

The interaction between stirring and osmosis. Part 1

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When pure solvent is separated from a solution of non-zero concentration C_b by a semi-permeable membrane, permeable to solvent (water) but not to solute, water flows osmotically across the membrane towards the solution. Its velocity J is given by $J = P\Delta C$, where P is a constant and ΔC is the concentration difference across the membrane. Because the osmotic flow advects solute away from the membrane, ΔC is usually less than C_b , by a factor γ which depends on the thickness of and flow in a concentration boundary layer. In this paper the layer is analysed on the assumption that the stirring motions in the bulk solution, which counter the osmotic advection, can be represented as two-dimensional stagnation-point flow. The steady-state results are compared with those of the standard physiological model in which the layer has a given thickness δ and the osmotic advection is countered only by diffusion. It turns out that the standard theory, although mechanistically inadequate, accurately predicts the value of γ over a wide range of values of the governing parameter $\beta = PC_b\delta/D$ (where D is the solute diffusivity) if δ is given by

$$\delta = 1.59 \left(\frac{D}{\nu}\right)^{\frac{1}{2}} \left(\frac{\nu}{\alpha}\right)^{\frac{1}{2}},$$

where ν is the kinematic viscosity of the fluid and α is the stirring parameter. The final approach to the steady state is also analysed, and it is shown to be achieved in a time scale $(D/\nu)^{\frac{1}{2}}/\alpha k'$ where k' is a dimensionless number whose dependence on β is computed. Moreover, if β exceeds a certain critical value (≈ 10), the approach to the steady state is not monotonic but takes the form of a damped oscillation (in practice, however, β is unlikely to rise significantly above 1). The theory is extended to the case where the solute concentration is non-zero on both sides of the membrane and in that case it is shown that J is bounded as $\beta \rightarrow \infty$.

1. Introduction

All biological membranes, and many synthetic ones, are to some extent semi-permeable; that is, they let through water and other small molecular species, but are impermeable to all solutes whose molecules exceed a certain size. Thus, when two solutions of such larger molecules are separated by a semi-permeable membrane, with a greater concentration of solute on one side than on the other, the fact that the chemical potentials on the two sides will tend to equilibrium causes water to flow across the membrane towards the side with greater solute concentration. This is the phenomenon of osmosis; it still occurs to a certain extent when the solute molecules can cross the membrane but experience more resistance in doing so than the water

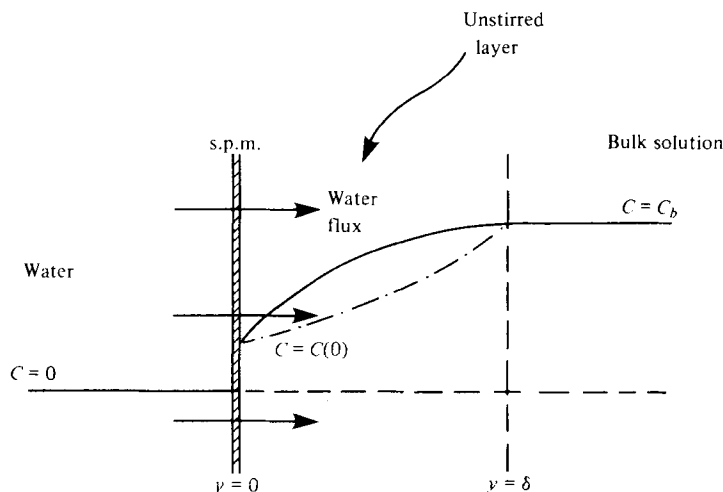


FIGURE 1. Sketch of the 'unstirred layer' separating pure water from a solution in which the solute concentration C far from the semi-permeable membrane (s.p.m.) is C_b . The osmotic water flow advects solute away from the membrane, thereby reducing the concentration at the membrane to a value $C(0)$ less than C_b ; hence the osmotic flux, given by (1.1) with $\Delta C = C(0)$ not C_b , is less than might have been predicted. The standard model treats the unstirred layer as a region of given thickness δ in which the osmotic advection is balanced only by back diffusion; the concentration distribution in the layer would then resemble the dash-dot curve rather than the anticipated solid curve.

molecules. Osmosis occurs not only across the membranes of all living cells but also into and out of organs in the body, such as kidney tubules, blood capillaries, the intestines, etc., whose walls are made up of one or more layers of cells (epithelia); in those cases the pathways followed by water or solute molecules may be rather complicated, involving passage either through or between the cells. Moreover osmosis is often associated with electrokinetic phenomena and with active transport, both important areas of modern biological research.

In order to be able to interpret experiments on these processes one needs to know the 'osmotic permeability' of the membranes concerned. If ΔC is the difference in osmolarity (effectively concentration) across a semi-permeable membrane and if J is the water flux per unit area (i.e. the average velocity) through it, then

$$J = P\Delta C, \quad (1.1)$$

where P is called the osmotic permeability of the membrane (equation (1.1) is based on the assumption that there are no other driving forces for water flux, such as a difference in hydrostatic pressure or in electrostatic potential). According to the standard equations of irreversible thermodynamics (see for example House 1974), P is a constant for an isothermal system, independent of the nature or the concentration of the impermeant solute. For such a membrane one might imagine that it would be quite straightforward to record the solute concentrations in the solutions on the two sides of the membrane, to measure J , and to use (1.1) to infer P .

However, consider an experiment in which a plane semi-permeable membrane separates two chambers, one containing pure water (solute concentration $C = 0$) and

one containing a solution of a non-electrolyte (e.g. sucrose) at concentration $C = C_b$. Then the osmotic flow will itself advect solute away from the membrane, and although this effect will be opposed, by diffusion and by stirring motions in the bulk solution, there will be a reduction in concentration at the membrane itself, a reduction in flow and hence an underestimate of P (figure 1). It is important to be able to predict the extent of this reduction.

The region in which C is reduced below C_b is referred to in the physiological literature as an 'unstirred layer'. The bulk solution in most experiments is stirred in some way, but near the membrane the stirring motion is inevitably parallel to it and is ineffective at mixing the solution. The standard physiological model (first expounded clearly by Dainty 1963) is of a layer of given thickness δ , which may be known from experiments in which the concentration distribution is measured directly (see Lerche 1976 for example). Then the effect of the layer is analysed on the assumptions that the only motion in the layer is the osmotic flux itself and that the solute concentration C in the layer depends only on the distance y from the membrane and on the time (see figure 1). This model is briefly outlined in § 2, but it gives no insight into how δ is actually determined; the model will be applicable only when there is known to be an *unstirred* layer next to the membrane; for example the layer of porous tissue on which many epithelia are mounted (Pedley & Fischbarg 1980). In other circumstances the stirring motions will interact with the osmotic flow in the formation of the layer, a concentration boundary layer. The main purpose of the present paper is to investigate this interaction. In particular we shall ask whether an effective boundary (or unstirred) layer thickness δ can be defined, and whether a formula similar to that derived from the standard model (equation (2.4) below) can be used to assess the effect of the layer on the osmotic flow.

It is clear that the thickness and effect of the concentration boundary layer will be determined by both the strength and the nature of the stirring motions; indeed, this was explicitly recognized by Dainty (1963) before he developed the simple model of § 2. In this paper we choose to represent the stirring motion as a simple stagnation-point flow against the infinite plane $y = 0$ occupied by the semi-permeable membrane. Such a model would be directly applicable in an experiment where the stirring was generated by two counter-rotating stirrers (figure 2*a*). Other types of flow could also be considered (and will be in subsequent papers). One possibility is that the membrane forms part of a solid wall past which the flow (in the absence of osmosis) is unidirectional, as in a tube or in a circular container with a concentric stirrer (figures 2*b*, *c*). Another possibility is that the stirring motion contains random fluctuations (figure 2*d*). Yet another is that the membrane is vertical and the buoyancy force associated with the lower solute concentration in the boundary layer drives a natural convection flow (figure 2*e*; cf. Everitt & Haydon 1969). However, the present model allows for a relatively simple complete solution to both the steady-state and the unsteady problems, and is expected to give results which are qualitatively similar to those of at least some of the other problems.

Theories similar to some of those proposed above have previously been performed in investigations into the desalination of sea-water by the 'reverse osmosis' method. In this method, sea-water is pumped through channels or tubes with semi-permeable walls; the pressure in the sea-water is greater than that in the fresh water outside by an amount that is sufficient to overcome the osmotic pressure difference between the

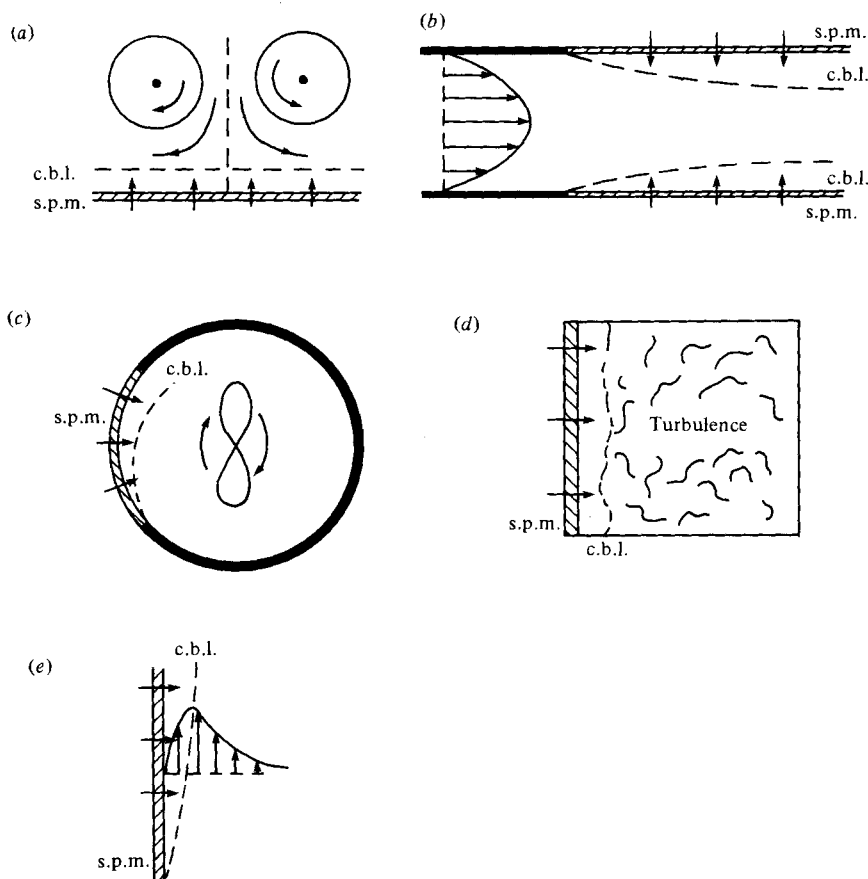


FIGURE 2. Sketches of various ways in which the bulk solution may be stirred so as to determine the concentration boundary layer (c.b.l.) next to the semi-permeable membrane (s.p.m.). (a) Stagnation-point flow, treated in this paper; (b) pipe or channel flow; (c) a single concentric stirrer; (d) random motions (turbulence); (e) natural convection driven by the relative buoyancy of the solute-poor fluid near the membrane.

two fluids, so an outward water flux takes place. Unfortunately, however, solute builds up in the concentration boundary layer ('concentration polarization'), increasing the concentration difference across the membrane, and inhibiting the waterflow. Such systems have been analysed by (among others) Hendricks & Williams (1971), Derzansky & Gill (1974) and Johnson & Acrivos (1969). However, these authors all considered the case of flow *towards* the membrane, causing a build-up of solute; the nonlinear nature of the fluid-mechanical problem means that the results are not directly applicable to the present case in which solute is swept away from the membrane by the osmosis. Furthermore, no one as far as I know has considered the case of stagnation-point flow, in which many details can be derived analytically.

2. The standard 'unstirred layer' model

According to this model, the solute concentration $C(y, t)$ in the unstirred layer ($0 \leq y \leq \delta$) satisfies the one-dimensional convection-diffusion equation, together with the boundary conditions that the membrane is impermeable to solute and that

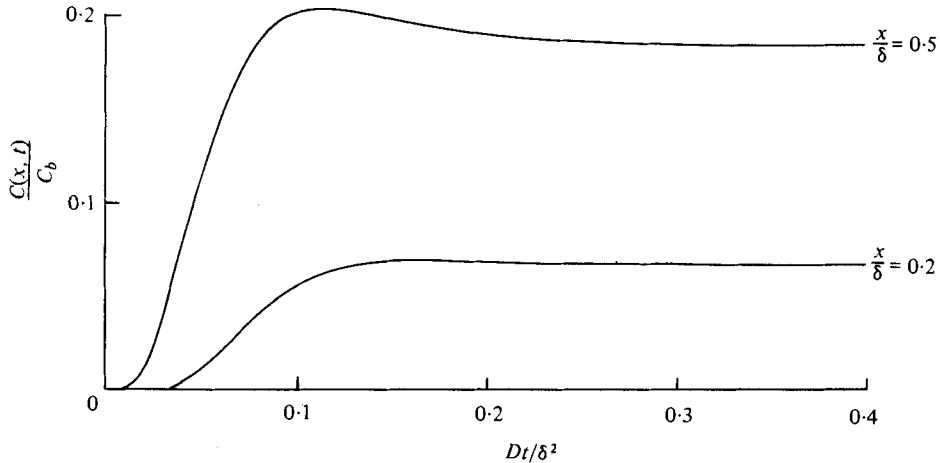


FIGURE 3. Solution of the initial-value problem defined by (2.1) to (2.3) with $C(y, 0) = 0$, in the case where $\beta = 100$. $C(x, t)/C_b$ is plotted against Dt/δ^2 for two values of x/δ . Note how the concentration overshoots its final steady-state value. (I am grateful to Dr D. L. S. McElwain for computing these results.)

$C(\delta, t) = C_b$; J is given by (1.1) with $\Delta C = C(0, t)$. There should also be an initial condition such as $C(y, 0) = 0$ ($0 \leq y \leq \delta$), which would apply if the concentration at $y = \delta$ were suddenly raised to the value C_b at time $t = 0$.

After an initial adjustment, the concentration distribution, and hence J , settles down to a steady state, in which

$$C = C_S(y) = C_b \exp[(J_S/D)(y - \delta)], \tag{2.1}$$

where D is the solute diffusivity, while (1.1) gives the steady water flux J_S :

$$J_S = PC_b \exp(-J_S \delta/D). \tag{2.2}$$

This is the solution that was given by Dainty (1963). Pedley & Fischbarg (1978) expressed it in dimensionless form, writing

$$\beta = PC_b \delta/D, \quad \gamma = J_S/PC_b = C_S(0)/C_b, \tag{2.3}$$

so that (2.2) became

$$\gamma = e^{-\beta\gamma}. \tag{2.4}$$

Here β is a parameter which is known when the system is known, while γ represents the derived osmotic flux; γ is equal to the ratio of the actual osmotic flux to the flux which would be expected if (1.1) were used with $\Delta C = C_b$. In an experiment one would measure J and infer P ; in terms of the dimensionless parameters this is equivalent to measuring the product $\beta\gamma$ and using (2.4) to infer γ (and hence β).

The steady concentration distribution in the layer has commonly been supposed to resemble the solid curve in figure 1 (see Dainty 1963, for example). However, the distribution (2.1) predicted by this model actually resembles the dash-dot curve in figure 1, with steepest concentration gradient at the outer edge of the layer not the wall. The present model does give a distribution resembling the solid curve.

The unsteady, initial-value problem has been solved numerically by Schafer, Patlak & Andreoli (1974); the time scale t_s for the decay of perturbations to the steady state

($\propto \delta^2/D$) was obtained analytically by Pedley & Fischbarg (1978), who made the interesting prediction that if β is large enough (greater than about 27) the approach to the steady state is not monotonic but involves an overshoot. This was not revealed by Schafer *et al.* (1974), but some more recent computations by Dr D. L. S. McElwain have confirmed it (figure 3). As Pedley & Fischbarg (1978) point out, the values of β appropriate to cell membranes are likely to be rather small (< 0.1 , say) so that the oscillatory behaviour is unlikely to be observed and γ would not fall below about 0.9, indicating an error of no more than 10% in estimating P . However, if the membrane is a (leaky) epithelium and sucrose the solute, β can rise as high as 7.5 ($\gamma = 0.21$), and in carefully controlled laboratory conditions it may be possible to raise it still higher.

3. Formulation and non-dimensionalization

The membrane is again taken to occupy the (infinite) plane $y = 0$. In the absence of osmosis the motion is taken to be two-dimensional stagnation-point flow so that far from the plane the velocity field is given by

$$u \sim \alpha x, \quad v \sim -\alpha y \quad \text{as } y \rightarrow \infty, \quad (3.1)$$

where x is measured along the plane, (u, v) are the velocity components in the (x, y) directions respectively, and α is a constant (the stirring parameter). The velocity field satisfies the unsteady two-dimensional Navier–Stokes equations for an incompressible viscous fluid, and the concentration $C(x, y, t)$ of solute satisfies

$$C_t + uC_x + vC_y = D(C_{xx} + C_{yy}). \quad (3.2)$$

The boundary conditions on u, v at the wall are

$$u = 0, \quad v = J(x, t) \quad \text{at } y = 0,$$

where $J(x, t)$ is once more the osmotic flux per unit area, given in terms of $C(x, 0, t)$ by

$$J(x, t) = PC(x, 0, t). \quad (3.3)$$

The boundary conditions on C are now

$$C \rightarrow C_b \quad \text{as } y \rightarrow \infty \quad \text{and} \quad JC - DC_y = 0 \quad \text{at } y = 0; \quad (3.4)$$

the second of these represents the impermeability of the wall to solute. An initial condition is also required for a fully posed initial-value problem.

The advantage of choosing stagnation-point flow is that the solution for v in the absence of osmosis is independent of x , so it is possible for the normal velocity at the wall, J , also to be independent of x without affecting the x dependence of u and v ; C too can now be independent of x . We accordingly eliminate x and non-dimensionalize the variables as follows:

$$\begin{aligned} u &= \alpha x f'(\eta, \tau), & v &= -(\alpha\nu)^{\frac{1}{2}} f(\eta, \tau), & C &= C_b g(\eta, \tau), \\ \eta &= (\alpha/\nu)^{\frac{1}{2}} y, & \tau &= \alpha t, \end{aligned}$$

where ν is the kinematic viscosity of the fluid and a prime denotes differentiation with respect to η . The problem is then transformed into the following pair of differential equations,

$$f''' + ff'' + 1 - f'^2 = f'_\tau, \tag{3.5a}$$

$$g'' + \sigma fg' = \sigma g_\tau, \tag{3.5b}$$

where $\sigma = \nu/D$ is the Schmidt number of the fluid; the boundary conditions are

$$f'(\infty, \tau) = 1, \quad f'(0, \tau) = 0, \quad f(0, \tau) = -V(\tau), \tag{3.6a}$$

$$g(\infty, \tau) = 1, \quad \sigma V(\tau)g(0, \tau) = g'(0, \tau), \tag{3.6b}$$

where

$$V(\tau) = (\alpha\nu)^{-\frac{1}{2}}J(t).$$

The final relation stems from (3.3), which becomes

$$\sigma V(\tau) = \beta_v g(0, \tau) \tag{3.7}$$

where

$$\beta_v = \frac{PC_b}{D} \left(\frac{\nu}{\alpha}\right)^{\frac{1}{2}}. \tag{3.8}$$

We may note that $(\nu/\alpha)^{\frac{1}{2}}$ is the length-scale for the viscous boundary-layer thickness in standard stagnation-point flow, so β_v is similar to β (equation 2.3) but with this scale in place of δ . To achieve a quantity truly analogous to β we should replace δ by a scale for the concentration boundary-layer thickness, δ_c . In the absence of osmosis and in the steady state this is given by a balance between the advection term vC_y of (3.2) and the diffusion term DC_{yy} . Now for all solutes of interest D is much smaller than ν (so $\sigma \gg 1$) with the consequence that the concentration boundary layer is much thinner than the viscous, and is confined to the inner part of it where

$$v \propto \alpha^{\frac{1}{2}}\nu^{-\frac{1}{2}}y^2$$

(when $J = 0$). The balance between advection and diffusion then gives

$$\delta_c = \sigma^{-\frac{1}{2}}(\nu/\alpha)^{\frac{1}{2}}, \tag{3.9}$$

so that a dimensionless parameter more appropriate than β_v may, in the case where osmosis is sufficiently weak, be

$$\beta' = \sigma^{-\frac{1}{2}}\beta_v. \tag{3.10}$$

Furthermore, this means that dimensionless y and t variables more appropriate than η and τ in solving for the concentration distribution may be

$$\zeta = y/\delta_c = \sigma^{\frac{1}{2}}\eta \quad \text{and} \quad \tau' = Dt/\delta_c^2 = \sigma^{-\frac{1}{2}}\tau. \tag{3.11}$$

Again, it should be remembered that these will be appropriate only when $\sigma \gg 1$ and when the osmosis is in some sense weak; however we shall see in §4 that the use of (3.11) will be justified for values of β' up to at least 10^3 .

In the steady state, we shall see that this regime corresponds to values of osmotic flux $V = O(\sigma^{-\frac{3}{2}})$. When β' is much larger, and hence so is V , the layer of greatest concentration gradient is no longer adjacent to the wall. The osmosis advects solution of very low concentration away from the wall, while the outer stagnation-point flow advects solution of concentration C_b towards the wall. At the location where the

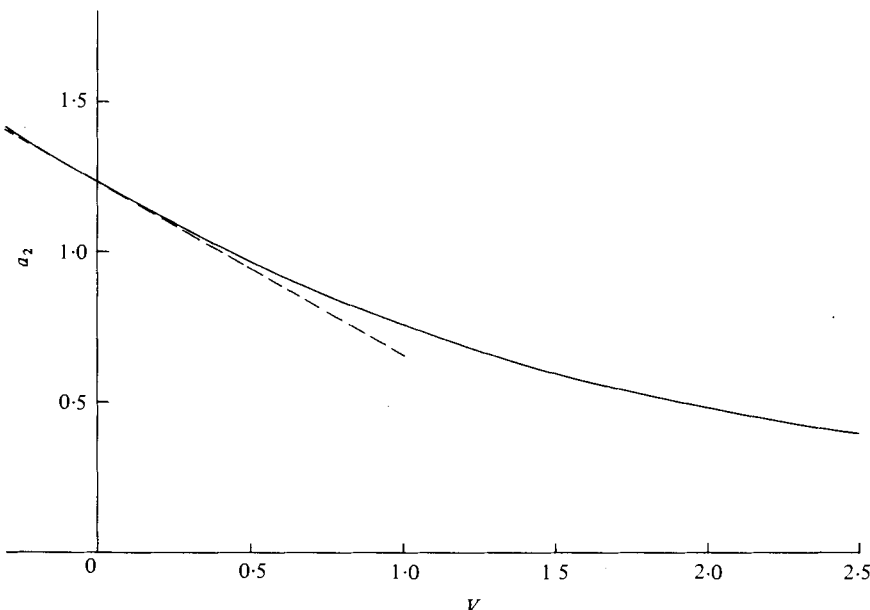


FIGURE 4. Plot of $a_2 = f''(0)$ against V . Broken line is the linear approximation (4.1).

normal velocity v is zero (say $\eta = \eta_0$), therefore, there is a region where the concentration gradient is large. The thickness δ_c of this region turns out to be $O[\sigma^{-\frac{1}{2}}(\nu/\alpha)^{\frac{1}{2}} V^{-\frac{1}{2}}]$ while V is still small ($\sigma^{-\frac{3}{2}} \ll V \ll 1$), and to be $O[\sigma^{-\frac{1}{2}}(\nu/\alpha)^{\frac{1}{2}}]$ when V is $O(1)$ or larger. The values of η_0 in the corresponding regimes are $O(V^{\frac{1}{2}})$ when $\sigma^{-\frac{3}{2}} \ll V \ll 1$, $O(1)$ when $V = O(1)$ and $O(V)$ when $V \gg 1$.

In § 4 we describe the eventual steady-state solution: first the flow field is calculated for arbitrary values of V , and then the concentration field is computed. In § 5 we perform a linearized time-dependent analysis to determine how and over what length of time the eventual steady state is achieved.

4. The steady solution

The flow field

In the steady state the right-hand sides of (3.5) are absent, so the dimensionless stream function is given by the same equation as in the absence of osmosis $f''' + ff'' + 1 - f'^2 = 0$ but with the modified boundary condition (3.6a), where V is a constant. This is therefore the problem of stagnation-point flow with blowing or suction, which was solved numerically by Schlichting & Bussman (1943); the case of blowing ($V > 0$) is the one posed here, whereas the case of suction is more relevant to reverse osmosis. The flow parameters of greatest importance for the concentration distribution are the value of η at which $f = 0$ (η_0), the value of u/x at that η ($a_1 = f'(\eta_0)$) and, especially, the shear stress on the wall, proportional to $a_2 = f''(0)$; a_2 is plotted against V in figure 4.

When V is small, the flow can be represented as a perturbation expansion about the $V = 0$ solution:

$$f(\eta) = \tilde{f}_0(\eta) + V\tilde{f}_1(\eta),$$

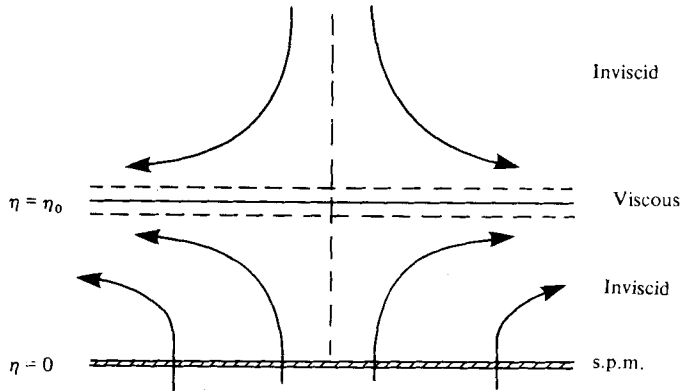


FIGURE 5. Structure of the flow field when $V \gg 1$. Vorticity is confined to a thin layer, of dimensionless thickness 1, around $\eta = \eta_0 = \frac{1}{2}\pi V$, where the normal velocity v is zero.

where $f_0(\eta)$ (the Hiemenz function) and its derivatives are tabulated in Rosenhead (1963, p. 232). The function f_1 and others like it can be derived by the numerical solution of linear ordinary differential equations, and for $V \ll 1$ we obtain

$$a_2 \approx 1.233 - 0.575V. \tag{4.1}$$

Higher-order terms have not been computed because the full nonlinear problem is not difficult to solve numerically.

When V is large (strong blowing) the solution is more interesting (Pretsch 1944; Proudman 1960), in that the blowing advects fluid away from the wall, while the outer stagnation-point flow advects it towards the wall. This means that viscous effects are confined to the neighbourhood of the value of η at which the normal velocity vanishes, i.e. at which $f = 0$ (figure 5). Suppose $f(\eta) = 0$ at $\eta = \eta_0$; then the inviscid flow outside the viscous layer is given by

$$f = \eta - \eta_0 \quad \text{for } \eta > \eta_0 \tag{4.2a}$$

and by

$$f = b \sin [(\eta - \eta_0)/b] \quad \text{for } \eta < \eta_0, \tag{4.2b}$$

where b is a constant (this is the relevant solution of $ff'' - f'^2 + 1 = 0$). The boundary condition $f'(0) = 0$ requires that $\eta_0 = \frac{1}{2}\pi b$, and then the condition $f(0) = -V$ shows that $b = V$. The values of a_1 and a_2 in this limit are thus 1 and $1/V$ respectively. We may note that there is continuity of f , f' and f'' at $\eta = \eta_0$ for the solutions (4.2a, b), so the boundary layer there will be a weak one; the viscous solution in the boundary layer is given by

$$f'' = -\frac{1}{2V^2} \left\{ \eta' \operatorname{erfc}(\eta'/\sqrt{2}) + \sqrt{\frac{2}{\pi}} e^{-\frac{1}{2}\eta'^2} \right\},$$

where

$$\eta' = \eta - \eta_0.$$

The concentration distribution

The general solution of the steady form of (3.5b) that satisfies the outer boundary condition $g(\infty) = 1$ is

$$g(\eta) = 1 - A \int_{\eta}^{\infty} \exp \left[-\sigma \int_0^{\eta'} f(s) ds \right] d\eta', \tag{4.3}$$

where A is a constant. Then the osmotic flow relation (3.7) and the second of the boundary conditions (3.6b) give

$$A = \sigma^2 V^2 / \beta_v \quad \text{and} \quad \beta_v = \sigma V(1 + \sigma VI), \quad (4.4)$$

where

$$I = \int_0^\infty \exp \left[-\sigma \int_0^\eta f(s) ds \right] d\eta. \quad (4.5)$$

As in §2, the quantity of greatest interest is the ratio of the actual osmotic flux to the flux that would be predicted if (1.1) were used with $\Delta C = C_b$: that is

$$\gamma = \frac{J}{PC_b} = \frac{\sigma V}{\beta_v} = g(0) = (1 + \sigma VI)^{-1}. \quad (4.6)$$

From this system of equations we can in principle predict γ as a function of β_v (cf. equation (2.4)). However, in an experiment the physical properties of the solvent and solute would be known, and from a measurement of J we would wish to derive the value of P ; in dimensionless terms we would know σ and V , and seek to predict β_v . In view of the fact that $f(\eta)$ depends on V , and hence I depends on σ and V , this is also the most convenient way to derive the theoretical results: given V and σ , deduce β_v and γ .

Given $f(\eta)$ from the flow field solution, it is in principle a simple matter to calculate I numerically from (4.5). However, since σ is always very large in practice it is both easier and more illuminating to perform an asymptotic analysis of the integral. Standard methods show that the dominant contribution to the integral in (4.5) (and in (4.3)) comes from the neighbourhood of $\eta = \eta_0$, where $f(\eta_0) = 0$, as predicted in §3. If V is not very small, so that η_0 is not too close to zero, we find that

$$I = \left(\frac{\pi}{2\sigma a_1} \right)^{\frac{1}{2}} e^{-\sigma F_0} \{1 + \operatorname{erf} Z + O[a_2(\sigma a_1^3)^{-\frac{1}{2}}]\}, \quad (4.7)$$

where

$$F_0 = \int_0^{\eta_0} f(\eta) d\eta \quad \text{and} \quad Z = \eta_0 \left(\frac{\sigma a_1}{2} \right)^{\frac{1}{2}}.$$

The dimensionless thickness of the layer of high concentration gradient around $\eta = \eta_0$ can be seen to be $O(\sigma^{-\frac{1}{2}})$, as long as $a_1 = O(1)$. When $V \gg 1$, so that $\eta_0 \approx \frac{1}{2}\pi V$, $a_1 \approx 1$, $a_2 \approx 1/V$ and $F_0 \approx -V^2$, equation (4.7) yields

$$I = \left(\frac{2\pi}{\sigma} \right)^{\frac{1}{2}} e^{\sigma V^2} [1 + O(1/V\sigma^{\frac{1}{2}})], \quad (4.8)$$

with the consequence that γ , from (4.6), is very small and β_v (4.4) is enormous. Such values are unlikely to be realistic.

When $V \ll 1$, η_0 is small and f can therefore be expanded in powers of η :

$$f(\eta) \sim -V + \frac{a_2}{2}\eta^2 + O(\eta^3), \quad (4.9)$$

where $a_2 \approx 1.233$ from (4.1). Thus we have

$$\eta_0 \approx (2V/a_2)^{\frac{1}{2}}, \quad a_1 \approx (2Va_2)^{\frac{1}{2}} \quad \text{and} \quad F_0 \approx -\frac{2}{3}V^{\frac{3}{2}} \left(\frac{2}{a_2} \right)^{\frac{1}{2}},$$

so that (4.7) becomes

$$I \approx \frac{1}{2} \left(\frac{\pi}{\sigma}\right)^{\frac{1}{2}} \left(\frac{2}{Va_2}\right)^{\frac{1}{2}} \exp\left\{\frac{2}{3} \sigma V^{\frac{1}{2}} \left(\frac{2}{a_2}\right)^{\frac{1}{2}}\right\} \{1 + \operatorname{erf}[\sigma^{\frac{1}{2}} V^{\frac{1}{2}} (2/a_2)^{\frac{1}{2}}] + O(\sigma^{-\frac{1}{2}} V^{-\frac{1}{2}})\}. \quad (4.10)$$

This expansion clearly breaks down when $V = O(\sigma^{-\frac{2}{3}})$, which is also the value at which the exponential in (4.10) becomes $O(1)$ and the error function ceases to be approximately equal to 1. At this value, too, $I = O(\sigma^{-\frac{1}{3}})$ and $\sigma VI = O(1)$, so that γ is now $O(1)$; $\beta_v = O(\sigma^{\frac{1}{3}})$ is still large.

A new expansion procedure is thus required when $V = O(\sigma^{-\frac{2}{3}})$. A rescaling of the variables on the lines envisaged in §3 makes the procedure less cumbersome. We introduce (cf. (3.10))

$$\beta' = \sigma^{-\frac{1}{3}} \beta_v, \quad V' = \sigma^{\frac{2}{3}} V, \quad I' = \sigma^{\frac{1}{3}} I, \quad (4.11)$$

which are all expected to be $O(1)$, and from (4.4) and (4.6) we obtain

$$\beta' = V'(1 + I'V')^{-1}, \quad \gamma = (1 + I'V')^{-1}. \quad (4.12)$$

The integral (4.5) can again be approximated using (4.9) for $f(\eta)$, and since η_0 is so small, of $O(\sigma^{-\frac{1}{3}})$, it becomes simpler to expand about $\eta = 0$ instead of $\eta = \eta_0$. Introducing $\zeta = \sigma^{\frac{1}{3}} \eta$ (cf. (3.11)) the integral becomes

$$I' = \int_0^\infty \exp\left[V'\zeta - \frac{a_2}{6} \zeta^3\right] d\zeta + O(\sigma^{-\frac{2}{3}}),$$

which can be denoted by

$$I' \approx \pi \left(\frac{2}{a_2}\right)^{\frac{1}{2}} \operatorname{Hi}\left[V'(2/a_2)^{\frac{1}{2}}\right] \quad (4.13)$$

(Abramowitz & Stegun 1964, p. 448). $\operatorname{Hi}(z)$ can be represented as an integral of Airy functions and it is readily shown that

$$\operatorname{Hi}(z) \sim \begin{cases} \frac{\Gamma(\frac{1}{3})}{3^{\frac{2}{3}}\pi} \left\{1 + \frac{2\pi z}{3^{\frac{1}{3}}\Gamma^2(\frac{1}{3})} + \frac{3^{\frac{2}{3}}z^2}{2\Gamma(\frac{1}{3})}\right\} & \text{as } z \rightarrow 0, \\ (\pi^2 z)^{-\frac{1}{2}} \exp\left(\frac{2}{3}z\right)^{\frac{3}{2}} & \text{as } z \rightarrow \infty. \end{cases} \quad (4.14)$$

$$\quad (4.15)$$

Thus, as $V' \rightarrow \infty$,

$$I' \sim \left(\frac{2\pi^2}{V'a_2}\right)^{\frac{1}{2}} \exp\left\{\frac{2}{3} V'^{\frac{1}{2}} (2/a_2)^{\frac{1}{2}}\right\}, \quad (4.16)$$

which is the same as (4.10) with $\operatorname{erf}[\] = 1$. The whole range of V is now covered: (4.13) must be used for $V = O(\sigma^{-\frac{2}{3}})$ ($V' = O(1)$), while (4.16) should be used for $\sigma^{-\frac{2}{3}} \ll V \ll 1$, and the full expression (4.7) is needed for $V = O(1)$, reducing to (4.8) when $V \gg 1$.

The above analysis shows that it is only when V is as small as $O(\sigma^{-\frac{2}{3}})$ that the scaling appropriate to a concentration boundary layer in the absence of osmosis, with thickness δ_c given by (3.9), is appropriate. For larger V the layer has dimensionless thickness $O(\sigma^{-\frac{1}{3}})$ (or $O(\sigma^{-\frac{1}{3}} V^{-\frac{1}{2}})$ when $V \ll 1$) and is centred on $\eta = \eta_0$.

The results of this analysis are presented in the form of a graph of γ against β' (log scale) in figure 6. Values of V' are also shown on the abscissa; it can be seen that V' is only 3 for a value of β' in excess of 1000, corresponding to a value of γ of about 0.005. Over this range of parameters, then, it is evident that V' is not large, and (4.13) was used to plot the solid curve on figure 6. The broken curves on figure 6 are the

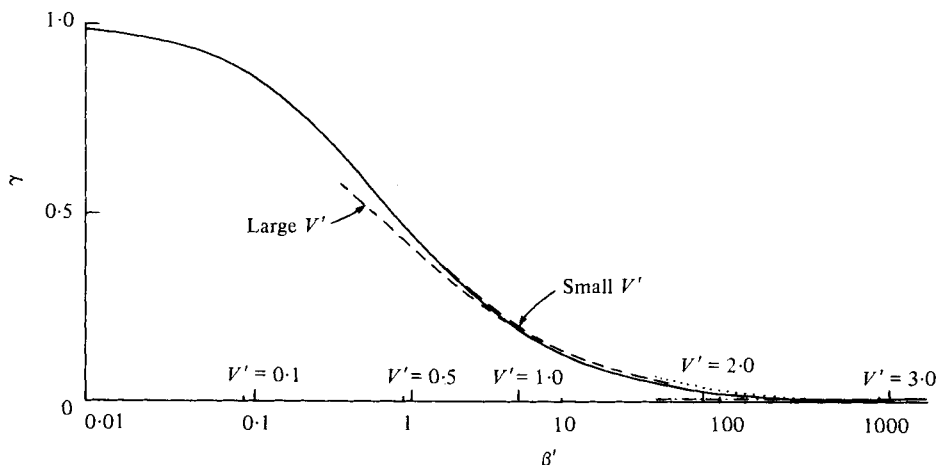


FIGURE 6. Graph of γ against β' , as computed from (4.12) and (4.13); values of V' are also marked on the abscissa. The broken curves are those obtained by using (4.14) to evaluate (4.13) at small values of V' , and by using (4.15) at large V' . The dash-dot curve and the dotted curve represent the asymptotic expansion (4.17), and its leading term, respectively.

approximate results for small and large V' , obtained by using (4.14) and (4.15) respectively as approximations to the function $\text{Hi}(z)$. It can be seen that together they give an extremely good overall approximation, and there would be little loss of accuracy if the small V' approximation were used for $V' < 1$ ($\beta' \lesssim 5.3$) and the large V' approximation for $V' > 1$. Since the result for large V' is the same as that for small V' , (4.10) with $\text{erf}[\] = 1$, the only possible source of inaccuracy in the solid curve of figure 6 is the fact that a_2 is not identically equal to 1.233. The results were recalculated with a_2 given by (4.1) and with V evaluated from V' on the assumption that $\sigma = 10^3$ (this is a realistic value; see § 6); figure 6 was not perceptibly changed.

Finally, we may note that the large V' expansion of I' in (4.16) can be inverted and used with (4.12) to give an asymptotic expansion for γ as a function of β' when β' is large (but not so large that (4.10) is invalid). When a_2 is put equal to 1.233 this expansion is

$$\gamma = \frac{1.115 (\log \beta')^{\frac{3}{2}}}{\beta'} \left\{ 1 - \frac{1}{\log \beta'} (2.128 + \frac{7}{9} \log \log \beta') + O \left[\frac{[\log \log \beta']}{(\log \beta')^2} \right] \right\}. \quad (4.17)$$

This expression is also plotted on figure 6 (the dash-dot curve), where it is seen to be considerably less useful for moderate V' than that obtained from the full large- V' expression (4.16). The leading term of (4.17) is plotted as a dotted curve; the curve representing the full solution passes between these two asymptotic representations.

5. The time-dependent problem

It is to be expected that the steady-state solution of the previous section will eventually be realized whatever the initial conditions, at least assuming that the stirring remains steady. The main question of experimental interest concerns the length of time required to establish the steady state. Suppose that the stirring is turned on and then, at time $t = 0$, solute is introduced into the stirred chamber so

that $C = C_b$ at some finite distance from the membrane. The development of the steady-state concentration distribution will then take place over two or three time scales. There will first be a rapid adjustment as solute is advected to the wall by the stirring motion; the relevant, convective time scale is α^{-1} . Then osmotic flow will begin and there may be a further convective period, as the diffusion layer around $\eta = \eta_0$ is set up, followed by a more gradual, diffusive settling down to the steady state. The time scales for these two stages are $\alpha^{-1}\eta_0/V$ for the first ($= O(\alpha^{-1})$ when $V \geq O(1)$; $= O(\alpha^{-1}V^{-\frac{1}{2}})$ when $V \ll 1$) and δ_c^2/D , where δ_c is the concentration layer thickness, for the second. In the most relevant case, in which $V = O(\sigma^{-\frac{1}{2}})$ and δ_c is given by (3.9), these two time scales merge into one, of $O(\alpha^{-1}\sigma^{\frac{1}{2}})$. What multiple of the diffusive time scale must be allowed before the steady state may be assumed can be deduced from a linearized theory in which the dependent variables are represented as small perturbations about their steady-state values.

The time-dependent problem is defined by equations (3.5) to (3.7) (plus an initial condition). If we represent the steady-state solutions of § 4 by the suffix zero, we can write

$$\begin{aligned} f(\eta, \tau) &= f_0(\eta) + f_1(\eta, \tau), \\ g(\eta, \tau) &= g_0(\eta) + g_1(\eta, \tau), \\ V(\tau) &= V_0 + V_1; \end{aligned}$$

substituting into the equations and linearizing, we obtain a homogeneous linear problem. This suggests that the solution may be represented as the sum of an infinite number of exponentially decaying terms of the form

$$f_1(\eta, \tau) = \bar{V}_1 F(\eta) e^{-k\tau}, \quad g_1(\eta, \tau) = \bar{V}_1 G(\eta) e^{-k\tau}, \quad V_1 = \bar{V}_1 e^{-k\tau},$$

where \bar{V}_1 is an arbitrary constant whose value will depend on the initial conditions but has no effect on the determination of k . We therefore assume such a form, and derive the following set of equations and boundary conditions (where a prime denotes $d/d\eta$):

$$\left. \begin{aligned} F''' + f_0 F'' - 2f_0' F' + f_0'' F &= -kF', \\ F(\infty) = F'(0) = 0, \quad F(0) &= -1; \end{aligned} \right\} \tag{5.1}$$

$$\left. \begin{aligned} \frac{1}{\sigma} G'' + f_0 G' + g_0' F &= -kG, \\ G(\infty) = 0, \quad 2\sigma V_0 G(0) &= G'(0), \quad G(0) = \sigma/\beta_v; \end{aligned} \right\} \tag{5.2}$$

in the derivation of this the relation $\sigma V_0 = \beta_v g_0(0)$, from (3.7), has been used. The problem defined by (5.1) and (5.2) is clearly an eigenvalue problem for k ; the final approach to the steady state will take place at a rate defined by the eigenvalue with smallest real part. We accordingly concentrate on evaluating this eigenvalue, for various values of σ and of β_r .

The first thing to notice is that the problem for G influences the problem for F only through the term involving k in (5.1). Thus, as long as k is not very large, the length scale for variations of F will be the same as the length scale for variations of f_0 . This means that η is the appropriate variable for F ; in particular, we will have

$$F(\eta) \approx -1 + \frac{1}{2}\alpha_2(k)\eta^2 \tag{5.3}$$

as $\eta \rightarrow 0$, where α_2 is a constant, depending on k , which remains $O(1)$ as long as k is not large.

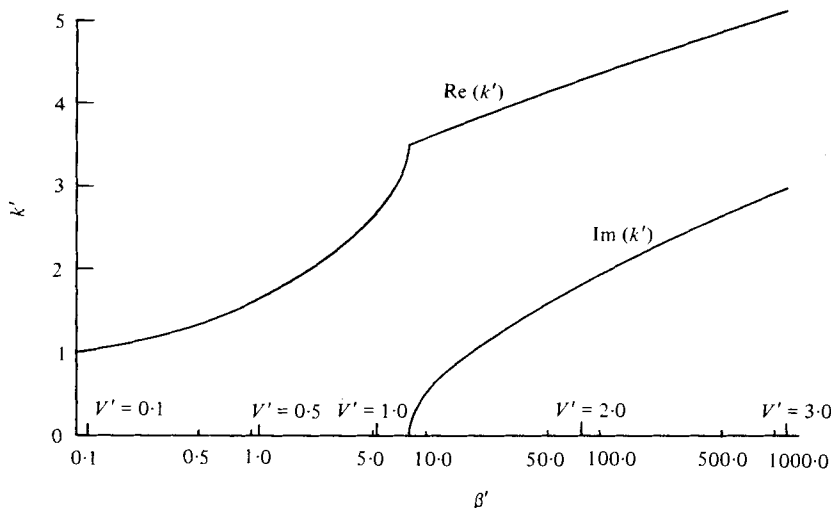


FIGURE 7. Graphs of the real and imaginary parts of the eigenvalue k' with smallest real part, against β' , computed from (5.4); values of V' are also marked on the abscissa.

Now we turn to the problem for $G(\eta)$. As in the previous section we assume that σ is large, and in view of those results (figure 6) we further restrict attention to $O(1)$ values of $V' = \sigma^{\frac{2}{3}}V_0$. This means that the dimensionless length scale for concentration variations is $\sigma^{-\frac{1}{3}}$, so that the independent variable should be ζ as defined in (3.11); this has the further implication that the appropriate time variable is τ' rather than τ . We therefore introduce the corresponding inverse time constant k' , where

$$k = \sigma^{-\frac{1}{3}}k';$$

hence k is expected to be small and (5.3) to be valid. Indeed, with this scaling, (5.3) gives

$$F(\eta) \approx -1 + \frac{1}{2}\sigma^{-\frac{2}{3}}\alpha_2\zeta^2 \approx -1,$$

so that the dependence of F on k has no effect on the determination of G .

If we make one further substitution, writing $G(\zeta) = (\sigma^{\frac{2}{3}}/\beta')H(\zeta)$, the problem reduces to the following:

$$\begin{aligned} H_{\zeta\zeta} + (-V' + \frac{1}{2}\alpha_2\zeta^2)H_{\zeta} + k'H &= V'^2 \exp(V'\zeta - \frac{1}{6}\alpha_2\zeta^3), \\ H(\infty) &= 0, \quad H(0) = 1, \quad H_{\zeta}(0) = 2V'. \end{aligned} \quad (5.4)$$

The objective is to calculate the eigenvalue k' with lowest real part for a range of values of V' , which should at least include $0 \leq V' \leq 3$ in order that all the β' values of figure 6 may be covered.

The problem defined by (5.4) is not amenable to analytical solution. When $V' = 0$ it is a simple matter to prove that k' is real, but even in that case there is no obvious solution for H ; in general it is not possible to prove that k' is real. The problem has therefore been solved numerically for $V' \leq 3$, by means of Runge-Kutta integration of the real and imaginary parts of the equation and a modified Newton-Raphson method for locating the relevant eigenvalue k' . When V' is less than the critical value $V'_c \approx 1.12$ then k' is real, but when V' exceeds this value k' becomes complex (the

second eigenvalue remains real until a higher critical value of V' is reached). A similar phenomenon was reported by Pedley & Fischbarg (1980) for the standard unstirred layer model; what it means is that the final approach to the steady state takes the form of a damped oscillation when $V' > V'_c$. The real and imaginary parts of k' are plotted against β' (log scale) in figure 7; the critical value of β' , corresponding to $V' = V'_c$, is $\beta'_c \approx 7.4$. It can be seen that, for values of β' in the range 0 to 1000, the real part of k' lies between about 1 and about 5. Thus for all β' in this range the time scale for decay of the perturbations is $O(\sigma^{1/2}/\alpha)$, the time scale for diffusion across the concentration boundary layer (cf. equation (3.11)). For the record, the numerical results yield the following expansion of k' when V' is small:

$$k' \approx 0.90 + 1.32V' + 0.28V'^2. \quad (5.5)$$

6. Discussion

Application of the above theory depends on the order of magnitude of the dimensionless parameters β_v and σ in an experiment. Let us consider experiments in which the solute is sucrose and the solvent water. Then $D \approx 5 \times 10^{-6} \text{ cm}^2 \text{ s}^{-1}$ while $\nu \approx 1 \times 10^{-2} \text{ cm}^2 \text{ s}^{-1}$ so that $\sigma \approx 2 \times 10^3$. Concentrations used in experiments *in vitro* vary widely; a value close to the top end of the commonly used range is 300 mmol per litre, or $C_b = 3 \times 10^{-4} \text{ mol cm}^{-3}$. Values of the (inferred) osmotic permeability P also vary widely according to the membrane under study; from House (1974) values of between 2.0×10^{-2} and $6.1 \times 10^{-1} \text{ cm s}^{-1} (\text{mol cm}^{-3})^{-1}$ can be inferred for cell membranes, while a value of $4.4 \text{ cm s}^{-1} (\text{mol cm}^{-3})^{-1}$ has been measured for rat kidney proximal tubule (an epithelium). The value may perhaps be made higher still by the careful manufacture of an artificial membrane, although in that case, as in the case of the proximal tubule, the membrane is so permeable that sucrose can leak back across it, cutting down the effective value of P .

Finally we must ascribe a value to the stirring parameter α . To do this, imagine a chamber stirred by two counter-rotating cylinders (or paddles), as depicted in figure 2(a). Assuming that the cylinders are not very close to the membrane, the flow near the stagnation point (but outside the viscous boundary layer) can be estimated from the irrotational motion due to two line vortices, located at the centres of the cylinders, in the presence of a plane boundary; their strength would be such as to give approximately the correct tangential velocity on the cylinder boundaries. If the cylinder radii are a , if their centres are distance $2b$ apart and distance d from the plane, and if they rotate with angular velocity Ω , then it is readily shown that

$$\alpha = \frac{8a^2bd\Omega}{(b^2+d^2)^2}.$$

This will be approximately valid as long as a/b , a/d and the inverse Reynolds number $\nu/\Omega ad$ are reasonably small. If we take $a = 0.2 \text{ cm}$, $b = 0.5 \text{ cm}$, $d = 1.0 \text{ cm}$ and $\Omega = 2\pi f \text{ s}^{-1}$, then $\alpha = 0.64f \text{ s}^{-1}$, so for a rotation rate of 10 cycles per second $\alpha = 6.4 \text{ s}^{-1}$, while for 1 cycle per second $\alpha = 0.64 \text{ s}^{-1}$.

Thus the value of β_r (equation (3.8)) is expected to range from 0.05 and below up to about 33. It follows that β' (equation (4.11)) is unlikely in biological experiments to rise above about 2.6, which is less than β'_c , so that the approach to the steady state is in general likely to be monotonic. However with $\beta' = 2.6$, the predicted value of γ

is about 0.29 (figure 6), which means that at values such as this the value of osmotic permeability inferred from (1.1) with $C = C_b$ is likely to be only 29% of the true value.

In view of the rather limited magnitude of β' in practice, and since the small V' or β' approximation is accurate for $V' \lesssim 1$, ($\beta' \lesssim 5.3$) from figure 6, it may be useful to set down the first few terms of the small V' expansion for γ (we give the expansion in powers of V' rather than β' because $V' = \beta'\gamma$ is the measurable quantity in practice). Using equations (4.12) to (4.14) we obtain

$$\gamma \approx 1 - 1.513V' + 0.992V'^2; \quad (6.1)$$

this is expected to be adequate for most practical applications.

We began this paper by asking whether an 'equivalent unstirred layer thickness' δ could be defined for an experiment in which the outward advection of solute by the osmotic flux is limited by the inward advection of the stirring rather than by back diffusion. A *scale* for the boundary-layer thickness, δ_c , has been shown to be $\sigma^{-\frac{1}{2}}(\nu/\alpha)^{\frac{1}{2}}$ (from (3.9)), but in view of the different mechanism at work and the different shape of the concentration profile (figure 1) it seems unlikely that merely putting δ_c into the definition of β (equation (2.6)) will allow γ to be predicted accurately by (2.4). However, the shape of the graph in figure 6 is very similar to that of the curve of γ against β obtained from (2.4) (Pedley & Fischbarg 1978, figure 4), and it turns out that a simple rescaling causes the two curves very nearly to coincide. If we take $\beta = b\beta''$, with b chosen so that the value of β' when $\gamma = 0.5$ from figure 6 is the same as the value of β'' when $\gamma = 0.5$ from (2.4) (that is, $b = 1.59$), then it can be seen from table 1 how close the coincidence is for $\beta' \leq 10^2$. This is equivalent to the statement that if we define an unstirred layer thickness

$$\delta = b\delta_c = 1.59 \left(\frac{D}{\nu}\right)^{\frac{1}{2}} \left(\frac{\nu}{\alpha}\right)^{\frac{1}{2}}, \quad (6.2)$$

then equation (2.4) can be used with acceptable accuracy to predict the effect of the layer on osmotic flow. This means that physiologists may continue to use their familiar formula, but without having to postulate a rather arbitrary value for δ , at least as long as the stirring is of the form proposed here. This may prove to be the most significant result of the present work. We may note that a similar result may be obtainable for the other types of stirring depicted in figure 2; the corresponding theory for some of these cases is in progress.

β' or β''	γ , present theory	γ , from (2.7)
0.01	0.988	0.984
0.05	0.935	0.929
0.1	0.874	0.871
0.5	0.613	0.614
1.0	0.472	0.472
5.0	0.198	0.201
10.0	0.125	0.129
50.0	0.038	0.040
100.0	0.019	0.024
500.0	0.008	0.006

TABLE 1. Comparison of the values of γ predicted for various values of β' from figure 6, and those predicted for the same values of $\beta'' (= \beta/1.59)$ from (2.7).

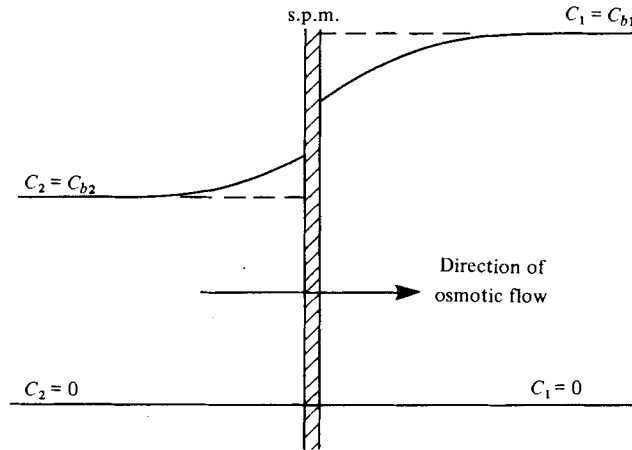


FIGURE 8. Sketch of the concentration distributions on the two sides of a semi-permeable membrane when the bulk concentration is non-zero on both sides. Note that solute is piled up against the membrane on the left, but swept away from it on the right.

This paper has examined the interaction of osmosis and stagnation-point stirring in one particular case, in which the membrane is a pure semi-permeable membrane (i.e. is impermeable to solute) and in which the solute concentration on one side of the membrane is identically zero. In a real experiment neither of these is likely to be the case. In order to generalize the theory one would need (a) to modify the boundary conditions (3.3) and (3.4), and (b) to consider another concentration boundary layer on the other side of the membrane. As far as (a) is concerned, there exist phenomenological equations relating the flux of water and salt across a leaky membrane to the concentration difference across it (House 1974). However, use of these requires knowledge of the solute permeability of the membrane as well as of two further dimensionless constants known as reflexion coefficients. These are usually taken to be equal to each other, but Hill (1979) has pointed out that there is no physical reason why they should be equal, with the consequence that we do not know the value of both and cannot apply realistic boundary conditions. There is therefore no point in constructing an elaborate theory for the case of a leaky membrane.

Nevertheless, most experiments with biological membranes will have non-zero solute concentrations on both sides, so in addition to sweeping away solute on the side towards which it is directed, the osmotic flow will cause solute to pile up on the other side. This will further cut down the concentration difference across the membrane, and means that the above estimate of γ is conservative. However, in the case of piling up, the concentration boundary layer will tend to be thinner than in the sweeping-away case, because both the stirring and the osmotic flow advect solute towards the membrane, so its effect will be smaller. The effect can be analysed in a manner similar to the above: let C_{b1}, C_{b2} be the bulk solutions on the two sides of the membrane (figure 8) with $C_{b1} > C_{b2}$ so that osmotic flow is to the right. Let the steady-state concentrations be $C_1 = C_{b1} g_1(\eta_1)$ and $C_2 = C_{b2} g_2(\eta_2)$, where $\eta_1 = -\eta_2 = (\alpha/\nu)^{1/2} y$, and it is assumed that the stirring parameter α is the same on the two sides of the membrane. The dimensionless boundary conditions corresponding to the second of (3.6b) are then

$$\sigma V g_1(0) = g_1'(0) \quad \text{and} \quad \sigma V g_2(0) = -g_2'(0),$$

while the osmosis condition (3.7) becomes

$$\sigma V = \beta_v[\theta_1 g_1(0) - (\theta_1 - 1) g_2(0)],$$

where β_v is defined as in (3.8) with $C_{b1} - C_{b2}$ for C_b , and where

$$\theta_1 = \frac{C_{b1}}{C_{b1} - C_{b2}}. \tag{6.3}$$

The case solved in § 4 is that with $\theta_1 = 1$. The solutions for g_1 and g_2 are both of the form (4.3), and it can readily be shown that the ratio γ of actual water flux to predicted flux is

$$\gamma = \frac{\theta_1}{1 + \sigma V I_1} - \frac{\theta_1 - 1}{1 - \sigma V I_2}, \tag{6.4}$$

where I_1 and I_2 are defined as in (4.5).

We now restrict attention to the case where $V' = \sigma^{\frac{1}{2}} V = O(1)$, so that the flow functions $f_1(\eta_1)$ and $f_2(\eta_2)$ are approximately given by

$$f_1(\eta_1) = -V + \frac{a_2}{2} \eta_1^2, \quad f_2(\eta_2) = +V + \frac{a_2}{2} \eta_2^2,$$

with the same value of a_2 in each case. We can again rescale the quantities I_1 , I_2 and β_v as in (4.11), and we obtain $\beta' \gamma = V'$ (as before) and (6.4) becomes

$$\gamma = \frac{\theta_1}{1 + V' I'_1} - \frac{\theta_1 - 1}{1 - V' I'_2}, \tag{6.5}$$

where I'_1 is given by (4.13), and I'_2 is the same but with $-V'$ for V' . Given θ_1 , (6.5) can be used in place of (4.12) to compute γ . The expansion for small V' , already seen to be most useful in practice, is in this case

$$\gamma \approx 1 - 1.513(2\theta_1 - 1) V' + 0.992 V'^2, \tag{6.6}$$

which should be compared with (6.1). These results show that the effect of a second concentration boundary layer on the other side of the membrane may be quite large if θ_1 is large, i.e. if the concentration difference during the osmotic flow is superimposed on an already large concentration on each side. Furthermore, rearrangement of (6.5) shows that for all $\theta_1 > 1$ there is an upper bound on the observable osmotic flow in this case, i.e. on V' , since $\gamma > 0$ and hence

$$1 - V'[\theta_1 I'_2 + (\theta_1 - 1) I'_1] > 0. \tag{6.7}$$

We know from (4.8) that $I'_1 \rightarrow \infty$ as $V' \rightarrow \infty$, so if $\theta_1 > 1$ then V' is bounded as β' increases. A similar bound arises in the standard model (§ 2) where the equivalent of (6.7) is

$$\gamma = \theta_1 e^{-\beta' \gamma} - (\theta_1 - 1) e^{\beta' \gamma} > 0. \tag{6.8}$$

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Note added in proof (3 November 1980).

After this paper went to press I was made aware of the paper ‘Ultrafiltration of proteins in stagnation flow’ by A. A. Kozinski & E. N. Lightfoot (*A.I.Ch.E.J.* **17**, 1981, 1971) in which ultrafiltration (similar to reverse osmosis) in the presence of stagnation point flow was examined numerically. The problem of this paper was not considered, however.