Separation Science and Technology
Publication details, including instructions for authors and subscription information:
http://www.informaworld.com/smpp/title~content=t713708471

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To cite this Article Shindo, Y. , Hakuta, T. , Yoshitome, H. and Inoue, H.(1985) 'Calculation Methods for Multicomponent Gas Separation by Permeation', Separation Science and Technology, 20: 5, 445 — 459
To link to this Article: DOI: 10.1080/01496398508060692
URL: http://dx.doi.org/10.1080/01496398508060692

SEPARATION SCIENCE AND TECHNOLOGY, **20**(5 & 6), pp. 445-459, 1985

Calculation Methods for Multicomponent Gas Separation by Permeation

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Abstract

Calculation methods for the single-stage permeation of a multicomponent gas mixture are presented for five flow patterns: cocurrent flow, countercurrent flow, cross flow, perfect mixing, and one-side mixing. The derivations are cast in a form suitable for computer calculation. The calculation methods presented are appropriate for systems with any number of components. Calculation results are shown for the separations of a NH_3, H_2, and N_2 gaseous mixture by means of a polyethylene membrane, and for a H_2, CH_4, CO, N_2, and CO_2 mixture through a microporous glass membrane.

INTRODUCTION

There are several models for a membrane module of gas permeation, and, depending on the flow patterns and operating conditions, these models are classified as cocurrent flow, countercurrent flow, cross flow, perfect mixing, and one-side mixing (*1,2*). Calculation methods for a binary system for these models have been developed by Kammermeyer et al. (*1, 3*) and other investigators (*2, 4-10*).

In membrane applications to actual gas separation systems, we often encounter a problem of calculation for the performance of a multi-

 0149-6395/85/2005-0445$3.50/0

component system (*11, 12*). There are, however, few reports regarding the calculation method for multicomponent gas separation by permeation. Calculation methods for ternary and quaternary systems with perfect mixing were developed by Brubaker and Kammermeyer (*13*). An iterative calculation method for multicomponent systems with perfect mixing has been described by Stern et al. (*14*). Pan and Habgood (*15*) developed a calculation method for a multicomponent mixture in the cross flow pattern. Sengupta and Sirkar (*16*) reported a numerical analysis of the separation of a ternary gas mixture by an asymmetric permeator.

The purpose of this paper is to develop calculation methods for a multicomponent system with the five kinds of flow patterns in a single permeation stage, and to show calculation results. Much higher selectivities are possible in permeation through membranes, and while cascades of a few stages or a continuous membrane column (*17–20*) may still be necessary for most practical separations, the calculations for and understanding of a single permeation stage are of prime importance.

ANALYTICAL STUDIES

Assumptions

Equations have been developed for each type of flow pattern. The assumptions utilized in this study are as follows:

(1) The rates of permeation obey Fick's law.

(2) The permeability of each gas component is the same as that of the pure gas, and is independent of pressure.

(3) The effective membrane thickness is constant along the length of the permeator.

(4) Concentration gradients in the permeation direction are negligible.

(5) Pressure drops of the feed and permeate gas streams are negligible.

(6) A plug flow situation exists in the feed and permeate streams, except in mixed flow and in the permeate streams in cross flow.

Cocurrent Flow

Cocurrent flow in a permeation stage is illustrated in Fig. 1. The overall material balance over the differential area dA is

$$-dF = dG \tag{1}$$

$$= dA \sum_{k=1}^{n} \frac{Q_k}{\delta}(P_h x_k - P_l y_k) \tag{2}$$

and the material balance for component i is

$$-d(x_i F) = d(y_i G) \tag{3}$$

$$= dA \frac{Q_i}{\delta}(P_h x_i - P_l y_i) \tag{4}$$

where F is the flow rate on the feed (high-pressure) stream; G is the flow rate on the permeate (low-pressure) stream parallel to the feed stream; n is the number of gaseous components; P_h and P_l are the pressures on the feed side and the permeate side, respectively; Q_i is the permeability of component i; and δ is the membrane thickness. x_i and y_i are the mole fractions of component i on the feed side and the permeate side, respectively, and there are the following conditions:

$$\sum_{k=1}^{n} x_k = 1 \tag{5}$$

$$\sum_{k=1}^{n} y_k = 1 \tag{6}$$

Solving for dx_i from Eq. (4), followed by substitution of Eq. (2), gives

$$dx_i = \frac{-dA}{F}\left[\frac{Q_i}{\delta}(P_h x_i - P_l y_i) - x_i \sum_{k=1}^{n} \frac{Q_k}{\delta}(P_h x_k - P_l y_k)\right] \tag{7}$$

Integration of Eqs. (1) and (3) from the inlet point to an arbitrary point yields

$$G = F_f - F \tag{8}$$

$$y_i = \frac{x_{fi}F_f - x_i F}{F_f - F}, \qquad G \neq 0 \tag{9}$$

where F_f is the feed flow rate at the inlet, and x_{fi} is the feed mole fraction of component i. It is considered that permeate flow rate G is zero at $A = 0$. The mole fraction y_i at $G = 0$ ($A = 0$) is obtained by a limiting process of the L'Hospital rule as $F \rightarrow F_f$.

$$y_i = \frac{\frac{Q_i}{\delta}(P_h x_i - P_l y_i)}{\sum_{k=1}^{n} \frac{Q_k}{\delta}(P_h x_k - P_l y_k)}, \qquad G = 0 \tag{10}$$

The mole fractions on the permeate stream at $G = 0$ ($A = 0$) can be obtained by solving the simultaneous equations, Eq. (10), for every component. The ratio of any two members of Eq. (10) becomes

$$\frac{y_i}{y_j} = \frac{Q_i(P_h x_i - P_l y_i)}{Q_j(P_h x_j - P_l y_j)} \tag{11}$$

Solving for y_j yields

$$y_j = \frac{x_j Q_j/Q_i}{P_l/P_h\{(Q_j/Q_i) - 1\} + (x_i/y_i)} \tag{12}$$

Substituting Eq. (12) into Eq. (6) gives

$$\sum_{k=1}^{n} \frac{x_k Q_k/Q_i}{P_l/P_h\{(Q_k/Q_i) - 1\} + (x_i/y_i)} = 1 \tag{13}$$

Equation (13) can be solved by Newton's iterative procedure. (See Appendix A.) The values of y for the other components can be calculated with the aid of Eq. (12).

We define the dimensionless variables as follows.

$$s = A\frac{Q_m P_h}{F_f \delta} \qquad s_t = A_t \frac{Q_m P_h}{F_f \delta} \tag{14}$$

$$f = F/F_f \qquad f_o = F_o/F_f \tag{15}$$

$$\theta = 1 - f_o \tag{16}$$

$$g = G/F_f \tag{17}$$

$$\gamma = P_l/P_h \tag{18}$$

$$q_i = Q_i/Q_m \tag{19}$$

where Q_m is the permeability of the base component, usually the most permeable component; F_0 is the flow rate of the reject stream; and θ is the stage cut. In terms of these dimensionless variables, the following governing equations for the cocurrent flow are obtained from Eqs. (2), (5) to (9), (12), and (13):

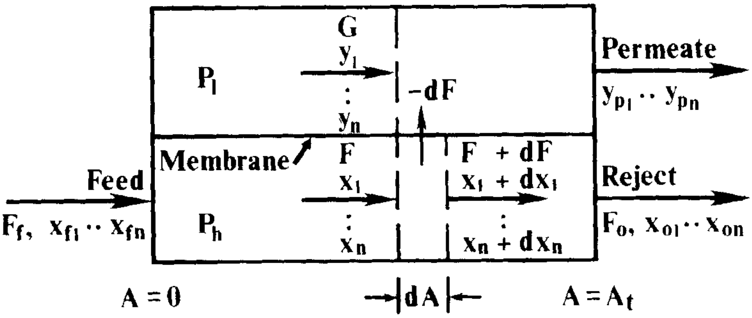

FIG. 1. Diagram of single permeation stage with cocurrent flow.

$$\frac{df}{ds} = -\sum_{k=1}^{n} q_k(x_k - \gamma y_k) \tag{20}$$

$$\frac{dx_i}{ds} = -\frac{q_i}{f}(x_i - \gamma y_i) + \frac{x_i}{f}\sum_{k=1}^{n} q_k(x_k - \gamma y_k) \quad (i = 1, \ldots, n-1) \tag{21}$$

$$x_n = 1 - \sum_{k=1}^{n-1} x_k \tag{22}$$

$$g = 1 - f \tag{23}$$

$$y_i = \frac{x_{if} - fx_i}{1 - f}, \quad g \neq 0 \ (i = 1, \ldots, n-1) \tag{24}$$

$$\sum_{k=1}^{n} \frac{x_k q_k / q_i}{\gamma\{(q_k/q_i) - 1\} + (x_i/y_i)} = 1, \quad g = 0 \tag{25}$$

$$y_j = \frac{x_j q_j / q_i}{\gamma\{(q_j/q_i) - 1\} + (x_i/y_i)}, \quad g = 0 \quad (j \neq i, n) \tag{26}$$

$$y_n = 1 - \sum_{k=1}^{n-1} y_k \tag{27}$$

The calculation for multicomponent gas separation through a membrane in the cocurrent flow pattern can be performed by the use of Eqs. (20) to (27). For example, we consider the problem of finding x_{oi}'s, y_{pi}'s, and θ for given x_{fi}'s, q_i's, γ, and s_t. x_{oi}'s and y_{pi}'s are the mole fractions of component i on the feed and permeate streams at the outlet, respectively. The values of x_{oi}'s, y_{pi}'s, and θ are obtained by the integration of the system of

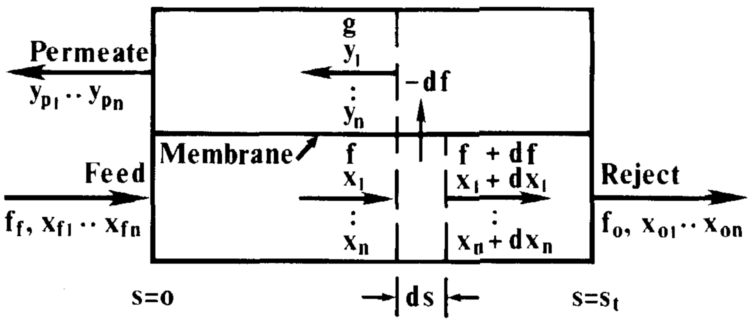

FIG. 2. Diagram of single permeation stage with countercurrent flow.

differential equations, Eqs. (20) and (21), in conjunction with Eqs. (22) to (27), with initial conditions

$$f = 1, \quad x_i = x_{fi} \quad (i = 1, \ldots, n - 1) \quad \text{at } s = 0$$

Countercurrent Flow

Consider a model as shown in Fig. 2. For such a model, one can derive a system of equations analogous to those for the cocurrent case. Note that the flow rate of the permeate stream, g, always possesses a negative value. Integrating Eqs. (1) and (3) from an arbitrary point to the outlet, and using dimensionless variables, yields the following equations:

$$g = f - (1 - \theta) \tag{28}$$

$$y_i = \frac{x_i f - x_{oi}(1 - \theta)}{f - (1 - \theta)}, \quad g \neq 0 \tag{29}$$

Equations (20) to (22) and (25) to (27) are still valid for countercurrent flow, and Eqs. (28) and (29) are used instead of Eqs. (23) and (24). For countercurrent flow, the integration is carried out backward. The solution procedure is to guess the values of x_{oi}'s $(i = 1, \ldots, n - 1)$ and θ, integrate from $s = s_t$ to $s = 0$, and check if the x_i's $(i = 1, \ldots, n - 1)$ and f at $s = 0$ are sufficiently close to x_{fi}'s $(i = 1, \ldots, n - 1)$ and 1, respectively. If they are not, new guesses have to be made at $s = s_t$ for x_{oi}'s $(i = 1, \ldots, n - 1)$ and θ, and the process repeated until the values of x_i's $(i = 1, \ldots, n - 1)$ and f obtained numerically are sufficiently close to the specified values of x_{fi}'s $(i = 1, \ldots, n - 1)$ and 1, respectively.

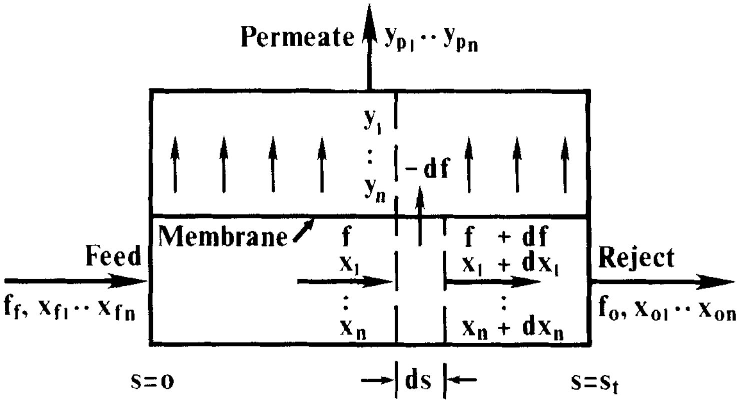

FIG. 3. Diagram of single permeation stage with cross flow.

Cross Flow

Permeation for the cross flow pattern is shown in Fig. 3. In this case, Eqs. (20) to (22) are valid. The permeate stream is in the direction vertical to the feed stream. Therefore, there is no parallel flow to the feed stream on the permeate side; i.e., $g = 0$ over all the membrane surface area. The mole fractions on the permeate side are given by the algebraic equations, Eqs. (25) to (27). Calculation for the cross flow can be performed in a manner similar to that for cocurrent flow by the use of Eqs. (20) to (22) and (25) to (27). The mass conservation yields

$$x_{fi} = x_{oi}(1 - \theta) + y_{pi}\theta \qquad (i = 1, \ldots, n) \tag{30}$$

The mole fractions of the permeate stream at the outlet are calculated by Eq. (30). Of course, Eq. (30) is valid for any flow pattern.

One-Side Mixing

A model for one-side mixing is shown in Fig. 4. In this case, Eqs. (20) to (22) are valid, and the mole fractions of the permeate phase are uniform. For one-side mixing the calculation procedure is to guess the values of y_{pi}'s $(i = 1, \ldots, n - 1)$ and integrate from $s = 0$ to $s = s_t$, and check if the values of x_{oi}'s, y_{pi}'s $(i = 1, \ldots, n - 1)$, and θ satisfy Eq. (30). The procedure is iterated until Eq. (30) is satisfied.

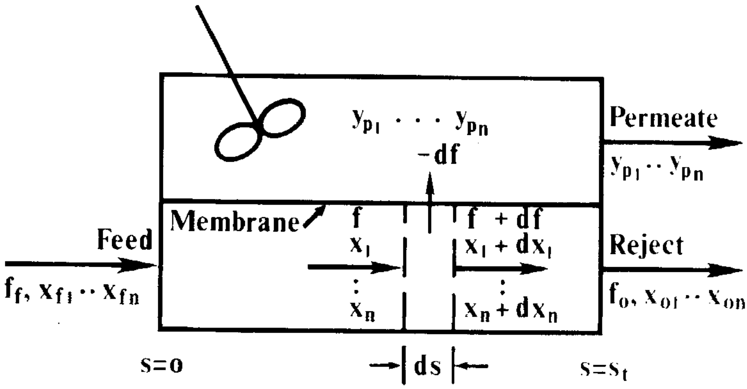

FIG. 4. Diagram of single permeation stage with one-side mixing.

Perfect Mixing

Figure 5 shows permeation for the perfect mixing case. The following relation between two components, similar to Eq. (11), is found to hold for the overall membrane surface area.

$$\frac{y_{pi}}{y_{pj}} = \frac{q_i(x_{oi} - \gamma y_{pi})}{q_j(x_{oj} - \gamma y_{pj})} \tag{31}$$

Eliminating x_{oi} and x_{oj} by the aid of Eq. (30), and solving for y_{pj} gives

$$y_{pj} = \frac{x_{fj}q_j/q_i}{(\gamma + \theta - \gamma\theta)\{(q_j/q_i) - 1\} + (x_{fi}/y_{pi})} \tag{32}$$

On substituting Eq. (32) into Eq. (6), one obtains

$$\sum_{k=1}^{n} \frac{x_{fk}q_k/q_i}{(\gamma + \theta - \gamma\theta)\{(q_k/q_i) - 1\} + (x_{fi}/y_{pi})} = 1 \tag{33}$$

Equation (33) expresses y_{pi} in terms of x_{fi}'s, and it can be solved by Newton's iterative procedure. (See Appendix A.) The values of y_p for the other components can be calculated by Eq. (32). The material balance for the overall membrane surface area leads to

$$\theta = s_t \sum_{k=1}^{n} q_k(x_{ok} - \gamma y_{pk}) \tag{34}$$

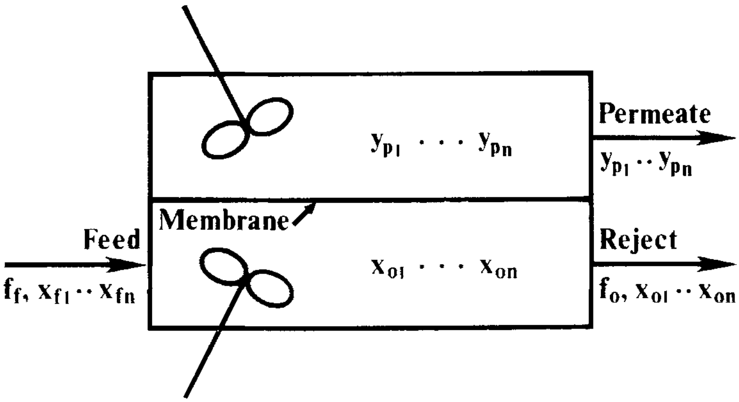

FIG. 5. Diagram of single permeation stage with perfect mixing.

The solution strategy for perfect mixing is to guess of value of θ, then calculate y_{pi}'s by Eqs. (33) and (32), and x_{oi}'s by Eq. (30), and check if the x_{oi}'s and y_{pi}'s satisfy Eq. (34). If they don't, a new guess has to be made, and the procedure is repeated until Eq. (34) is satisfied.

CALCULATION RESULTS

A membrane module in every case is characterized by the variables: x_{fi}'s, q_i's, γ, s_t, x_{oi}'s, y_{pi}'s, and θ. Therefore, many situations can be considered. We treat two problems here.

Problem 1. To find x_{oi}'s, y_{pi}'s, and θ for given x_{fi}'s, q_i's, γ, and s_t.
Problem 2. To find x_{oi}'s, y_{pi}'s, and s_t for given x_{fi}'s, q_i's, γ, and θ.

The calculation methods shown above are used for Problem 1. The system of equations required for the calculation of Problems 2 can be obtained by rearrangement of those used for Problem 1. The calculation methods for Problem 2 are shown in Appendix B.

Calculations were performed for the separation of a NH_3, H_2, and N_2 mixture by permeation through a polyethylene membrane. The permeabilities of gases through a polyethylene membrane were determined by Brubaker and Kammermeyer (*13*). The permeability data used are shown in Table 1. The calculation conditions and the numerical results are shown in Tables 2 and 3. Other calculations were made for the separation of a H_2, CH_4, CO, N_2, and CO_2 mixture through a microporous

TABLE 1
Permeability of Gases Through a Polyethylene Membrane at 50°C

No.	Gas	Q (mol/s · m · Pa)	q (-)
1	NH_3[a]	36.9×10^{-15}	1.000
2	H_2	11.7×10^{-15}	0.317
3	N_2	2.41×10^{-15}	0.065

[a]Base component.

TABLE 2
Calculation Results of Gas Permeation through a Polyethylene Membrane[a]

	Permeate mole fraction, y_p (-)			
	NH_3	H_2	N_2	θ (-)
Countercurrent flow	0.7368	0.2010	0.0622	0.3745
Cross flow	0.7338	0.2035	0.0627	0.3726
One-side mixing	0.7325	0.2046	0.0629	0.3717
Cocurrent flow	0.7300	0.2067	0.0632	0.3702
Perfect mixing	0.6991	0.2226	0.0783	0.3345

[a]Calculation conditions: $\gamma = 0.13$, $s_t = 1.0$, q as shown in Table 1. Feed compositions: $NH_3 = 0.45$, $H_2 = 0.25$, $N_2 = 0.30$.

TABLE 3
Calculation Results of Gas Permeation through a Polyethylene Membrane[a]

	Permeate mole fraction, y_p (-)			
	NH_3	H_2	N_2	s_t (-)
Countercurrent flow	0.7054	0.2200	0.0746	1.4603
Cross flow	0.7003	0.2241	0.0756	1.4740
One-side mixing	0.6961	0.2273	0.0766	1.4864
Cocurrent flow	0.6923	0.2303	0.0774	1.4963
Perfect mixing	0.6393	0.2492	0.1115	1.8000

[a]Calculation conditions: $\gamma = 0.13$, $\theta = 0.5$, q as shown in Table 1. Feed compositions: $NH_3 = 0.45$, $H_2 = 0.25$, $N_2 = 0.30$.

glass membrane. The permeabilities of gases for a microporous glass membrane were measured in the same way as previously reported (*21, 22*), and the permeability data are indicated in Table 4. The calculation conditions and the numerical results are shown in Tables 5 and 6. Calculations were performed for the five kinds of flow pattern. The Runge-Kutta-Gill numerical method was used to integrate the differential equations. A trial-and-error procedure was necessary for countercurrent flow, perfect mixing in Problem 1, and one-side mixing patterns. Powell's nonlinear optimization method was used for the trial-and-error procedure. For perfect mixing, the initial estimates were generated on the basis of cocurrent flow, while for countercurrent flow and one-side mixing they were based on cross flow. The calculation was made in double precision by means of a FACOM-M-380 System at the Tsukuba Research Center of the Agency of Industrial Science and Technology (AIST Japan).

DISCUSSION

Oishi et al. (*2*) showed by numerical calculation and theoretical analysis that the countercurrent flow was the best among the five flow patterns for binary systems. Blaisdell and Kammermeyer (1, 3) compared the five models for O_2–N_2 separation by a silicone-rubber membrane in terms of numerical calculation, and they also showed that countercurrent flow was the most advantageous. Countercurrent flow also shows the best performance in the present calculations. Separations in the countercurrent flow mode show the highest composition of the most permeable component, the largest cut in Tables 2 and 5, and the least membrane area in Tables 3 and 6.

Stern et al. (*14*) described a calculation method for perfect mixing in Problem 2 by a trial-and-error procedure. The use of the present method, as shown in Appendix B, permits direct solution by numerical calculation. Pan and Habgood (*15*) developed a calculation method for cross flow that differs from the present method. Their derivations for calculation are more complicated than those presented in this study, and they showed concrete solutions for only binary and ternary systems. The calculation methods developed in this study can be used for systems consisting of any number of components. The present methods are limited only by computer time.

SUMMARY

Calculation methods for multicomponent gas separation by a single permeation stage have been developed for the five kinds of flow pattern:

TABLE 4
Permeability of Gases through a Microporous Glass Membrane at 25°C

No.	Gas	Q (mol/s · m · Pa)	q (-)
1	H_2[a]	48.0×10^{-12}	1.000
2	CH_4	19.1×10^{-12}	0.398
3	CO	14.0×10^{-12}	0.292
4	N_2	13.8×10^{-12}	0.288
5	CO_2	14.8×10^{-12}	0.308

[a]Base component.

TABLE 5
Calculation Results of Gas Permeation through a Microporous Glass Membrane[a]

	Permeate mole fraction, y_p (-)					
	H_2	CH_4	CO	N_2	CO_2	θ (-)
Countercurrent flow	0.4742	0.0905	0.1793	0.1065	0.1495	0.4146
Cross flow	0.4707	0.0910	0.1806	0.1072	0.1505	0.4131
One-side mixing	0.4692	0.0913	0.1811	0.1075	0.1510	0.4125
Cocurrent flow	0.4662	0.0917	0.1821	0.1081	0.1518	0.4112
Perfect mixing	0.4310	0.0945	0.1959	0.1165	0.1621	0.3964

[a]Calculation conditions: $\gamma = 0.1$, $s_t = 1.0$, q as shown in Table 4. Feed composition: $H_2 = 0.3$, $CH_4 = 0.1$, $CO = 0.25$, $N_2 = 0.15$, $CO_2 = 0.2$.

TABLE 6
Calculation Results of Gas Permeation through a Microporous Glass Membrane[a]

	Permeate mole fraction, y_p (-)					
	H_2	CH_4	CO	N_2	CO_2	s_t (-)
Countercurrent flow	0.4544	0.0927	0.1867	0.1109	0.1553	1.231
Cross flow	0.4502	0.0933	0.1882	0.1118	0.1565	1.236
One-side mixing	0.4475	0.0937	0.1891	0.1123	0.1573	1.240
Cocurrent flow	0.4441	0.0942	0.1904	0.1131	0.1582	1.244
Perfect mixing	0.4038	0.0967	0.2065	0.1230	0.1700	1.296

[a]Calculation conditions: $\gamma = 0.1$, $\theta = 0.5$, q as shown in Table 4. Feed composition: $H_2 = 0.3$, $CH_4 = 0.1$, $CO = 0.25$, $N_2 = 0.15$, $CO_2 = 0.2$.

cocurrent flow, countercurrent flow, cross flow, perfect mixing, and one-side mixing. The calculation methods presented for these flow patterns can be used for systems consisting of any number of components. Calculation results are shown for gas separations of a NH_3, H_2, and N_2 gas mixture by a polyethylene membrane, and for a H_2, CH_4, CO, N_2, and CO_2 mixture by a microporous glass membrane.

APPENDIX A

Function $f(y_{pi})$ is defined as

$$f(y_{pi}) = \sum_{k=1}^{n} \frac{x_{fk}q_k/q_i}{(\gamma + \theta - \gamma\theta)\{(q_k/q_i) - 1\} + (x_{fi}/y_{pi})} - 1 \quad \text{(A-1)}$$

Differentiation of $f(y_{pi})$ with respect to y_{pi} yields

$$f'(y_{pi}) = \sum_{k=1}^{n} \frac{(x_{fk}q_k/q_i)x_{fi}/y_{pi}^2}{[(\gamma + \theta - \gamma\theta)\{(q_k/q_i) - 1\} + (x_{fi}/y_{pi})]^2} \quad \text{(A-2)}$$

The value of y_{pi} is obtained by the following iterative procedure with an appropriate initial value.

$$y_{pi}^{(r+1)} = y_{pi}^{(r)} - \frac{f(y_{pi}^{(r)})}{f'(y_{pi}^{(r)})} \quad \text{(A-3)}$$

where r is the iteration number. The iteration is continued until the value calculated is within a specified tolerance. When $\theta = 0$, Eq. (33) is analogous to Eq. (13).

APPENDIX B

Dividing Eq. (21) by Eq. (20) gives

$$\frac{dx_i}{df} = \frac{q_i(x_i - \gamma y_i) - x_i \sum_{k=1}^{n} q_k(x_k - \gamma y_k)}{f \sum_{k=1}^{n} q_k(x_k - \gamma y_k)} \quad (i = 1, \ldots, n - 1) \quad \text{(B-1)}$$

From Eq. (20),

$$\frac{ds}{df} = \frac{-1}{\sum_{k=1}^{n} q_k(x_k - \gamma y_k)} \quad \text{(B-2)}$$

For cocurrent flow, cross flow, and one-side mixing, the values of x_{oi}'s, y_{pi}'s, and s_t can be obtained by the integration of Eqs. (B-1) and (B-2) from $f = 1$ to $f = 1 - \theta$ in conjunction with Eqs. (22) to (27). For countercurrent flow, the integration of Eqs. (B-1) and (B-2) with Eqs. (22) and (25) to (29) is carried out backward. For one-side mixing and countercurrent flow, of course, a trial-and-error procedure is necessary. For perfect mixing, the values of y_{pi}'s and x_{oi}'s can be directly calculated by Eqs. (33), (32), and (30). Then the total membrane area s_t is given by

$$s_t = \frac{\theta}{\sum_{k=1}^{n} q_k(x_{ok} - \gamma y_{pk})} \quad \text{(B-3)}$$

SYMBOLS

A	membrane area (m^2)
A_t	total membrane area (m^2)
F	flow rate on the feed stream (mol/s)
F_f	feed flow rate (mol/s)
F_o	reject flow rate (mol/s)
f	dimensionless flow rate on the feed stream
f_o	dimensionless flow rate on the feed stream at the outlet
G	flow rate on the permeate stream parallel to the feed stream
g	dimensionless flow rate on the permeate stream
n	number of components
P_h	pressure of the feed stream (Pa)
P_l	pressure of the permeate stream (Pa)
Q	permeability (mol/s · m · Pa)
q	ratio of permeability
r	iteration number
s	dimensionless membrane area defined by Eq. (14)
s_t	dimensionless total membrane area defined by Eq. (14)
x	mole fraction of the gas component in the feed stream
x_f	mole fraction of the gas component in the feed stream at the inlet

x_o mole fraction of the gas component in the feed stream at the outlet

y mole fraction of the gas component in the permeate stream

y_p mole fraction of the gas component in the permeate stream at the outlet

Greek

γ pressure ratio, P_l/P_h

δ thickness of membrane (m)

θ stage cut defined by Eq. (16)

Subscripts

i, j, k component indication

m base component

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Received by editor December 22, 1984