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The time-dependent deformation of a capsule freely suspended in a linear shear flow

By D. BARTHÈS-BIESEL
Université de Technologie de Compiègne, France
AND J. M. RALLISON
Department of Applied Mathematics and Theoretical Physics,
University of Cambridge, England

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An analysis is presented of the dynamics of a small deformable capsule freely suspended in a viscous fluid undergoing shear. The capsule consists of an elastic membrane which encloses another viscous fluid, and it deforms in response to the applied external stresses and the elastic forces generated within the membrane. Equations are derived which give its time-dependent deformation in the limit that the departure of the shape from sphericity is small. The form of the shear flow is arbitrary and a general (two-dimensional) elastic material is considered. Limiting forms are obtained for highly viscous capsules and for membranes which are area-preserving, and earlier results for surface tension droplets and incompressible isotropic membranes are derived as particular cases. Results for the viscosity of a dilute suspension of capsules are also given.

The theoretical prediction for the relaxation rate of the shape is derived for an interface which has elastic properties appropriate for a red-blood-cell membrane, and is compared with experimental observations of erythrocytes.

1. Introduction

Barthès-Biesel (1980) has recently proposed the use of the term 'capsule' to describe a particle consisting of an elastic membrane which encloses a drop of an incompressible Newtonian fluid. Such a particle will deform when freely suspended in a second fluid undergoing shear, and provides a mathematical model to describe the flow-induced deformation of red blood cells and of emulsions stabilized by interfacial cross-linking polymerization. Previous work in this area has modelled the particles as elastic solids (Lighthill 1968) or as liquid droplets (Hyman & Skalak 1972); here we incorporate more of the known particle structure by restricting the elastic response to a thin surface membrane whose properties are arbitrary but supposed known. This approach was taken also by Brennen (1975) in calculating the viscosity of blood. His model was restricted, however, to the case of instantaneously *spherical* capsules so that he was unable to calculate either their time-dependent deformation or their steady shape. Both these defects are remedied in the present analysis.

The problem thus consists in finding the motion and deformation of the capsule under the influence of the viscous fluid forces. It involves a free surface where the boundary conditions (continuity of velocity and a balance between elastic and viscous forces) are applied and whose position is *a priori* unknown. Earlier work by Barthès-Biesel (1980) has determined the steady deformation of an almost spherical capsule freely suspended in a simple shear flow. It is predicted that the particle reaches a steady shape, although its membrane rotates around that shape continuously. However, the lack of time-dependence in the analysis means that it cannot be used to interpret relaxation experiments such as those of Schmid-Schönbein (1975) on red blood cells. Furthermore the membrane of the capsule is treated as the two-dimensional limit of an isotropic, incompressible, three-dimensional elastic solid as is also the case for the linear time-dependent analysis of Brunn (1980). This restricts the applicability of the model, which cannot reproduce the surface-tension droplet case, nor encompass a wide class of two-dimensional membrane rheologies, such as that proposed by Skalak *et al.* (1973) for red blood cells.

Consequently we here generalize the analysis of capsule deformation to include unsteady response to flow, and general two-dimensional elastic membranes. We assume only that the membrane has no bending resistance, is homogeneous, and has isotropic properties in the plane. It is then possible to recover as special cases the surface-tension-controlled interface, the infinitely thin three-dimensional solid, and the constant surface area interface postulated to describe the red-blood-cell membrane. It should be pointed out, however, that the analysis remains deficient for red blood cells in two important respects. First, it is probable (Brennen 1975; Chien *et al.* 1978) that the erythrocyte membrane is *viscoelastic* rather than instantaneously elastic. Second, we consider here only near-spheres and thus only 'sphered' (osmotically swollen) red blood cells are appropriately modelled. We note later (§ 8) how both these complications may be incorporated into a more sophisticated framework.

An inherent difficulty in problems involving interaction between a fluid and a deformable solid arises from the two different formulations most naturally used: Eulerian for the fluid, and Lagrangian for the solid. This difficulty is compounded by the fact that classical membrane theory is expressed in a curvilinear co-ordinate system fixed in the membrane and which deforms with it. Although the twofold formulation cannot be avoided altogether, we have been led (in §§ 2 and 3) to reformulate the membrane theory in the same frame of reference as the fluid problem. This has the advantage that Cartesian tensors can be used, and furthermore the interaction between viscous and elastic forces can be expressed without difficulty. This approach is also taken by Secomb & Skalak (1982) in a similar problem.

In § 4 we apply this theory to consider the time-dependent behaviour of an initially spherical capsule in the limiting case where its deformation remains small. The resulting shape-evolution equations are then studied for different membrane rheological behaviours, for weak flows (§ 5), as well as for very viscous particles (§ 6). In particular, the result for the relaxation time of the shape of a constant-area membrane is compared with experimental data on red blood cells. Finally (§ 7), the constitutive equation for a dilute suspension of such particles is derived and shown to be of the viscoelastic type.

2. Determination of the stresses within the membrane

In this section we derive an appropriate formulation to describe the local elastic response of a membrane under finite deformation. We consider a membrane whose thickness is vanishingly small, so that it has no bending resistance, and which is *transversely isotropic* (about its local normal $\mathbf{n}$) i.e. unchanged by rotations in its local plane surface. On the further assumptions that the membrane is purely elastic, and that the elasticity is local and instantaneous, its mechanical response is characterized by a strain energy function $w \equiv w(\lambda_1, \lambda_2, \lambda_3)$, where λ_3 is the principal stretch along $\mathbf{n}$, and $\lambda_{1,2}$ are the principal planar stretches. In view of the isotropy, w is a symmetric function of λ_1, λ_2. Finally we suppose that the 'thinning' λ_3 is determined locally by λ_1, λ_2 so that $\lambda_3 = \lambda_3(\lambda_1, \lambda_2)$ and hence that the strain energy can be written as the symmetrical function $w = w(\lambda_1, \lambda_2)$.

Expressions for the components of stress referred to general non-orthogonal co-ordinate systems have been derived for such a membrane (see Green & Adkins 1960, §4), but in order to facilitate the connection with the fluid-mechanical problem which follows we prefer to rederive the results using Cartesian tensors throughout. For a three-dimensional solid, the results are well known (see e.g. Leigh 1968), and we use a similar technique here to derive analogous formulae for the two-dimensional case.

Strain analysis

In order to keep track of the positions of material points in the membrane, let the position of each point in some reference configuration be labelled by $\mathbf{X}$, and suppose that at time t its position is $\mathbf{x}(\mathbf{X}, t)$. Let

$$\mathsf{C} = \partial \mathbf{x}/\partial \mathbf{X}$$

be the relative deformation tensor. This is defined only for fibres $\mathbf{dX}$ which lie in the reference surface. In consequence, if $\mathbf{N}(\mathbf{X})$ is a normal to the initial surface, let

$$\mathsf{A} = \mathsf{C}.(\mathsf{I} - \mathrm{NN})$$

so that $\mathsf{A}.\mathbf{dX} = 0$ if $\mathbf{dX}$ is parallel to N, and $\mathsf{A}.\mathbf{dX} = \mathsf{C}.\mathbf{dX}$ if $\mathbf{dX}$ is perpendicular to N. It follows from the assumption of two-dimensionality that $\mathbf{n}.\mathsf{A} = 0$, where $\mathbf{n}$ is the normal to the final surface, and so

$$\mathsf{A} = (\mathsf{I} - \mathbf{nn}).\partial \mathbf{x}/\partial \mathbf{X}.(\mathsf{I} - \mathrm{NN}). \tag{2.1}$$

Thus A is the two-dimensional relative displacement gradient for the surface. (Note that if the initial and final states coincide $\mathsf{C} = \mathsf{I}$ and $\mathsf{A} = \mathsf{I} - \mathbf{nn}$.) The Cauchy–Green strain tensor is then

$$\Lambda^2 = \mathsf{A}^T.\mathsf{A},$$

(and the metric tensor G of Green & Adkins (1960, equation 4.1.2) corresponds to $\Lambda^2 + \mathrm{NN}$), with eigenvalues $\lambda_1^2, \lambda_2^2, 0$ and corresponding eigenvectors $\mathbf{e}_1, \mathbf{e}_2, \mathrm{N}$. Finally, by the polar decomposition theorem we may put

$$\mathsf{A} = \mathsf{R}.\Lambda,$$

with R a planar rotation ($\mathsf{R}^T.\mathsf{R} = \mathsf{I}$).

Stress analysis

In the same way as in the three-dimensional case, the principal stress resultants σ_1, σ_2 are given by

$$\sigma_i = \frac{1}{\lambda_1 \lambda_2} \lambda_i \frac{\partial w}{\partial \lambda_i}, \quad i = 1, 2 \quad \text{(no summation)},$$

and further the principal axes of the Cauchy stress are coaxial with those of $\mathbf{A}.\mathbf{A}^T$. It is convenient to define new strain invariants

$$a = \log \lambda_1\lambda_2 = \tfrac{1}{2}\log\{\tfrac{1}{2}[\mathrm{tr}\,(\mathbf{A}.\mathbf{A}^T)]^2 - \tfrac{1}{2}\mathrm{tr}\,[(\mathbf{A}.\mathbf{A}^T)^2]\}, \tag{2.2}$$

$$b = \tfrac{1}{2}(\lambda_1^2+\lambda_2^2) - 1 = \tfrac{1}{2}\mathrm{tr}\,(\mathbf{A}.\mathbf{A}^T) - 1, \tag{2.3}$$

so that e^a is the local membrane area change, and then we have

$$\begin{aligned}\boldsymbol{\sigma} &= e^{-a}\mathbf{R}.\left[\frac{\partial w}{\partial a}(\mathbf{e}_1\mathbf{e}_1+\mathbf{e}_2\mathbf{e}_2)+\frac{\partial w}{\partial b}(\lambda_1^2\mathbf{e}_1\mathbf{e}_1+\lambda_2^2\mathbf{e}_2\mathbf{e}_2)\right].\mathbf{R}^T\\ &= e^{-a}\left[\frac{\partial w}{\partial a}(\mathbf{I}-\mathbf{nn})+\frac{\partial w}{\partial b}\mathbf{A}.\mathbf{A}^T\right].\end{aligned}$$

This result corresponds to equations (4.3.12–16) of Green & Adkins (1960) in which the dependence of w on λ_3 has been suppressed here in view of the constitutive assumption $\lambda_3 = \lambda_3(\lambda_1, \lambda_2)$.

Load vector

Finally, the local vector $\mathbf{f}$ exerted by the membrane is given by

$$\mathbf{f} = \nabla^s.\,\boldsymbol{\sigma},$$

where ∇^s is the surface gradient operator $\nabla^s \equiv (\mathbf{I}-\mathbf{nn}).\nabla$. The expression for $\mathbf{f}$ may then be rewritten as

$$\begin{aligned}\mathbf{f} = -e^{-a}\left(\frac{\partial w}{\partial a}+\frac{\partial w}{\partial b}\right)\mathbf{n}\nabla.\mathbf{n} + (\mathbf{I}-\mathbf{nn}).\nabla\left[e^{-a}\left(\frac{\partial w}{\partial a}+\frac{\partial w}{\partial b}\right)\right]\\ +(\mathbf{I}-\mathbf{nn}).\nabla.\left[e^{-a}\frac{\partial w}{\partial b}(\mathbf{A}.\mathbf{A}^T-\mathbf{I}).(\mathbf{I}-\mathbf{nn})\right].\end{aligned} \tag{2.4}$$

The first term in this expression corresponds to a surface tension: it is directed along the normal, and is proportional to the surface curvature. The surface-tension coefficient $\gamma \equiv e^{-a}(\partial w/\partial a + \partial w/\partial b)$ depends upon the surface stretches through a, b. The second term has no normal component and arises from the variation of γ in the surface. The third has both normal and tangential components and is generated by surface distortions which, in the limit of small deformations, are area preserving.

Small deformations

The expression above is valid for finite deformations of the membrane, but a simpler form is available, and is more convenient, when the deformations are small. We therefore consider

$$\mathbf{C} = \mathbf{I}+\epsilon\mathbf{D}, \quad \mathbf{n} = \mathbf{N}+O(\epsilon)$$

in which $\epsilon \ll 1$. It is convenient also to replace b by the alternative invariant $c \equiv b-a$, and to regard $w = w(a,c)$. Then

$$a = \tfrac{1}{2}\epsilon\,\mathrm{tr}\,[(\mathbf{I}-\mathbf{nn}).(\mathbf{D}+\mathbf{D}^T).(\mathbf{I}-\mathbf{nn})]+O(\epsilon^2)$$

and $c = O(\epsilon^2)$. The expression for the load becomes

$$\begin{aligned}\mathbf{f} &= -e^{-a}\frac{\partial w}{\partial a}\mathbf{n}\nabla.\mathbf{n}+(\mathbf{I}-\mathbf{nn}).\nabla\left(e^{-a}\frac{\partial w}{\partial a}\right)+(\mathbf{I}-\mathbf{nn}).\nabla.\left[e^{-a}\frac{\partial w}{\partial c}(\mathbf{A}.\mathbf{A}^T-\mathbf{I}).(\mathbf{I}-\mathbf{nn})\right]\\ &= -e^{-a}\frac{\partial w}{\partial a}\mathbf{n}\nabla.\mathbf{n}+(\mathbf{I}-\mathbf{nn}).\nabla\left(e^{-a}\frac{\partial w}{\partial a}\right)\\ &\quad +\epsilon(\mathbf{I}-\mathbf{nn}).\nabla.\left[e^{-a}\frac{\partial w}{\partial c}(\mathbf{I}-\mathbf{nn}).(\mathbf{D}+\mathbf{D}^T).(\mathbf{I}-\mathbf{nn})\right]+O(\epsilon^2)\end{aligned}$$

and, on expanding about $\epsilon = 0$,

$$w = w_0 + \alpha_1 a + \tfrac{1}{2}(\alpha_2 + \alpha_1) a^2 + \alpha_3 c + O(\epsilon^3)$$

so that

$$e^{-a}\frac{\partial w}{\partial a} = \alpha_1 + \alpha_2 a + O(\epsilon^2), \quad e^{-a}\frac{\partial w}{\partial c} = \alpha_3 + O(\epsilon),$$

giving finally

$$\mathbf{f} = -\alpha_1 \mathbf{n}\nabla.\mathbf{n} - \alpha_2 a\mathbf{n}\nabla.\mathbf{n} + \alpha_2(\mathbf{I}-\mathbf{nn}).\nabla a + \epsilon\alpha_3(\mathbf{I}-\mathbf{nn}).\nabla.[(\mathbf{I}-\mathbf{nn}).(\mathbf{D}+\mathbf{D}^T).(\mathbf{I}-\mathbf{nn})] + O(\epsilon^2). \quad (2.5)$$

Connection with three-dimensional elasticity

If the membrane is considered to consist of an elastic material which is isotropic in *three* dimensions, then its strain energy function may be written $w = w(I_1, I_2, I_3)$, where

$$I_1 = \tfrac{1}{2}(\lambda_1^2 + \lambda_2^2 + \lambda_3^2), \quad I_2 = \tfrac{1}{2}(\lambda_1^{-2} + \lambda_2^{-2} + \lambda_3^{-2}), \quad I_3 = \lambda_1 \lambda_2 \lambda_3.$$

If further the material is incompressible, then $I_3 = 1$, and the elastic properties are defined by the parameters

$$\Phi = \partial w/\partial I_1, \quad \Psi = \partial w/\partial I_2.$$

For small deformations, it may then be shown that the constants α_i for this class of materials are given by

$$\alpha_1 = 0, \quad \alpha_2 = 2(\Phi + \Psi) = \tfrac{2}{3}E, \quad \alpha_3 = \Phi + \Psi = \tfrac{1}{3}E,$$

where Φ, Ψ are evaluated at zero deformation, and E is the Young's modulus of the material.

3. Specification of the capsule distortion

We now turn to the problem of determining the *global* capsule deformation. We consider a membrane whose material properties are uniform, for which it is helpful to distinguish at the outset between two forms of material motion. When a capsule is placed in a shear flow its external shape changes in response to the flow forces until either a steady shape is set up or the deformation grows without bound and the particle breaks (or, exceptionally, a steady oscillation is set up). This time-dependent motion of the shape may be separated into a solid body rotation, and a stretching. In addition, even in a *steady* state for the external shape, the material of the membrane will in general be rotating ('tank-treading') so that the deformation at the fixed Eulerian point $\mathbf{x}$ is constant in time, but that at a fixed point $\mathbf{X}$ of the material is not.

Since the solid body rotation of the membrane generates no deformation and thus no elastic force, it is natural to subtract it out of the problem. This may be achieved by defining the capsule distortion relative to a frame of reference which rotates at a rate given by the sum of the solid body and tank-treading rotations. Because material points can move within the membrane, however, there is no uniquely specified angular velocity for the membrane as a whole; in any definition of a *mean* rotation rate for the points of the membrane, the weight to be attached to each element of surface depends upon whether the original or deformed surface area is considered. The final results for stresses and shapes cannot of course depend on this choice (though the intermediate mathematical details will), and we find it convenient to use the undeformed elements of surface to define an average rotation rate. This is therefore the instantaneous mean angular velocity of marked points evenly distributed on the reference surface.

In the case where the membrane is initially spherical, we want to consider as a reference state the sphere which has at each instant of time the same angular velocity $\boldsymbol{\omega}$ as the membrane (defined above, and to be determined as part of the solution). This may be achieved by the following device: define the rotation matrix $\mathbf{Q}(t)$ $(\mathbf{Q}^T.\mathbf{Q} = \mathbf{I})$ by

$$\mathbf{Q}(0) = \mathbf{I}, \quad \dot{\mathbf{Q}}(t) = \boldsymbol{\omega}.\mathbf{Q}(t),$$

so that $\mathbf{Q}(t)$ is the total rotation of the membrane up to time t; then, starting from the 'fixed' reference sphere $\mathbf{Y}.\mathbf{Y} = 1$, let $\mathbf{X} = \mathbf{Q}.\mathbf{Y}$. It follows that $\mathbf{X}.\mathbf{X} = 1$, and

$$\dot{\mathbf{X}} = \boldsymbol{\omega}.\mathbf{X} \tag{3.1}$$

as required. Since the material of the membrane is uniform and instantaneously elastic, we may in computing the elastic load use $\mathbf{X}$ rather than $\mathbf{Y}$ to define the position of the material point before displacement.

By analogy with the problem of determining the deformation of a surface-tension drop in a shear flow, we seek a deformation field which involves only second harmonics at leading order and consider

$$\mathbf{x} = \mathbf{X}+\epsilon\mathbf{K}.\mathbf{X}+\epsilon\mathbf{X}\mathbf{X}.(\mathbf{J}-\mathbf{K}).\mathbf{X}+\epsilon^2\mathbf{g}(\mathbf{X})+O(\epsilon^3), \tag{3.2}$$

in which $\mathbf{J}$ and $\mathbf{K}$ are functions of time only. The instantaneous external shape of the capsule is then

$$r \equiv |\mathbf{x}| = 1+\epsilon\mathbf{X}.\mathbf{J}.\mathbf{X}+O(\epsilon^2) = 1+\epsilon\mathbf{x}.\mathbf{J}.\mathbf{x}/r^2+O(\epsilon^2), \tag{3.3}$$

with normal

$$\mathbf{n} = \mathbf{x}/r+2\epsilon\mathbf{x}\mathbf{x}.\mathbf{J}.\mathbf{x}/r^3-2\epsilon\mathbf{J}.\mathbf{x}/r+O(\epsilon^2) \tag{3.4}$$

indicating that at leading order in ϵ $\mathbf{K}$ represents motion of fibres *within* the surface, whereas $\mathbf{J}$ involves overall deformation of the surface too.

We suppose further that $\mathbf{J}$, $\mathbf{K}$ are symmetric and traceless second-rank tensors, so that they involve only second-harmonic distortions. In the case of $\mathbf{J}$, symmetry may be imposed without loss of generality, and tracelessness follows from conservation of the drop volume. For $\mathbf{K}$ we check *a posteriori* that the equations for its time evolution preserve these requirements.

There is some interest in pursuing the calculation to $O(\epsilon^2)$ in particular for very high viscosity capsules. It is then convenient to regard $\mathbf{J}$ and $\mathbf{K}$ as describing the second harmonic component of the distortion at *all* orders in ϵ, though they define the *full* deformation only at $O(\epsilon)$. Then $\mathbf{g}(\mathbf{x})$ involves only harmonics of orders 0 and 4. The 0th-order harmonics may be immediately computed from the volume conservation requirement:

$$\mathbf{g}(\mathbf{x}) = g_0\mathbf{x}+\mathbf{g}_4(\mathbf{x}),$$

with

$$g_0 = -\tfrac{2}{15}\mathbf{J}:\mathbf{J}-\tfrac{1}{10}\mathbf{K}:\mathbf{K}+\tfrac{2}{5}\mathbf{J}:\mathbf{K}, \tag{3.5}$$

and we may put

$$\mathbf{g}_4(\mathbf{x}) = \mathbf{K}_4\vdots\mathbf{x}\mathbf{x}\mathbf{x}+\mathbf{x}\mathbf{x}:(\mathbf{J}_4-\mathbf{K}_4)\vdots\mathbf{x}\mathbf{x}\mathbf{x}, \tag{3.6}$$

with $\mathbf{J}_4$, $\mathbf{K}_4$ symmetric and traceless fourth-rank tensor functions of time. It should be noted that at $O(\epsilon^2)$ the external shape is *not* specified by $\mathbf{J}$ alone; it involves also $\mathbf{K}$ and $\mathbf{g}$.

4. Governing equations for capsule deformation

We consider a viscous capsule filled with an incompressible fluid of viscosity $\lambda\mu$ bounded by a two-dimensional elastic membrane and immersed in an incompressible fluid of viscosity μ. We suppose that the outer fluid is sheared at a rate $G(\mathbf{E}+\mathbf{\Omega})$ with $\mathbf{E}$ and $\mathbf{\Omega}$ the symmetric and antisymmetric parts of the velocity gradient, and that the dynamics are inertialess. We seek to determine the time-dependent deformation of the particle in the limit of small shape distortions. This is the case when the elastic restoring forces are much larger than the viscous deforming stresses, or when the internal viscosity is high ($\lambda \gg 1$) and the vorticity non-zero.

We non-dimensionalize the problem as follows: lengths are scaled by the undisturbed particle radius ρ and times by the shear time G^{-1}. If a typical surface elastic modulus for the material is α, then the ratio of elastic restoring forces to viscous deforming stresses is given by

$$k = \alpha/\mu G\rho.$$

In non-dimensional form, the governing equations for the (time-dependent) drop deformation are

$$\left.\begin{array}{llll} \nabla.\mathbf{u}=0, & \nabla.\boldsymbol{\sigma}=0, & \boldsymbol{\sigma}=-p\mathbf{I}+\lambda(\nabla\mathbf{u}+\nabla\mathbf{u}^T) & \text{in } \; V, \\ \nabla.\mathbf{u}=0, & \nabla.\boldsymbol{\sigma}=0, & \boldsymbol{\sigma}=-p\mathbf{I}+(\nabla\mathbf{u}+\nabla\mathbf{u}^T) & \text{in } \; \hat{V}, \\ & & [\mathbf{u}]_S=0, & \end{array}\right\} \tag{4.1}$$

$$\mathbf{u} \sim (\mathbf{E}+\mathbf{\Omega}).\mathbf{x} \quad \text{as} \quad |\mathbf{x}|\rightarrow\infty, \tag{4.2}$$

$$[\boldsymbol{\sigma}.\mathbf{n}]_S = -k\mathbf{f}, \tag{4.3}$$

in which $\mathbf{f}$ is the elastic load exerted by the membrane on the fluid, and $[\,]_S$ denotes the jump of the bracketed quantity across S. By the linearity of the Stokes equations above we may put

$$\mathbf{u} = \mathbf{u}^e + \mathbf{u}^h,$$

in which $\mathbf{u}^e$ and $\mathbf{u}^h$ represent respectively the velocity determined (instantaneously) by the elastic forces, and by the hydrodynamic forces. Each satisfies (4.1), and $\mathbf{u}^e$ satisfies (4.3) with $\mathbf{u}^e \rightarrow 0$ as $|\mathbf{x}|\rightarrow\infty$; $\mathbf{u}^h$ obeys (4.2) with $[\boldsymbol{\sigma}^h.\mathbf{n}]_S = 0$.

4.1. *Determination of* $\mathbf{u}^e$

We have shown above how the load vector $\mathbf{f}$ may be computed from the local deformation gradient. It is straightforward to show that the displacement field (3.2) gives a deformed surface of curvature

$$\nabla.\mathbf{n} = 2+4\epsilon\mathbf{x}.\mathbf{J}.\mathbf{x}+O(\epsilon^2)$$

and that

$$\mathbf{D} = \mathbf{K}+\mathbf{I}\mathrm{X}.(\mathbf{J}-\mathbf{K}).\mathrm{X}/X^2+2\mathrm{X}.(\mathbf{J}-\mathbf{K})\mathrm{X}/X^2-2\mathrm{XXX}.(\mathbf{J}-\mathbf{K}).\mathrm{X}/X^4,$$

giving a local area change characterized by

$$a = \epsilon\mathbf{x}.(2\mathbf{J}-3\mathbf{K}).\mathbf{x}.$$

It follows after substitution into (2.5) that the load vector $\mathbf{f}$ is given by

$$\mathbf{f} = -2\alpha_1\mathbf{n}+\epsilon\mathbf{L}.\mathbf{x}+\epsilon\mathbf{xx}.\mathbf{M}.\mathbf{x}+O(\epsilon^2), \tag{4.4}$$

where

$$\mathbf{L} = 4(\alpha_2+\alpha_3)\,\mathbf{J}-(6\alpha_2+10\alpha_3)\,\mathbf{K}, \tag{4.5}$$

and

$$\mathbf{M} = -4(\alpha_1+2\alpha_2+2\alpha_3)\,\mathbf{J}+(12\alpha_2+16\alpha_3)\,\mathbf{K}. \tag{4.6}$$

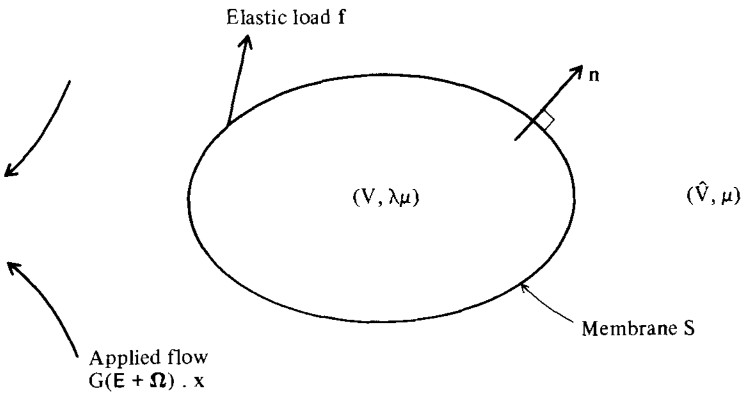

FIGURE 1. Definition sketch for capsule.

Thus $\mathbf{L}$ and $\mathbf{M}$ are both symmetric and traceless and so involve only second harmonics. Now the load $-2\alpha_1\,\mathbf{n}$ does not generate any velocity field, it merely raises the pressure inside the drop, and in consequence we need only find the fluid velocities produced by the $O(\epsilon)$ terms above. Further, correct to $O(\epsilon)$, this load can be considered to act on the unperturbed surface $r = 1$ rather than S, and so the solution of the Stokes' equations is conveniently accomplished by means of Lamb's decomposition into spherical harmonics. We have

$$\mathbf{u}^e = \begin{cases} 6\mathbf{T}^e.\mathbf{x} - \frac{2}{7}\mathbf{xx}.\mathbf{S}^e.\mathbf{x} + \frac{5}{7}r^2\mathbf{S}^e.\mathbf{x}, & \mathbf{x}\in V, \\ 6\hat{\mathbf{T}}^e.\mathbf{x}/r^5 - 15\mathbf{xx}.\hat{\mathbf{T}}^e.\mathbf{x}/r^7 - \mathbf{xx}.\hat{\mathbf{S}}^e.\mathbf{x}/r^5, & \mathbf{x}\in \hat{V}, \end{cases}$$

and, using the four continuity requirements on $\mathbf{u}$, $\boldsymbol{\sigma}.\mathbf{n}$ at $r = 1$, the four unknown tensors $\mathbf{S}^e$, $\hat{\mathbf{S}}^e$, $\mathbf{T}^e$, $\hat{\mathbf{T}}^e$ may be determined. We find

$$\mathbf{T}^e = k(\tfrac{1}{6}b_0\,\mathbf{L} + b_3\,\mathbf{M}), \quad \hat{\mathbf{T}}^e = k(\tfrac{1}{6}b_0\,\mathbf{L} + \tfrac{1}{3}b_1\mathbf{M}),$$
$$\mathbf{S}^e = -\tfrac{7}{2}kb_0\,\mathbf{M}, \quad \hat{\mathbf{S}}^e = -k(\tfrac{5}{2}b_0\,\mathbf{L} + b_0\,\mathbf{M}).$$

In particular, for $\mathbf{x}\in S$, we have

$$\mathbf{u}^e = k\epsilon[b_0\,\mathbf{L}.\mathbf{x} + b_1\mathbf{M}.\mathbf{x} + b_2\,\mathbf{xx}.\mathbf{M}.\mathbf{x} + O(\epsilon)], \tag{4.7}$$

where

$$b_0 = 1/(2\lambda+3); \quad b_1 = 2(3\lambda+2)/(19\lambda+16)(2\lambda+3); \quad b_2 = 2/(19\lambda+16),$$

and

$$b_3 = (16\lambda+19)/(2\lambda+3)(19\lambda+16).$$

4.2. *Determination of* $\mathbf{u}^h$

Taking for S the shape

$$r = 1 + \epsilon\mathbf{x}.\mathbf{J}.\mathbf{x}/r^2 + O(\epsilon^2),$$

the velocity field $\mathbf{u}^h$ for this problem is identical with that for the corresponding surface tension droplet, and may be found by the same method as that above. We find in particular that, for $\mathbf{x}\in S$,

$$\mathbf{u}^h = \boldsymbol{\Omega}.\mathbf{x} + a_0\,\mathbf{E}.\mathbf{x} + \epsilon[a_1 Sd(\mathbf{E}.\mathbf{J}).\mathbf{x} + a_2(\mathbf{E}.\mathbf{J} - \mathbf{J}.\mathbf{E}).\mathbf{x}] + O(\epsilon^2), \tag{4.8}$$

where

$$a_0 = 5/(2\lambda+3); \quad a_1 = 60(\lambda-1)/7(2\lambda+3)^2 \quad \text{and} \quad a_2 = 2(\lambda-1)/(2\lambda+3)$$

and

$$Sd(\mathbf{E}.\mathbf{J}) \equiv \tfrac{1}{2}(\mathbf{E}.\mathbf{J}+\mathbf{J}.\mathbf{E}-\tfrac{2}{3}\mathbf{E}:\mathbf{J}\mathbf{I})$$

denotes a symmetric deviator of order two. (This expression plainly reduces to

$$\mathbf{u}^h = (\mathbf{E}+\mathbf{\Omega}).\mathbf{x} \quad \text{when} \quad \lambda = 1$$

for then fluid of the same viscosity may be regarded as filling all space.)

4.3. *Evolution of the shape*

The evolution of the drop shape (through the variation of $\mathbf{J}$ and $\mathbf{K}$ in time) may now be determined from the kinematic condition

$$\dot{\mathbf{x}} = \mathbf{u}^e+\mathbf{u}^h \quad \text{for each} \quad \mathbf{x}\in S. \tag{4.9}$$

In order to allow for the possibility that the capsule shape adjusts on the elasticity time scale $\mu\rho/\alpha$ rather than the shear time G^{-1} so that $\partial/\partial t = O(k)$ (which is $O(\epsilon^{-1})$ for sufficiently large k), we retain $\epsilon\,\partial/\partial t$ terms. Consequently consistency demands that, whereas $\mathbf{u}^e, \mathbf{u}^h$ have been determined correct to $O(\epsilon)$, the expression for $\mathbf{x}$ must include $O(\epsilon^2)$ terms as well. It follows from (3.2) that

$$\dot{\mathbf{x}} = \dot{\mathbf{X}}+\epsilon[\dot{\mathbf{K}}.\mathbf{X}+\mathbf{K}.\dot{\mathbf{X}}+\dot{\mathbf{X}}\mathbf{X}.(\mathbf{J}-\mathbf{K}).\mathbf{X}+\mathbf{X}\dot{\mathbf{X}}.(\mathbf{J}-\mathbf{K}).\mathbf{X} +\mathbf{X}\mathbf{X}.(\mathbf{J}-\mathbf{K}).\dot{\mathbf{X}}+\mathbf{X}\mathbf{X}.(\dot{\mathbf{J}}-\dot{\mathbf{K}}).\mathbf{X}]+\epsilon^2[\dot{g}_0\mathbf{X}+g_0\dot{\mathbf{X}}+\dot{\mathbf{g}}_4]+O(\epsilon^3),$$

in which

$$\dot{\mathbf{X}} = \boldsymbol{\omega}.\mathbf{X}.$$

Then, substituting the known expressions for $\mathbf{u}^e, \mathbf{u}^h$ into (4.9), and replacing $\mathbf{x}$ by $\mathbf{X}$, we have

$$\begin{aligned}\boldsymbol{\omega}.\mathbf{X}+\epsilon\frac{\mathscr{D}^*}{\mathscr{D}t}\mathbf{K}.\mathbf{X}+\epsilon\mathbf{X}\mathbf{X}.\frac{\mathscr{D}^*}{\mathscr{D}t}(\mathbf{J}-\mathbf{K}).\mathbf{X}+\epsilon^2\left\{\frac{\mathscr{D}^*}{\mathscr{D}t}g_0\mathbf{X}+\frac{\mathscr{D}^*}{\mathscr{D}t}\mathbf{g}_4\right\}+O(\epsilon^2\,\partial/\partial t)\\ = \mathbf{\Omega}.\mathbf{X}+a_0\mathbf{E}.\mathbf{X}+\epsilon\{(\mathbf{\Omega}-\boldsymbol{\omega}).\mathbf{K}.\mathbf{X}+(\mathbf{\Omega}-\boldsymbol{\omega}).\mathbf{X}\mathbf{X}.(\mathbf{J}-\mathbf{K}).\mathbf{X}\\ +a_0\mathbf{E}.\mathbf{K}.\mathbf{X}+a_0\mathbf{E}.\mathbf{X}\mathbf{X}.(\mathbf{J}-\mathbf{K}).\mathbf{X}+a_2(\mathbf{E}.\mathbf{J}-\mathbf{J}.\mathbf{E}).\mathbf{X}+a_1Sd(\mathbf{E}.\mathbf{J}).\mathbf{X}\}\\ +k\epsilon\{b_0\mathbf{L}.\mathbf{X}+b_1\mathbf{M}.\mathbf{X}+b_2\mathbf{X}\mathbf{X}.\mathbf{M}.\mathbf{X}\}+O(\epsilon^2, k\epsilon^2)\end{aligned} \tag{4.10}$$

in which $\mathscr{D}^*/\mathscr{D}t$ is a Jaumann derivative which rotates with $\boldsymbol{\omega}$, the particle angular velocity.

A useful aside may be made at this point concerning the limiting case of very viscous capsules, $\lambda\to\infty$. As noted by Rallison (1980) in connection with very viscous surface tension droplets, the error terms on the right-hand side of (4.10) arise from stretching rather than rotation of the capsule shape. Since the time scale for such stretching is given by the larger of the fluid viscosities $\mu, \lambda\mu$ and not G^{-1} directly, the error terms are not larger than $O(\epsilon^2, k\epsilon^2)/(1+\lambda)$, which is a useful improvement on the error estimate when $\lambda = O(\epsilon^{-1})$ (see §6).

For modest λ, it may be seen without difficulty that $\boldsymbol{\omega} = \mathbf{\Omega}+O(\epsilon)$ so that, to the order of accuracy given, the third and fourth terms of the right-hand side of (4.10) are negligible. The result is then to hold everywhere on the unit sphere $\mathbf{X}.\mathbf{X} = 1$, and the equation may be solved by the technique of Frankel & Acrivos (1970), which involves integrating the expression over the unit sphere, and taking advantage of the orthogonality relations between spherical harmonics. Multiplying (4.10) by $\mathbf{X}$ and integrating, we obtain

$$\boldsymbol{\omega}+\epsilon\frac{\mathscr{D}^*}{\mathscr{D}t}\mathbf{K}+\tfrac{2}{5}\epsilon\frac{\mathscr{D}^*}{\mathscr{D}t}(\mathbf{J}-\mathbf{K}) = \boldsymbol{\Omega}+a_0\,\mathbf{E}+\epsilon\{(\tfrac{2}{5}a_0+a_1)\,\mathrm{Sd}\,(\mathbf{E}.\mathbf{J}) + \tfrac{3}{5}a_0\,\mathrm{Sd}\,(\mathbf{E}.\mathbf{K})+(\tfrac{1}{5}a_0+a_2)\,(\mathbf{E}.\mathbf{J}-\mathbf{J}.\mathbf{E})+\tfrac{3}{10}a_0(\mathbf{E}.\mathbf{K}-\mathbf{K}.\mathbf{E})\} + k\epsilon\{b_0\,\mathbf{L}+(b_1+\tfrac{2}{5}b_2)\,\mathbf{M}\}+\epsilon\mathbf{I}\left[\tfrac{2}{15}a_0\,\mathbf{E}:\mathbf{J}+\tfrac{1}{5}a_0\,\mathbf{E}:\mathbf{K}-\epsilon\frac{\mathscr{D}^*}{\mathscr{D}t}g_0\right]+O(\epsilon^2,k\epsilon^2). \quad (4.11)$$

Similarly, multiplying (4.10) by $\mathbf{XXX}$. and integrating, we find

$$\epsilon\frac{\mathscr{D}^*}{\mathscr{D}t}\mathbf{J} = a_0\,\mathbf{E}+\epsilon\{\tfrac{3}{7}a_0\,\mathrm{Sd}\,(\mathbf{E}.\mathbf{K})+(a_1+\tfrac{4}{7}a_0)\,Sd(\mathbf{E}.\mathbf{J})\}+k\epsilon\{b_0\,\mathbf{L}+(b_1+b_2)\,\mathbf{M}\} + \epsilon\mathbf{I}\left\{\tfrac{1}{2}a_0\,\mathbf{E}:\mathbf{K}+\tfrac{1}{3}a_0\,\mathbf{E}:\mathbf{J}-\tfrac{5}{2}\epsilon\frac{\mathscr{D}^*}{\mathscr{D}t}g_0\right\}+O(\epsilon^2,k\epsilon^2). \quad (4.12)$$

Separating the symmetric and antisymmetric parts in (4.11) and rearranging, we have

$$\epsilon\frac{\mathscr{D}^*}{\mathscr{D}t}(\mathbf{J}-\mathbf{K}) = \tfrac{2}{7}\epsilon a_0[\mathrm{Sd}\,(\mathbf{E}.\mathbf{J})-\mathrm{Sd}\,(\mathbf{E}.\mathbf{K})]+k\epsilon b_2\,\mathbf{M}+O(\epsilon^2,k\epsilon^2), \quad (4.13)$$

$$\boldsymbol{\omega} = \boldsymbol{\Omega}+\epsilon[(\tfrac{1}{5}a_0+a_2)\,(\mathbf{E}.\mathbf{J}-\mathbf{J}.\mathbf{E})+\tfrac{3}{10}a_0(\mathbf{E}.\mathbf{K}-\mathbf{K}.\mathbf{E})]+O(\epsilon^2). \quad (4.14)$$

It may be shown from the incompressibility condition (3.5) that the isotropic terms in (4.11) and (4.12) are identically zero as they should be, for since

$$\epsilon\frac{\mathscr{D}^*}{\mathscr{D}t}\mathbf{J} = \epsilon\frac{\mathscr{D}^*}{\mathscr{D}t}\mathbf{K} = a_0\,\mathbf{E}+O(\epsilon),$$

$$\epsilon\frac{\mathscr{D}^*}{\mathscr{D}t}g_0 = a_0[\tfrac{1}{5}\mathbf{E}:\mathbf{K}+\tfrac{2}{15}\mathbf{E}:\mathbf{J}], \quad \text{as required.}$$

Finally, the fourth-order harmonics may be separated to give

$$\epsilon\frac{\mathscr{D}^*}{\mathscr{D}t}\mathbf{K}_4 = \epsilon\frac{\mathscr{D}^*}{\mathscr{D}t}\mathbf{J}_4 = \frac{a_0}{3}[\mathrm{Sd}_4\,(\mathbf{EJ})-\mathrm{Sd}_4\,(\mathbf{EK})], \quad (4.15)$$

where the fourth-order symmetric deviator is defined in indicial notation by

$$\mathrm{Sd}_4\,(A_{ijab}) = \tfrac{1}{8}\,[A_{ijab}+A_{ijba}+22\text{ others} - \tfrac{2}{7}\,\{\delta_{ab}(A_{ijll}+A_{ilj l}+10\text{ others})+5\text{ others}\} + \tfrac{8}{35}(\delta_{ij}\,\delta_{ab}+\delta_{ia}\,\delta_{bj}+\delta_{ib}\,\delta_{ja})\,(A_{llmm}+A_{lmlm}+A_{lmml})].$$

It may be seen from (4.14) that the membrane angular velocity $\boldsymbol{\omega}$ is independent of its elastic properties except insofar as they have determined $\mathbf{J}$ and $\mathbf{K}$ in past time; the elastic stresses are plainly unable to provide any net couple on the fluid here. It may at first sight seem surprising that $\boldsymbol{\omega}$ depends on the in-surface deformation $\mathbf{K}$ (if $\mathbf{K}$ and $\mathbf{E}$ are not coaxial). The reason is that the *mean* rotation rate of the membrane is defined relative to the original undeformed state and so at $O(\epsilon)$ is biased by $\mathbf{K}$. This dependence could be eliminated by an implicit definition of $\boldsymbol{\omega}$ relative to the deformed surface.

It may be seen from (4.12) and (4.13) that equilibria are possible within the domain of validity of the analysis only if the forcing $a_0\,\mathbf{E}$ is comparable in magnitude to either $\epsilon\boldsymbol{\Omega}$ or $k\epsilon b_0$. This occurs if either k is large (weak flows) or λ is large in a shear flow. The situation here is exactly analogous to that for a surface tension droplet. As noted by Rallison (1980), ϵ may be regarded as being small independently of the magnitudes of k and λ, but the cases of interest arise when the smallness of the distortion (ϵ) is *induced* by either weak flow (k^{-1}) or high viscosity (λ^{-1}), and then ϵ can be identified with the small parameter. The physics in each case is different and we discuss them separately.

5. Weak flows

This case corresponds to $k \to \infty$ with $\lambda = o(k)$. The deformation of the particle is limited by the large elastic modulus α, or by small values of the shear rate G or by both. Then, identifying ϵ with k^{-1}, equations (4.12) and (4.13) give

$$\left.\begin{aligned} \epsilon \frac{\mathscr{D}}{\mathscr{D}t}\mathbf{K} &= a_0\,\mathbf{E} + b_0\,\mathbf{L} + b_1\,\mathbf{M} + O(\epsilon), \\ \epsilon \frac{\mathscr{D}}{\mathscr{D}t}(\mathbf{J}-\mathbf{K}) &= b_2\,\mathbf{M} + O(\epsilon), \end{aligned}\right\} \tag{5.1}$$

in which $\mathscr{D}/\mathscr{D}t$ is a Jaumann derivative rotating with the vorticity $\mathbf{\Omega}$. In fact, at this order $\mathscr{D}/\mathscr{D}t$ may be replaced by $\partial/\partial t$ but greater numerical accuracy is probably obtained by retaining the contribution from $\mathbf{\Omega}$.

As might have been expected, the particle displays a viscoelastic behaviour with two relaxation times corresponding to the two deformation modes $\mathbf{J}$, $\mathbf{K}$ (overall shape and in-surface stretching respectively). The relaxation times depend upon the viscosity ratio and the elastic interface properties. They are computed later for two particular choices of membrane elasticity.

5.1. *Equilibrium solutions*

At equilibrium for any steady flow, neglecting the vorticity terms, we find that

$$\mathbf{M} = 0, \quad \mathbf{L} = -\frac{a_0}{b_0}\mathbf{E} = -5\mathbf{E}.$$

It follows that

$$\left.\begin{aligned} \mathbf{J} &= \frac{5}{2}\frac{3\alpha_2 + 4\alpha_3}{\alpha_1(3\alpha_2+5\alpha_3)+2\alpha_3(\alpha_2+\alpha_3)}\mathbf{E}, \\ \mathbf{K} &= \frac{5}{2}\frac{\alpha_1 + 2\alpha_2 + 2\alpha_3}{\alpha_1(3\alpha_2+5\alpha_3)+2\alpha_3(\alpha_2+\alpha_3)}\mathbf{E}, \end{aligned}\right\} \tag{5.2}$$

so that both $\mathbf{J}$ and $\mathbf{K}$ are proportional to and coaxial with $\mathbf{E}$. In addition, they are independent of λ. This independence from λ is to be expected for an extensional flow at all orders in ϵ since the steady shape for the capsule involves no internal motion, the external shear stress is supported entirely by the elastic forces in the membrane and thus the distortion cannot depend upon the viscosity of the inner fluid, and so is independent of λ. When vorticity is included, even in the steady state there is a tank-treading motion of the membrane, and, since its shape is non-spherical, an internal motion involving shear is generated and thus the steady deformation does depend on λ. This dependence, however, appears only at $O(\epsilon^2)$, since the solution here is a perturbation away from a sphere.

If the vorticity terms in (5.1) are included, the equations for the components of $\mathbf{J}$, $\mathbf{K}$ remain linear, but have a more complex structure, and a numerical procedure for the matrix inversion is more appropriate. The equilibrium values do then depend upon λ.

5.2. *Stability of equilibrium*

The question arises as to the range of elastic parameters for which the equilibrium found above is stable. It is a straightforward matter to perturb the solution and to determine from the corresponding eigenvalue problem whether the perturbation grows or decays. We find that necessary conditions for stability are

$$\left.\begin{aligned} &20(\lambda+1)\,\alpha_1+(23\lambda+32)\,\alpha_2+7(7\lambda+8)\,\alpha_3 > 0 \\ \text{and}\quad &\alpha_1(3\alpha_2+5\alpha_3)+2\alpha_3(\alpha_2+\alpha_3) > 0. \end{aligned}\right\} \tag{5.3}$$

It is clear that the coefficient α_1 represents the pre-stress of the membrane (i.e. the isotropic tension in the membrane in the absence of an applied load); it is therefore determined by the pressure difference between the interior and exterior of the undeformed membrane. It can be seen from the stability criteria (5.3) that if $3\alpha_2+5\alpha_3 > 0$ sufficient (positive) pre-stress is always stabilizing, and that throughout the parameter régime sufficient negative pre-stress always destabilizes the equilibrium.

The signs of α_2, α_3 depend upon whether the material of the membrane strain-hardens or softens on deformation near its equilibrium. For a strain-hardening material, $\alpha_2, \alpha_3 > 0$ and thus under positive surface tension the equilibrium solutions above are stable. If either is negative, however, it is physically clear that there is a possibility of a run-away phenomenon whenever a fluid load is applied (e.g. $\alpha_2 < 0$, $\alpha_1 = \alpha_3 = 0$).

In the case where the membrane is not prestressed, $\alpha_1 = 0$, the equilibrium is given by

$$\mathbf{J} = \frac{5}{2}\frac{3\alpha_2+4\alpha_3}{2\alpha_3(\alpha_2+\alpha_3)}\mathbf{E}, \quad \mathbf{K} = \frac{5}{2\alpha_3}\mathbf{E}$$

with stability only if $\alpha_3 > 0$ and $\alpha_2 > -\alpha_3$ for all values of λ. Thus in this case α_2, α_3 may be determined by measured membrane deformation at equilibrium.

The above results can be explored in more detail for three particular types of membrane behaviour.

5.3. *Three-dimensional isotropic incompressible membrane*

A case of interest is a membrane which is the infinitely thin limit of a three-dimensional, isotropic incompressible material. Then the elastic coefficients α_i are given in non-dimensional form by

$$\alpha_1 = 0, \quad \alpha_2 = \tfrac{2}{3}, \quad \alpha_3 = \tfrac{1}{3}.$$

The characteristic elastic modulus α is identified with the Young's modulus. In this case the differential system defining the shape evolution of the particle becomes

$$\epsilon\frac{\partial}{\partial t}\begin{pmatrix}\mathbf{K}\\ \mathbf{J}-\mathbf{K}\end{pmatrix} = \frac{1}{3(2\lambda+3)(19\lambda+16)}\begin{pmatrix}-2(47\lambda+48) & 12(7\lambda+8)\\ 32(2\lambda+3) & -48(2\lambda+3)\end{pmatrix}\begin{pmatrix}\mathbf{K}\\ \mathbf{J}-\mathbf{K}\end{pmatrix}+\begin{pmatrix}\dfrac{5\mathbf{E}}{2\lambda+3}\\ 0\end{pmatrix}.$$

The solution of this system is straightforward:

$$\mathbf{K} = \tfrac{15}{2}\,\mathbf{E}+\frac{1}{\epsilon(2\lambda+3)(19\lambda+16)}[\mathbf{C}_1(\lambda+24+\Delta)\,e^{-t/\tau_1}+\mathbf{C}_2(\lambda+24-\Delta)\,e^{-t/\tau_2}],$$

$$\mathbf{J}-\mathbf{K} = 5\mathbf{E}+\frac{32}{(19\lambda+16)\,\epsilon}[\mathbf{C}_1\,e^{-t/\tau_1}+\mathbf{C}_2\,e^{-t/\tau_2}],$$

where

$$\Delta^2 = 5377\lambda^2 + 14256\lambda + 9792,$$

and where the two characteristic relaxation times of the system are given by

$$\tau_i = \frac{3(19\lambda+16)(2\lambda+3)\epsilon}{5(19\lambda+24)+(-)^i\Delta}, \quad i = 1, 2,$$

in agreement with Brunn (1980). It should be noted that the steady-state solution is independent of λ:

$$\mathbf{J} = \tfrac{25}{2}\mathbf{E}, \quad \mathbf{K} = \tfrac{15}{2}\mathbf{E},$$

which is identical to the result obtained earlier by Barthès-Biesel (1980). Furthermore, this equilibrium solution is always stable.

5.4. *Pure surface tension membrane*

Another particular case which is included in the model is the surface tension droplet. Then the membrane surface energy is proportional to its area, so that $w = e^a$, and $\alpha_1 = \alpha$, $\alpha_2 = \alpha_3 = 0$. In consequence, $\mathbf{L} = 0$, $\mathbf{M} = -4\alpha\mathbf{J}$ (of course the elastic stresses are independent of the in-surface deformation represented by $\mathbf{K}$ in this case) and thus, neglecting the vorticity terms,

$$\epsilon\frac{\partial\mathbf{K}}{\partial t} = a_0\mathbf{E} - 4b_1\mathbf{J},$$

$$\epsilon\frac{\partial\mathbf{J}}{\partial t} = a_0\mathbf{E} - 4(b_1+b_2)\mathbf{J} = \frac{5\mathbf{E}}{2\lambda+3} - \frac{40(\lambda+1)\mathbf{J}}{(2\lambda+3)(19\lambda+16)}.$$

The latter equation is in agreement with the standard result; the former does not appear in such analyses, since it involves motions within the surface which are irrelevant to the surface tension response. Indeed at equilibrium

$$\frac{\partial\mathbf{J}}{\partial t} = 0 \quad \text{so that} \quad \mathbf{J} = \frac{19\lambda+16}{8(\lambda+1)}\mathbf{E},$$

but plainly $\partial\mathbf{K}/\partial t \neq 0$: there is still continuous shearing of the fibres within the membrane. In consequence, the equilibrium (5.2) is not appropriate here.

We note in passing that the stability of this equilibrium plainly demands that the coefficient of $\mathbf{J}$ be positive, i.e. that $\alpha_1 > 0$ so that the surface tension is positive; it is physically clear that if $\alpha_1 < 0$ the deformation will grow without bound.

5.5. *Constant area membrane*

A particular case of importance occurs when the elastic properties of the membrane are such that its area is preserved to a close approximation. Since the sphere is the shape of minimal surface area for given volume, its overall area is stationary with respect to small (linear) perturbations to the shape, and since the local area change is given by

$$a = \epsilon\mathbf{x}.(2\mathbf{J} - 3\mathbf{K}).\mathbf{x},$$

as noted following equation (2.3), the constancy of area requires $\mathbf{J} = \tfrac{3}{2}\mathbf{K}$ to a good approximation specified below.

Mathematically, the limit of interest is $\alpha_2 \gg \alpha_1, \alpha_3$, and as may be seen in the equilibrium solutions (5.2), this gives $\mathbf{J}, \mathbf{K} \sim \mathbf{E}/\alpha_1$ *not* $\mathbf{E}/\alpha_2$ by virtue of the stationarity property mentioned above. An example is the constitutive equation proposed by Skalak *et al.* (1973) for the red-blood-cell membrane. They choose

$$w = \tfrac{1}{8}B[4(1+b)^2 - 4(1+b) - 2e^{2a}] + \tfrac{1}{8}C(e^{4a} - 2e^{2a}), \tag{5.4}$$

in our notation, and for small deformations this gives

$$\alpha_1 = 0, \quad \alpha_2 = C, \quad \alpha_3 = \tfrac{1}{2}B.$$

With the suggested numerical values $B = 0{\cdot}005$ dyne/cm, $C = 5$ dyne/cm we have $\alpha_2 \gg \alpha_1, \alpha_3$ as required.

Now in the time-dependent case we put

$$\mathbf{J} = \tfrac{3}{2}\mathbf{K} + \mathbf{F}, \tag{5.5}$$

so that $\mathbf{F}$ becomes small ($O(\alpha_2^{-1})$) on a very rapid time scale. Then

$$\mathbf{L} = -4\alpha_3\,\mathbf{K} + 4\alpha_2\,\mathbf{F},$$
$$\mathbf{M} = (4\alpha_3 - 6\alpha_1)\,\mathbf{K} - 8\alpha_2\,\mathbf{F}.$$

Since $\mathbf{F}$ is small, after the initial transient $\mathscr{D}\mathbf{F}/\mathscr{D}t \ll \mathscr{D}\mathbf{K}/\mathscr{D}t$, hence on elimination of the $\alpha_2\,\mathbf{F}$ terms (which are $O(1)$ in view of the largeness of α_2), we find

$$\epsilon\frac{\mathscr{D}\mathbf{J}}{\mathscr{D}t} = \frac{60}{23\lambda+32}\,\mathbf{E} - \frac{8}{23\lambda+32}\,(2\alpha_3 + 3\alpha_1)\,\mathbf{J}. \tag{5.6}$$

This equation has exactly the same structure as that for the time evolution of a surface-tension drop, in which the surface-tension coefficient has become $2\alpha_3 + 3\alpha_1$, and with a slightly different dependence on the viscosity ratio λ. In particular, the equilibrium solution here (at $\mathbf{\Omega} = 0$) is independent of λ, whereas it is not in the surface-tension case.

Because at $O(\epsilon^2)$ any perturbation of the spherical shape involves an overall area increase, the range of validity of the equation above is small. Terms of order $\alpha_2\epsilon^2$ have been neglected, and thus the result is formally correct only for $\epsilon \ll \alpha_1/\alpha_2 \ll 1$.

On the other hand, the estimate of the relaxation rate τ for the shape given by (5.6), namely (in dimensional terms)

$$\tau = \frac{(23\lambda+32)\,\mu\rho}{8(2\alpha_3 + 3\alpha_1)\,\alpha},$$

should still be appropriate for analysing the longer time scale of relaxation for constant area membranes which are substantially deformed and not close to being spherical. For instance, for red blood cells, Schmid-Schönbein (1975) has measured the relaxation of the shape to (non-spherical) equilibrium after the cessation of a steady shear flow, and obtains a return to equilibrium after a time of about 0·6 seconds (corresponding say to 3τ or 4τ). Theoretically, the constitutive equation (5.4) proposed by Skalak *et al.* (1973) can be used to estimate the elastic parameters in $\tau: \alpha_3 \sim 2{\cdot}5\times10^{-3}$ dyne/cm; $\alpha_1 = 0$ (no pre-stress); the ambient fluid viscosity in the experiments was 60 cp; taking for ρ the radius of the sphere of red-blood-cell volume, we have $\rho \sim 3\,\mu$m; and choosing $\lambda \sim 0{\cdot}1$ we obtain $\tau \sim 0{\cdot}2$ s, which is consistent with Schmid-Schönbein's observation. It should be noted however that a viscoelastic component of the membrane response could also generate a relaxation time constant. It is not clear at present which mechanism is principally responsible for the erythrocyte data.

6. Highly viscous capsules

We consider in this section the case of highly viscous capsules $\lambda \to \infty$ for which the strength of the flow is unlimited except that the vorticity is non-zero. We suppose the capsule has been deformed by the flow and take $\epsilon = \lambda^{-1}$. Then reverting to equation (4.10), with the improved error estimates discussed in the paragraph which follows it, the coefficients a_i, b_i may be replaced by their asymptotic values as $\lambda \to \infty$, and the equation may be solved by the Frankel & Acrivos technique as before, noting that on this occasion the terms involving $\boldsymbol{\omega} - \boldsymbol{\Omega}$ are *not* negligible.

We find that the rotation rate of the capsule is now

$$\boldsymbol{\omega} = \boldsymbol{\Omega} + \frac{1}{\lambda}(\mathbf{E}.\mathbf{J} - \mathbf{J}.\mathbf{E}) + O\left(\frac{1}{\lambda^2}\right), \tag{6.1}$$

and that

$$\begin{aligned}\mathscr{D}^*\mathbf{J}/\mathscr{D}t = \tfrac{5}{2}\mathbf{E} + \frac{1}{\lambda}[-\tfrac{15}{4}\mathbf{E} + \tfrac{15}{14}\mathrm{Sd}(\mathbf{E}.\mathbf{K}) + \tfrac{25}{7}\mathrm{Sd}(\mathbf{E}.\mathbf{J}) + \mathrm{Sd}(\mathbf{J}.\mathbf{E}.\mathbf{K}) \\ - \mathrm{Sd}(\mathbf{E}.\mathbf{J}.\mathbf{K})] + \frac{k}{\lambda}(\tfrac{1}{2}\mathbf{L} + \tfrac{5}{19}\mathbf{M}) + O\left(\frac{1}{\lambda^2}, \frac{k}{\lambda^2}\right),\end{aligned} \tag{6.2}$$

$$\begin{aligned}\mathscr{D}^*(\mathbf{J} - \mathbf{K})/\mathscr{D}t = \frac{1}{\lambda}[\tfrac{5}{7}\mathrm{Sd}\{\mathbf{E}.(\mathbf{J} - \mathbf{K})\} + \tfrac{2}{3}\mathrm{Sd}\{\mathbf{E}.\mathbf{J}.(\mathbf{J} - \mathbf{K})\} \\ - \tfrac{2}{3}\mathrm{Sd}\{\mathbf{J}.\mathbf{E}.(\mathbf{J} - \mathbf{K})\}] + \frac{k}{\lambda}(\tfrac{2}{19}\mathbf{M}) + O\left(\frac{1}{\lambda^2}, \frac{k}{\lambda^2}\right).\end{aligned} \tag{6.3}$$

Similarly, the evolution equations for the fourth-order tensors are

$$\frac{\mathscr{D}^*}{\mathscr{D}t}\mathbf{J}_4 = \frac{\mathscr{D}^*}{\mathscr{D}t}\mathbf{K}_4 = \tfrac{5}{6}\mathrm{Sd}_4\{\mathbf{E}(\mathbf{J} - \mathbf{K})\} + O\left(\frac{1}{\lambda}, \frac{k}{\lambda}\right), \tag{6.4}$$

and thus do not involve the elasticity of the membrane at this order.

The $O(1)$ version of these equations is

$$\frac{\mathscr{D}\mathbf{J}}{\mathscr{D}t} = \frac{\mathscr{D}\mathbf{K}}{\mathscr{D}t} = \tfrac{5}{2}\mathbf{E} + O\left(\frac{1}{\lambda}, \frac{k}{\lambda}\right),$$

and hence at this order the shape is unaffected by the membrane properties altogether (and so is identical to the surface-tension droplet). These equations represent a periodic motion with no tendency toward equilibrium, the higher-order $O(k/\lambda)$ terms being necessary to drive the shape toward equilibrium at large times.

In two special cases the equations above may be further simplified.

6.1. *Weak flows*

If k is of the same order of magnitude as λ, we have

$$\left.\begin{aligned}\frac{\mathscr{D}\mathbf{J}}{\mathscr{D}t} = \tfrac{5}{2}\mathbf{E} + \frac{k}{\lambda}(\tfrac{1}{2}\mathbf{L} + \tfrac{5}{19}\mathbf{M}) + O\left(\frac{1}{\lambda}\right), \\ \frac{\mathscr{D}}{\mathscr{D}t}(\mathbf{J} - \mathbf{K}) = \frac{k}{\lambda}\tfrac{2}{19}\mathbf{M} + O\left(\frac{1}{\lambda}\right),\end{aligned}\right\} \tag{6.5}$$

and the same result may be obtained from the high-λ limit of equation (5.1) provided that a Jaumann derivative is used there. The position of the equilibrium given by (6.5) does depend at leading order on the vorticity $\mathbf{\Omega}$, whereas, for modest λ, the equilibrium for weak flows is independent of $\mathbf{\Omega}$.

6.2. *Constant area membrane*

A second double limit of interest occurs when the capsule is both highly viscous, and has a constant area membrane ($\alpha_2 \gg \alpha_1, \alpha_3$). It is then straightforward to show either from (6.1)–(6.3) or from (5.6) (again provided that a Jaumann derivative is used) that

$$\frac{\mathscr{D}\mathbf{J}}{\mathscr{D}t} = \tfrac{60}{23}\mathbf{E} - \frac{8k}{\lambda}\frac{2\alpha_3 + 3\alpha_1}{23}\mathbf{J} + O\left(\frac{1}{\lambda}\right),$$

where the smaller elastic modulus is relevant as regards the definition of k.

7. Dilute solution rheology

We turn finally to examine the constitutive equation for a dilute suspension of identical capsules. Batchelor (1970) has shown that the deviatoric bulk stress in the suspension is given by

$$\boldsymbol{\sigma} = 2\mu(\mathbf{E} + \phi\hat{\mathbf{S}}),$$

where ϕ is the volume concentration of particles, and $\hat{\mathbf{S}}$ is the average stresslet exerted by a single particle on the fluid. We have determined $\hat{\mathbf{S}}$, with neglect of the interactions between particles, as

$$\hat{\mathbf{S}} = \frac{5(\lambda - 1)}{2\lambda + 3}\mathbf{E} - \frac{5}{2}\frac{1}{2\lambda + 3}k\epsilon\mathbf{L} - \frac{1}{2\lambda + 3}k\epsilon\mathbf{M} + O(\epsilon, k\epsilon^2). \tag{7.1}$$

The first term in (7.1) arises from the failure of the interior fluid to deform with the ambient rate of strain $\mathbf{E}$ when $\lambda \neq 1$, and the last two from the elastic stresses generated by the membrane. The constitutive equation for the suspension then follows by relating $\mathbf{L}$, $\mathbf{M}$ to $\mathbf{J}$, $\mathbf{K}$ through (4.5, 6) and using the time-evolution equations for $\mathbf{J}$, $\mathbf{K}$ given in earlier sections as appropriate for the physical properties of the capsule.

Not surprisingly, the constitutive equations have the same structure as the results of Roscoe (1967) and Goddard & Miller (1967) for suspensions of elastic and visco-elastic homogeneous particles whose deviation from sphericity is small. The most obvious difference here is that the suspension has a viscoelastic behaviour with two time constants. The structure of the suspension is described by two second-rank tensors $\mathbf{J}$, $\mathbf{K}$ at leading order, and thus the constitutive equation is a generalization of that proposed by Hand (1962) and discussed by Barthès-Biesel & Acrivos (1973) in the context of suspensions of particles with ellipsoidal anisotropy.

If the equilibrium values (5.2) of $\mathbf{J}$, $\mathbf{K}$ are incorporated into (7.1) we find a zero-shear-rate intrinsic viscosity for the suspension of $\frac{5}{2} + O(\epsilon)$. This agreement with the Einstein result for rigid spheres is of course to be expected, since at equilibrium the particle appears to act as a rigid sphere as far as the external flow is concerned.

8. Discussion

We have shown in this paper how the theory of membrane deformation can be cast in a form suitable for problems involving free solid–fluid interfaces, and, by exploiting the linearity of Stokes equations, how a straightforward analysis of the time evolution of the capsule is possible. The analysis here has been restricted, however, to small deviations from sphericity, and to tackle problems in which such deviations are large (notably for red blood cells where the basic state is a biconcave disk) a more general framework is needed. In addition, we have considered only elastic membranes, and in general a viscoelastic response is involved. The technique presented here can usefully be combined to tackle these harder problems with the method of Rallison & Acrivos (1978) which provides a numerical solution for finite deformations of the surface, and in addition follows in time the positions of Lagrangian points in the surface, thereby permitting more complex constitutive relations to be incorporated into the membrane dynamics. These generalizations will appear in a later study.

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