

MASS TRANSPORT IN REVERSE OSMOSIS IN CASE OF VARIABLE DIFFUSIVITY

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Abstract—This paper analyzes the problem of concentration polarization in reverse osmosis in cases of concentration dependent diffusivity. A numerical solution of the mass transport equation for the laminar flow through two equally permeating, flat, parallel membranes is obtained. Three types of diffusivity-concentration relationships; a linear, an exponential, and a parabolic—and two values of solute rejections are investigated. The decrease in diffusivity with an increase in concentration is found to increase the value of concentration polarization modulus over that obtained in the case of constant diffusivity under the same system conditions. The increment is found to be larger for the case of a stronger diffusivity-concentration relationship and for a larger magnitude of membrane wall concentration. A method is proposed by which the effect of variable diffusivity on the value of concentration polarization modulus can be calculated for a wide range of practical conditions using the existing theoretical results for the case of constant diffusivity.

NOMENCLATURE

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| <p>c, solute concentration [lbm/ft³];</p> <p>C, dimensionless solute concentration (c/c_0);</p> <p>D, molecular diffusion coefficient of solute [ft²/h];</p> <p>h, half-width of channel [ft];</p> <p>N_s, solute flux through membrane [lbm/ft² h];</p> <p>S, fractional solute rejection = $1 - N_s/c_w v_w$;</p> <p>u, velocity component in the x-direction [ft/h];</p> <p>\bar{u}, average value of u over the channel at a given value of x [ft/h];</p> <p>U, dimensionless axial velocity (u/\bar{u}_0);</p> <p>v, velocity component in the y-direction [ft/h];</p> <p>V_w, dimensionless transverse velocity (v/v_{w0});</p> <p>\bar{V}_w, average value of V_w over the channel length;</p> <p>x, longitudinal distance from channel inlet [ft];</p> <p>X, dimensionless longitudinal distance (v_{w0}/\bar{u}_0) (x/h);</p> | <p>y, transverse distance from channel mid-plane [ft];</p> <p>Y, dimensionless transverse distance (y/h).</p> <p>Greek symbols</p> <p>α, normalized diffusion coefficient ($D/v_w h$);</p> <p>β, constant in equation (1);</p> <p>γ, constant in equation (1);</p> <p>λ, constant defined by equation (13);</p> <p>ξ, constant defined by equation (11);</p> <p>$\bar{\xi}$, average value of ξ over the channel length;</p> <p>Γ, concentration polarization modulus defined by equation (10);</p> <p>Δ, fraction of inlet solvent flow removed through membrane wall;</p> <p>$\Delta\pi$, difference in osmotic pressure across membrane, p.s.i.;</p> <p>ΔP, pressure drop across membrane, p.s.i.</p> <p>Subscripts</p> <p>0, channel inlet, i.e. $x = 0$;</p> <p>w, channel wall, i.e. at the membrane surface.</p> |
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INTRODUCTION

IN AN ultrafiltration or reverse osmosis process, the aqueous slurry is concentrated by flowing it through a semi-permeable membrane which rejects the dissolved solids. The convective flow of the solution carries solute up to the membrane surface, and since solute is rejected by the membrane it must diffuse back into the bulk aqueous slurry. Thus, the solute concentration at the membrane surface builds up to a value exceeding the bulk concentration until the back diffusion of solute produced by this concentration gradient plus any solute passing through the membrane just counter-balances the convection of solute to the membrane surface by the solvent flowing through the membrane. This building up of solute concentration at the membrane surface has proven to have, in general, detrimental effect on the separation process [1-4]. Thus, the problem of solute concentration polarization is important in the design of an effective membrane separation process. In the case of reverse osmosis, this problem has been studied and discussed by Merten [2], Merten *et al.* [3], Gill *et al.* [5], Sherwood *et al.* [6] and Brian [7]. Strathmann [13] has recently summarized the published information available in this area.

The existing theoretical studies on concentration polarization in reverse osmosis have assumed constant transport properties. This assumption, which is good in some cases such as desalination by reverse osmosis, falls short of reality in many other industrial applications of membrane separation processes (e.g. concentrations of aqueous sucrose solution and egg white solution). In these cases transport properties such as viscosity and diffusivity vary with concentration. Thus, a design of a membrane module requires the knowledge of concentration polarization in case of variable fluid properties.

Ginette and Merson [8] have recently carried out theoretical analysis of mass transfer in laminar flow of a viscous solution whose viscosity varies with concentration. The purpose of the present paper is to present the results

of a theoretical study on mass transport in reverse osmosis for the case of variable diffusivity. The effects of practical ranges of linear, parabolic and exponential diffusivity-concentration relationships on the concentration polarization in laminar flow have been investigated. The theoretical results are interpreted in terms of their design application.

THEORETICAL

The problem analyzed is concentration polarization in an aqueous solution flowing in a two-dimensional channel between flat parallel membranes when diffusivity varies with concentration. The diffusivity-concentration relationships examined in the present study can be written in the form of the following equation [9-11]:

$$D = D_0(1 + \beta C + \gamma C^2). \quad (1)$$

By assigning the proper values to the coefficients β and γ , equation (1) can be used to describe a linear, a parabolic or the approximate series expansion (up to the second order term) of an exponential diffusivity-concentration relationship. One such typical system of relationships is described in Fig. 1.

The continuity equation for solute conservation in this problem would be very similar to one used by Brian [1]. This equation in a dimensionless form can be written as

$$\frac{\partial(UC)}{\partial X} + \frac{\partial}{\partial Y} \left[VC - \alpha_0(1 + \beta C + \gamma C^2) \times \frac{\partial C}{\partial Y} \right] = 0. \quad (2)$$

The above equation considers convective flow in the longitudinal direction and diffusion and convection in the transverse direction, but longitudinal diffusion is neglected. The normalized solute concentration, C , is the local solute concentration divided by the solute concentration in the feed at the channel inlet, and U and V represent the normalized velocity components in the longitudinal and transverse directions, respectively. The dimensionless coordinates,

X and Y , represent dimensionless distances in the longitudinal and transverse directions, respectively: X would be numerically equal to the fractional solvent removal at a given longitudinal position if the solvent flux through the membrane were to remain constant at its value at the channel inlet. The origin of the coordinates is at the channel half width of the inlet. The normalized diffusion coefficient, $\alpha_0(1 + \beta C + \gamma C^2)$ is defined as the diffusion coefficient divided by the channel half-width and the flux through the membrane at the channel inlet.

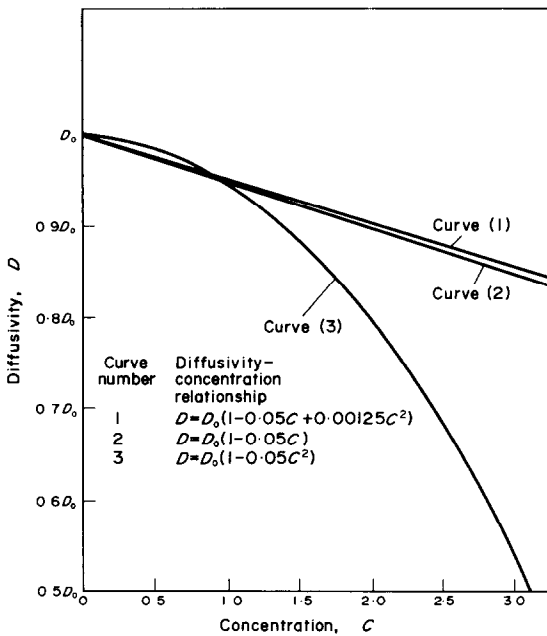


FIG. 1. Various types of diffusivity-concentration relationship investigated in the present study

The boundary conditions to equation (2) in the dimensionless form are

at $X = 0$, any Y : $C = 1$ (3)

at $Y = 0$, any X : $\frac{\partial C}{\partial Y} = 0$ (4)

at $Y = 1$, any X : $\alpha_0(1 + \beta C + \gamma C^2) \times \frac{\partial C}{\partial Y} = SVC$. (5)

Equation (3) represents the assumption that the solute concentration is uniform at the channel inlet, and equation (4) implies symmetry with respect to the mid-plane. This latter condition is based upon the assumption that the two membranes forming the channel walls are identical in properties and are therefore permeating at equal rates. Equation (5) relates solute diffusion and convection at the membrane surface to the solute rejection, S , defined as 1 minus the ratio of the solute flux through the membranes to the product of the permeation velocity at the membrane surface and the solute concentration at the membrane surface.

In order to integrate the non-linear differential equation (2), the velocity field must be known. In the present study, a solution of Berman [12] for the velocity field for the case in which the solvent flux through the membrane is uniform has been used. This solution for a small permeation Reynolds number (permeation Reynolds number based upon the half-width of the channel and the permeation velocity) in the dimensionless form can be written as [1]

$$U = \frac{3}{2}(1 - \Delta)(1 - Y^2) \quad (6)$$

$$V = V_w \left(\frac{Y}{2}\right)(3 - Y^2). \quad (7)$$

The above equations assume that the parabolic velocity profile is already developed at $X = 0$. In equation (7), V_w is the local value of the permeation velocity divided by the permeation velocity at the channel inlet. The quantity Δ is the fractional solvent removal, obtained by integrating the permeation velocity with respect to longitudinal position [1].

$$\Delta = \int_0^x V_w dx'. \quad (8)$$

For the case of a constant permeation velocity V_w is equal to unity and equation (8) simplifies to

$$\Delta = X. \quad (9)$$

In the analysis presented here, the volume change upon mixing solutions of different

solute concentrations is assumed to be negligible. In considering membranes with incomplete solute rejection, the solute rejection, S , is assumed to be constant. The permeation flux in reverse osmosis decreases with increasing recovery rate because the osmotic pressure of the solution increases with the increase in solute concentration [2]. However, Brian [7] has shown for the case of constant fluid properties that the average polarization over the length of the membrane is very nearly the same for cases of constant and variable permeation fluxes, if the average permeation fluxes in both cases are equal in magnitude. Based upon this finding, he has also suggested a simplified procedure for predicting polarization effects upon pressure drop requirements and the solute concentration in the product solvent using the theoretical results based upon the assumption of constant permeation flux. Since this procedure should be able to use for the case of concentration-dependent diffusivity, the assumption of constant permeation flux is made in the present analysis.

The measure of concentration polarization is designated by a parameter commonly called concentration polarization modulus, Γ , which is defined as [7]:

$$\Gamma = (1 + \xi)(1 - \Delta) - 1 \quad (10)$$

where

$$\xi = C_w - 1. \quad (11)$$

The parameter Γ as a function of X can be obtained with the knowledge of the concentration at the membrane wall, C_w , as a function of X . The latter relationship can be obtained from the solution of equations (1)–(9) for the concentration profile within the channel.

RESULTS

The system of equations (1)–(7) was solved by a finite difference method on an IBM 360 digital computer. A two-step, linearized, X -centered finite-difference method was used. This method is a modification of that proposed by Douglas [13]. The non-linear term in equation

(2) was linearized by evaluating the coefficients at the previous grid point.

The convergence of the finite-difference solution was tested by changing the longitudinal and transverse increments. For several values of α_0 , the computer solution with the values of β and γ equal to zero in equation (1), was checked against the numerical solution of Brian [7]. They were found to be in good agreement. Based upon these considerations, the finite difference solutions are believed to be convergent within less than 0.5 per cent.

For the three types of diffusivity-concentration relationships examined in the present study the value of coefficient β was varied between -0.01 and -0.2 and that of γ between -0.05 and $+0.02$. These values of β and γ are believed to represent the diffusivity-concentration relationships in the range of the most practical interest [9–11].

The theoretical results obtained in the present study are described in terms of parameters which are important in the design of a membrane separation process. These are Γ , concentration polarization modulus; Δ , fractional solvent removal; and α , normalized diffusion coefficient. The results are plotted as the curves of Γ vs. $\Delta/3\alpha_0^2$ for the various values of α . This method of data representation enables one to show a comparison between the theoretical results of the present study and the ones obtained by Brian [1] for the case of constant diffusivity.

Figure 2 shows comparison between a plot of Γ vs. $\Delta/3\alpha_0^2$ for the cases of constant and variable diffusivity. The results are shown for the three different values of α_0 . For the variable diffusivity case, an approximate exponential diffusivity-concentration relationship with the exponent value of -0.05 was chosen. The results shown on this figure indicate that the deviation in the values of concentration polarization modulus for the two cases increases with an increase in value of $\Delta/3\alpha_0^2$. Also, for a given value of $\Delta/3\alpha_0^2$ the value of concentration polarization modulus in case of variable diffusivity is always higher than one for the case of constant diffusivity.

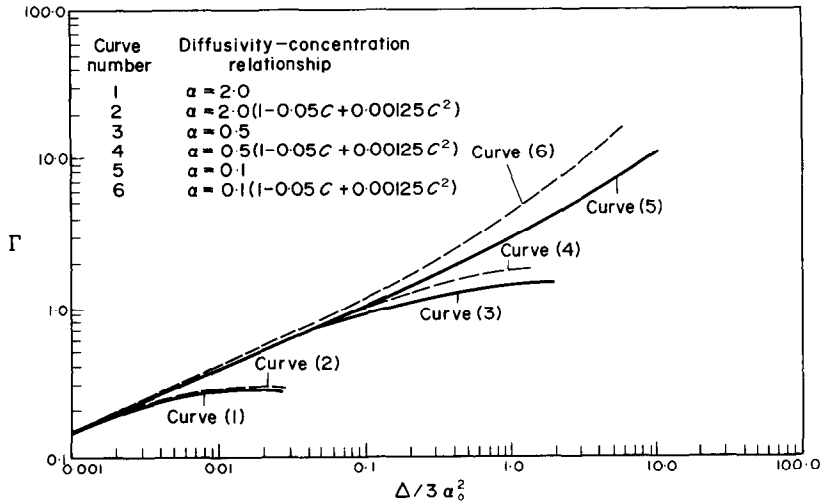


FIG. 2. Effect of variable diffusivity on concentration polarization moduli at various α_0 , for the ease of constant permeation flux and complete solute rejection.

The results on the effect of various types of diffusivity-concentration relationships on the values of concentration polarization modulus is described in Fig. 3. Three types of diffusivity-concentration relationships; a linear, a parabolic and an exponential, with a value of α_0 equal to 0.5 were examined. These results indicate that in all three cases the deviation between the values of Γ for the constant and variable diffusivity cases increases with an increase in

value of $\Delta/3\alpha_0^2$. Also, the deviation is larger for a stronger diffusivity-concentration relationship. The same results in terms of a plot of dimensionless wall concentration vs. dimensionless axial distance are described in

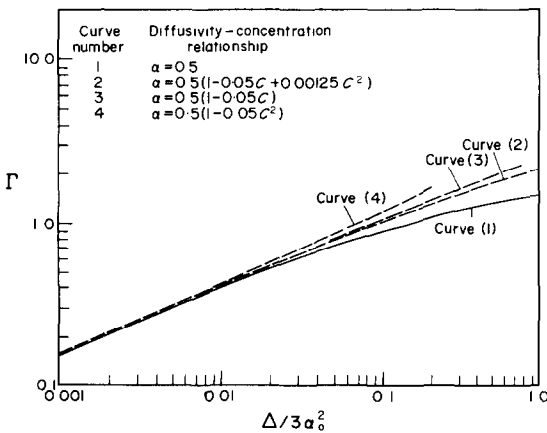


FIG. 3. Effect of various types of diffusivity-concentration relationship on concentration polarization moduli for the case of constant permeation flux and complete solute rejection.

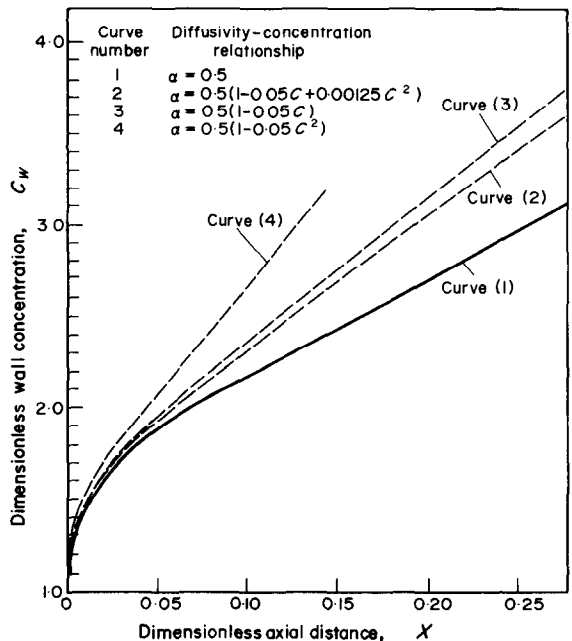


FIG. 4. Effect of variable diffusivity on wall concentration for the case of constant permeation flux and complete solute rejection

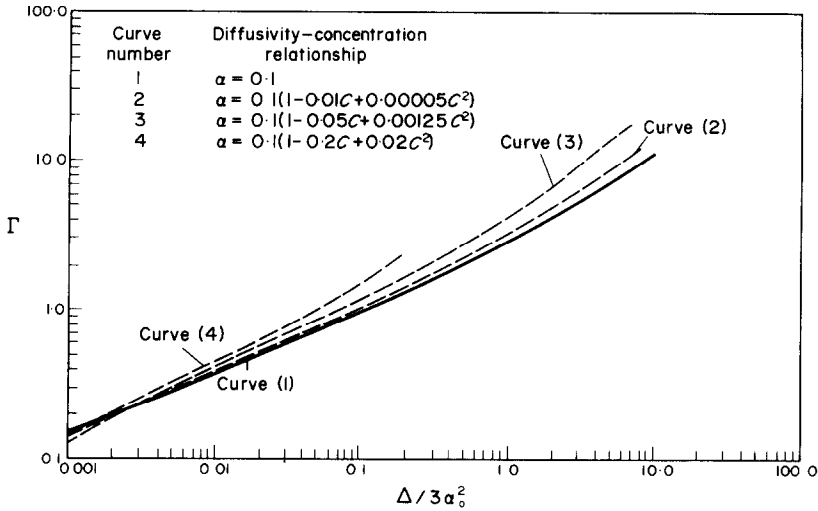


FIG. 5. Effect of exponent coefficient of diffusivity-concentration relationship on concentration polarization moduli for the case of constant permeation flux and complete solute rejection.

Fig. 4. These results once again indicate that the stronger the diffusivity-concentration relationship, the larger the rate at which wall concentration builds up.

Since an exponential diffusivity-concentration relationship is the most common one encountered in practice [9-11], the effect of

exponent coefficient on concentration polarization was investigated. Three values of exponent coefficient; -0.01, -0.05 and -0.20 were examined. The results of this study for a typical value of α_0 and various values of $\Delta/3\alpha_0^2$ are shown in Fig. 5. These results indicate that for a large value of $\Delta/3\alpha_0^2$, the value of exponent

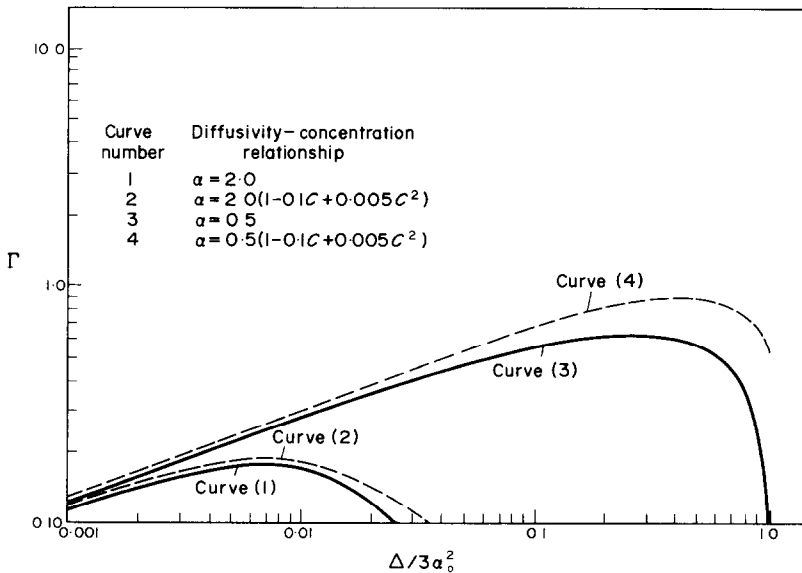


FIG. 6. Effect of variable diffusivity on concentration polarization moduli for the case of constant permeation flux and solute rejection = 0.8.

coefficient strongly affects the value of concentration polarization modulus.

The effect of variable diffusivity on the values of concentration polarization modulus was also examined for a value of solute rejection equal to 0.8. The results of this study in terms of a plot of Γ vs. $\Delta/3\alpha_0^2$ are shown in Fig. 6. These results are described for two different values of α . The results once again indicate that the deviation between values of concentration polarization modulus in two cases increases with an increase in the values of $\Delta/3\alpha_0^2$.

DISCUSSION

Since concentration polarization is caused by lack of mixing in the fluid, the diffusivity should have an important role in determining the extent of concentration build-up at the membrane surface. Obviously, the smaller the diffusivity, the larger the concentration build-up should be at the membrane wall. For the case of constant diffusivity this has been verified by Sherwood *et al.* [6] and Brian [1, 7]. In the case of variable diffusivity, as membrane wall concentration builds up, the effective diffusion coefficient decreases and this should further increase the concentration build-up rate at the membrane surface. Thus, the net effect of decrease in diffusivity with concentration is to increase the concentration polarization at any downstream position over one obtained for the case of constant diffusivity under the same system conditions.

Based upon the above explanation, the deviation in the values of concentration polarization modulus for the variable and the constant diffusivity cases should increase with an increase in the difference in the values of diffusivity. Hence, for the given values of α_0 and $\Delta/3\alpha_0^2$, the deviation should be larger in the case of a stronger diffusivity-concentration relationship. For the same values of the pertinent coefficients in equation (1) and those of α_0 and $\Delta/3\alpha_0^2$, a parabolic diffusivity-concentration relationship should thus predict a much larger deviation in the values of concentration polarization

modulus than a linear or an exponential diffusivity-concentration relationship. The results of Fig. 3 indicate this to be the case.

With the same line of reasoning as described above, one should expect the difference in the values of Γ predicted for the variable and the constant diffusivity cases to be a strong function of the actual magnitude of wall concentration. This is because the larger the magnitude of this concentration, the larger would be the difference in the effective values of diffusivity in the two cases. The results of Figs. 2, 4-6 validate this expectation.

The above described explanation of the theoretical results appear to be compatible with the physics of the process. It also suggests that one may be able to calculate concentration polarization modulus in the case of variable diffusivity with the use of the theoretical results of Brian [1, 7], if one takes proper account of variation in α with the downstream position. Based upon this premise, the following alternate four step method to obtain the plots of Γ vs. $\Delta/3\alpha_0^2$ at various values of α in the cases of variable diffusivity was examined.

1. Calculate $\Delta/3\alpha_0^2$ and obtain Γ for given values of α_0 and $\Delta/3\alpha_0^2$ using the theoretical results of Brian [3].
2. Obtain a value of dimensionless wall concentration from the calculated values of Γ and Δ .
3. Using the calculated value of wall concentration, obtain an actual value of α from the diffusivity-concentration relationship.
4. Calculate a new value of Γ at the actual values of α and $\Delta/3\alpha^2$ using the theoretical results of Brian [1, 7]. This will be then the actual value of Γ corresponding to the value of $\Delta/3\alpha_0^2$ in a case of variable diffusivity.

For several cases examined in the present study, the plots of Γ vs. $\Delta/3\alpha_0^2$ were obtained using the above described proposed method. These results are compared in Fig. 7 with the ones obtained from numerical solution of equations (1)-(7). As indicated in the figure, the

comparison was made for various diffusivity-concentration relationship and for the two values of solute rejection. The results indicate that at least within the range of diffusivity-concentration relationships examined in the present study, the proposed method works quite well. At present, there is no mathematical proof available for the success of the proposed method. However,

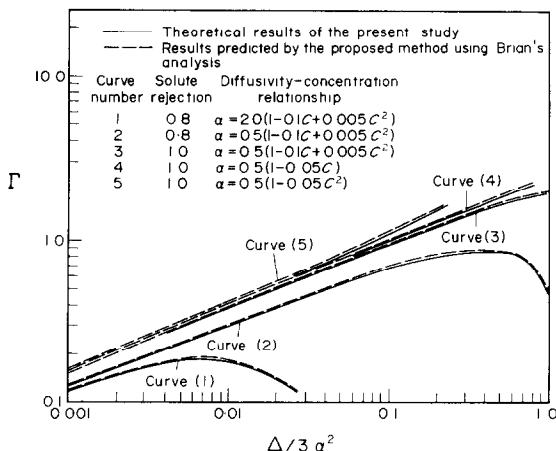


FIG. 7. Comparison between the theoretical results of the present study with the ones predicted using Brian's analysis by the proposed method.

it is believed that for the cases examined in the present research, the proper accounts for the variation in the magnitude of α with the downstream positions were taken by the proposed method.

UTILITY OF PRESENT ANALYSIS IN THE DESIGN OF PRACTICAL REVERSE OSMOSIS UNITS

In the design of a practical reverse osmosis unit, the knowledge of the operating pressure required to obtain a specified solvent production with a given membrane is of great importance. This is because operating costs for running a reverse osmosis unit depends significantly upon the operating pressure. In the present analysis, the solute concentration at the membrane surface, C_w , was evaluated for the various system conditions when the solute diffusion

coefficient is dependent upon the solute concentration. Let us now, with the help of an illustrative example, look at how the information obtained in the present analysis can be used to obtain the operating pressure for a specific reverse osmosis unit.

Brian [7] reports that assuming the osmotic pressure is directly proportional to the solute concentration and that pressure drop across membrane, ΔP , is essentially constant, the following relationship can be written for the average permeation flux, \bar{V}_w , over the entire channel

$$\bar{V}_w = 1 - \lambda \bar{\xi} \quad (12)$$

where

$$\lambda = \frac{S\pi_0}{\Delta P - S\pi_0} \quad (13)$$

and

$$\bar{\xi} = \frac{1}{X} \int_0^X (C_w - 1) dX' \quad (14)$$

In equation (13), π_0 is the osmotic pressure at the inlet feed concentration. For the given values of \bar{V}_w , X , α_0 , S and from the results shown in Figs. 2-7, ΔP , the required design parameter, can be calculated from equations (12)-(14). This type of calculation for an hypothetical reverse osmosis unit used for the concentration of sucrose-water solution is briefly outlined below. The following data are used in this calculation.

$$S = 1.0$$

$$\alpha_0 = 0.5 \text{ (evaluated at the average permeation flux)}$$

$$\bar{V}_w = 0.30$$

$$\pi_0 = 36 \text{ p.s.i. (obtained from [9] assuming the molality of the feed solution to be 0.1)}$$

$$\alpha = \alpha_0 e^{-0.01C} \text{ (an approximate relation obtained from the diffusivity-concentration data for the sucrose-water system reported in [9])} \quad (15)$$

$$X = 0.25.$$

Assuming α to be constant at the value of α_0 , from equation (14) and Fig. 4, $\bar{\xi}$ can be calculated to be equal to 1.266. Similarly assuming α to be function of concentration as given by equation

(15), $\bar{\xi}$ can be calculated to be 1.390. The membrane wall concentration as a function of X for the diffusivity-concentration relationship defined by equation (15) was obtained by the linear interpolations between the C_w vs. X curves for $\alpha = 0.5 \exp(-0.05 C)$ and $\alpha = 0.5$ shown in Fig. 4. Thus, from equations (12) and (13) and using the data listed above, the values of ΔP for the two cases can be calculated to be approximately equal to 101 p.s.i. and 108 p.s.i. respectively. This means that even for a small value of X examined here, there would be approximately 7 per cent error in the calculation of ΔP if the diffusivity were to be assumed constant at its channel inlet value. It should be noted that the difference in the ΔP values for the constant and the variable diffusivity cases will increase rapidly with increase in X because as shown in Fig. 4, the membrane wall concentrations for the two cases diverges from each other more and more with increasing value of X . It should also be noted that the calculation of $\bar{\xi}$ can also be carried out with the help of results similar to ones shown in Figs. 2, 3, 5-7 because Γ and ξ are related to each other as shown by equation (10).

CONCLUSION

It is concluded from the present study that the net effect of decrease in diffusivity with concentration is to increase solute build-up at the membrane surface in a reverse osmosis process. This increment is larger for a stronger diffusivity-concentration relationship. The theoretical results of Sherwood *et al.* [6] and Brian [1, 7] for the case of constant diffusivity can be used to calculate concentration polarization moduli at various downstream positions in the case of variable diffusivity by the method proposed in this paper.

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TRANSPORT MASSIQUE DANS L'OSMOSE INVERSE DANS LE CAS D'UNE DIFFUSIVITÉ VARIABLE

Résumé—Cet article analyse le problème de polarisation par concentration dans l'osmose inverse lorsque la diffusivité dépend de la concentration. On obtient une solution numérique de l'équation du transport massique pour l'écoulement laminaire à travers deux membranes parallèles, plates et également perméables.

On considère trois types de relations entre diffusivité et concentration, une linéaire, une exponentielle et une parabolique. On trouve que la décroissance de la diffusivité avec un accroissement de concentration, augmente la valeur du module de polarisation par concentration par rapport à celle obtenue dans le cas d'une diffusivité constante dans les mêmes conditions du système. L'augmentation est plus grande dans le cas d'une relation entre diffusivité et concentration plus forte, et pour une plus grande concentration à la paroi de la membrane. On propose une méthode par laquelle l'effet de diffusivité variable sur la valeur du module de polarisation par concentration peut être calculé pour une large gamme de conditions pratiques en utilisant les résultats théoriques existants dans le cas d'une diffusion constante.

STOFFÜBERTRAGUNG BEI UMGEKEHRTER OSMOSE FÜR DEN FALL VERÄNDERLICHER DURCHLÄSSIGKEIT

Zusammenfassung—Die Arbeit analysiert das Problem der Konzentrationspolarisation bei umgekehrter Osmose für den Fall konzentrationsabhängiger Durchlässigkeit. Das Ergebnis ist eine numerische Lösung der Stofftransportgleichung für die laminare Strömung durch 2 gleich durchlässige, flache, parallele Membranen. Es werden 3 Arten der Abhängigkeit der Durchlässigkeit von der Konzentration untersucht: eine lineare, eine exponentielle und eine parabolische Abhängigkeit und 2 Werte von Lösungsundurchlässigkeit. Die Abnahme der Durchlässigkeit mit dem Ansteigen der Konzentration erhöht den Konzentrations-Polarisations-Modul gegenüber den Werten für den Fall konstanter Durchlässigkeit bei sonst gleichen Systembedingungen. Für eine stärkere Abhängigkeit der Durchlässigkeit von der Konzentration und für höhere Werte der Konzentration an der Membran ist diese Erhöhung grösser.

Es wird eine Methode vorgeschlagen, mit deren Hilfe die Wirkung der veränderlichen Durchlässigkeit auf die Grösse des Konzentrations-Polarisations-Moduls in einem weiten Bereich praktisch vorkommender Bedingungen berechnet werden kann, wobei die vorhandenen theoretischen Lösungen für den Fall konstanter Durchlässigkeit benützt werden.

ПЕРЕНОС МАССЫ ПРИ ОБРАТНОМ ОСМОСЕ В СЛУЧАЕ ПЕРЕМЕННОГО КОЭФФИЦИЕНТА ДИФФУЗИИ

Аннотация—В данной работе анализируется задача концентрационной поляризации при обратном осмосе для случая коэффициента диффузии, зависящего от концентрации. Получено численное решение уравнения переноса массы для ламинарного течения через две плоские параллельные мембраны с одинаковой проницаемостью. Исследуется три типа зависимости между коэффициентом диффузии и концентрацией—линейная, экспоненциальная и параболическая. Найдено, что уменьшение коэффициента диффузии с увеличением концентрации вызывает увеличение модуля концентрационной поляризации по сравнению со значением, полученным в случае постоянного коэффициента диффузии при тех же условиях. Найдено, что увеличение больше для случая более сильной зависимости коэффициента диффузии от концентрации на стенке мембраны. Предполагается метод, с помощью которого влияние переменного коэффициента диффузии на значение модуля концентрационной поляризации может рассчитываться для широкого диапазона условий, используя теоретические результаты для случая постоянного коэффициента диффузии.