

# Application of Modified Correction-Factor Analysis to Solvent Extraction in Rectangular Membrane Modules with External Recycle

Ho-Ming Yeh\* and Ching-Chun Hsu

*Energy and Opto-Electronic Materials Research Center, Department of Chemical and Materials Engineering, Tamkang University, Tamsui, Taiwan 251, R.O.C.*

## Abstract

The expressions of mass transfer rate for membrane extraction through a rectangular module with external recycle have been derived under cocurrent-flow, countercurrent-flow and cross-flow operations based on the modified correction-factor analysis coupled with the mass balances. These expressions are explicit and the results can be readily calculated without using try-and-error method, which should be employed in the classical correction-factor analysis for designing heat and mass exchangers. For cross-flow operation, the correction factors are function of flow rate, mass-transfer area, distribution coefficient and overall mass-transfer coefficient, and some values of them are given graphically. Considerable improvement in mass transfer rate is achievable if the devices are operated with recycle, leading to increase the mass transfer coefficient.

**Key Words:** Membrane Extraction, Rectangular Module, Modified Correction Factor, Internal Recycle

## 1. Introduction

A conventional solvent extractor can operate efficiently only within a small flow range; large flow variation leads to flooding [1–3]. Further, there has to be a density difference between the aqueous and the organic streams. In addition to the limitations mentioned above, the new dispersion-free microporous membrane-based solvent extraction technique [4,5] also overcomes several other limitations of conventional liquid extraction, such as the need for intimate mixing to form emulsions of the two phases, the limitation on independent phase flow rate variation and inability to handle particulates. Recently, extensive studies on dispersion-free solvent extraction using microporous membranes have been carried out [6–10].

The mass transfer for membrane solvent extraction through rectangular equipments with and without recycle have been analyzed in the previous works [11,12]. The ordinary differential equations for solute concentra-

tion distributions in raffinate and extract phases for cocurrent and countercurrent flows were derived based on mass balance with the assumptions of uniform concentrations and velocities over the cross section of flow, and the concentration distributions as well as the outlet concentrations were then obtained by solving simultaneously the first-order equations. The theory of the membrane solvent extraction in cross-flow system is rather complicated since the concentrations over the cross sections of flow are nonuniform. Therefore, instead of ordinary differential equations, two partial differential equations were derived, and the solute concentration distributions as well as the outlet concentrations were obtained by solving these equations simultaneously even with the method of Laplace transform. These theories though are not difficult to be understood but the results are rather complicated and cumbersome to be applied.

Actually, the analysis of mass transfer in mass exchangers can be analogous to heat transfer in heat exchangers with the use of correction factor. However, this conventional correction-factor analysis results in the expressions for heat and mass-transfer rates in implicit

\*Corresponding author. E-mail: hmyeh@mail.tku.edu.tw

form [13], and the method of try-and-error must be employed for calculation. A modified correct-factor analysis for the device without recycle has been introduced in previous work [14], in which the expressions of mass transfer rate were derived in explicit form for convenient applications. It is the purpose of present study to investigate the effect of external recycle on membrane extraction in rectangular devices by modified correction-factor analysis. The order of magnitude of mass transfer rates in the devices of three flow types, as well as the improvements in performance by recycle operation, will be discussed.

### 2. Correction-Factor Analysis

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* [2].

The extraction rate  $W$  in any one of the recycle-type rectangular membrane modules, as shown in Figures 1–3 with length  $L$  and width  $B$ , may be expressed from the mass-transfer point of view by

$$W = KBL \times (\text{averaged concentration difference}) \quad (1)$$

where  $K$  denotes the overall mass-transfer coefficient, while the averaged concentration differences between the raffinate (fluid a) and extract (fluid b) phases are varied with the flow patterns and membrane systems. For convenience, we may take its logarithmic means,  $(\Delta C)_{lm,1}$  or  $(\Delta C)_{lm,2}$ , analogous to heat transfer in heat exchangers, as

$$W = KBLF_1(\Delta C)_{lm,1} \quad (2)$$

$$= KBLF_2(\Delta C)_{lm,2} \quad (3)$$

where

$$(\Delta C)_{lm,1} = \frac{(H_{ac}C_{a,e} - H_{bc}C_{b,e}) - (H_{ac}C_{a,i}^o - H_{bc}C_{b,i})}{\ln[(H_{ac}C_{a,e} - H_{bc}C_{b,e}) / (H_{ac}C_{a,i}^o - H_{bc}C_{b,i})]} \quad (4)$$

$$(\Delta C)_{lm,2} = \frac{(H_{ac}C_{a,i}^o - H_{bc}C_{b,e}) - (H_{ac}C_{a,e} - H_{bc}C_{b,i})}{\ln[(H_{ac}C_{a,i}^o - H_{bc}C_{b,e}) / (H_{ac}C_{a,e} - H_{bc}C_{b,i})]} \quad (5)$$

and  $F_1$  and  $F_2$  are the so-called correction factors, to be determined, while  $H_{ac}$  and  $H_{bc}$  are the distribution coefficients of solute at the phase interfaces. It is noted

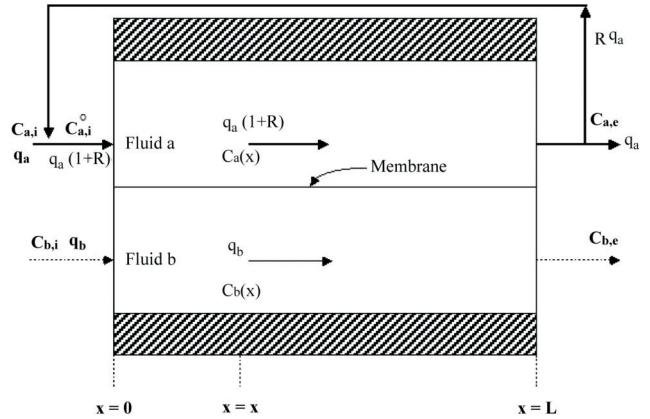


Figure 1. Schematic diagram of a cocurrent-flow membrane extractor with external recycle.

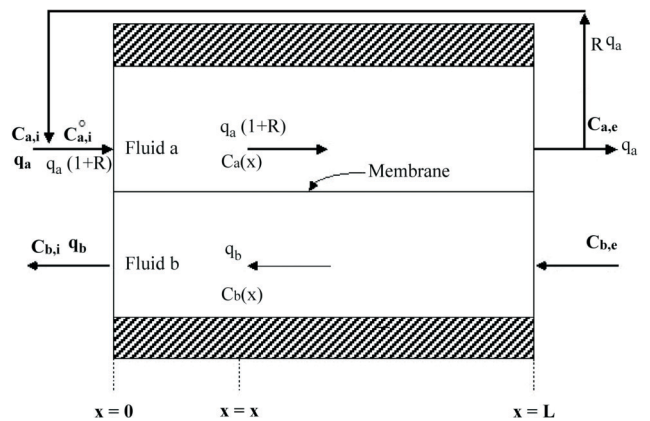


Figure 2. Schematic diagram of a countercurrent-flow membrane extractor with external recycle.

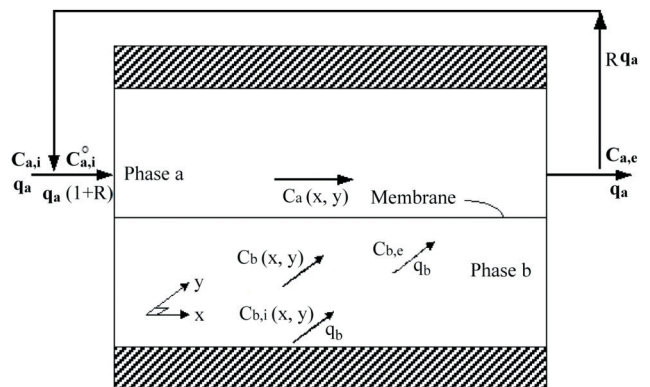


Figure 3. Schematic diagram of membrane solvent extraction in a cross-flow membrane extractor with external recycle.

that the overall mass-transfer coefficients in Eqs. (2) and (3) are assumed to be equal for mathematical simplification.

In addition to the mixed inlet concentration ( $C_{a,i}^0$ ), the outlet concentrations ( $C_{a,e}$  and  $C_{b,e}$ ) are also not specified prior. Therefore, the calculation of the overall mass-transfer rate  $W$  should be also conducted with the use of the try-and-error method, and the procedure is rather cumbersome [11,15].

### 3. Modified Correction-Factor Analysis

#### 3.1 Mass-Transfer Rate

The following is the overall mass balance:

$$W = q_a(1+R)(C_{a,i}^o - C_{a,e}) \quad (6)$$

$$= q_b(C_{b,e} - C_{b,i}) \quad (7)$$

where  $R$  is the reflux ratio while  $q_a$  and  $q_b$  are the volumetric flow rates in the raffinate and extract phases, respectively. Eqs. (6) and (7) can be rewritten as

$$C_{a,e} = C_{a,i}^o - W/[q_a(1+R)] \quad (8)$$

$$C_{b,e} = W/q_b + C_{b,i} \quad (9)$$

Substituting Eqs. (4), (8) and (9) into Eq. (2) to eliminate  $C_{a,e}$  and  $C_{b,e}$ , one has, after rearrangement

$$W = \frac{(H_{ac}C_{a,i}^o - H_{bc}C_{b,i})\{1 - \exp[-KBLF_1(H_{ac}/q_a(1+R) + H_{bc}/q_b)]\}}{(H_{ac}/q_a(1+R)) + (H_{bc}/q_b)} \quad (10)$$

Similarly, combination of Eqs. (2), (5), (8) and (9) yields

$$W = \frac{(H_{ac}C_{a,i}^o - H_{bc}C_{b,i})\{1 - \exp[KBLF_2(H_{ac}/q_a(1+R) - H_{bc}/q_b)]\}}{(H_{bc}/q_b) - \{H_{ac}/q_a(1+R)\}\exp[KBLF_2(H_{ac}/q_a(1+R) - H_{bc}/q_b)]} \quad (11)$$

#### 3.2 Outlet Concentrations

The mass-transfer equipment for membrane solvent extraction may be extremely different in design and construction, but in principle the only difference are those of relative direction of the two fluids, phases a and b. Accordingly, distinction is made between parallel flow and cross flow. The two fluids in parallel flow may be co-

current or countercurrent, while the direction of two fluids in cross flow may generally be different by 90 angular degrees.

Define the following dimensionless groups

$$Q_a = \frac{q_a(1+R)}{KBLH_{ac}} \quad (12)$$

$$Q_b = \frac{q_b}{KBLH_{bc}} \quad (13)$$

$$\zeta_a = \frac{H_{ac}C_a - H_{ac}C_{a,i}^o}{H_{ac}C_{a,i}^o - H_{bc}C_{b,i}} \quad (14)$$

#### 3.2.1 Cocurrent-Flow Operation

The schematic diagram shown in Figure 1 may serve to explain the nomenclature to be employed for the cocurrent-flow device with external recycle. The system consists of two channels for fluids a (raffinate phase) and b (extract phase), respectively, which are separated by a microporous membrane sheet through which the solute is extracted and transferred perpendicularly to its exposed surface. The mass balances for the solute in phases a and b of differential length  $dx$  are

$$-q_a(1+R)dC_a = KB(H_{ac}C_a - H_{bc}C_b)dx \quad (15)$$

$$-q_a(1+R)dC_a = q_b dC_b \quad (16)$$

Integrating Eq. (16) from  $x = 0$  ( $C_a = C_{a,i}^o$  and  $C_b = C_{b,i}$ ) to  $x = x$ , one obtains

$$-q_a(1+R)(C_a - C_{a,i}^o) = q_b(C_b - C_{b,i}) \quad (17)$$

Rearrangement of Eq. (15) yields

$$-d(H_{ac}C_a - H_{ac}C_{a,i}^o) = \frac{KBH_{ac}}{q_a(1+R)}[(H_{ac}C_a - H_{ac}C_{a,i}^o) - (H_{bc}C_b - H_{bc}C_{b,i}) + (H_{ac}C_{a,i}^o - H_{bc}C_{b,i})]dx \quad (18)$$

Substitution of Eq. (17) into Eq. (18) to eliminate the term  $(H_{bc}C_b - H_{bc}C_{b,i})$  and with the use of Eqs. (12)–(14) results in

$$-\frac{d\zeta_a}{d\xi} = \left(\frac{1}{Q_a}\right)[\{1 + (Q_a/Q_b)\}\zeta_a + 1] \quad (19)$$

where

$$\xi = x / L \tag{20}$$

Solving Eq. (19) with the use of the following boundary conditions:

$$\zeta_a = 0 \quad (C_a = C_{a,i}^o) \quad \text{at} \quad \xi = 0 \quad (x = 0) \tag{21}$$

$$\zeta_a = \zeta_{a,e} \quad (C_a = C_{a,e}) \quad \text{at} \quad \xi = 1 \quad (x = L) \tag{22}$$

where

$$\zeta_{a,e} = \frac{H_{ac} C_{a,e} - H_{ac} C_{a,i}^o}{H_{ac} C_{a,i}^o - H_{bc} C_{b,i}} \tag{23}$$

The result is

$$\zeta_{a,e} = [Q_a / (Q_a + Q_b)] [e^{-\{(1/Q_a) + (1/Q_b)\}} - 1] \tag{24}$$

or

$$\zeta_{a,e} = [e^{-(1+Q_a/Q_b)/Q_a} - 1] / [1 + (Q_a / Q_b)] \tag{25}$$

### 3.2.2 Countercurrent-Flow Operation

Figure 2, which shows a schematic diagram of the countercurrent-flow device with external recycle, is almost the same as Figure 1, except that in this figure the flow directions of the two fluids are opposite. The mass balances for the solute in phases a and b of differential length  $dx$  are

$$-q_a(1+R)dC_a = KB(H_{ac}C_a - H_{bc}C_b)dx \tag{26}$$

$$-q_a(1+R)dC_a = -q_b dC_b \tag{27}$$

Solving Eqs. (26) and (27) with the use of same boundary conditions, as indicated in Eqs. (21) and (22) and shown in Figure 2, and following the same procedure performed from Eq. (15) through Eq. (25), one has [11]

$$\zeta_{a,e} = \left[ \frac{e^{-\{(1/Q_a) - (1/Q_b)\}} - 1}{1 - (Q_b / Q_a) e^{-\{(1/Q_a) - (1/Q_b)\}}} \right] = \left[ \frac{e^{-(1-Q_a/Q_b)/Q_a} - 1}{1 - (Q_a / Q_b) e^{-(1-Q_a/Q_b)/Q_a}} \right] \tag{28}$$

### 3.2.3 Cross-Flow Operation

The schematic diagram for the cross-flow device with external recycle shown in Figure 3, is also almost the same as those shown in Figures 1 and 2, except that in

this figure, the flow directions of the two fluids, instead of parallel, cross each other. By taking the mass balances for membrane solvent extraction through a differential area,  $dxdy$ , in a cross-section mass exchanger, two partial differential equations for solute concentrations,  $C_a(x, y)$  and  $C_b(x, y)$ , are obtained

$$[KB / q_a(1+R)](H_{ac}C_a - H_{bc}C_b) = -(\partial C_a / \partial x) \tag{29}$$

$$(KL / q_b)(H_{ac}C_a - H_{bc}C_b) = (\partial C_b / \partial y) \tag{30}$$

The exact solution for outlet concentration can be analogous to that derived in the previous work [11] with  $q_a$  and  $C_{a,i}$  replaced  $q_a(1+R)$  and  $C_{a,i}^o$ , respectively.

The result is

$$\zeta_{a,e} = (-1/Q_a) \int_0^1 e^{-(1/Q_a)\xi} \int_0^1 e^{-(1/Q_b)\phi} I_0[2(1/Q_a)\sqrt{(Q_a/Q_b)\xi\phi}] d\phi d\xi = -(1/Q_a) \int_0^1 e^{-(1/Q_a)\xi} \int_0^1 e^{-(Q_a/Q_b)(1/Q_a)\phi} I_0[2(1/Q_a)\sqrt{(Q_a/Q_b)\xi\phi}] d\phi d\xi \tag{31}$$

in which  $I_0$  is the zeroth-order modified Bessel function [11].

### 3.3 Correction Factor

The expressions of correction factors may be rewritten from Eqs. (2) and (3) with the use of Eq. (6) as

$$F_1 = \frac{q_a(1+R)(C_{a,i}^o - C_{a,e})}{KBL(\Delta C)_{lm,1}} \tag{32}$$

$$F_2 = \frac{q_a(1+R)(C_{a,i}^o - C_{a,e})}{KBL(\Delta C)_{lm,2}} \tag{33}$$

With the use of the definitions of Eqs. (12), (13) and (23), as well as employing Eqs. (6) and (7) for replacing  $(C_{b,e} - C_{b,i})$  by  $(q_a(1+R) / q_b)(C_{a,i} - C_{a,e})$ , Eqs. (4) and (5) may be rewritten as

$$(\Delta C)_{lm,1} = \frac{(H_{ac}C_{a,i}^o - H_{bc}C_{b,i})\{(Q_a / Q_b) + 1\}\zeta_{a,e}}{\ln[1 + (Q_a / Q_b)\zeta_{a,e} + 1]} \tag{34}$$

$$(\Delta C)_{lm,2} = \frac{(H_{ac}C_{a,i}^o - H_{bc}C_{b,i})\{(Q_a / Q_b) - 1\}\zeta_{a,e}}{\ln \frac{1 + (Q_a / Q_b)\zeta_{a,e}}{1 + \zeta_{a,e}}} \tag{35}$$

Substitution of Eqs. (34) and (35) into Eqs. (32) and (33), respectively, with the use of Eqs. (6), (12) and (23)

yields

$$F_1 = -\frac{\ln[1 + \{1 + (Q_a / Q_b)\} \zeta_{a,e}]}{[1 + (Q_a / Q_b)] / Q_a} \quad (36)$$

$$F_2 = \frac{\ln[\{1 + (Q_a / Q_b)\} \zeta_{a,e} / (1 + \zeta_{a,e})]}{(1 - Q_a / Q_b) / Q_a} \quad (37)$$

### 3.3.1 Cocurrent Flow

Substitution of Eq. (25) into Eqs. (36) and (37) to eliminate  $\zeta_{a,e}$  gives the correction factors as

$$F_1 = 1 \quad (38)$$

$$F_2 = \{Q_a / (1 - Q_a / Q_b)\} \times \ln \left[ \left\{ 1 + \left[ \frac{Q_a / Q_b}{(Q_a / Q_b) + 1} \right] \left[ e^{-(1 + Q_a / Q_b) / Q_a} - 1 \right] \right\} / \left\{ 1 + \left[ \frac{1}{(Q_a / Q_b) + 1} \right] \left[ e^{-(1 + Q_a / Q_b) / Q_a} - 1 \right] \right\} \right] \quad (39)$$

It is shown that the first logarithmic mean of concentration difference  $(\Delta C)_{lm,1}$  defined by Eq. (4) is exactly the averaged concentration difference for cocurrent-flow operation, which is verified by Eq. (38) with  $F_1 = 1$ .

### 3.3.2 Countercurrent Flow

Substitution of Eq. (28) into Eqs. (36) and (37) yields

$$F_1 = -[Q_a / (1 + Q_a / Q_b)] \ln[1 + \{1 + (Q_a / Q_b)\} \{e^{-(1 - Q_a / Q_b) / Q_a} - 1\} / \{1 - (Q_a / Q_b)e^{-(1 - Q_a / Q_b) / Q_a}\}] \quad (40)$$

$$F_2 = 1 \quad (41)$$

It is noted that the second logarithmic mean of concentration difference  $(\Delta C)_{lm,2}$  defined by Eq. (5) is exactly the averaged concentration difference for countercurrent-flow operation, which is verified by Eq. (41) with  $F_2 = 1$ .

### 3.3.3 Cross Flow

Substitution of Eq. (31) into Eqs. (36) and (37) results in two correction factors of integration form. The calculations though are rather cumbersome and the expressions are somewhat longer and complicated, the results are presented graphically in Figures 4 and 5 for convenient use.

### 3.4 Mass Transfer Rate with Inlet Concentration in Recycled Device

Inspection of Eqs. (10) and (11) shows that the mixed inlet concentration  $C_{a,i}^0$  due to the recycle is not

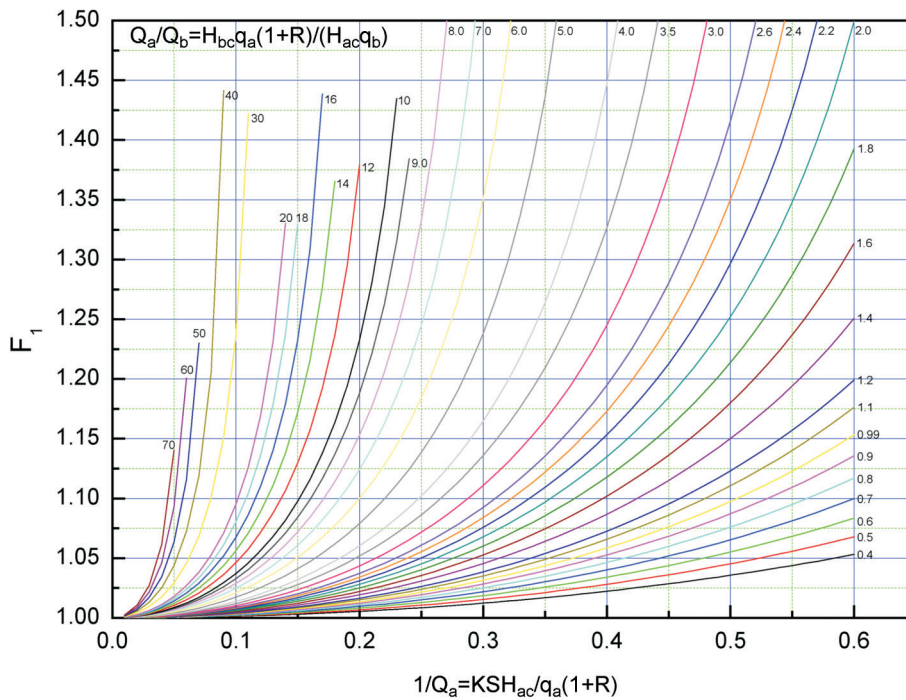


Figure 4. Modified correction factor  $F_1$  vs.  $1/Q_a$  for cross-flow operation.

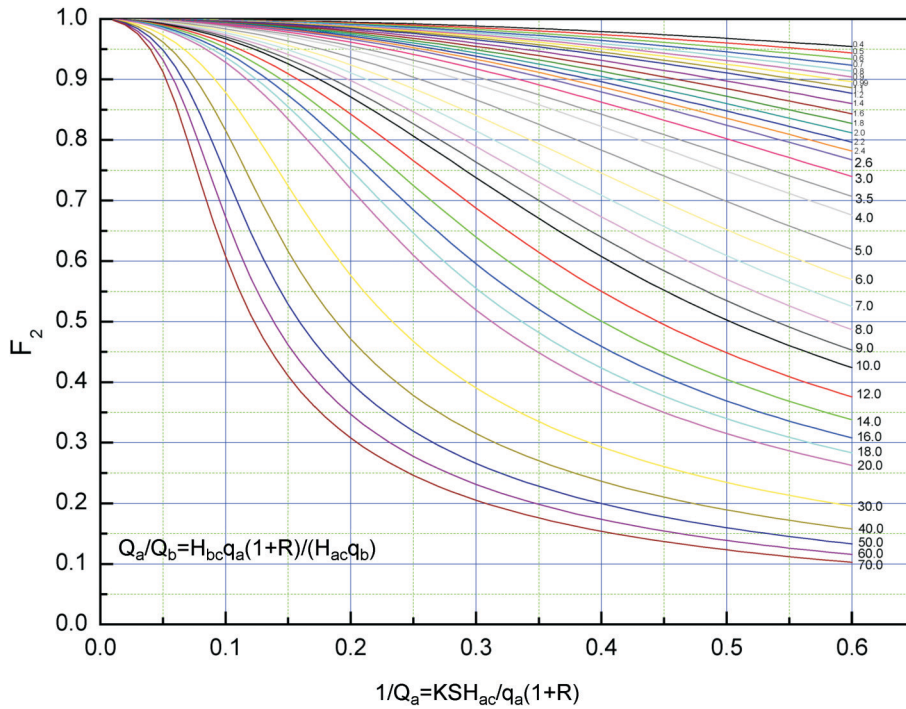


Figure 5. Modified correction factor  $F_2$  vs.  $1/Q_a$  for cross-flow operation.

specified a priori. Mathematically, one more relation for the mixing effect at the inlet is needed for determination of this value. For this reason, let us take a mass balance at the inlet with the use of Eq. (8):

$$(1 + R)C_{a,i}^o = C_{a,i} + RC_{a,e} = C_{a,i} + RC_{a,i}^o - WR/q_a(1 + R) \quad (42)$$

or

$$C_{a,i}^o = C_{a,i} - RW/q_a(1 + R) \quad (43)$$

Substitution of Eq. (43) into Eqs. (10) and (11) to eliminate  $C_{a,i}^o$  results in, respectively

$$W = \frac{(H_{ac}C_{a,i} - H_{bc}C_{b,i})\{1 - \exp[-KBLF_1\{(H_{ac}/q_a)(1 + R) + (H_{bc}/q_b)\}]\}}{H_{ac}/q_a(1 + R) + H_{bc}/q_b + [H_{ac}R/q_a(1 + R)]\{1 - \exp[-KBLF_1\{(H_{ac}/q_a)(1 + R) + (H_{bc}/q_b)\}]\}} \quad (44)$$

$$W = \frac{(H_{ac}C_{a,i} - H_{bc}C_{b,i})\{1 - \exp[KBLF_2\{(H_{ac}/q_a)(1 + R) - (H_{bc}/q_b)\}]\}}{(H_{bc}/q_b) + H_{ac}R/q_a(1 + R) - (H_{ac}/q_a)\exp[KBLF_2\{(H_{ac}/q_a)(1 + R) - (H_{bc}/q_b)\}]} \quad (45)$$

### 3.5 Mass Transfer Rate without Recycle

The equations for estimating the mass transfer rate

without recycle  $W_0$ , may be obtained either from Eqs. (10) and (11) by replacing  $C_{a,i}^o$  and  $R$  with  $C_{a,i}$  and zero, respectively, or from Eqs. (44) and (45) by merely setting  $R = 0$ :

$$W_0 = \frac{(H_{ac}C_{a,i} - H_{bc}C_{b,i})\{1 - \exp[-KBLF_1\{(H_{ac}/q_a) + (H_{bc}/q_b)\}]\}}{(H_{ac}/q_a) + (H_{bc}/q_b)} \quad (46)$$

$$W_0 = \frac{(H_{ac}C_{a,i} - H_{bc}C_{b,i})\{1 - \exp[-KBLF_2\{(H_{ac}/q_a) - (H_{bc}/q_b)\}]\}}{(H_{bc}/q_b) - (H_{bc}/q_a)\exp[KBLF_2\{(H_{ac}/q_a) - (H_{bc}/q_b)\}]} \quad (47)$$

in which  $F_1$  and  $F_2$  are determined also with  $R = 0$ . Actually, Eqs. (46) and (47) were already derived in the previous work [14].

## 4. Results and Discussion

### 4.1 Use of Correction Factors

Since  $F_1 = 1$  for cocurrent-flow operation and  $F_2 = 1$  for countercurrent-flow operation, it is concluded that Eqs. (44) and (45) are properly applicable to the cocurrent-flow and countercurrent-flow devices with recycle, respectively, with  $F_1$  and  $F_2$  just taken as unity,

while either Eq. (44) or (45) may be employed for the design of a cross-flow device with recycle. On the other words, the logarithmic means of concentration differences defined in Eqs. (4) and (5) are exactly the real average concentration difference for cocurrent-flow and countercurrent-flow operations, respectively. For practical applications, some values of correction factors for cross-flow operation were calculated and plotted in Figures 4 and 5 for convenient use. For the devices without recycle, Eqs. (44) and (45) are replaced by Eqs. (46) and (47), respectively, with  $Q_a$  in  $F_1$  and  $F_2$  reduced from Eq. (12) to  $KBLH_{ac} / q_a$  by setting  $R = 0$ .

#### 4.2 Mass-Transfer Efficiency

The mass-transfer efficiency in the rectangular membrane extractors with external recycle may be defined as

$$\eta_{eff} = \frac{W}{KBL(H_{ac}C_{a,i}^o - H_{bc}H_{b,i})} \quad (48)$$

Substitution of Eqs. (10) and (11) into Eq. (48) with the use of Eqs. (12) and (13) yields, respectively

$$\eta_{eff} = \frac{1 - \exp[-F_1(1 + Q_a/Q_b)/Q_a]}{(1 + Q_a/Q_b)/Q_a} \quad (49)$$

$$\begin{aligned} \eta_{eff} &= \frac{1 - \exp[F_2(1 - Q_a/Q_b)/Q_a]}{[(Q_a/Q_b) - \exp\{F_2(1 - Q_a/Q_b)/Q_a\}]/Q_a} \\ &= \frac{Q_a}{1 - (Q_a/Q_b)} + (Q_a/Q_b) \quad (50) \\ &= \frac{Q_a}{1 - \exp[-F_2(1 - Q_a/Q_b)/Q_a]} \end{aligned}$$

For the device without recycle,  $Q_a$  in Eqs. (49) and (50) should be reduced from Eq. (12) to  $KBLH_{ac} / q_c$  by setting  $R = 0$ .

#### 4.3 Effect of Flow Type on Performance

Since  $F_1 = 1$  for cocurrent-flow operation while  $F_1 > 1$  for cross-flow operation, as show in Figure 4, it is noted from Eq. (49) that the performance in a cross-flow device overcomes that in a cocurrent-flow one of same size, especially for larger value of  $Q_a / Q_b$ . On the other hand, since  $F_2 = 1$  for countercurrent-flow operation while  $F_2 < 1$  for cross-flow operation, as shown in Figure 5, and since  $[1 - (Q_a / Q_b)] / [1 - \exp\{-F_2(1 - Q_a / Q_b) / Q_a\}]$  decreases when  $F_2$  increases whatever  $(1 - Q_a / Q_b)$  is positive or negative, Eq. (50) indicates that the performance in a countercurrent-flow device is better than that

in a cross-flow one, especially also for higher value of  $(Q_a / Q_b)$ . Accordingly, we may conclude that the order of magnitude of extraction efficiencies obtained in the device of three flow types either with or without external recycle, is

Countercurrent flow > cross flow > cocurrent flow (51)

#### 4.4 Theoretical Predictions

The modified correction-factor analysis of mass transfer for membrane extraction in cocurrent-flow and countercurrent-flow devices are easy to follow with  $F_1 = F_2 = 1$ , while that for cross-flow operation is rather complicated and the results should be checked by experiment. The theoretical predictions of outlet concentration in the cross-flow devices without recycle were already confirmed well [14] with Chen's experimental data [16]. The experiment was carried out with the use of a membrane sheet ( $L = B = 16.5$  cm) made of microporous polypropylene as a permeable barrier to extract acetic acid from aqueous solution by methyl isobutyl ketone ( $H_{ac} = 0.524$  at  $25^\circ\text{C}$  and  $H_{bc} = 1$ ) in the flow channel of height 0.19 cm.

The theoretical predictions for the mass transfer rates in the external-recycle devices has been calculated from the appropriate equations with the use of above numerical values as well as the following correlation equations for the overall mass-transfer coefficients [14,16,17]:

$$\begin{aligned} K \times 10^4 \text{ (cm/s)} &= 7.256v_a^{0.14}v_b^{0.02}, \text{ for} \\ C_{a,i} &= 5 \times 10^{-4} \text{ mol/cm}^3, \text{ and} \quad (52) \end{aligned}$$

$$\begin{aligned} K \times 10^4 \text{ (cm/s)} &= 7.256v_a^{0.124}v_b^{0.02}, \text{ for } C_{a,i} = 2 \times 10^{-3} \text{ mol/cm}^3 \\ & \quad (53) \end{aligned}$$

where the velocities were calculated by

$$v_a = \frac{q_a(1+R)}{hB} \quad (54)$$

and

$$v_b = \frac{q_b}{hB} \text{ or } v_b = \frac{q_b}{hL} \quad (55)$$

for parallel-flow or cross-flow operation. Some prediction results are presented in Tables 1–3 for cocurrent, countercurrent and cross flows, respectively.

**Table 1.** Improvement of performance by recycle in cocurrent-flow module for  $C_{a,i} = 5 \times 10^{-4}$  mol/cm<sup>3</sup>,  $C_{b,i} = 0$  and  $q_b = 0.8$  cm<sup>3</sup>/s

$q_a$ cm <sup>3</sup> /s	$W_0 \times 10^5$ mol/s	R = 1		R = 3		R = 5		R = 10	
		$W \times 10^5$ mol/s	I (%)	$W \times 10^5$ mol/s	I (%)	$W \times 10^5$ mol/s	I (%)	$W \times 10^5$ mol/s	I (%)
0.1	2.1669	2.1178	-2.27	2.1415	-1.17	2.1746	0.36	2.2399	3.37
0.2	2.6868	2.7250	1.42	2.8266	5.20	2.9039	8.08	3.0356	12.98
0.4	3.1549	3.2918	4.34	3.4802	10.31	3.6068	14.32	3.8125	20.84
0.8	3.5870	3.8119	6.27	4.0798	13.74	4.2519	18.54	4.5266	26.19
1.6	4.0026	4.2991	7.41	4.6335	15.76	4.8442	21.02	5.1775	29.35
3.2	4.4178	4.7717	8.01	5.1610	16.82	5.4036	22.32	5.7858	30.97
6.4	4.8439	5.2456	8.29	5.6813	17.29	5.9511	22.86	6.3744	31.60

**Table 2.** Improvement of performance by recycle in countercurrent-flow module for  $C_{a,i} = 5 \times 10^{-4}$  mol/cm<sup>3</sup>,  $C_{b,i} = 0$  and  $q_b = 0.8$  cm<sup>3</sup>/s

$q_a$ cm <sup>3</sup> /s	$W_0 \times 10^5$ mol/s	R = 1		R = 3		R = 5		R = 10	
		$W \times 10^5$ mol/s	I (%)	$W \times 10^5$ mol/s	I (%)	$W \times 10^5$ mol/s	I (%)	$W \times 10^5$ mol/s	I (%)
0.1	2.2137	2.1424	-3.22	2.1553	-2.64	2.1846	-1.31	2.2462	1.47
0.2	2.7265	2.7474	0.77	2.8398	4.16	2.9138	6.87	3.0420	11.57
0.4	3.1850	3.3098	3.92	3.4913	9.62	3.6151	13.51	3.8180	19.88
0.8	3.6084	3.8252	6.01	4.0882	13.30	4.2583	18.01	4.5309	25.57
1.6	4.0172	4.3083	7.25	4.6394	15.49	4.8487	20.70	5.1806	28.96
3.2	4.4275	4.7780	7.92	5.1650	16.66	5.4068	22.12	6.7879	30.72
6.4	4.8504	5.2498	8.23	5.6840	17.19	5.9532	22.74	6.3759	31.45

**Table 3.** Improvement of performance by recycle in cross-flow module for  $C_{a,i} = 5 \times 10^{-4}$  mol/cm<sup>3</sup>,  $C_{b,i} = 0$  and  $q_b = 0.8$  cm<sup>3</sup>/s

$q_a$ cm <sup>3</sup> /s	$W_0 \times 10^5$ mol/s	R = 1		R = 3		R = 5		R = 10	
		$W \times 10^5$ mol/s	I (%)	$W \times 10^5$ mol/s	I (%)	$W \times 10^5$ mol/s	I (%)	$W \times 10^5$ mol/s	I (%)
0.1	2.1899	2.1300	-2.737	2.1483	-1.898	2.1796	-0.472	2.2431	2.428
0.2	2.7065	2.7362	1.098	2.8332	4.682	2.9088	7.478	3.0388	12.279
0.4	3.1698	3.3008	4.131	3.4858	9.967	3.6110	13.916	3.8152	20.361
0.8	3.5976	3.8185	6.139	4.0840	13.518	4.2551	18.274	4.5287	25.881
1.6	4.0099	4.3037	7.326	4.6365	15.625	4.8464	20.861	5.1791	29.157
3.2	4.4226	4.7748	7.963	5.1630	16.740	5.4052	22.217	5.7868	30.845
6.4	4.8472	5.2477	8.263	5.6826	17.236	5.9522	22.797	6.3751	31.523

#### 4.5 Effect of Reflux on Performance

The improvement in performance by recycle operation is best calculating the percentage increase in membrane extraction based on the device without recycle, i.e.

$$I = \frac{W - W_0}{W_0} \quad (56)$$

in which  $W_0$  is calculated from Eqs. (46) and (47), while

$W$  is calculated from Eqs. (44) and (45). With the use of the numerical values given in section 3.4, some results of  $W_0$ ,  $W$  and  $I$  are calculated and listed in Tables 1–3. It is shown in these tables that recycle operation really improves the performance, and the improvement increases with the reflux ratio. It must be mentioned that the order of magnitude of improvements  $I$  is opposite to that of extraction rates  $W$  shown in Eq. (51). This is be-



cause that the cocurrent-flow operation though yields the lower mass-transfer rate, but has larger room to be improved.

Actually, the recycle operation has two conflict effects on performance. The application of recycle operation on mass transfer not only creates the desirable effect of increase of fluid velocity, leading to enhance overall mass-transfer coefficient, but also lower the concentration difference (mass-transfer driving force) due to the undesirable effects of remixing and dilution of inlet fluid. At small inlet volume rate  $q_a$ , the fluid velocity,  $v_a = q_a(1 + R) / hB$ , is small, therefore, the production of increase in fluid velocity created by applying the recycle with reflux ratio  $R$  which is not large enough, may not compensate for the situation that the driving force of mass transfer in the mass exchangers decreases. Thus, the mass transfer rate  $W$  in the recycle devices of small  $q_a$  with low  $R$  cannot be over that  $W_0$  in the devices of the same size but without recycle. Taking a critical case of the given numerical values for example, if a counterflow membrane extractor is operated (see Table 1) with  $q_a = 0.1 \text{ cm}^3 / \text{s}$ ,  $q_b = 0.8 \text{ cm}^3 / \text{s}$ ,  $C_{a,i} = 5 \times 10^{-4} \text{ mol} / \text{cm}^3$  and  $C_{b,i} = 0$ , the performances obtained are  $W_0 = 2.1669 \text{ mol} / \text{s}$  for  $R = 0$  and  $W = 2.1178 \text{ mol} / \text{s}$  for  $R = 1$ , resulting in minus sign of improvement,  $I = -2.27\%$ . However, the introduction of reflux still has positive effects on membrane extraction for larger inlet volume rate. This is due to the increase of fluid velocity having more influence here than decrease of concentration difference, and the performance in a recycled device is over that in a device of the same size without recycle. It is seen in the tables that more than 30% of improvement  $I$  is achieved in the present study of interest.

## 5. Conclusion

The expressions, Eqs. (44) and (45), for calculating the mass-transfer rates in rectangular membrane extractors with external recycle were derived, analogous to heat transfer in heat exchangers with the use of modified correction-factor analysis. Eq. (44) is properly applicable to the cocurrent-flow operation with  $F_1 = 1$  while for countercurrent-flow operation, Eq. (45) with  $F_2 = 1$  is suitable to be used. When operating with cross-flow, either Eq. (44) or (45) is applicable with  $F_1$  and  $F_2$  given in Figures 4 and 5. Once the correction factors are determined with given system properties and apparatus and operating conditions, the mass-transfer rates,  $W$ , are readily

calculated from Eqs. (44) or (45). The expressions for calculating the mass transfer rates  $W_0$  in the devices without recycle are also presented in Eqs. (46) and (47), which are reduced from Eqs. (44) and (45) with  $R = 0$ .

It is found that application of the external recycle on rectangular membrane extractors really improves the performance, and the improvement in performance increases when the reflux ratio increases. The order of magnitude of mass transfer rates obtained in the devices of three flow types either with or without recycle is: countercurrent-flow operation > cross-flow operation > cocurrent-flow operation, while the order of magnitude of the improvements in performance is opposite. This is because that the device with lower mass transfer rate has larger room to be improved.

In the conventional correction-factor analysis, the equations of mass-transfer rate for mass transfer in mass exchangers, as shown in Eqs. (2) and (3), are implicit with all  $F_1$ ,  $F_2$ ,  $(\Delta C)_{lm,1}$ ,  $(\Delta C)_{lm,2}$  including the unknown outlet concentrations,  $C_{a,e}$  and  $C_{b,e}$ . The calculation is cumbersome because the methods of tri-and-error and even iteration should be employed. Unlike the conventional correction-factor analysis, the expressions of mass-transfer rate obtained by the present modified correction-factor analysis are explicit, instead of being implicit. Therefore, they are convenient to be used and the results are readily calculated. The modified correction-factor approach developed in this work can be extended for application to a hollow-fiber system, to a multipass membrane system, to other recycled membrane systems, or to different mass-transfer devices with or without chemical reaction. However, some of these extensions would require the use of additional mathematical techniques, and could be rather cumbersome.

## 6. List of Symbols

$B$	membrane width (m)
$C_a, C_b$	Solute concentration distribution in phase a, in phase b, respectively ( $\text{kg mol} / \text{m}^3$ )
$C_{a,i}, C_{b,i}$	$C_a, C_b$ , respectively, at the inlet ( $\text{kg mol} / \text{m}^3$ )
$C_{a,i}^0$	$C_{a,i}$ after mixing with recycling stream ( $\text{kg mol} / \text{m}^3$ )
$C_{a,e}, C_{b,e}$	$C_a, C_b$ , respectively, at the outlet ( $\text{kg mol} / \text{m}^3$ )
$(\Delta C)_{lm,1}, (\Delta C)_{lm,2}$	logarithmic-mean concentration dif-

	ference, defined by Eq. (4), by Eq. (5) (kg mol/m <sup>3</sup> )
$F_1, F_2$	correction factor defined by Eqs. (2) and (3) and given in Eqs. (36) and (37)
$H_{ac}, H_{bc}$	distribution coefficient between phase a and phase c, between phase b and phase c, respectively.
h	height of flow channel (m)
I	improvement in performance defined by Eq. (56)
K	overall mass-transfer coefficient (m/s)
L	length of the membrane sheet (m)
$Q_a, Q_b$	dimensionless group defined by Eq. (12), by Eq. (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 membrane extractor with recycle (kg mol/s)
W <sub>0</sub>	W without recycle (kg mol/s)
x, y	rectangular coordinates (m)

### 6.1 Greek Letters

$\zeta_a, \zeta_{a,e}$	dimensionless solute concentration difference defined by Eq. (14), by Eq. (23)
$\xi$	x/L
$\phi$	y/B
$\eta_{eff}$	extractor efficiency

### References

- [1] Treybal, R. E., "Liquid Extraction," 2nd ed., McGraw-hill, New York, Chapter 11 (1963).
- [2] Lo, T. C. and Baird, M. H. I., "Liquid-Liquid Extraction" in M. Grayson (Ed.), *Kirktothmer Encycloedia of Chemical Technology*, Vol. 9, 3rd ed., Wiley, New York (1980).
- [3] Porter, M. C., *Handbook of Industrial Membrane Technology*, Noyes Publications, New Jersey, pp. 1–3, 175 (1990).
- [4] Kiani, A., Bhawe, R. R. and Sirkar, K. K., "Solvent Extraction with Immobilized Interfaces in Microporous Membrane," *J. Membr. Sci.*, Vol. 20, p. 125 (1984).
- [5] Prasad, R., Kiani, A., Bhawe, R. R. and Sirkar, K. K., "Further Studies on Solvent Extraction with Immobilized Interfaces in a Microporous Hydrophobic Membrane," *J. Membr. Sci.*, Vol. 26, p. 76 (1986).
- [6] Yang, M. C. and Cussler, E. L., "Designing Hollow-Fiber Contractors," *AIChE J.*, Vol. 32, p. 1910 (1986).
- [7] Prasad, R. and Sirkar, K. K., "Dispersio-Free Solvent Extraction with Microporous Hollow-Fiber Modules," *AIChE J.*, Vol. 34, p. 177(1998).
- [8] Ding, H. and Cussler, E. L., "Fractional Extraction with Hollow Fibers with Hydrogelfilled Walls," *AIChE J.*, Vol. 37, p. 855 (1991).
- [9] Yeh, H. M., Peng, Y. Y. and Chen, Y. K., "Solvent Extraction through a Double-Pass Parallel-Plate Membrane Channel with Recycle," *J. Membr. Sci.*, Vol. 163, p. 177 (1999).
- [10] Yeh, M. H. and Chen, Y. K., "The Effect of Multipass Arrangement on the Performance in a Membrane Extraction of Fixed Configuration," *Chem. Eng. Sci.*, Vol. 55, p. 5873 (2001).
- [11] Yeh, H. M. and Hsu, Y. S., "Analysis of Membrane Extraction through Rectangular Mass Exchangers," *Chem. Eng. Sci.*, Vol. 54, p. 897 (1999).
- [12] Yeh, H. M., "Membrane Extraction in Rectangular Module with External Recycle," *J. Taiwan Inst. Chem. Engrs.*, Vol. 39, p. 679 (2008).
- [13] Bowman, R. A., Mueller, A. C. and Nagle, W. M., "Mean Temperature Difference in Design," *Trans. Am. Mech. Engrs.*, Vol. 62, p. 283 (1940).
- [14] Yeh, H. M., "Modified Correction-Factor Analysis of Solvent Extraction in Rectangular Membrane Modules," *J. Chinese Inst. Chem. Engrs.*, Vol. 38, p. 385 (2007).
- [15] Yeh, H. M. and Huang C. H., "Solvent Extraction in Multipass Parallel-Flow Mass Exchangers of Microporous Hollow-Fiber Modules," *J. Membr. Sci.*, Vol. 103, p. 135 (1995).
- [16] Chen, Y. K., "The Influence of Aspect Ratio on Separation Performance in Flat-Plate Cross-Flow Membrane Extractors," *Master Thesis*, Tamkang University, Tamsui, Taiwan (1999).
- [17] Yeh, H. M., "Effect of Reflux and Reflux-Barrier Location on Solvent Extraction through Cross-Flow Flat-Plate Membrane Modules with Internal Reflux," *J. Membr. Sci.*, Vol. 269, p. 133 (2006).

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