Convective Diffusion in Stagnation Flow with an Imperfect Semipermeable Interface

DALE W. ZEH and WILLIAM N. GILL

Clarkson College of Technology, Potsdam, N. Y.

In this note convective diffusion in stagnation flow is discussed in the context of membrane separation processes, reverse osmosis in particular. It is shown that this is a desirable configuration for determining the rejection parameter R, a measure of how imperfect the semipermeable membrane interface is, because the dimensionless concentration function depends only on η.

A simple approximate analytical solution is developed for (w_{sw}/w_{se}) which applies to separation processes in general. For reverse osmosis, $N_{Sc} \simeq 560$ and in this case the approximate results are compared with exact numerical calculations. It is found that the analytical solution is very accurate up to a polarization of $(w_{sw}/w_{se}) \simeq 4$ which constitutes a wide range of practical interest. For completeness, numerical results are reported up to a polarization of about 30.

There has been considerable interest in recent years in the possibility of purifying saline solutions by the reverse osmosis or hyperfiltration process. The process is especially attractive because of its simplicity. A membrane permeable to water but not salt separates water from a saline solution; the saline solution is maintained at a pressure sufficiently high to overcome the osmotic pressure and to cause water to flow through the membrane from the saline side to the pure water side; hence the term *reverse osmosis*. A major problem in this process is that the rejected salt accumulates near the membrane surface, thereby increasing the osmotic pressure and decreasing the driving force for water production.

Several analytical studies have been published which predict the polarization and water production to be expected in parallel plate systems (*1* to *3*, *6*) and in tubes and annuli (*4*). Another system of interest is that of stagnation flow. This system has the useful feature that the defining equations may be expressed in terms of a single independent variable, which greatly simplifies the analysis. More importantly, perhaps, to study incomplete salt rejection, it is not necessary to make an assumption about the behavior of the salt flux at the membrane surface, as is necessary in other geometries, since it is a constant. It will be shown that a very accurate approximate closed-form solution, relating the water flux to the polarization, is easily derived. Incomplete salt rejection by the membrane is easily accounted for in an exact fashion, and a method of experimentally determining membrane properties is described.

ANALYSIS

The coordinate system used is sketched in Figure 1. For dilute isothermal saline solutions it is reasonable to assume that the physical properties and density are constant. The boundary-layer equations describing such a system are, then

$$\frac{\partial u}{\partial x} + \frac{\partial v}{\partial y} = 0 \qquad (1)$$

$$u\frac{\partial u}{\partial x} + v\frac{\partial u}{\partial y} = -\frac{1}{\rho}\frac{\partial P}{\partial x} + \nu\frac{\partial^2 u}{\partial y^2} \qquad (2)$$

$$u\frac{\partial w_s}{\partial x} + v\frac{\partial w_s}{\partial y} = D\frac{\partial^2 w_s}{\partial y^2} \qquad (3)$$

with the boundary conditions

$$y = 0: \quad u = 0, \quad v = v_w, \quad w_s = w_{sw} \qquad y = \infty: \quad u = U_e, \quad w_s = w_{se} \qquad (4)$$

Flow through the membrane is described by

$$-v_w = \frac{A}{1 - w_{sp}}\left[\Delta P - \pi_o\frac{(w_{sw} - w_{sp})}{w_{se}}\right] = A\Delta P\left[1 - B\frac{w_{sw}}{w_{se}}\right] \qquad (5)$$

The mass flux of salt in the y direction is given by

$$n_s = w_s\rho v - \rho D\frac{\partial w_s}{\partial y} \qquad (6)$$

so that at the wall we can write

$$\frac{D}{w_{sw}}\frac{\partial w_s}{\partial y}\bigg|_w = v_w - \frac{n_{sw}}{\rho w_{sw}} = \left[1 - \frac{n_{sw}}{w_{sw}\rho v_w}\right]v_w = Rv_w \qquad (7)$$

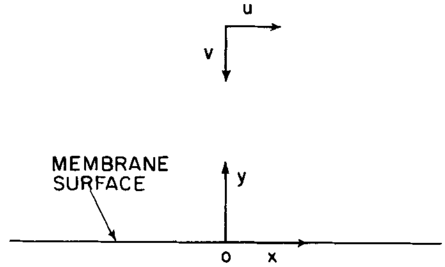

Fig. 1. Coordinate system.

D. W. Zeh is with Jet Propulsion Laboratory, Pasadena, California.

The parameter R will be a measure of the salt rejection and is truly a constant in the present problem. Consequently it is not necessary to speculate about the variation of n_{sw} as one is usually required to do when considering incomplete rejection in other geometries. For an ideal membrane, $n_{sw} = 0$ and $R = 1$.

The following transformations are introduced:

$$u = \frac{\partial \psi}{\partial y}, \quad v = -\frac{\partial \psi}{\partial x}$$

$$\eta = \sqrt{\frac{U_e}{\nu x}}\, y, \quad \psi = \sqrt{U_e \nu x}\; F(\eta), \quad \phi = \frac{w_s}{w_{sw}} \tag{8}$$

For stagnation flow $U_e \propto x$ (5). For practical purposes, this means we must restrict attention to configurations such that, with an ideal fluid, the velocity along the surface varies linearly with distance. This is the case, for example, near the forward stagnation point of a cylinder in crossflow. Equations (1) to (3) then reduce to

$$F''' + FF'' - (F')^2 + 1 = 0 \tag{9}$$

$$\phi'' + N_{Sc} F \phi' = 0 \tag{10}$$

with the boundary conditions

$$\eta = 0: \quad F = F(0), \quad F' = 0, \quad \phi = 1 \qquad\qquad \eta \to \infty: \quad F' = 1, \quad \phi = \phi_e \tag{11}$$

Equation (7) provides another relation to be satisfied at $\eta = 0$:

$$\phi'(0) = -R\, N_{Sc}\, F(0) \tag{12}$$

Twice integrating Equation (10) and letting $\eta \to \infty$, we obtain

$$\phi_e = 1 - R N_{Sc} F(0) \int_0^\infty e^{-N_{Sc}\int_0^\eta F d\eta}\, d\eta$$
$$= 1 - R(1 - \phi_e|_{R=1}) \tag{13}$$

It is clear that with the solution of Equation (13) for $R = 1$ we can obtain solutions for ϕ_e for arbitrary R. Furthermore Equation (13) can be used to develop a relation for R which will prove to be quite useful:

$$R = 1 - \frac{n_{sw}}{w_{sw}\rho v_w} = 1 - \frac{n_{sw}}{n_{tw} w_{sw}} = 1 - \frac{w_{sp}}{w_{se}}\phi_e$$
$$= 1 - \frac{w_{sp}}{w_{se}}\left(1 - R\left(1 - \phi_e|_{R=1}\right)\right) = 1 - \frac{w_{sp}}{w_{sw}} \tag{14a}$$

where w_{sp} is the mass fraction of salt in the product water. Solving for R, we obtain

$$R = \frac{1 - \dfrac{w_{sp}}{w_{se}}}{1 - \dfrac{w_{sp}}{w_{se}}(1 - \phi_e|_{R=1})} \tag{14b}$$

One of the significant aspects of stagnation flow systems is that they should enable one to delineate more precisely the functional dependence of R on water flux and ΔP through Equations (13) and (14).

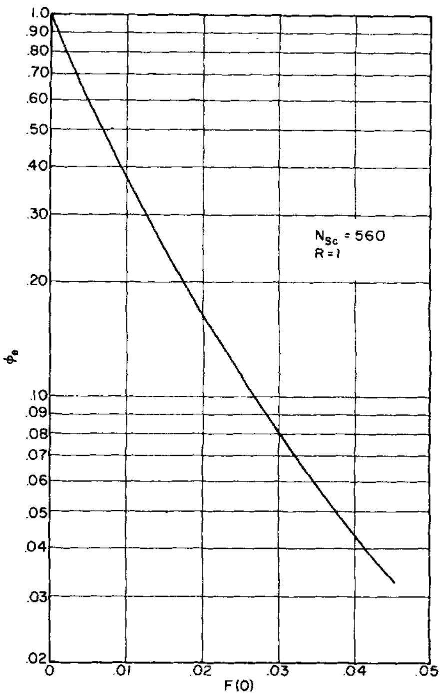

Fig. 2. ϕ_e **vs. F(0) with** $N_{Sc} = 560, R = 1$.

Equations (9), (11), and (13) may be solved directly by a numerical procedure, and exact values of ϕ_e for $R = 1, N_{Sc} = 560$, for $F(0)$ up to 0.045 are provided in Figure 2. However, an accurate yet simple closed form solution, applicable for large N_{Sc}, can be obtained as follows: For large N_{Sc} the diffusion boundary layer is much thinner than the momentum boundary layer, so it is reasonable to approximate F in the diffusion equation by

$$F \cong F(0) + F'(0)\eta + \frac{F''(0)}{2}\eta^2$$
$$\cong F(0) + \frac{F''(0)}{2}\eta^2$$

If one inserts this result in Equation (13) we get

$$\phi_e \cong 1 - R\, N_{Sc} F(0) \int_0^\infty \left[e^{-N_{Sc}F(0)\eta}\, e^{-\frac{N_{Sc}}{6}F''(0)\eta^3} \right] d\eta \tag{15}$$

Now let

$$e^{-N_{Sc}F(0)\eta} = \sum_{i=0}^{\infty} \frac{[-N_{Sc}F(0)]^i}{i!}\eta^i \tag{16}$$

and substitute Equation (16) into (15) to obtain

$$\phi_e \cong 1 + R \sum_{i=0}^{\infty} \left\{ -N_{Sc}^{2/3} F(0) \left(\frac{6}{F''(0)} \right)^{1/3} \right\}^{i+1} \frac{\Gamma\left(\dfrac{i+1}{3}\right)}{3\Gamma(i+1)} \tag{17}$$

It will be shown that a major range of polarizations of interest will correspond to $0 < F(0) < 0.02$. In this range $F''(0)$ is not a strong function of $F(0)$, as shown below:

$F(0)$	$F''(0)$
0	1.2326
0.005	1.2354
0.02	1.2441

This occurs because the small rate of mass transfer through the membrane does not markedly affect the momentum characteristics of the system. Hence it is reasonable to use a constant value of $F''(0)$ equal to 1.233. Equation (17) can be rewritten as

$$\phi_e = 1 + R\sum_{i=0}^{\infty} a_i \{N_{Sc}^{2/3} F(0)\}^{i+1} \qquad (18)$$

$$a_i = (-1)^{i+1}\left\{\frac{6}{F''(0)}\right\}^{\frac{i+1}{3}} \frac{\Gamma\left(\frac{i+1}{3}\right)}{3\Gamma(i+1)}$$

where Γ denotes the gamma function. With $F''(0) = 1.233$ the first seven values of a_i are

$$\begin{aligned} a_0 &= -1.5133 \\ a_1 &= 1.2937 \\ a_2 &= -0.8110 \\ a_3 &= 0.4091 \\ a_4 &= -0.1749 \\ a_5 &= 0.0658 \\ a_6 &= -0.0221 \end{aligned} \qquad (19)$$

One would expect good results with these many terms as long as $F(0) \lesssim N_{Sc}^{-2/3}$. If one lets $N_{Sc} = 560$ and $R = 1$, Equations (18) and (19) yield the following results

$F(0)$	ϕ_e	$\phi_{e\text{Exact}}$
0.001	0.903	0.902
0.005	0.608	0.605
0.1	0.382	0.379
0.015	0.240	0.246
0.020	0.107	0.164

In the above tabulation, the $\phi_{e\text{Exact}}$ were obtained by solving Equation (9), (11), and (13) exactly. The approximate results are thus seen to be very good for $F(0) \leq 0.015$. Since this corresponds to a polarization of $w_{sw}/w_{se} \cong 1/0.25 = 4$, the approximate solution is seen to be valid over a wide range of interest with the seven terms of the series given.

A final relation between the parameters of the system follows from Equation (5):

$$-v_w = \sqrt{\frac{U_e \nu}{x}}\, F(0)\, \frac{A\pi_o}{1 - w_{sp}} \left[\frac{1}{B} - \frac{R}{\phi_e}\right] \qquad (20)$$

Equations (14), (18) (or Figure 2 for $N_{Sc} = 560$), and (20) provide a means of determining experimentally the parameters A and R for a membrane, or, if the parameters are known, of specifying a system.

For example, suppose it is desired to determine experimentally the values of A and R for a membrane. For a given run one would measure ΔP, v_w, w_{sp}, and U_e/x (for example, for a cylinder of radius a in cross flow, $U_e/x = 2U_\infty/a$, where U_∞ is the velocity of the feed stream at a large distance from the stagnation point). The value of $F(0)$ follows from Equation (20), and $\phi_e|_{R=1}$ can be evaluated from Equation (18) or Figure 2. R is then evaluated from Equation (14) and ϕ_e is obtained from Equation (13). Finally, A is then obtained from Equation (20). Note that the difficult task of measuring w_{sw} is avoided and that this value arises as a result of the analysis.

Suppose, on the other hand, A and R are known, and we wish to specify the water production and quality. Equations (20) and (14) provide a direct relation between B and ϕ_e. Then, selecting a feasible operating pressure, that is, selecting B, one obtains a corresponding value of ϕ_e. This in turn fixes $F(0)$, which then via Equation (20) indicates the required value of U_e/x. Equation (14) can then be used to estimate the mass fraction of salt that will appear in the product water. There are of course alternate ways to specify the system.

CONCLUSION

The reverse osmosis process in stagnation flow has been examined and shown to be a straightforward analytical problem. The analysis presented should be useful in both practical applications and in the experimental determination of membrane properties. The approximate analysis given applies to stagnation flow mass transfer processes in general and need not be restricted only to reverse osmosis systems. The only restriction is that the phenomenological law given by Equation (5) must be a valid approximation.

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NOTATION

a = radius of a cylinder
A = membrane constant as defined by Equation (5)
B = $\pi_o/\Delta P$
D = diffusion coefficient
F = stream function as defined in Equation (8)
n_s = mass flux of salt as defined by Equation (6)
n_t = total mass flux through membrane
ΔP = pressure drop across membrane
R = measure of salt rejection by membrane as defined in Equation (14)
N_{Sc} = Schmidt number, ν/D
U_e = velocity at surface of body for a hypothetical inviscid fluid
u = velocity in x direction
v = velocity in y direction
w_s = mass fraction of salt in saline solution
x = distance parallel to surface
y = distance normal to surface
$\sim$ = approximately equal

Greek Letters

ρ = density
π_o = osmotic pressure of original saline solution
ϕ = w_s/w_{se}
ν = kinematic viscosity
η = dimensionless coordinate, $\sqrt{\frac{U_e}{\nu x}}\, y$
ψ = stream function as defined in Equation (8)
Γ = gamma function

Subscripts

e = value in the free stream
p = value in the product solution
w = value at the membrane surface

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