

## INFLUENCE OF PERMEABILITY ON THE RATE OF MUTUAL APPROACH OF MEMBRANES

D.S. DIMITROV\*

*Department of Chemical Engineering, Carnegie—Mellon University, Pittsburgh, PA 15213 (U.S.A.)*

I.B. IVANOV

*Laboratory of Physicochemical Hydrodynamics and Thermodynamics, University of Sofia, Sofia 1126 (Bulgaria)*

and T.T. TRAYKOV

*Department of Physics and Biophysics, Medical Academy, Sofia 1431 (Bulgaria)*

(Received March 28, 1983; accepted in revised form May 18, 1983)

### Summary

A general equation describing the hydrodynamic behavior of thin liquid films between permeable, tangentially mobile, deformable membranes has been derived. The solution of this equation for the case of circular plane-parallel films with tangentially immobile surfaces has been obtained and considered in details. The final formulae for the thinning rate and the hydrodynamic pressure distribution are presented in an analytical form. It has been shown that the permeability significantly affects the thinning rate when the dimensionless parameter  $\epsilon^2 = 12\mu LR^2/h^3$  is of the order of, or greater than unity. Here  $\mu$  is the bulk film viscosity,  $L$  the hydraulic permeability coefficient, and  $R$  and  $h$  the radius and thickness of the film, respectively. Analytical expressions for the rate of mutual approach of two different spherical membranes in the limiting cases of low and high permeabilities have been also obtained. The numerical estimates have shown that the permeability is important at very close membrane approach. The possible role of the permeability effect for explanation of important stages in the process of cell (or vesicle) adhesion and fusion is pointed out.

---

### Introduction

The rate of mutual approach of two particles (bubbles, drops, solid particles, cells, vesicles, etc.) in a liquid medium strongly depends on the geometrical and the physicochemical properties of the particle surfaces (surface mobility, deformability, etc.). These effects have been investigated intensively in the last decade (see e.g. [1–4]). Relatively little attention has

---

\*On leave for a year from the Central Laboratory of Biophysics, Bulgarian Academy of Sciences, Sofia 1113, Bulgaria.

been paid to the influence of the hydraulic permeability of the surfaces, i.e., the possibility of the fluid between the surfaces flowing through them. This effect does not play any role when the particle surfaces separate two immiscible fluids or non-porous solids. Hence, it seems that in most colloidal systems the hydraulic permeability is not important. However, in some biological processes, especially in the case of close approach of two membranes, the surface permeability could strongly increase the approach rate. This may be important when considering the kinetics of cell (or vesicle) adhesion and fusion [4, 5]. The permeability effect can also play a role in different processes involving dynamic interactions between artificial membranes.

The basic purpose of this paper is to present a general formulation of the problem for the dynamic interactions between approaching permeable and deformable membranes and to derive simple formulae for the rate of approach of systems of practical use.

First, we will derive a general equation, valid for the case of approach of two different particles with deformable, permeable and tangentially mobile surfaces. Further on, to simplify the mathematical treatment, we will consider in detail only the case of two circular plane-parallel membranes. The basic result is a formula for the rate of mutual approach as a function of the driving force,  $F$ , the film viscosity,  $\mu$ , the membrane radius,  $R$ , the film thickness,  $h$ , and the permeability coefficient,  $L$ . The approach rate of spherical membranes is also examined, but because of mathematical difficulties, it is calculated in an analytical form only for the limiting cases of low and high permeabilities.

### General equation of the hydrodynamics of thin liquid films between permeable membranes

We consider the system shown in Fig. 1 — two different particles A and B with deformable, tangentially mobile surfaces and hydraulic permeability coefficients  $L_A$  and  $L_B$  approach each other along the axis of symmetry,  $z$ .

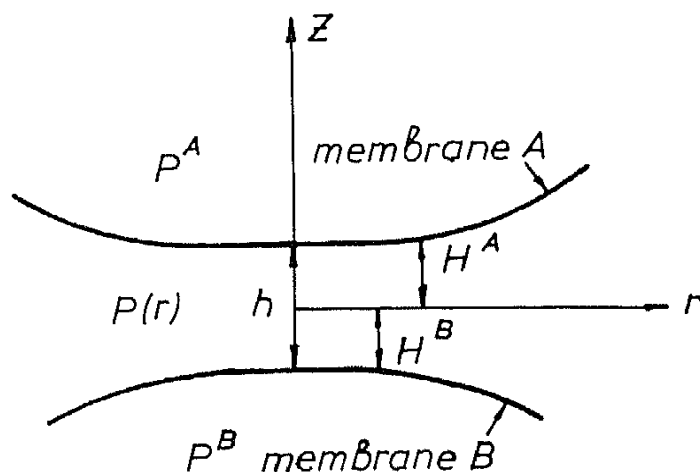


Fig. 1. Sketch of a film between two different deformable membranes.

The liquid flow in the region, where the derivative  $\partial H/\partial r$  ( $H = H^A + H^B$  is the local distance between the particle surfaces and  $r$  the radial coordinate) is much smaller than unity, is given by the lubrication theory equation (for details see e.g. [1,4])

$$\partial p/\partial r = \mu(\partial^2 v_r/\partial z^2) \quad (1)$$

$$\partial p/\partial z = 0 \quad (2)$$

$$\nabla_r v_r + \partial v_z/\partial z = 0, \quad \nabla_r v_r = [\partial(v_r r)/\partial r]/r, \quad (3)$$

where  $p$  is the hydrodynamic pressure and  $v_r$  and  $v_z$  are the radial and the axial velocity components, respectively.

We assume the following general boundary conditions:

$$v_r = U^A \quad \text{at } z = H^A \quad (4a)$$

$$v_r = U^B \quad \text{at } z = H^B \quad (4b)$$

$$v_z = \partial H^A/\partial t + U^A(\partial H^A/\partial r) + L_A(p - p^A) \quad \text{at } z = H^A \quad (4c)$$

$$v_z = -\partial H^B/\partial t - U^B(\partial H^B/\partial r) - L_B(p - p^B) \quad \text{at } z = -H^B, \quad (4d)$$

where  $p^A$  and  $p^B$  are the pressures in the particles A and B, respectively. The hydraulic permeability coefficients  $L_A$  and  $L_B$  represent the velocity of the fluid through the membrane per unit pressure difference across the membrane [6]. Taking into account eqns. (2), (4a) and (4b) we have

$$v_r = (z^2/2\mu)(\partial p/\partial r) + Az + B, \quad (5)$$

where

$$A = (U^A - U^B)/H + (H^B - H^A)(\partial p/\partial r)/2,$$

$$B = U^B + H^B(U^A - U^B)/H - H^B H^A(\partial p/\partial r)/2.$$

Inserting eqn. (5) into eqn. (3) and integrating over  $z$  from  $-H^B$  to  $H^A$ , by means of eqns. (4c) and (4b), we obtain

$$-\partial H/\partial t - Lp + V_f = \nabla_r [H(U^A + U^B)/2 - H^3(\partial p/\partial r)/12\mu], \quad (6)$$

where  $L = L_A + L_B$  and  $V_f = L_A p^A + L_B p^B$  (the subscript f means filtration). This expression (6) is the general differential equation of the dynamics of thin viscous liquid films with deformable, tangentially mobile and permeable surfaces.

### Thinning of cylindrical plane-parallel films

In this section we consider the particular case of thinning of a film between circular, plane-parallel, tangentially immobile membranes with radius  $R$  (Fig. 2), i.e.,

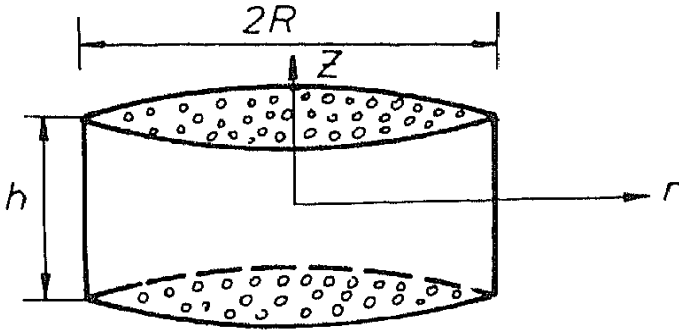


Fig. 2. A cylindrical film formed between two plane-parallel membranes.

$$H = h, \quad \partial H / \partial r = 0, \quad V = -dh/dt, \quad U^A = U^B = 0.$$

Then eqn. (6) reduces to

$$x^2 (\partial^2 \tilde{p} / \partial x^2) + x (\partial \tilde{p} / \partial x) - \epsilon^2 x^2 \tilde{p} + 8x^2 (\tilde{V} + \tilde{V}_f) = 0, \quad (7)$$

where  $x = r/R$ ,  $\tilde{p} = p\pi R^2/F$ ,  $\epsilon^2 = 12\mu LR^2/h^3$ ,  $\tilde{V} + \tilde{V}_f = (V + V_f)/V_{Re}$ .

Here,  $F$  is the driving force and  $V_{Re} = 2Fh^3/3\pi\mu R^4$  is the Reynolds velocity of thinning of a film with non-permeable surfaces (see e.g. [4]). We suppose that the hydrodynamic pressure,  $p$ , is equal to the pressure in the bulk liquid (set equal to zero) at the film perimeter, i.e.,

$$\tilde{p} = 0 \quad \text{at } x = 1. \quad (8)$$

The driving force,  $F$ , must be equal to the hydrodynamic resistance force

$$1 = \int_0^1 \tilde{p} dx^2. \quad (9)$$

The solution of eqn. (7), finite at  $r=0$  and satisfying the boundary condition (8), is

$$\tilde{p} = 8(\tilde{V} + \tilde{V}_f) [1 - I_0(\epsilon x)/I_0(\epsilon)] / \epsilon^2, \quad (10)$$

where  $I_n$  is the modified Bessel function of the first kind,  $n$ th order (in eqn. (10),  $n = 0$ ). The substitution of eqn. (10) into eqn. (9) yields the following formula for the thinning rate,  $\tilde{V}$ :

$$\tilde{V} = -\tilde{V}_f + \epsilon^2 I_0(\epsilon) / 8I_2(\epsilon). \quad (11)$$

For membranes of low permeability ( $\epsilon \ll 1$ ), eqn. (11) reduces to

$$V = -V_f + 1 + \epsilon^2/6. \quad (12)$$

In the opposite case ( $\epsilon \gg 1$ ), eqn. (11) yields

$$\tilde{V} = -\tilde{V}_f + \epsilon/4 + \epsilon^2/8. \quad (13)$$

The combination of eqns. (10) and (11) gives the following formula for the pressure distribution:

$$\tilde{p} = [1 - I_0(\epsilon x)/I_0(\epsilon)]I_0(\epsilon)/I_2(\epsilon). \quad (14)$$

The asymptotic expressions for low ( $\epsilon \ll 1$ ) and high ( $\epsilon \gg 1$ ) permeabilities are

$$\tilde{p} = 2(1-x^2) + (-1/24 + x^2/6 - x^4/8)\epsilon^2 \quad \text{for } \epsilon \ll 1 \quad (15a)$$

$$\tilde{p} = 1 - \exp[\epsilon(x-1)]/x^{1/2} \quad \text{for } \epsilon \gg 1, x \sim 1 \quad (15b)$$

$$\tilde{p} = 1 - (2\pi x)^{1/2} \exp(-\epsilon) \quad \text{for } \epsilon \gg 1, \epsilon x \ll 1 \quad (15c)$$

### Rate of approach of spherical membranes

In the thin film approximation, the spherical surface in the region near to the axis of symmetry (the region of high dissipation of energy) is approximated by a parabola (see e.g. [3])

$$H = h + r^2/2\bar{R}_s, \quad \bar{R}_s = R_s^A R_s^B / (R_s^A + R_s^B), \quad (16)$$

where  $R_s^A$  and  $R_s^B$  are the radii of the spheres. The substitution of this expression (16) into eqn. (6) at  $U^A = U^B = 0$  yields an equation

$$V + V_f + \nabla_r [(h + r^2/2\bar{R}_s)^3 (\partial p/\partial r)]/12\mu = Lp, \quad V = -dh/dt, \quad (17)$$

from which the hydrodynamic pressure,  $p$ , can be determined. Since the general solution of eqn. (17) is not easy to obtain, we use an iteration procedure to get the approach rate at small permeabilities. The zeroth approximation (right hand side of eqn. (17) equal to zero) leads to results obtained earlier (see e.g. [3])

$$p = 3\mu(V + V_f)\bar{R}_s/(h + r^2/2\bar{R}_s)^2, \quad (18)$$

excluding the introducing of the constant rate  $V_f$ . The substitution of eqn. (18) into the right hand side of eqn. (17) and the subsequent integration over  $r$  leads to the following expression for the pressure gradient (including the zeroth and the first approximations)

$$\partial p/\partial r = -6\mu(V + V_f)r/(h + r^2/2\bar{R}_s)^3 + 18\mu^2(V + V_f)L\bar{R}_s r/h(h + r^2/2\bar{R}_s)^4, \quad (19)$$

where the condition for the lack of singularity at  $r = 0$  is used to determine the integration constant.

The approach rate,  $V$ , should be determined from the integral balance of forces, eqn. (9), which can be represented in a more convenient form for the present consideration as:

$$F = \pi \int_0^\infty p dr^2 = \pi p r^2 \Big|_0^\infty - \pi \int_0^\infty r^2 (\partial p/\partial r) dr = -\pi \int_0^\infty r^2 (\partial p/\partial r) dr, \quad (20)$$

where the condition of vanishing dynamic pressure at  $r \rightarrow \infty$  is used instead of eqn. (8).

The substitution of eqn. (19) into eqn. (20) gives the final expression for the approach rate

$$V + V_f = (Fh/6\pi\mu\bar{R}_s^2)(1 + 15\sqrt{2}\pi\mu L\bar{R}_s^{3/2}/128h^{5/2}), \quad (21)$$

where the term due to the permeability should be smaller than unity. It is interesting to note that, due to the changed geometry of the approaching membranes, the permeability influences the approach rate in different functional ways: at fixed radii of the film,  $R$ , and the sphere,  $R_s$ , the permeability effect increases slower with decreasing  $h$  for the case of spherical membranes (as  $h^{-5/2}$  instead of  $h^{-3}$  for the case of plane-parallel films).

For great permeabilities, eqn. (17) gives a formula, analogous to that for plane-parallel films (eqn. (13) at  $\epsilon \gg 1$ )

$$V + V_f = FL/\pi\bar{R}_s^2. \quad (22)$$

The expression (22) is derived by neglecting the viscous resistance (the last term in the left hand side of eqn. (17) is set equal to zero) and integrating to finite film radius  $R = \bar{R}_s$  when using the equation of the balance of forces (9). It should be pointed out that at great permeabilities the lubrication theory approximation is questionable and can only be used with caution.

## Discussion

One can see from the initial differential equation (eqn. (7)) and the formulae obtained (10—15) that the basic parameter determining the relative effect of the permeability on the hydrodynamic behavior of thin plane-parallel films is

$$\epsilon^2 = 12\mu LR^2/h^3.$$

When  $\epsilon^2 < 1$  this influence is small. The opposite is true when  $\epsilon^2 \gtrsim 1$ . It is interesting to note that this dimensionless group does not depend on the driving force  $F$ . It is expressed through the geometric ( $h$  and  $R$ ) and the dynamic ( $\mu$ ) characteristics of the system, as well as through the membrane permeability,  $L$ .

Let us suppose that at  $\epsilon^2 > 0.1$  the permeability must be taken into account. This is fulfilled at  $L > 10^{-2}h^3/R^2$ . If  $h \sim 10^{-6}$  cm,  $\mu \sim 10^{-2}$  p and  $R \sim 10^{-3}$  cm,  $L$  should be larger than  $10^{-13}$  cm<sup>3</sup>/dyn-sec. This is a quite reasonable value — two orders of magnitude higher than the permeability coefficient of the erythrocyte membrane ( $L \sim 10^{-11}$  cm<sup>3</sup>/dyn-sec, [6]). If the fluid flow occurs through pores with radius  $r_0$  and length  $d$  and the Poiseuille formula is valid, the permeability coefficient  $L$  is given by the simple formula:  $L = \alpha r_0^2/8\mu d$ , where  $\alpha$  is the ratio of the area occupied by the pores to the total membrane area ( $\alpha = n\pi r_0^2$ ,  $n$  being the number of the pores in 1 cm<sup>2</sup>). Assuming  $\alpha \sim 10^{-4}$ ,  $r_0^2 \sim 10^{-15}$  cm<sup>2</sup>,  $\mu \sim 10^{-1}$  p and  $d \sim 10^{-7}$  cm, we obtain  $L \sim 10^{-12}$  cm<sup>3</sup>/dyn-sec, which is a reasonable value for cell membranes.

Figure 3 shows the dependence of the dimensionless rate  $\tilde{V} + \tilde{V}_f$  on the parameter  $\epsilon$  according to eqn. (11). The pressure distribution  $\tilde{p}(x)$  is shown in Fig. 4 for three different values of  $\epsilon$ :  $\epsilon = 0.1, 1$  and  $10$ . It can be seen that at small  $\epsilon$  the pressure distribution is identical with that given by the Reynolds law. With increasing permeability, the pressure decreases and becomes almost constant, so that the liquid is squeezed out through the membranes. In this respect, it is important to note that according to eqn. (13) at  $\epsilon \gg 1$ , the rate of thinning,  $V$ , is given by an analogue of eqn. (22) (with  $R$  instead of  $R_s$ ), i.e., it is strongly increased and does not depend on the thickness and the viscosity.

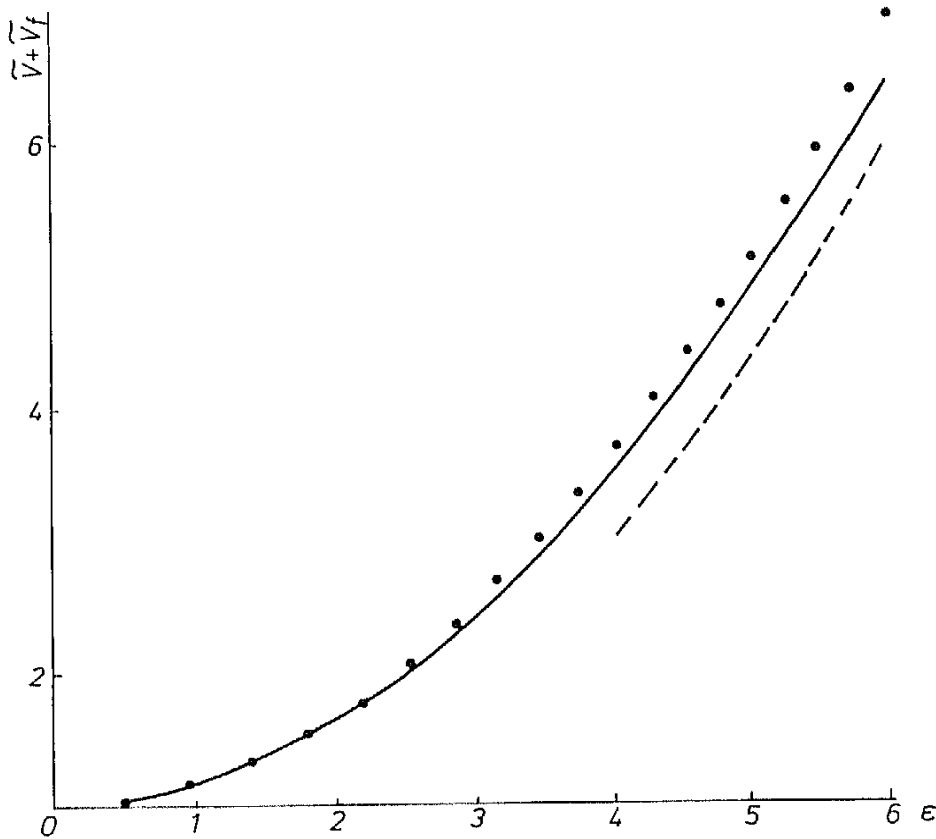


Fig. 3. Dimensionless thinning rate,  $\tilde{V} + \tilde{V}_f$ , vs. dimensionless permeability parameter  $\epsilon$  (solid line). The broken lines are calculated from the asymptotic formulae (12) and (13): the dotted line corresponds to  $\epsilon \ll 1$  and the dashed line to  $\epsilon \gg 1$ .

The calculations carried out on the basis of eqn. (21) show that for spherical membranes the permeability influence can be significant (for  $R_s > 10^{-3}$  cm and  $L > 10^{-11}$  cm-sec/g) at very close membrane approach ( $h < 10^{-7}$  cm). With increasing permeability, this effect could also play a role for larger separations and smaller particles.

The permeability mechanisms could play a role in membrane adhesion and fusion, particularly in the electric field induced cell fusion [7]. Recently, we have proposed a dynamic thin film model for the kinetics of the electric field induced membrane fusion [5, 8]. Considering the interacting membranes and

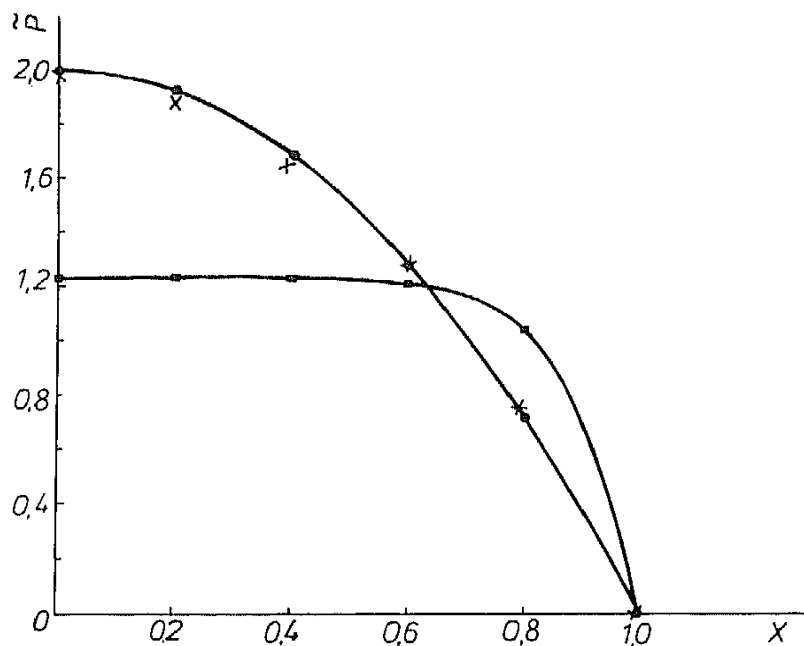


Fig. 4. Dimensionless pressure,  $\tilde{p}$ , vs. dimensionless distance,  $x$ , calculated from eqn. (14) for  $\epsilon = 0.1$  ( $\bullet$ ),  $\epsilon = 1$  ( $\times$ ) and  $\epsilon = 10$  ( $\blacksquare$ ).

the intervening fluid as thin films, we have estimated the time-course of three possible stages of the fusion process: (1) formation of pores in the interacting membranes, leading to an increased membrane permeability [8]; (2) destabilization and breaking of the liquid film between the membranes, which is to a major extent influenced by the increased membrane permeability [5]; and (3) extrusion of the fluid by the fused membranes [4]. Consequently, the membrane permeability effect can strongly increase the rate of cell and vesicle fusion.

### Acknowledgement

We thank Prof. E.F. Leonard and Prof. R.K. Jain for useful discussions. This work was supported by the National Science Foundation, Grant No. INT 8209490.

### References

- 1 D.S. Dimitrov and I.B. Ivanov, Hydrodynamics of thin liquid films. On the rate of thinning of microscopic films with deformable interfaces, *J. Colloid Interface Sci.*, 64 (1978) 97.
- 2 I.B. Ivanov and R.K. Jain, Formation and thinning of thin liquid films, in: T.S. Sorensen (Ed.), *Dynamics and Instability of Fluid Interfaces*, Springer-Verlag, Berlin, 1979, p. 120.
- 3 I.B. Ivanov and D.S. Dimitrov, Thin film drainage, in: I.B. Ivanov (Ed.), *Thin Liquid Films*, Marcel Dekker, New York, 1983.
- 4 D.S. Dimitrov, Dynamic interactions between approaching surfaces of biological interest, in: S.G. Davison (Ed.), *Progress in Surface Science*, Pergamon Press, Oxford, 1983.
- 5 D.S. Dimitrov and R.K. Jain, Stability of thin viscoelastic films between permeable membranes. An application to the membrane fusion, submitted to *J. Colloid Interface Sci.*, 1983.



- 6 A. Katchalsky and P.F. Curran, *Nonequilibrium Thermodynamics in Biophysics*, Harvard University Press, Cambridge, 1967.
- 7 U. Zimmermann, Electric field-mediated fusion and related electrical phenomena, *BBB*, 694 (1982) 227.
- 8 D.S. Dimitrov, Electric field-induced breakdown of lipid bilayers and cell membranes — a thin viscoelastic film model, submitted to *J. Membrane Biol.*, 1983.