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Effect of constitutive laws for two-dimensional membranes on flow-induced capsule deformation

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Three constitutive laws (Skalak *et al.*'s law extended to area-compressible interfaces, Hooke's law and the Mooney–Rivlin law) commonly used to describe the mechanics of thin membranes are presented and compared. A small-deformation analysis of the tension–deformation relation for uniaxial extension and for isotropic dilatation allows us to establish a correspondence between the individual material parameters of the laws. A large-deformation analysis indicates that the Mooney–Rivlin law is strain softening, whereas the Skalak *et al.* law is strain hardening for any value of the membrane dilatation modulus. The large deformation of a capsule suspended in hyperbolic pure straining flow is then computed for several membrane constitutive laws. A capsule with a Mooney–Rivlin membrane bursts through the process of continuous elongation, whereas a capsule with a Skalak *et al.* membrane always reaches a steady state in the range of parameters considered. The small-deformation analysis of a spherical capsule embedded in a linear shear flow is modified to account for the effect of the membrane dilatation modulus.

1. Introduction

A capsule consists of a liquid internal medium enclosed by a solid deformable interface. Such capsules are found in nature (e.g. cells, eggs) or are manufactured for various industrial applications (cosmetic, food or biomedical industries). The capsules are typically suspended in another liquid. When the suspension is subjected to flow, viscous stresses are exerted on the capsule interface, and this may lead to large deformations and eventual burst. A major issue is the determination of the mechanical properties of the membrane, as these control the resistance of the capsule to applied stresses. In some cases (biological cells, polymerized interfaces), the interface thickness is so small compared to the capsule dimensions that the membrane may be considered as a two-dimensional solid with hyperelastic or viscoelastic properties.

A constitutive law for a two-dimensional membrane can be obtained in two different ways. In the first approach, the constitutive law is an extrapolation of a three-dimensional elastic relation to a thin material (Green & Adkins 1970). For example, the behaviour of a membrane consisting of an isotropic volume-incompressible material may be described by the two-dimensional equivalent of the Mooney–Rivlin law. Another approach is to postulate directly a two-dimensional constitutive law. This has been accomplished by different authors to describe biological membranes which are nearly area incompressible (Skalak *et al.* 1973; Evans 1973). Although it is

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usually used for area-incompressible membranes, the Skalak *et al.* law accounts separately for both shear deformation and area dilatation, and is thus a good candidate to model membranes even when they are not area incompressible.

In the case of capsules, previous models (e.g. Li, Bartès-Biesel & Helmy 1988; Pozrikidis 1990; Ramanujan & Pozrikidis 1998; Eggleton & Popel 1998; Quéguiner & Barthès-Biesel 1997) have considered either a membrane that allows for area dilatation (with a two-dimensional Mooney–Rivlin law or equivalent) or an area-incompressible membrane (with the Skalak *et al.* law or an equivalent form). If the capsule deformation is sufficiently small, all interface constitutive relations reduce to a linear stress–strain relation, yielding the two-dimensional equivalent of Hooke's law. However, when a capsule undergoes large deformations, the relation between the capsule deformation and the applied stress depends not only on the values of some elastic parameters, but also on the mathematical form of the membrane constitutive law. This observation leads to the following important questions:

how should different laws be compared?

how does the choice of the constitutive law affect the motion of a capsule in flow and thus the solution of the inverse problem, i.e. the evaluation of the membrane properties from experimental measurements?

The objective of this paper is to compare three hyperelastic laws, the two-dimensional Hooke and Mooney–Rivlin laws, and the Skalak *et al.* law extended to area-compressible membranes, and to predict how they influence the flow-induced deformation of a capsule. The reponses of these laws to elementary deformation (uniaxial extension and isotropic dilatation) are compared. The influence of the membrane constitutive law on the global motion of a capsule in a linear shear flow is illustrated by two examples. First, the classical model for small deformations of a spherical capsule suspended in a viscous shear flow is adapted to account for variable membrane area-dilatation modulus. Then, the large deformation of a capsule suspended in a purely straining flow is computed for several membrane laws. It is found that, for membranes with identical small-deformation elastic parameters, the mathematical form of the constitutive law has a strong effect on the overall capsule behaviour.

2. Membrane mechanics

The membrane is assumed to be an infinitely thin sheet of a material that is isotropic in its plane and without bending resistance. It is also assumed that the membrane reacts instantaneously to stress, so that its response is hyperelastic. The case of a viscoelastic membrane is usually treated by adding a linear viscous contribution to the elastic stress (Hochmuth & Waugh 1987; Barthès-Biesel & Sgaier 1985). The equations of membrane mechanics may be expressed in terms of surface curvilinear coordinates (Green & Adkins 1970) or of general Cartesian coordinates (Barthès-Biesel & Rallison 1981, hereafter denoted I). This last approach is briefly presented.

The position of a membrane point is denoted by $\boldsymbol{X}$ in a reference configuration, and by $\boldsymbol{x}(\boldsymbol{X}, t)$ in the deformed state. The surface displacement gradient $\boldsymbol{A}$ is defined as:

$$\boldsymbol{A} = (\boldsymbol{I} - \boldsymbol{nn}) \cdot \frac{\partial \boldsymbol{x}}{\partial \boldsymbol{X}} \cdot (\boldsymbol{I} - \boldsymbol{NN}), \tag{2.1}$$

where $\boldsymbol{N}$ and $\boldsymbol{n}$ are the unit normal vectors to the membrane in the reference and deformed configuration respectively, and $\boldsymbol{I}$ is the three-dimensional identity matrix.

The surface Green–Lagrange deformation tensor $\boldsymbol{e}$ is defined as

$$\boldsymbol{e} = \tfrac{1}{2}[\boldsymbol{A}^T \cdot \boldsymbol{A} - (\boldsymbol{I} - \boldsymbol{NN})]. \tag{2.2}$$

Note that $\boldsymbol{e} = 0$ at the reference state. The tensor $\boldsymbol{A}^T \cdot \boldsymbol{A}$ has two non-zero eigenvalues λ_1^2 and λ_2^2, associated with two orthogonal eigenvectors corresponding to local principal axes of deformation in the tangential membrane plane. The principal extension ratios λ_1 and λ_2 are thus measured along the principal lines of deformation:

$$\lambda_i = \frac{\mathrm{d}s_i}{\mathrm{d}S_i}, \quad i = 1, 2, \text{ no summation over i,}$$

where $\mathrm{d}S_i$ and $\mathrm{d}s_i$ denote line elements in the reference and deformed states. The principal non-zero strain components e_1 and e_2 follow from (2.2):

$$e_i = \tfrac{1}{2}(\lambda_i^2 - 1) \quad i = 1, 2. \tag{2.3}$$

The ratio J_s between the deformed and undeformed local surface area is given by

$$J_s = \lambda_1 \lambda_2 = \sqrt{\det(\boldsymbol{A}^T \cdot \boldsymbol{A} + \boldsymbol{NN})}. \tag{2.4}$$

Following Skalak *et al.* (1973), two-dimensional strain invariants may be defined as

$$I_1 = 2\mathrm{tr}(\boldsymbol{e}) = \lambda_1^2 + \lambda_2^2 - 2 \quad \text{and} \quad I_2 = J_s^2 - 1 = \lambda_1^2 \lambda_2^2 - 1, \tag{2.5}$$

where the definition of I_1 is classical and I_2 is a measure of local area dilatation.

Because the membrane is very thin, the three-dimensional stresses may be replaced by two-dimensional elastic tensions. For example, the Cauchy tensions are forces per unit length measured in the *deformed* membrane plane. The equilibrium of the membrane then relates the Cauchy tension tensor $\boldsymbol{T}(\boldsymbol{x}, t)$ to the external load $\boldsymbol{q}$ (force per unit area of the deformed surface) exerted on the membrane:

$$\left[(\boldsymbol{I} - \boldsymbol{nn}) \cdot \frac{\partial}{\partial \boldsymbol{x}}\right] \cdot \boldsymbol{T} + \boldsymbol{q} = 0. \tag{2.6}$$

In the case of a capsule suspended in a flowing liquid, the load is related to the jump of the hydrodynamic stress $\boldsymbol{\sigma}$ across the interface by $\boldsymbol{q} = \lfloor \boldsymbol{\sigma} \rfloor \cdot \boldsymbol{n}$ (I; Pozrikidis 2001). To close the problem, the elastic tension tensor is related to the deformation by means of the strain energy function $w(I_1, I_2)$ per unit of *initial* membrane area:

$$\boldsymbol{T} = \frac{1}{J_s} \boldsymbol{A} \cdot \frac{\partial w}{\partial \boldsymbol{e}} \cdot \boldsymbol{A}^T, \tag{2.7}$$

or

$$\boldsymbol{T} = \frac{2}{J_s} \left\{ \frac{\partial w}{\partial I_1} \boldsymbol{A} \cdot \boldsymbol{A}^T + \frac{\partial w}{\partial I_2} J_s^2 (\boldsymbol{I} - \boldsymbol{nn}) \right\}. \tag{2.8}$$

In view of the assumed isotropy, the principal directions of tension and deformation are colinear.

The generality of the above equations allows them to be used to describe arbitrary membrane deformations. When the local radius of curvature becomes comparable to the interface thickness, or when the main mode of deformation is due to curvature changes, the membrane bending resistance cannot be neglected. Accounting for the bending resistance complicates the formulation of the membrane mechanics. The contribution of bending moments must be added to the equilibrium relation (2.6) and a constitutive equation for the bending moments must be postulated. A comprehensive presentation of the bending mechanics of thin membranes is given by Pozrikidis (2001).

3. Classical constitutive laws

For each constitutive law, we discuss the tension component T_1 in the principal 1-direction. The other component is obtained by interchanging the indices 1 and 2.

3.1. *Two-dimensional Hooke's law*

The simplest law stems from the assumption of linear dependence of tension on surface deformations. This is the two-dimensional equivalent of Hooke's law (H), restricted to small deformations:

$$T_1^H = \frac{2G_s}{(1-\nu_s)}(e_1 + \nu_s e_2) = \frac{G_s}{(1-\nu_s)}[\lambda_1^2 - 1 + \nu_s(\lambda_2^2 - 1)], \tag{3.1}$$

where G_s denotes the surface shear modulus (expressed in $\mathrm{N\,m^{-1}}$), and $\nu_s (\nu_s \neq 1)$ is the surface Poisson ratio.

3.2. *Mooney–Rivlin law*

Another classical assumption is that the membrane is a very thin sheet of an isotropic volume-incompressible rubber-like material with initially uniform thickness. This leads to the two-dimensional Mooney–Rivlin (MR) law (Green & Adkins 1970, chap. 4 and 9) with strain energy and tension given by

$$w^{MR} = \frac{G_{MR}}{2}\left[\Psi\left(I_1 + 2 + \frac{1}{I_2+1}\right) + (1-\Psi)\left(\frac{I_1+2}{I_2+1} + I_2 + 1\right)\right], \tag{3.2}$$

$$T_1^{MR} = \frac{G_{MR}}{\lambda_1\lambda_2}\left(\lambda_1^2 - \frac{1}{(\lambda_1\lambda_2)^2}\right)\left[\Psi + \lambda_2^2(1-\Psi)\right], \tag{3.3}$$

where G_{MR} is an elastic modulus, and Ψ is a scalar coefficient varying in the range $[0, 1]$ (the neo-Hookean case corresponds to $\Psi = 1$). The area dilatation is unrestricted and is compensated by a corresponding thinning of the membrane.

3.3. *Skalak law*

To model the large deformations of a red blood cell membrane, Skalak *et al.* (1973) introduced the law (SK)

$$w^{SK} = \frac{G_{SK}}{4}(I_1^2 + 2I_1 - 2I_2 + CI_2^2), \tag{3.4}$$

$$T_1^{SK} = \frac{G_{SK}}{\lambda_1\lambda_2}\{\lambda_1^2(\lambda_1^2 - 1) + C(\lambda_1\lambda_2)^2[(\lambda_1\lambda_2)^2 - 1]\}, \tag{3.5}$$

which accounts for shear deformations (first term on the right-hand side of (3.5)) and area dilatation (second term on the right hand side of (3.5)), with associated moduli G_{SK} and CG_{SK}. The red blood cell membrane has a lipid bilayer structure and is thus almost area incompressible but easy to shear. Accordingly, Skalak *et al.* (1973) postulated $C \gg 1$ and studied in detail the predictions of their law in this limit. However, the SK law is very general and can be used to model a two-dimensional membrane whether or not it is area incompressible.

The question now arises as to how to relate the elastic moduli of these different laws for the purpose of comparison.

4. Comparison of laws

In the asymptotic limit of small deformation ($|e_i| \ll 1$, $i = 1, 2$), all hyperelastic laws reduce to Hooke's law (3.1), which thus arises as a reference common limit. The asymptotic form of the MR (3.3) and SK (3.5) laws lead to the following correspondence between parameters:

for any value of Ψ, the Mooney–Rivlin modulus G_{MR} is equal to the shear modulus G_s of a Hookean membrane with Poisson ratio $\nu_s = 1/2$;

the Skalak modulus G_{SK} is equal to the shear modulus G_s of a Hookean membrane, provided that the coefficients ν_s and C are related by $\nu_s = C/(1 + C)$.

However, the three laws predict different behaviour at large deformation under simple strains.

4.1. *Uniaxial extension*

A membrane sample is stretched in direction 1 only: $T_1 \neq 0$, $T_2 = 0$. The condition $T_2 = 0$ allows λ_2 to be expressed in terms of λ_1.

Hooke's law leads to the tension–strain relation:

$$T_1^H = G_s(1 + \nu_s)(\lambda_1^2 - 1) = E_s e_1, \tag{4.1}$$

where E_s is the surface Young modulus, related to G_s by

$$E_s = 2G_s(1 + \nu_s). \tag{4.2}$$

The surface Poisson ratio relates the transversal deformation e_2 to the extension e_1:

$$e_2 = -\nu_s e_1.$$

MR and SK lead, respectively, to

$$T_1^{MR} = \frac{G_{MR}}{\lambda_1^{3/2}}(\lambda_1^3 - 1)[\Psi + (1 - \Psi)/\lambda_1], \tag{4.3}$$

$$T_1^{SK} = G_{SK}\lambda_1(\lambda_1^2 - 1)\sqrt{\frac{1 + C\lambda_1^2}{1 + C\lambda_1^4}}\left[\frac{1 + C\lambda_1^4}{1 + C\lambda_1^2} + \frac{C}{1 + C\lambda_1^4}\right]. \tag{4.4}$$

MR and SK predict complex nonlinear relations between the tension and elongation λ_1. In the limit of small deformations, the surface Young modulus is given by

$$E_s = 2G_s(1 + \nu_s) = 3G_{MR} = 2G_{SK}\frac{1 + 2C}{1 + C}. \tag{4.5}$$

4.2. *Isotropic tension*

A membrane sample is stretched by isotropic tensions ($T_1 = T_2 = T$) and the extension ratios are equal ($\lambda_1 = \lambda_2 = \lambda$). This type of deformation occurs during the inflation of a spherical capsule under an isotropic positive pressure difference between the internal and external liquids. Hooke's law (3.1) leads to

$$T^H = G_s\frac{1 + \nu_s}{1 - \nu_s}(\lambda^2 - 1) = K(\lambda^2 - 1) = K\Delta A/A, \tag{4.6}$$

where K is the area dilatation modulus and $\Delta A/A$ is the relative area change. Then, MR and SK lead to

$$T^{MR} = \frac{G_{MR}(\lambda^4 + \lambda^2 + 1)}{\lambda^6}(\lambda^2 - 1)[\Psi + \lambda^2(1 - \Psi)], \tag{4.7}$$

$$T^{SK} = G_{SK}(\lambda^2 - 1)[1 + C\lambda^2(\lambda^2 + 1)], \tag{4.8}$$

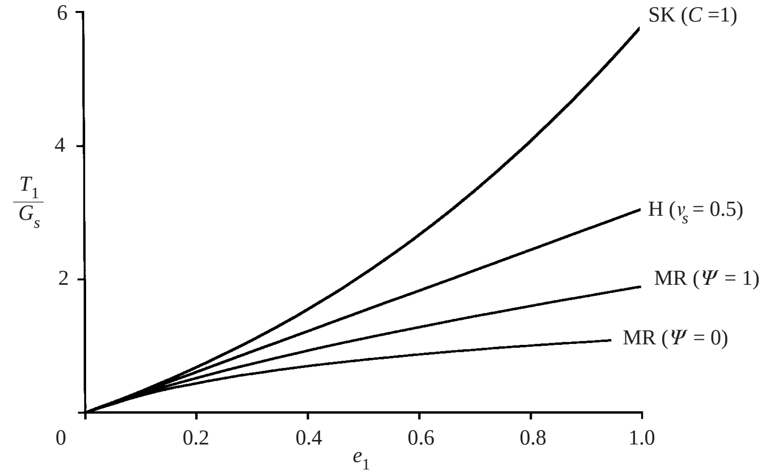

FIGURE 1. Uniaxial extension: principal tension T_1 as a function of principal strain e_1. Comparison of Hooke (H), Mooney–Rivlin (MR) and Skalak *et al.* (SK) laws for the same value of the small-extension Young modulus.

In the limit of small deformations, the area dilatation modulus is given by

$$K = G_s \frac{1+\nu_s}{1-\nu_s} = 3G_{MR} = G_{SK}(1+2C). \tag{4.9}$$

Consequently, an area-incompressible membrane is obtained for a surface Poisson ratio equal to unity (contrary to the three-dimensional case, where an incompressible solid corresponds to a Poisson ratio equal to 1/2).

4.3. *Large-deformation behaviour*

The predictions of the three laws for uniaxial extension are studied by plotting the tension T_1/G_s as a function of the principal strain component e_1 given in (2.3). Two MR membranes (3.3) with either $\Psi = 1$ or 0 are compared to a SK membrane (3.5) with $C = 1$ and to a Hookean membrane (3.1) with $\nu_s = 1/2$ (figure 1). All membranes thus have identical elastic parameters at small deformation. It appears that an MR membrane is easy to extend and is strain softening. Adding a nonlinear term ($\Psi = 0$) promotes the strain softening effect. The SK membrane, on the other hand, is strain stiffening and thus requires larger tensions to achieve the same extension. The effect of the coefficient C is illustrated in figure 2. It is interesting to note that even for $C = 0$, the SK law is still strain stiffening. The curves for $C = 10$ and 100 are almost superimposed. In the case of uniaxial extension, a value of C larger than 1 thus ensures area incompressibility ($C \gg 1$).

The behaviour of the membranes under isotropic extension is illustrated in figure 3, where T/G_s is plotted as a function of the relative area dilatation. The MR, H ($\nu_s = 1/2$) and SK ($C = 1$) membranes have the same area-dilatation modulus. Because the membrane thickness is reduced during deformation, the MR membrane is easy to dilate. This leads again to strain softening even for this type of load, irrespective of the value Ψ. On the other hand, the SK membrane is strain hardening (except for $C = 0$ where the variation of T/G_s with $\Delta A/A$ is linear). For the same value of area dilatation, the tension in an SK membrane increases with C. However, when scaled by the area-dilatation modulus, the tension T/K of an SK membrane becomes nearly independent of C for $C > 10$. This comparison shows that

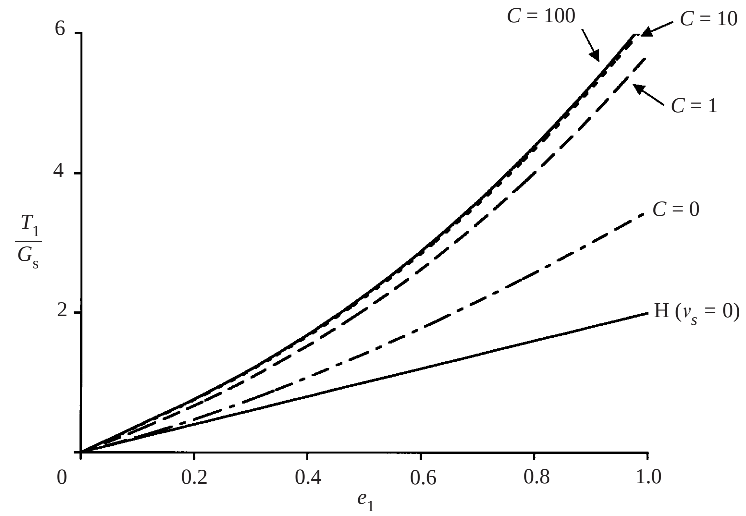

FIGURE 2. Uniaxial extension: effect of the area-dilatation parameter C. Hooke's law is shown only for $v_s = 0$ and is tangent to the $C = 0$ Skalak *et al.* curve. For $C > 10$, an asymptotic state is reached.

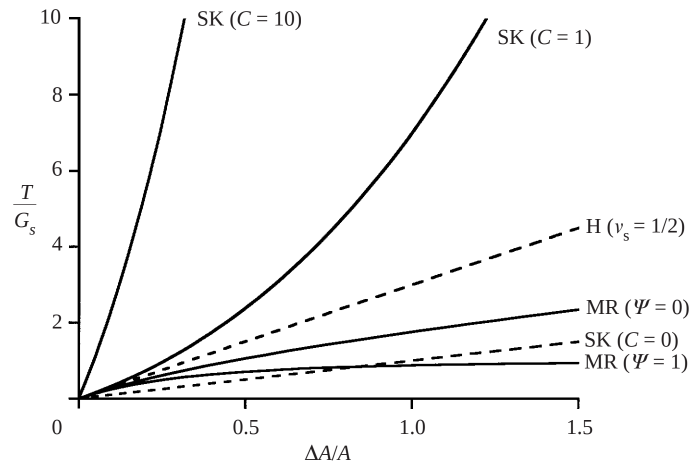

FIGURE 3. Isotropic tension: principal tension T/G_s as a function of relative area change. Comparison of Hooke (H), Mooney–Rivlin (MR) and Skalak *et al.* (SK) laws. For a given level of area change, T increases with C.

in situations where the local area change is large, the choice of a Mooney–Rivlin or a Skalak *et al.* constitutive law leads to quite different predictions.

For physical stability reasons, the area dilatation modulus K must be positive (like the three-dimensional dilatation modulus). Consequently, the two-dimensional Poisson ratio ranges in the interval $]-1, +1[$ (equation (4.9)). Zero or slightly negative values of v_s have been measured for two-dimensional membranes (Pieper, Rehage & Barthès-Biesel 1998). Monte Carlo simulations and mean field calculations for special polymerized networks (termed 'auxetic') have been performed by Boal, Seifert & Shillock (1993) predicting negative values of v_s for intermediate deformations. This can be understood by envisioning the two-dimensional membrane as being wrinkled in the direction perpendicular to its plane. The wrinkles are first smoothed out, and this leads to an expansion in the lateral direction under uniaxial extension.

More complex constitutive laws can be considered where the elastic parameters G_{MR}, G_{SK} or C depend on the strain invariants.

5. Motion of a capsule freely suspended in linear shear flow

We turn now to the question of the influence of the membrane constitutive law on the motion of a capsule in shear flow. The general problem is to determine the motion and deformation of a capsule that is freely suspended in an unbounded linear shear flow, with velocity $\boldsymbol{u}^\infty$ in a reference frame centred on the capsule centre of mass and moving with the capsule:

$$\boldsymbol{u}^\infty = (\boldsymbol{E} + \boldsymbol{\Omega}) \cdot \boldsymbol{x}, \tag{5.1}$$

where $\boldsymbol{E}$ and $\boldsymbol{\Omega}$ are the rate of strain and vorticity tensors. This is a classical problem for which the governing equations are well established (I; Pozrikidis 2001). The flow of the internal liquid and suspending fluid is governed by the Stokes equations. The boundary conditions require continuity of fluid and membrane velocity at the capsule surface. As mentioned in § 2, the load $\boldsymbol{q}$ is equal to the jump in the viscous traction across the interface. The fluid motion and interface deformation are thus strongly coupled.

5.1. *Small deformation of a spherical capsule*

Barthès-Biesel & Rallison (I) have studied the small deformations of an initially spherical capsule of radius a, freely suspended in the flow (5.1). To first order in deformation, the equation of the deformed capsule profile does not depend on the internal liquid viscosity and is given by

$$r^2 = a^2 + 2\mu a \boldsymbol{x} \cdot \boldsymbol{J} \cdot \boldsymbol{x},$$

where μ is the viscosity of the suspending fluid and $\boldsymbol{J}$ is a second-order tensor to be determined as part of the solution. This linear model has been used to analyse experiments performed on artificial capsules (Chang & Olbricht 1993*a*, *b*) or to validate numerical models (Eggleton & Popel 1998; Ramanujan & Pozrikidis 1998). Two constitutive laws for the capsule membrane were considered in I: a Mooney–Rivlin law, corresponding to $\nu_s = 1/2$, and an area-incompressible law, corresponding to $\nu_s = 1$. It is of interest to extend the earlier results to arbitrary values of ν_s and thus to arbitrary values of the membrane area-dilatation modulus.

In the limit of small deformations, the elastic strain energy function of the membrane was assumed in I to be given by

$$w = w_0 + \frac{1}{2}\alpha_2(\ln \lambda_1\lambda_2)^2 + \alpha_3 \left[\frac{1}{2}(\lambda_1^2 + \lambda_2^2) - 1 - \ln \lambda_1\lambda_2\right]. \tag{5.2}$$

The term (coefficient α_1 in I) corresponding to constant isotropic surface tension has been ignored. Making a correspondence between this law and the two-dimensional Hooke's law, we find

$$\alpha_2 = \frac{2\nu_s G_s}{1-\nu_s} = \frac{\nu_s E_s}{1-\nu_s^2}, \quad \alpha_3 = G_s = \frac{E_s}{2(1+\nu_s)}. \tag{5.3}$$

After replacing α_2 and α_3 using (5.3), the equation of the capsule deformed profile is given by

$$r^2 = a^2 + 2\frac{5}{2}\frac{\mu a}{G_s}\frac{2+\nu_s}{1+\nu_s}\boldsymbol{x} \cdot \boldsymbol{E} \cdot \boldsymbol{x} = a^2 + 2\frac{\mu a}{E_s}5(2+\nu_s)\boldsymbol{x} \cdot \boldsymbol{E} \cdot \boldsymbol{x}.$$

It is clear that ν_s has a non-negligible effect on the overall capsule deformation (this effect can be quite large if ν_s takes negative values). However, it is not possible to separate the respective roles of G_s (or E_s) and ν_s on the basis of a single measure of the global deformation of a capsule in shear flow. To obtain the values of G_s and ν_s, it is necessary to perform another independent experiment where the membrane is stressed differently. For example, one may consider measuring the deformation of the capsule due to centrifugal forces exerted in a spinning drop apparatus. In this case, the capsule deformed profile depends on the ratio $(5+\nu_s)/E_s$ (Pieper *et al.* 1998). When feasible, another experiment consists in creating a flat sample of membrane and in shearing it in a surface rheometer. The value of G_s is then obtained directly (Pieper *et al.* 1998).

5.2. *Large deformation of a spherical capsule*

A simple way of examining the effect of the membrane law when the capsule undergoes large deformations is to consider an initially spherical capsule of radius a, freely suspended in an axisymmetric straining flow. Thus, in (5.1), the vorticity tensor is zero, and the only non-zero components of the rate-of-strain tensor are

$$E_{11} = 2\dot{\gamma}, \quad E_{22} = E_{33} = -\dot{\gamma}. \tag{5.4}$$

This problem has been solved numerically for a capsule with a MR membrane (Li *et al.* 1988; Diaz, Pelekasis & Barthès-Biesel 2000), and for a capsule with an area-incompressible membrane (Pozrikidis 1990). The deformation is determined by the capillary number, measuring the ratio of viscous to elastic forces based on either the shear modulus or the Young modulus:

$$\varepsilon_s = \mu\dot{\gamma}a/G_s \quad \text{or} \quad \varepsilon_Y = \mu\dot{\gamma}a/E_s.$$

The relation between the two capillary numbers involves the area-dilatation modulus:

$$\varepsilon_Y = \varepsilon_s \frac{1}{2(1+\nu_s)} = \varepsilon_s \frac{1+C}{2(1+2C)}.$$

Here, the numerical technique of Diaz *et al.* (2000) is used to compute the capsule deformation $D = (L-B)/(L+B)$, where L and B are respectively the length and breadth of the deformed profile in a meridional plane. The method involves following the time-dependent response of a capsule subjected to the sudden start of the flow (5.4). Steady state is assumed to have been reached when the time derivative of D is less than $5 \times 10^{-4}\dot{\gamma}$. For very elongated shapes, it was found necessary to discretize a meridian curve into 113 elements. Details on the numerics are given by Diaz *et al.* (where their expression for the elongational flow field is erroneous and shoud be the same as (5.4)).

The steady deformation of two capsules with an MR membrane ($\Psi = 1$) or an SK membrane with $C = 1$, are first compared in figure 4. The two membranes have the same small-deformation behaviour. However, the membrane constitutive law has a significant effect when the overall capsule deformation is larger than 0.2. Indeed, for an MR membrane, a critical value of ε_s (of order 0.25) exists beyond which there is no steady state and the capsule then elongates indefinitely until burst occurs. Decreasing Ψ from 1 to 0, leads to the same effect, but the critical value of ε_s decreases as shown by Li *et al.* (1988) for prolate ellipsoidal capsules. This phenomenon does not occur for an SK capsule, which always reaches a steady state for all values of ε_s tested here. A capsule with an SK membrane requires larger values of the shear rate to reach the same deformation as an MR capsule. This is due to the shear stiffening behaviour of

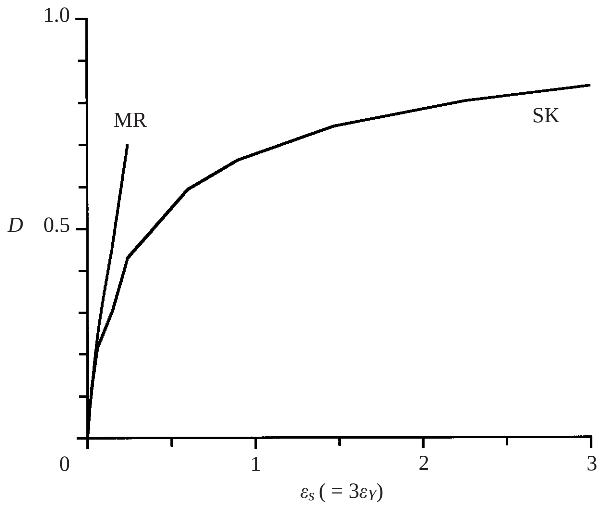

FIGURE 4. Numerical computation of the steady deformation of a spherical capsule in an elongational flow. For a capsule with an MR membrane ($\Psi = 1$) there exists a critical value of ε_s past which no steady state exists and burst occurs. The capsule with an SK membrane ($C = 1$) reaches an asymptotic deformation.

an SK material. As ε_s increases, the SK capsule seems to reach an asymptotic value of deformation of order 0.85 without showing any sign of an impending process of continuous elongation. The existence of this asymptotic state may be explained in the following way. For a given capsule, the increase of ε_s is obtained by increasing the shear rate $\dot{\gamma}$, while keeping G_s constant. As $\dot{\gamma}$ increases, so does the deformation of the membrane. Because of the strain hardening effect, the *apparent* current value G_{sa} of G_s also increases (the way G_{sa} is computed is immaterial). It is possible that the two effects compensate in such a way that the apparent value $\varepsilon_{sa} = \mu\dot{\gamma}a/G_{sa}$ of ε_s remains roughly constant as $\dot{\gamma}$ increases. Similarly, for the strain-softening MR membrane, as $\dot{\gamma}$ and the deformation increase, the apparent current value G_{sa} of G_s decreases. The apparent value ε_{sa} then increases more steeply than that of ε_s. This may be the cause of the continuous elongation process.

The asymptotic maximum extension ratio of the meridian is of the order of 3.5, which is large. Values of ε_s larger than 3 lead to numerical problems due to shape oscillations near the poles. In this region and for very elongated profiles, the curvature becomes large and difficult to compute with precision. The influence of different values of the membrane area-dilatation modulus C is investigated for SK capsules (figure 5*a*, *b*). When D is plotted as a function of ε_s, the effect of C appears clearly: it is easier to deform the capsule with $C = 0$ than it is with $C = 10$. However, when the same deformation is plotted against ε_Y, the three curves are almost superimposed. This indicates that the global deformation of the capsule is governed by the Young modulus and corresponds essentially to a uniaxial extension.

In practice, burst occurs because some break-up criterion (based on either deformation or stress) for the membrane material has been exceeded. Since the numerical model computes the deformation and tension distribution in the membrane for each value of ε_s, it is possible to determine the maximum value of ε_s beyond which an MR or SK capsule suspended in elongational flow will burst. However, an MR capsule will always burst when ε_s exceeds the critical value of 0.25 (for $\Psi = 1$) because of the continuous elongation. It should be noted that a strain-hardening behaviour of the

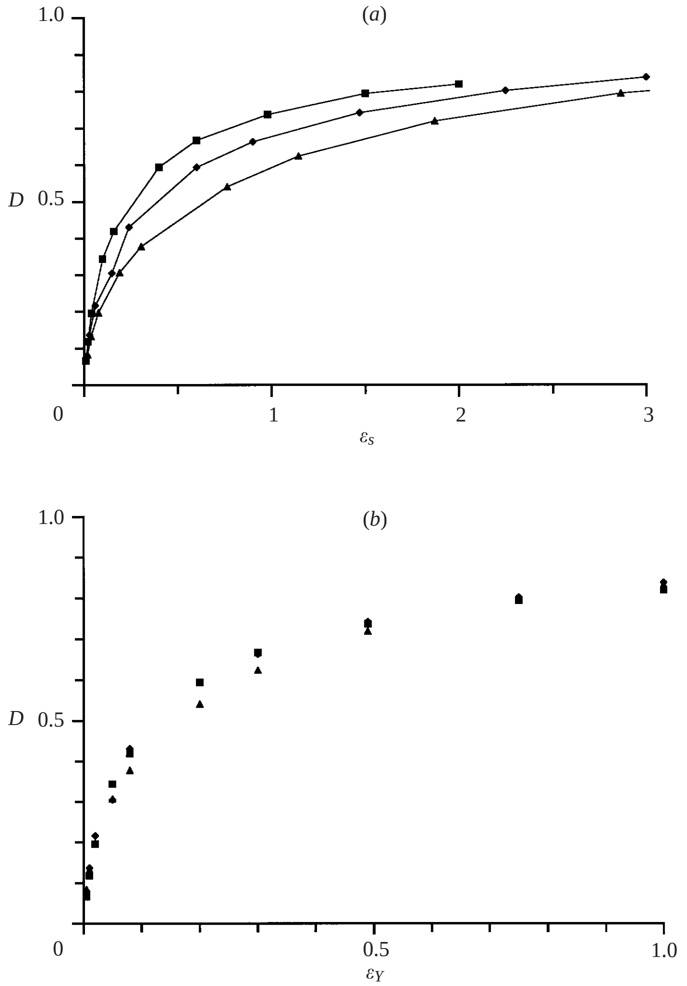

FIGURE 5. Effect of C on the steady deformation of a capsule with an SK membrane. The capillary number is based on the shear modulus (a) or on the Young modulus (b). ■, $C = 0$; ◆, $C = 1$; ▲, $C = 10$.

membrane material is not sufficient to prevent burst. Burst through continuous elongation occurs because the elastic tensions past the critical deformation (corresponding to the critical value of ε_s) can no longer balance the viscous tractions due to flow. Strain hardening can prevent this type of burst if the rate of increase of the elastic tensions with deformation is large enough to allow equilibrium between elastic and viscous forces as the shear rate (i.e. ε_s) increases. As pointed out earlier, this seems to be the case for an SK membrane.

The large deformation of a capsule suspended in a simple shear flow has been simulated numerically by Ramanujan & Pozrikidis (1998) for particles with different initial geometry. They used two different laws for the capsule membrane with strain energies given either by (3.2) with $\Psi = 1$ or by (5.2) with coefficients α_2 and α_3 corresponding to v_S=1/2. Since (5.2) is essentially to a polynomial expansion of (3.2), the capsule deformation depends weakly on the type of law, as observed by Ramanujan & Pozrikidis.

6. Conclusion

The small-deformation analysis allows us to establish a consistent correspondence between the material parameters of the different laws used in the literature to model infinitely thin membranes. When a membrane is subjected to large deformations, these laws predict quite different behaviours under uniaxial extension and isotropic dilatation. The popular two-dimensional Mooney–Rivlin law is strain softening, due to its three-dimensional origin, where any area dilatation is compensated by a decrease of the membrane thickness. The Skalak *et al.* law has been devised to account for shear and area dilatation. It is well suited to membranes with a bilayer structure, which are almost area incompressible ($C \gg 1$). However, for $O(1)$ (or even negative) values of C, the Skalak *et al.* law can also be used to model membranes obtained by interfacial polymerization. A strain stiffening behaviour is predicted, but this effect might be realistic when a polymerized network is deformed so much that the macromolecules are fully extended.

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