(H_{bc}/H_{bc})C_{b,i}\xi_{a,e} + C_{a,e}}{1 + R\xi_{a,e}} \tag{B.3}$$

Substitution of Eq. (B.3) into Eq. (16) results in Eq. (17) of the outlet concentration, $C_{a,e}$.