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POLARIZATION ADJACENT TO AN INHOMOGENEOUS MEMBRANE

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ABSTRACT

The diffusion field over the surface of a membrane has been analyzed in a simplified manner responsive to the microporous nature of the actual membrane. The membrane surface is modeled as having intermittent strips of active sites (ribbons) separated by inactive sites (islands). The two-dimensional representation of the concentration field omits the third (transverse) direction of diffusion and assumes a regular and systematic surface arrangement. The equations of flow and concentration are shown to be reducible to a set of linear equations, solvable by standard methods of finite differencing and algebraic solution.

The results show systematic variations of a significant magnitude locally around the ribbons (pores) of membrane. The magnitude, for example, assumes a concentration value 1.15 times the local concentration to occur from the upstream to the downstream side of a given ribbon for 33% surface porosity. Smaller ribbons are expected to produce larger jumps. The effect of the discontinuous membrane is isolated. That is, the rapid increases and decreases in concentration are in proportion to the local concentration value.

INTRODUCTION

Membranes are modern devices used to accomplish separation of solutes from solvents. The solute retained by an operating membrane becomes concentrated near the surface of the membrane and diffuses back into the stream. The solute concentration and diffusion process is frequently referred to as solute polarization. The polarization process has heretofore been modeled as a uniform process near an idealized membrane surface. But, on the scale of thickness of the polarization layer of about a few micrometers, the membrane is anything but homogeneous. Cast polymeric membranes have active flux sites of about 0.5 μm separated by several μm; formed-in-place membranes formed on steel supports have a sea of membrane punctuated by 10μm impermeable islands; and ceramic membranes commonly have pores of 0.1μm spaced at 0.3μm intervals. So it is clear that the transverse flow of solvent through the membrane is non-homogeneous on a microscopic (but not on a molecular) scale. This effort aspires to study a model responsive to the degree of non-uniformity expected and predict the behavior as it depends on the surface.

Polymeric membranes (1,2) are sometimes microporous, as in the case of the usual ultrafilters, and sometimes asymmetric, having small regions of very thin material near which the solvent flow will be locally higher. The effect of the asymmetric structure will be regarded as the same as a pore in that it produces a small region of high solvent flow, surrounded by a region of low or zero flow. Ceramic membranes (3) tend to exhibit much greater porosity than the polymeric membranes. In these cases of much higher pore densities the pores are obviously much closer together probably leading to a nearer approach to the homogeneous surface. The formed-in-place (also known as dynamic) membrane (4) is formed on a porous metal tube resulting in a deposited gel layer or polymer layer which forms a "sea" of membrane interspersed with impermeable "islands" of stainless steel. Thus the membrane surface is not directly comparable with those surfaces having a structure of pores in impermeable material.

In any of the surfaces mentioned, an observer riding on a streamline would view, looking directly at the membrane surface

with a narrow field of view, the three-dimensional membrane surface as intermittent patches of active membrane and inactive surface with the active pores having sizes from as small as1nm to as large as 10μm. The patches of membrane would be separated by distances of less than one pore dimension to distances of about 100 pore dimensions. If the pores are regularly arranged and diffusion transverse to the motion of the observer is not considered, the result should be well represented by a two-dimensional solution wherein the pores are slots or ribbons. This easier two-dimensional problem is addressed.

MODELING AND ANALYSIS

Desiring to determine the effect of the inhomogeneous nature of the membrane on the details of the membrane process, the following model has been constructed. The membrane surface is the boundary of a channel idealized as having a fully developed and uniform velocity field, having a very slight amount of flow withdrawn as permeate and having little pressure drop compared with the pressure. Under such assumptions the flux of permeate and the shear stress at the membrane surface can be considered constant. The fluid is allowed to permeate the surface in a regular arrangement of strips or ribbons of membrane separated by islands of impermeable material. The membrane ribbons are selectively permeable allowing almost complete retention of solute, which in turn returns to the fluid by diffusion. In this model the ribbons of membrane are aligned on the surface so as to be perpendicular to the crossflow direction allowing for the diffusion parallel to the ribbon to be neglected. Such problems are characterized by large Schmidt numbers and very thin layers of concentration polarization. However, the common assumption to neglect diffusion in the stream flow direction due to the thinness of the concentration layer must be re-examined due to the smallness of the pore spacing in the stream direction. For most cases it continues to be negligible, as will be shown.

The domain for solution is the thin region totally encompassing the zone in which concentration polarization occurs, but much less than the entire channel. Boundary conditions are

asymptotic at infinity, meaning the functions approach asymptotic conditions and their derivatives vanish. The thickness of the diffusive zone and the layer containing the focusing the membrane flow are independently scaled. Coordinates adopted have the x-direction parallel to the surface and aligned with the primary fluid velocity, u. The y-coordinate is normal to the membrane surface; this is parallel to the filter flow velocity, v. The z-coordinate is mutually perpendicular to the others. The problem is formulated to avoid involvement of this third direction in the interest of simplicity and with the hope that essential information is not lost.

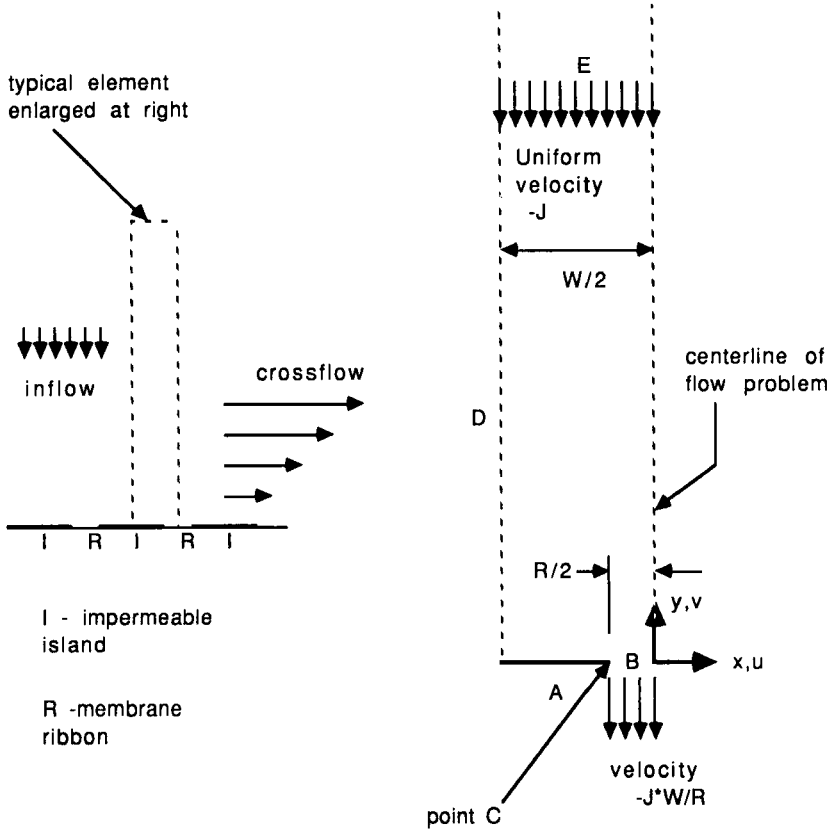


Figure 1 Schematic Diagram of the Solution Block

Since the membrane is regularly arranged there are building blocks which will be referred to as modules. One module is the smallest representative element of the whole and comprises the width of one ribbon-island pair and which extends far enough from the membrane that the streamlines are straight and parallel. There is no motive for flow in the direction (z) of the edge of the ribbon strip, and the (x,y) cross section of one-half of one module is sketched in the magnified view of Figure 1. The other half is the mirror image of the half shown.

In the diagram the surface A represents the surface ot an island and surface B is the ribbon of membrane. Surface D is adjoining another such block at left, while surface E represents the condition far enough from the membrane that the flow, (v), is essentially straight and parallel. The flow problem, as indicated, is symmetric about the line extending from the center of the ribbon element. The main crossflow is from left to right (x-direction) and the membrane flow (negative y-direction) is from top to bottom. The v-velocity far from the membrane surface has the value -J, the same as volume flow per unit of projected surface.

With the shear field fully developed, and because the scale of the problem is small (maximum dimension of A+B=W/2 about 0.05mm) and the velocities are low (v of order 10^{-5} m/s) the flow is dominated by viscous forces and all convective terms in the equation of motion are negligible. This observation leads to equations of motion:

$$\frac{\partial u}{\partial x} + \frac{\partial v}{\partial y} = 0$$

$$0 = -\frac{\partial P}{\partial x} + \mu \left(\frac{\partial^2 u}{\partial x^2} + \frac{\partial^2 u}{\partial y^2} \right)$$

$$0 = -\frac{\partial P}{\partial y} + \mu \left(\frac{\partial^2 v}{\partial x^2} + \frac{\partial^2 v}{\partial y^2} \right) \quad .$$

The first is the equation of continuity and the second pair represent the equations for momentum in the directions of the cross flow and perpendicular to the membrane surface, respectively. The boundary

conditions are:

$u = 0$ on A and B
$v = 0$ on A
$v = -J^*W/R$ on B
$u = \tau/\mu\, y$ on D and E
$v = -J$ on E
v on D^+ = v on D^- (symmetry)

The problem thusly posed is observed to be linear and is facilitated by using a superposition: $u,v = u_1,v_1 + u_2,v_2$. The first component is a pure shear flow and the second is the converging flow where the uniform volume flow is funneled into the individual pores of the surface. Each component problem will satisfy the set of differential equations and the boundary conditions for each become:

$u_1 = \tau/\mu\, y$ in the entire region
$v_1 = 0$

$u_2 = 0$ on the entire boundary
v_2 = symmetric with respect to centerline and D
$v_2 = 0$ on A
$v_2 = -J^*W/R$ on B
$v_2 = -J$ on E

The solution to the second problem is facilitated by introduction of a stream function ,ψ, such that

$$u_2 = \partial\psi/\partial y \quad \text{and} \quad v_2 = -\partial\psi/\partial x.$$

With the introduction of vorticity, $\omega = \partial v/\partial x - \partial u/\partial y$, it is well known (5) that the equation of continuity will be solved exactly, and that two equations not having pressure explicitly will govern the flow. These are

$$\frac{\partial^2\omega}{\partial x^2} + \frac{\partial^2\omega}{\partial y^2} = 0 \quad \text{and}$$

$$\frac{\partial^2\psi}{\partial x^2} + \frac{\partial^2\psi}{\partial y^2} = -\omega.$$

The boundary conditions on these equations follow from physical reasoning based on the velocity relations presented already and are:

$\omega = 0$ on the boundary except on A. At point C the vorticity, ω, is singular, positive on the corner shown and negative on its image.

$\psi = 0$ on D and A

$\psi = J^*(W/2-x)$ on E

$\psi = J^*W/R^*(R-x)$ on B

$\psi = W^*J$ on the line of symmetry over the ribbon.

These equations were programmed for numerical solution. A grid was selected as 6 mesh spaces in the x-direction and 20 to 300 divisions of essentially the same size in the y-direction. The vorticity equation has been solved by alternating direction implicit (ADI) method and the stream function equation by succcessive over-relaxation method (SOR). The singular condition is represented by a value of ω at point C. The value used was adjusted to afford consistency with the value of J. Intuitively there is one and only one value of J for a given value of the strength of the circulation at point C. The proper value gives zero $\partial v/\partial y$ at the center of the outlet of the pore (midpoint of A). The physical angle of convergence of the streamlines is affected mildly by the selection, with more

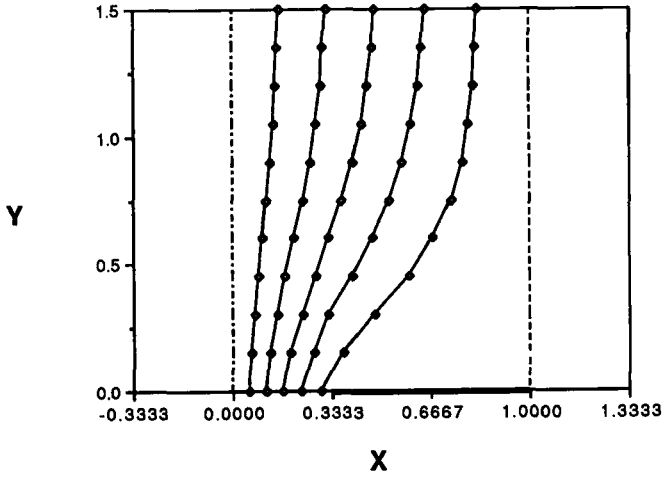


Figure 2 Streamlines of the Solution

convergence for lower magnitudes of the value of ω at C. If too large a values of vorticity at point C is selected, the centerline velocity (v-direction) overshoots, then returns to the value of JW/R. At the same time a recirculation region appears in the corner; such a pattern is not expected and not valid for a flow without inertial terms included. Solutions with great depth in the y-direction prove to be identical with those of less depth; the effective region in which the streamlines converge is about the same as the width of the region. Figure 2 shows the pattern of streamlines calculated. There are no unanticipated features of the flow.

It is assumed that the diffusion is in two directions only; it will be further assumed that the diffusion coefficient and density are constant. Under these assumptions the equation governing the concentration of solute is:

$$u\frac{\partial c}{\partial x} + v\frac{\partial c}{\partial y} = D\left(\frac{\partial^2 c}{\partial x^2} + \frac{\partial^2 c}{\partial y^2}\right). \qquad (1)$$

This equation is subject to boundary conditions as follows:

$$c(0,y) = c_B \qquad (2a)$$

$$c(x,\infty) = c_B \qquad (2b)$$

$$\partial c/\partial y = (v/D)c \quad \text{at } y=0 \qquad (2c)$$

Also some condition may be required at x=L. The condition represented by (2c) is total exclusion of solute by the membrane. The start of the field (x=0) is located in the middle of the first island.

According to previous analysis the velocities are given by the sum of two solutions:

$$u_1 = \frac{\tau}{\mu}y \quad \text{and } v_1 = 0.$$

and

$u_2 = J\,U(x',y')$ and $v_2 = J\,V(x',y')$. Here $x'=x/W-n$ and $y'=y/W$ and the normalized solutions U and V depend on the coordinates (x',y') and on the parameter R/W representing the area of the membrane per surface area. The value of n is the number of flow modules upstream of the current one.

The magnitude of u_1 compares to u_2 as τ/μ to JW/R^2. In most cases u_1 is several orders of magnitude greater so that u_2 is insignificant. Therefore $u \cong u_1 = \tau/\mu$ and $v \cong v_2$; $v/J = V(x',y')$. These terms are installed into the diffusion equation (1).

Substitute into Equations (1 and 2) the variable changes: $C=c/cB$, $Y=Jy/D$, $X=x\mu J^3/\tau D^2$. There results:

$$Y\frac{\partial C}{\partial X} + \frac{v}{J}\frac{\partial C}{\partial Y} = \frac{\partial^2 C}{\partial Y^2} + \frac{\mu^2 J^4}{\tau^2 D^2}\frac{\partial^2 C}{\partial X^2}. \qquad (3)$$

$$C(0,Y) = 0 \qquad (4a)$$

$$C(x,\infty) = 0 \qquad (4b)$$

$$\partial C/\partial Y = -(W/R)\,C \quad \text{at } Y=0 \qquad (4c)$$

The importance of the longitudinal diffusion term in Equation (3) depends on the value of the parameter μ^2J^4/τ^2D^2. Values of τ/μ, the shear rate, will typically be from 100 to 100,000 sec^{-1}, while J may range from 0.05 to $1x10^{-4}$ m/s, and D may be from 10^{-11} to $1.5x10^{-9}$ m^2/s. Nominal values of $\tau/\mu = 10^3$, $J=10^{-5}$, and $D=10^{-10}$ yield $\mu J^2/\tau D=10^{-3}$, a small value. Whenever this parameter is very small, the last term in the equation is of negligible importance, since the value of the longitudinal derivative will at most be approximately the same as that for the transverse derivative. Lower values of D may usually be identified with lower values of J, and higher values of shear are used to avert low values of J, hence not with large J-values. It appears that most of the time the longitudinal diffusion term may be safely neglected.

Based on observations of computations (6) for the homogeneous membrane, the thickness of the diffusive zone is approximately 2D/J. The corresponding thickness of the flow solution in the y-direction (*ie* the zone in which the convergence of the streamlines is consummated) is shown in Figure 2 to be approximately W. The ratios of the thicknesses of the flow and diffusion zones is WJ/2D. Whenever the parameter is significantly greater than unity the diffusive zone is declared to be thin, and the velocity, v, forms strips having values alternating between zero and JW/R over the islands and ribbons, respectively. If the value of

WJ/2D is unity or smaller, the diffusive zone is declared to be thick. For the calculations herein the value of WJ/2D is 1/4. A complete book of solutions would have computations for several values of WJ/2D.

The ramifications of the size of JW/2D concern the magnitude of the effort to obtain a solution. In the simplest (thin case) type of calculations, the velocity v is constant over a pore and the velocity field is not dependent upon the concentration field. A single velocity solution applies to every ribbon module and there are needed about 1 to 10-million steps each requiring the calculation of 300 concentration points. In the most complicated case osmotic resistance or gel retardation is important and the velocity and concentration fields will depend mutually upon each other and the solutions will be extremely tedious. There will be 10,000 to 100,000 blocks of 100x300 concentration points interacting iteratively with the flow solution. The former calculation has proven to be challenging to the endurance or budget of a modest computer (non supercomputer). The latter represents at least an order of magnitude more expense.

To effect a solution, finite difference techniques have been used to form sets of algebraic equations from the differential equation. Since the longitudinal diffusion term has been eliminated from consideration, the procedure is of marching -solution type. The concentration equation (3) has been broken into finite difference forms using central differences in the Y-direction and forward-difference formulation in the X-direction. The maximum-guaranteed-stable step size is $\Delta X = (\Delta Y)^3/2$ for this procedure. Normally such an equation would be solved using implicit techniques, since the step size in the X-direction can be much larger than with explicit techniques. However, for this problem the steps must be small to account for the physical arrangement. In many cases the step size and maximum step size for stabilty of the explicit method are quite close. The explicit technique has been selected as the calculations may be made rapidly. Attempts to speed the solution are resisted because there are many steps. A typical physical problem has a flow module 10^{-5}m in length, and the module is divided into perhaps 100 segments. If one calculates the conditions along one meter of membrane, the result

requires 10 million steps. Enough grids in the y-direction have been selected (typically 300) to ensure the edge of the diffusion zone does not exceed the range.

Results of calculations are in the form of

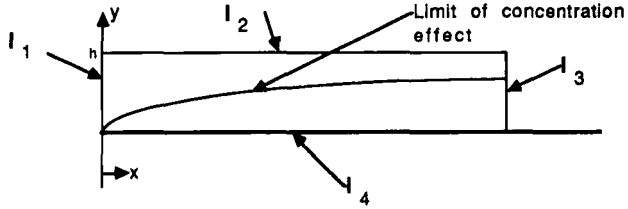
- Distribution of wall concentration $C(x,0)=C_w$
- The integral **C:**

$$\mathbf{C} = \int_0^L v(x,0)\, C(x,0)\, dx$$

- Concentration profiles C(y) at selected places, particularly at the ends of ribbons and islands
- The mass excess integral **B:**

$$\mathbf{B} = \int_0^\infty u(x,y)\, \{\, c(x,y) - c_B \,\}\, dy$$

The control volume stretching from the start of the membrane to x=L and extending to the value of y where the diffusion ceases is sketched in Figure 3. The flow of solvent entering the front face at x=0 is the same as that leaving the rear at x=L.



$$I_1 = \int_0^h u(0,y) c_B dy \; ; \quad I_2 = \int_0^L J c_B dx \; ; \quad I_3 = \int_0^h u(x,y)\, c(x,y) dy \; ;$$

$$I_4 = \int_0^L v(x,0)\, c(x,0)\, s\, dx \; ; \; s = \text{passage}$$

For passage=0, $I_4 \cong 0$, and u(0,y)=u(x,y), giving

$$I_2 = J c_B L = I_3 - I_1 = B = \int_0^\infty u(x,y)\, \{\, c(x,y) - c_B \,\}\, dy \; .$$

Figure 3 Control Volume Illustrating the Solute Mass Flow Integrals

The solvent entering the top is the same as that leaving into the permeate, its amount is $\int Jdx = JL$. The difference in solute exiting the rear, $\int ucdy$, and the front, $\int uc_B dy$, is the mass excess integral **B** and it must equal Jc_BL. Two membranes are alike except in microdetail if they have the same transport variables and the same Jc_BL. They must have the same **B**-values consequently. However, this does not mean they have the same **C**-values, since the concentration profiles may be quite different. Yet the integrals represented by **C** are proportional to the amount of material passed to the permeate; the ratio of **C** of two membranes having equal **B**-values and other properties is a primary result.

A few observations are in order concerning technical details of the numerical work and the apparent accuracy. Herein accuracy is judged partly by the calculated disparity between the integrals **B** and Jc_BL. The disparity between the values is typically reduced to under 15% only with considerable attention. The same measure of accuracy as for homogeneous membrane calculations, about 2 to 3%, was not possible to obtain. Errors appear significantly only at positions several hundred modules downstream, making the effort for computational experiments relatively large. The study of error centered on the simulation of the membrane boundary condition, the mesh size, and the interpretation of velocity field at the points of discontinuity.

Various techniques have been used for the calculation of the concentration at the membrane surface,*ie* the wall boundary condition. The best boundary condition approach found assumes constant v-velocity to integrate the boundary condition (4c) from 0 to ΔY resulting in:

$$C(X,0) = C(X,\Delta Y) \exp[(W/R)\Delta Y]$$

In all the formulations the ratio of the membrane surface concentration to the concentration of the adjacent transverse node is dependent on the parameters but not on the upstream concentration value. Use of the exponential form resulted uniformly in superior accuracy to the other techniques.

The size of mesh proved to be effective in producing improved accuracy. The nondimensional transverse step size ΔY was varied from 0.0625 to 0.01 resulting in improvements in accuracy. Similarly step sizes in the flow direction were effective in improving accuracy, but to a lesser degree.

The velocity at the membrane has been interpreted in two ways. In the first interpretation, the velocity at the membrane surface y=0 assumes the Dirichlet notion of the average value of the sum of limit from the left and limit from the right. This notion interprets the velocity as half the center velocity at the edge points of the membrane ribbon. At all other points the velocity will be the center velocity and zero at all positions of the island. In the second interpretation, the velocity at the membrane surface is the limit from the left at all points. The velocity will be thereby zero at the first station of the ribbon and full velocity at all points in the ribbon including the last point of the ribbon, where transition to the island occurs. The net flow is identical, and the results for the solute excess mass flow are only slightly different. The first interpretation tends to smooth the concentration at the wall and thus to be slightly less abrupt in nature. The second probably serves the mathematical statement more faithfully; it has been used for the calculations herein.

RESULTS

A particular case has been evaluated to illustrate the calculations outlined above. It has R/W=1/3, or one-third membrane. The basic calculations have $J\Delta y/D=0.0375$ and a module length $W\mu J^3/\tau D^2=0.000075$. As mentioned already the ratio of WJ/2D is 1/4. For reference purposes this corresponds to a case of flux of $6x10^{-5}$m/s, a diffusion coefficient $2.4x10^{-10}m^2/s$, a shear rate of $1000s^{-1}$, and a module length (pore spacing) of 20 μm. Calculations at 100, 200, and 1000 modules are reported, corresponding to positions of 2mm to 2 cm from the starting point of the membrane edge. Other reference conditions may also correspond to the same non-dimensional variables.

The distribution of concentration at the surface (denoted C_w) of the membrane is shown in Figure 4 for selected modules. The concentration values generally have the expected feature of rising from the start of the modular unit to the end of the unit. This simply reflects the accumulation of solute by the membrane action. A homogeneous membrane case would be expected to have a smooth and steady increase. For the non-homogeneous membrane the concentration declines over the islands as the solute is free to diffuse with no countering bulk motion and rises at the membrane ribbons. As the membrane is exposed to more and more concentrated fluid the relative rise is seen to grow.

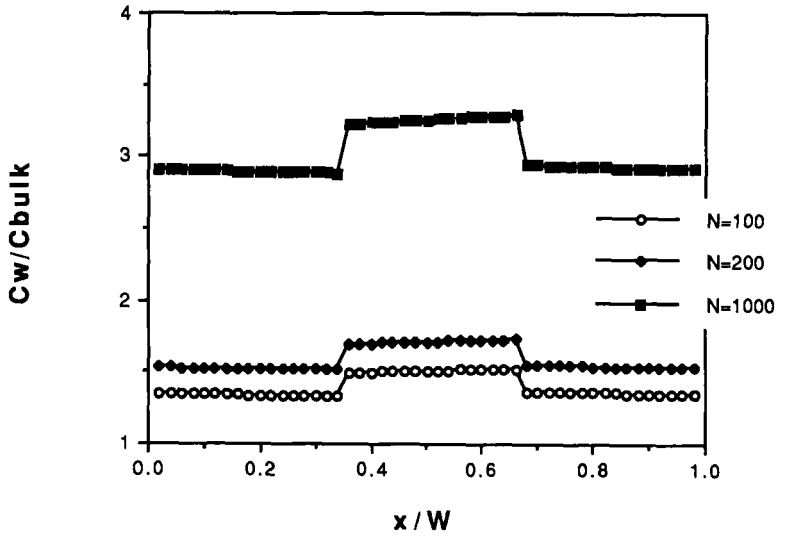


Figure 4 Distributions of Concentration at the Surface of the Membrane

The profiles for concentration at points not on the membrane surface corresponding to the same selected modules are shown in Figure 5. At each module position the profiles at the end of the membrane ribbon and at the end of the inactive island are displayed. These profiles display the local maximum and the local minimum for the concentration at the membrane; the difference between them

shows the rise due to passing over one pore. At the more downstream position, the concentrations are higher and, as noted for the distributions in the preceeding paragraph, increase more in absolute value over the membrane ribbon.

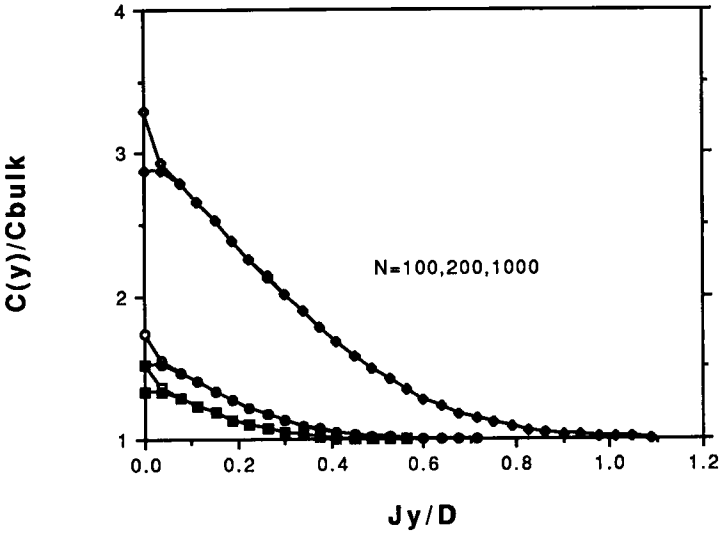


Figure 5 Concentration Profiles at Various Positions

The value of C_w at the most upstream position of each module is designated $C_{entering}$. The distributions of Figure 4 and the difference in the profiles of Figure 5 may be scaled, or normalized, by this local value, $C_{entering}$. By so doing the results of Figure 6 and 7 are obtained. The normalized distribution of concentration over a module is seen to form a universal plot. Also the profiles of concentration at the end of island and end of the membrane ribbon also are caused to collapse into one plot. This observation means that the perturbation in the concentration near one pore is only dependent on the upstream concentration. The extent (thickness) and relative rise of the concentration perturbation are therefore effectively disconnected from the relative position. Suppose one can calculate, using independent methods, the concentration expected upstream of a pore located some distance downstream. The details near that pore may be calculated without calculating the first 1000 pores separately. If the thickness and shape of the

concentration layer outside the locally affected region are the same as for a homogeneous layer, the homogeneous layer model may be used to provide the independent estimate of concentration.

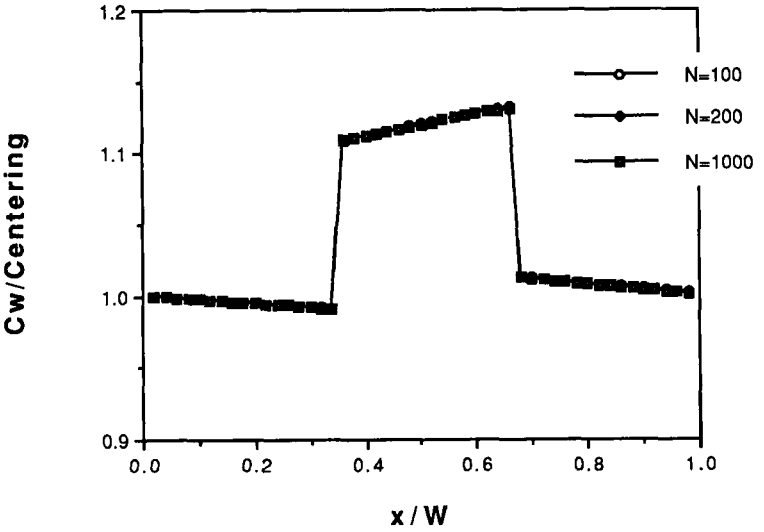


Figure 6 Normalized Distributions of Concentration at the Surface of the Membrane

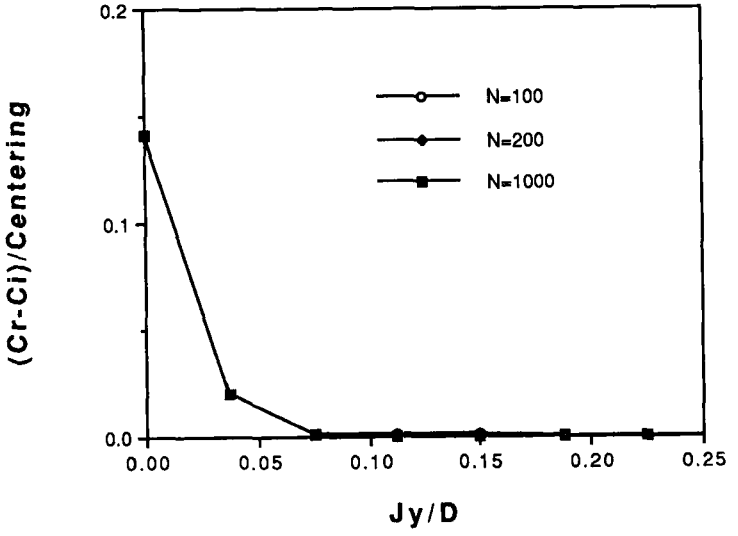


Figure 7 Normalized Concentration Profiles

Formally the result intimated above may be expressed as

$$C(X,Y) = C_s(X,Y) + C_{s,w}(X)\, C'(x/W,Y)$$

Here C_s is a "smooth" concentration function having a local value of concentration at the membrane surface, $C_{s,w}$. The C' function is the perturbation function which has a magnitude and extent which may vary systematically with R/W, or with the non-dimensional value of W.

Not included in the governing equations herein are the effect of osmotic pressure or gel deposit on the surface of the membrane. Such calculations are possible, but involve an interactive computation of the flow and concentration fields. Such an interactive computation has not been entered into here. However, it is possible to speculate concerning a possible effect. Suppose the concentration over a homogeneous membrane were found to have a value midway between the ribbon and island portions of a typical calculation such as in Figure 4. It would have a osmotic resistance based on $1.42*C_B$ at N=100. The mean concentration indicated over the non-homogeneous membrane at N=100 in Figure 4 is $1.5*C_B$. The osmotic pressure is approximately linearly related to the concentration so the difference in the values of osmotic resistance in the two cases is $0.08*C_B$ and approximately 6 percent of the osmotic resistance value. This modification is not very severe. However, inasmuch as some membranes may have porosities of the order of a few percent compared with the 33% case of Figure 4, the effect might be considerably more. The basis for the difference is the increase in the concentration as the flux becomes more focused over a smaller pore.

In summary the results are applicable to real three-dimensional situations by extrapolation from the idealized two-dimensional results shown. These results are clear and valuable from their form as much as from their actual detail.
1. For most membrane situations the longitudinal diffusion term is expected to be negligible, allowing solution by marching methods. Otherwise solutions would be considerably more expensive.

2. Criteria for the relative thickness of the converging flow pattern relative thickness of the converging flow pattern and the diffusive zone are shown to depend on the magnitude of WJ/2D compared to unity. 3. The membrane has an effect on the concentration field causing relatively rapid increases in concentration immediately over the membrane pore, followed by relaxation of the concentration. These details are not shown in any previous publication even though they are easily anticipated. 4. The distribution of concentration along a pore and its adjoining islands is shown to conform to a single function when suitably scaled. The scaling is proportioned according to the local concentration level. Thus the pulse in concentration over any pore is locally scaled, independent of other pores. This important, unanticipated result implies that the local effects surrounding a single pore may be analyzed without solving an entire membrane surface in the manner needed for microscopic analysis as was done here. 5. Though not shown, the enlarged swings in concentration may affect the osmotic resistance and hence the estimate of productivity of a membrane modeled including such effects.

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