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Capsule motion in flow: Deformation and membrane buckling

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Abstract

The objective of this review is to present the salient features of capsule mechanical behaviour under the influence of viscous deforming forces due to a flowing fluid. We focus on artificial capsules that are initially spherical with an internal liquid core and that are enclosed by a very thin hyperelastic membrane. Different constitutive laws, commonly used to describe the rheological behaviour of thin membranes, are presented. The motion and deformation of a single capsule freely suspended in a simple shear flow is presented as a function of membrane constitutive law and of initial pre-stress. The limitations of classical membrane models that neglect bending effects are discussed. ***To cite this article: D. Barthès-Biesel, C. R. Physique 10 (2009).***

Résumé

Mouvement d'une capsule dans un écoulement : Déformation de la membrane. L'objectif de cette revue est de présenter les aspects essentiels du comportement mécanique d'une capsule soumise aux efforts visqueux d'un fluide en écoulement. On se focalise sur les capsules artificielles qui ont une forme initiale sphèrique, un milieu interne liquide et qui sont entourées d'une membrane mince hyperélastique. Différentes lois constitutives usuelles sont présentées pour décrire le comportement mécanique de la membrane. On étudie ensuite le mouvement et la déformation d'une capsule isolée suspendue librement dans un écoulement de cisaillement, en fonction de la loi de paroi et de précontraintes éventuelles. On discute enfin de la limite de validité d'un modèle de membrane qui néglige les effets de résistance à la flexion. ***Pour citer cet article : D. Barthès-Biesel, C. R. Physique 10 (2009).***

Keywords: Capsule; Hyperelastic membrane; Stokes flow; Hydrodynamic interaction; Suspension

Mots-clés : Capsule ; Membrane hyperélastique ; Écoulement de Stokes ; Interaction hydrodynamique ; Suspension

1. Introduction

A capsule consists of some internal medium enclosed by a semi-permeable membrane that controls exchanges between the environment and the internal contents and has thus a protection role. Natural capsules are cells, bacteria or eggs. Artificial capsules are widely used in many industries such as pharmaceutical, cosmetic and food industries for controlled release of active principles, aromas or flavours. They are also used for bioengineering applications like drug targeting or encapsulated cell culture for artificial organs [1].

Artificial capsules can be obtained through interfacial polymerisation of a liquid droplet. The process thus leads to approximately spherical particles enclosed by a thin polymerised membrane with mechanical properties that de-

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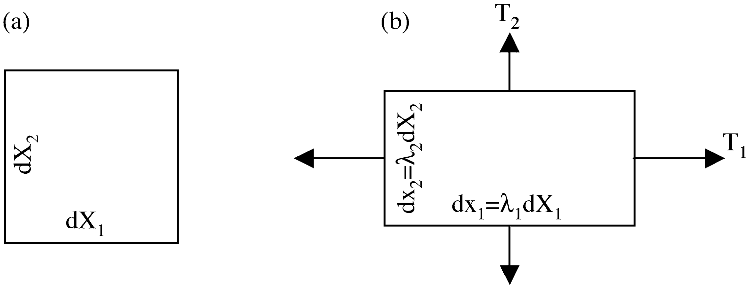

Fig. 1. Principal deformations of a membrane element. (a) Reference state with dimensions $dX_1 \times dX_2$; (b) Deformed state with extension ratios λ_1 and λ_2, under principal tractions T_1, T_2.

pend on the fabrication procedure. For biological applications, the typical membranes that are used are natural or synthetic polymers such as poly-L-lysine, alginate or polyacrylates. A liquid droplet can also be encapsulated by an adsorbed layer of proteins that confers visco-elastic properties to the interface [2]. Such particles are different from phospho-lipid vesicles that are enclosed by a bi-layer membrane with constant area. Indeed a vesicle can take different geometrical shapes at low energy cost, provided its initial shape is not spherical. Conversely, capsule deformation is achieved through stretching of the membrane and costs elastic energy.

In most situations, capsules are suspended into another liquid and are thus subjected to hydrodynamic forces when the suspension is flowing. The motion of the suspending and internal liquids creates viscous stresses on the membrane and may lead to capsule break-up. The control of this process is, of course, essential for the design of artificial capsules or for the protection of natural capsules, but is difficult to achieve unless we have models of the underlying mechanics.

The objective of this review is to present the salient features of the mechanical behaviour of a capsule, under the influence of viscous deforming forces due to a flowing fluid. This situation is encountered in very dilute suspensions or in devices specially designed to measure the deformability of capsules [3,4]. We focus on artificial capsules that are initially spherical with an internal liquid core, enclosed by a very thin hyperelastic membrane. The mechanical properties of the membrane are essential in determining the motion and deformation of the capsule. We thus first present different constitutive laws that are commonly used to describe the rheological behaviour of thin membranes. We then consider the motion and deformation of a single capsule freely suspended in a simple shear flow and discuss the effect of the membrane constitutive law and of initial pre-stress.

2. Capsule properties

We consider an initially spherical capsule with radius a, filled with a Newtonian incompressible liquid with viscosity $\mu^{(1)}$.

2.1. Membrane mechanics

When the capsule wall is very thin compared to a, it can be treated as a hyperelastic surface with surface shear elastic modulus G_s and area dilation modulus K_s. Resistance to bending is ignored as well as shear transverse forces across the wall. Elastic stresses in the membrane are replaced by elastic tensions $\mathbf{T}$ (forces per unit arclength measured in the membrane plane) while deformation is measured by the relative extension and distortion of lines in the membrane plane. If the membrane is further assumed to be isotropic in its plane, the principal directions of deformation and stress are co-linear. A simple way to express the membrane constitutive law is thus to relate the two principal tensions T_1 and T_2 to the two principal extension ratios λ_1 and λ_2 (Fig. 1).

A number of laws have been proposed to model thin membranes, but only simple ones with constant material coefficients are considered here. Depending on the law, quite different material behaviours can be modelled under large deformations. In particular, it is possible to represent strain softening features appropriate for gelled membranes that exhibit rubber-like elasticity or strain hardening behaviour that correspond to membranes made of a polymerised network with strong covalent links. In the limit of small deformations, all laws reduce to Hooke's law (H)

$$T_1 = \frac{G_s}{1-\nu_s}\left[\lambda_1^2 - 1 + \nu_s\left(\lambda_2^2 - 1\right)\right] \tag{1}$$

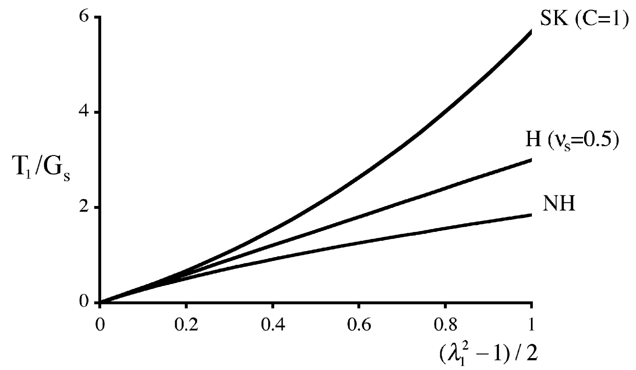

Fig. 2. Uniaxial traction of a membrane element ($T_1 \neq 0$, $T_2 = 0$). Hooke's law is linear, SK law is strain hardening and NH law is strain softening.

where ν_s is the surface Poisson ratio that varies between -1 and $+1$. The area dilation modulus is $K_s = G_s(1+\nu_s)/(1-\nu_s)$. An area incompressible membrane thus corresponds to $\nu_s = 1$.

For large deformations, a widely used law is the neo-Hookean law (NH) that represents the behaviour of an infinitely thin sheet of a three-dimensional isotropic material that is volume incompressible

$$T_1 = \frac{G_s}{\lambda_1\lambda_2}\left[\lambda_1^2 - \frac{1}{(\lambda_1\lambda_2)^2}\right] \tag{2}$$

Owing to the hypothesis of volume incompressibility, area dilation is balanced by membrane thinning and the area dilation modulus K_s is then shown to be $3G_s$ [5]. Another law (SK) has been derived by Skalak et al. [6] for two-dimensional materials

$$T_1 = \frac{G_s}{\lambda_1\lambda_2}\left[\lambda_1^2(\lambda_1^2 - 1) + C(\lambda_1\lambda_2)^2\left[(\lambda_1\lambda_2)^2 - 1\right]\right] \tag{3}$$

The area dilation modulus is given by $K_s = G_s(1+2C)$. The SK law was initially designed to model the area incompressible membrane of biological cells such as red blood cells, corresponding to $C \gg 1$. However, this law is very general and can be used also to model other types of membranes for which K_s and G_s are of the same order of magnitude as is the case for alginate membranes [7].

The expression for T_2 is obtained by interchanging the roles of indices 1 and 2 in (1) to (3). When $C = 1$, NH and SK laws predict the same small deformation behaviour of the membrane with $K_s = 3G_s$, corresponding to $\nu_s = 1/2$. However, they lead to different nonlinear tension-strain relations under large deformations. In particular, it is easily checked that the NH law is strain softening under uniaxial stretching ($T_1 \neq 0$, $T_2 = 0$), whereas SK law is strain hardening as shown in Fig. 2 [5].

2.2. Osmotic effects and membrane pre-stress

A positive pressure difference may occur between inside and outside of the capsule, particularly in bioengineering applications where the membrane is semi-permeable, i.e., permeable to small molecules such as water or small ions but impermeable to large molecules [1]. The pressure difference is then due to osmotic effects. For example, in the case of a simple capsule consisting of a drop of saline solution enclosed by an alginate membrane, some partial dissolution of the membrane occurs and leads to an unknown concentration of large molecules that are trapped inside the capsule [8]. As a consequence, the internal concentration is usually underestimated. Thus when the capsule is suspended in a saline solution with supposedly the same concentration as the internal medium, there exists a concentration jump across the membrane that leads to osmotic effects. We thus now assume that the capsule is subjected to such a positive osmotic pressure difference $p^{(0)}$ between the internal and external phases. Consequently, since the capsule is spherical, the membrane is pre-stressed by an isotropic elastic tension $T^{(0)}$ given by the Laplace law

$$T_1 = T_2 = T^{(0)} = \frac{ap^{(0)}}{2} \tag{4}$$

where a is the radius of the inflated capsule. The membrane is stretched with an initial isotropic elongation $\lambda_1 = \lambda_2 = a/a_0 = 1+\alpha$, where a_0 is the capsule radius in the unstressed configuration. The relation between $T^{(0)}$ and α depends

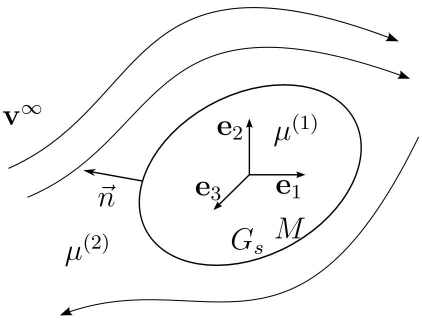

Fig. 3. Schematics of an isolated capsule freely suspended in a simple shear flow.

on the membrane constitutive law [9]. In particular, for a neo-Hookean membrane, $T^{(0)}$ is obtained from (2) and, in the limit of small inflation, is given by $T^{(0)} = 6\alpha G_s$.

2.3. Measurement of membrane properties

Although essential, the membrane constitutive law is quite difficult to determine owing to the smallness and fragility of artificial capsules. Different methods have been proposed to measure the mechanical properties of capsule membranes. They are all based on the measurement of the capsule deformation under a well defined stress. Large millimeter-sized artificial capsules are fairly easy to manipulate. A widely used technique consists in squeezing the particle between two rigid parallel plates and in measuring simultaneously the distance between the plates and the compression force [10,11,7,12,13]. It is also possible to subject the capsule to hydrodynamic forces and to measure the corresponding deformation. This technique has been used for capsules freely suspended in shear flow [3,14] or subjected to centrifugal forces created in a spinning rheometer [15,16]. A limitation of this last technique is linked to the rather low level of mechanical stress that can be applied. In all cases, the membrane mechanical properties are obtained by means of an inverse analysis based on a complete model of the mechanical response of a capsule to the imposed stress. It is also possible to obtain a break-up criterion for the membrane if the deforming forces are increased until burst occurs.

The manipulation of small micron size capsules and cells is much more difficult as it requires the use of microtechniques such as partial aspiration of the membrane in a micropipette under a given pressure or microcompression by various means such as AFM (see the recent comprehensive review [17]). Another technique consists of flowing a suspension of capsules in a channel with cross dimensions of the same order as the capsule size, and in measuring simultaneously the velocity and the deformed profile of the capsules. Then a numerical model of the axisymmetric motion and deformation of an initially spherical liquid-filled capsule of radius a flowing inside a cylindrical tube with radius R is used to infer the membrane elastic properties [18,19].

In conclusion, a capsule can usually deform under applied forces. In order to determine the membrane mechanical properties from the experimentally measured deformation under a given force, a complete mechanical model of the capsule response is needed. Unless the deformation is small, such models are difficult to formulate as capsule mechanics usually involve strong fluid-structure coupling.

3. Capsule dynamics in flow

3.1. Problem description

An initially spherical capsule is suspended in an unbounded shear flow with far field velocity $\mathbf{v}^\infty(\mathbf{x})$ and characteristic velocity V^∞ (Fig. 3). The unknown deformed surface of the capsule is denoted M. The internal (superscript 1) and external (superscript 2) liquids are Newtonian and have equal density ρ. The flow Reynolds number based on the capsule dimension is assumed to be very small $\rho V^\infty a/\mu^{(2)} \ll 1$, so that the motion of the internal and external liquids is governed by the Stokes equations

$$\nabla p^{(\alpha)} = \mu^{(\alpha)} \nabla^2 \mathbf{v}^{(\alpha)}, \qquad \nabla.\mathbf{v}^{(\alpha)} = 0, \quad \alpha = 1, 2 \tag{5}$$

where we use a reference frame linked to the external fluid at infinity and centred on the capsule centre of mass (Fig. 3). The internal and external velocities are equal to the membrane velocity on M

$$\mathbf{v}^{(1)}(\mathbf{x},t) = \mathbf{v}^{(2)}(\mathbf{x},t) = \partial\mathbf{x}(\mathbf{X},t)/\partial t, \quad \mathbf{x} \in M \tag{6}$$

where $\mathbf{x}$ is the current position of a membrane material point that was at position $\mathbf{X}$ in the reference state. Finally, the elastic tension tensor $\mathbf{T}$ is balanced by the external load due to the viscous tractions across the interface

$$\nabla_s.\mathbf{T} + \left[\boldsymbol{\sigma}^{(2)}(\mathbf{x}) - \boldsymbol{\sigma}^{(1)}(\mathbf{x})\right].\mathbf{n} = 0, \quad \mathbf{x} \in M \tag{7}$$

where $\boldsymbol{\sigma}$ denotes the stress tensor in one of the liquids, $\mathbf{n}$ the outward unit normal vector to M and ∇_s the gradient along M. An important parameter of the problem is the ratio between viscous and elastic forces

$$\varepsilon = \mu^{(2)} V^{\infty}/G_s \tag{8}$$

that acts as an equivalent capillary number where surface tension is replaced by the membrane shear elastic modulus. For a given capsule, ε may also be viewed as a non-dimensional shear rate.

The solution of Eqs. (5) to (7) with constitutive equation (2) or (3) is difficult to get, because the problem involves a strong coupling between fluid and solid mechanics, in the domain where the shell deformations may be large and where the hydrodynamic forces are due to pressure and viscous shear stresses. Another difficulty is linked to the simultaneous use of Lagrangian ($\mathbf{X}$) and Eulerian ($\mathbf{x}$) descriptions for the membrane and fluid motion, respectively. The two descriptions are related by (6).

3.2. *Stability issues*

As the fluid motion is described by the Stokes equation, the inertia of the system is neglected. This means that, at any time, the capsule deformation results from the balance between the membrane elastic tensions and the viscous stresses exerted by the flowing liquids. Consequently, there is no guarantee on the *stability* of the equilibrium state that is found by solving the problem equations. This issue is well known for liquid droplets sedimenting in another liquid. In the case of capsules, we have to verify that the solution to the problem is compatible with the initial hypotheses. Specifically, since the bending modulus of the membrane has been neglected, the capsule wall should be under tension everywhere, otherwise, it may buckle locally in the regions where the elastic tensions are compressive. This phenomenon is well known for thin elastic sheets, see for example [20,21]. In that case, a full shell model including bending moments and transverse shear forces is necessary to describe properly the mechanics of the capsule wall.

3.3. *Small deformations*

When the flow strength is weak ($\varepsilon \ll 1$), the capsule deformation is small and asymptotic solutions can be developed for an hyperelastic [22] or linearly visco-elastic membrane [23]. The profile distortion from sphericity is first assumed to be small. Then, the membrane being subjected to small deformations, a linear Hooke's law (1) is appropriate to describe its behaviour. To first order in ε, the asymptotic solution shows that the capsule takes a steady ellipsoidal shape, inclined at an angle $\Phi = 45°$ with respect to the far field streamlines, while the membrane continuously rotates around the steady profile. In the case of a purely elastic membrane, the deformed profile equation is then given by

$$r^2 = x_1^2 + x_2^2 + x_3^2 = a^2 + \frac{5\varepsilon}{\dot{\gamma}} \frac{2+\nu_s}{1+\nu_s} \mathbf{x} \cdot \mathbf{e}^{\infty} \cdot \mathbf{x} \tag{9}$$

where $\mathbf{e}^{\infty} = 1/2(\nabla \mathbf{v}^{\infty T} + \nabla \mathbf{v}^{\infty})$ is the rate of strain of the unperturbed flow. It is clear that ν_s can have a quite large effect on the capsule deformation, particularly if it takes negative values. Such negative values of ν_s occur for membranes that are wrinkled in the direction perpendicular to their plane [24,15].

The predictions from the asymptotic theory are useful to validate numerical models of capsule motion in the range of small deformations or to evaluate the membrane elastic properties from experimental measurements. For example, in a simple shear flow in the $\mathbf{e}_1, \mathbf{e}_2$ plane, given by

$$v_1^{\infty} = \dot{\gamma} x_2; \qquad v_2^{\infty} = v_3^{\infty} = 0 \tag{10}$$

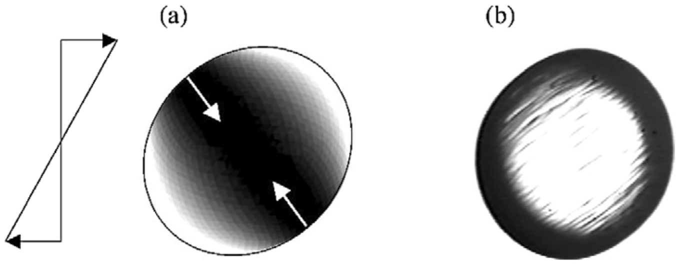

Fig. 4. Small deformation of a capsule freely suspended in a simple shear flow. (a) Asymptotic theory: the membrane undergoes compression in the direction of the arrows. The gray scale measures the relative intensity of compressive tensions. (b) Experimental deformation of a capsule with an organosiloxane membrane: folds appear in the direction of compression (reprinted with permission from Walter et al. [14], © 2001 Elsevier).

the Taylor deformation D of the capsule follows from (9) and is given by

$$D = \frac{L-B}{L+B} = \frac{5}{4}\left(\frac{2+\nu_s}{1+\nu_s}\right)\frac{\mu^{(2)}\dot{\gamma}a}{G_s} \tag{11}$$

where L and B denote respectively the maximum and minimum profile diameters in the shear plane. Eq. (11) has been used to infer an evaluation of the membrane shear elastic modulus of artificial capsules with different membranes [25, 3,4]. The procedure consists in suspending a capsule of known radius a in a simple shear flow and in measuring the deformation D as a function of shear rate $\dot{\gamma}$. Assuming a value for ν_s (usually $\nu_s = 1/2$), it is easy to deduce G_s.

Note though that the asymptotic model predicts that negative tensions occur in the vicinity of the equatorial plane that is orthogonal to the main extension direction (Fig. 4). This means that the equilibrium solution (11) is mechanically unstable and that the membrane may buckle. A physical membrane always has a finite resistance to bending, which prevents buckling when the load is small enough and does not exceed the critical buckling value. This may explain why no buckling is observed on most capsules. However, capsules with an organosiloxane membrane ($\nu_s \simeq 0$) do exhibit buckling, even at very low shear rates [14]. The direction of the folds corresponds to the direction of compression, as predicted by the asymptotic theory. The reason for the early appearance of such folds on a organosiloxane membrane is still an open question. Obviously, part of the answer is that the bending resistance of such a membrane is extremely low. The analysis of the folds can then lead to the value of the bending rigidity [26].

3.4. Numerical models

In the case of large deformations, it is necessary to resort to numerical models. The usual technique of resolution consists in injecting the undeformed capsule in the flow field and in following numerically the time evolution of the capsule motion and deformation until a steady state is reached, if any. At a given time, the position $\mathbf{x}(\mathbf{X}, t)$ of the membrane material points is thus known. By comparison with the initial reference state, the deformation and extension ratios of the capsule membrane are easily computed. The elastic tensions $\mathbf{T}$ follow from the constitutive law (2) or (3). The equilibrium equation (7) leads to the value of the traction jump $[\boldsymbol{\sigma}^{(2)} - \boldsymbol{\sigma}^{(1)}].\mathbf{n}$ on the membrane. Then, the solution of the Stokes equations (5) gives the velocity $\mathbf{v}(\mathbf{x})$ of the membrane points. The time integration of (6) leads to the new position of the membrane material points, and the process is repeated. From a purely numerical point of view, different discretisation techniques have been used, but the general principle is the same.

One technique is based on the boundary integral method that consists in first recasting the Stokes equations (5) in integral form. For the particular case where the viscosity of the two liquids are equal, $\mu^{(1)} = \mu^{(2)} = \mu$, as will be assumed hereafter, the interfacial velocity is given by [27]

$$\mathbf{v}(\mathbf{x}) = \mathbf{v}^{\infty}(\mathbf{x}) - \frac{1}{8\pi\mu}\oint_M \mathbf{J}(\mathbf{x},\mathbf{y}).\left[\boldsymbol{\sigma}^{(2)}(\mathbf{y}) - \boldsymbol{\sigma}^{(1)}(\mathbf{y})\right].\mathbf{n}(\mathbf{y})\,\mathrm{d}S(\mathbf{y}) \tag{12}$$

where $\mathbf{v}^{\infty}(\mathbf{x})$ is the undisturbed flow and $\mathbf{J}$ the Oseen tensor for Stokes flow

$$J_{ij}(\mathbf{x},\mathbf{y}) = \frac{\delta_{ij}}{r} + \frac{r_i r_j}{r^3} \tag{13}$$

with $\mathbf{r} = \mathbf{y} - \mathbf{x}$ and $r = \|\mathbf{r}\|$. The integration is performed on the capsule deformed profile M at time t.

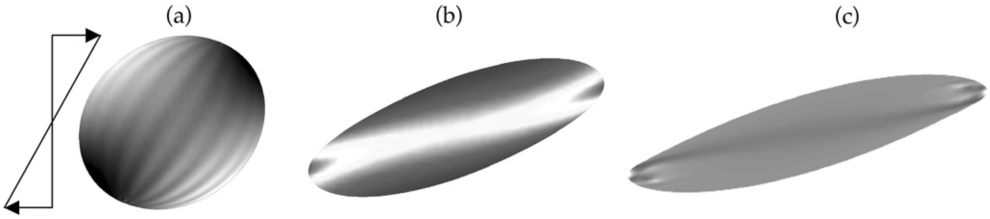

Fig. 5. Deformed profiles of a capsule in simple shear flow. (a) For low flow strength ($\varepsilon < \varepsilon_L$), the membrane is under compression and buckles. (b) For medium flow strength, the equilibrium profile is stable and the membrane rotates around the steady shape. (c) For high flow strength ($\varepsilon > \varepsilon_H$), high curvature tips appear where the membrane is compressed. The gray scale measures the relative intensity of compression forces.

Within the framework of this integral representation, the fluid (12) and solid (7) equations can then be solved on the same grid, thus reducing the geometric dimension of the problem by one. This technique has been used extensively over the years [28–36] and has been shown to be very precise. It is however, restricted to Stokes flows and thus excludes inertial or non-Newtonian effects. Conversely, it is also possible to solve the fluid flow problem by means of the immersed boundary method [37–39]. Two grids are then used: a stationary 3D grid for the fluid flow and a moving 2D boundary grid for the interface. The forces exerted by the membrane on the fluid and the flow velocity convecting the membrane are applied locally from one grid to the other using approximate Dirac functions. However, since the method does not treat the membrane as a physical boundary, it results in a lack of precision of the Lagrangian interface tracking. For the solution of the membrane mechanics problem (7), with any constitutive relation (2) or (3), two approaches may be considered: the equations of the force equilibrium on the capsule wall may either be written locally at each point (strong form used in [28–31,37,32–36,39]) or multiplied by a test function and integrated over the capsule surface in order to define a variational problem that is solved by means of finite elements techniques [37, 38,40]. A prototypical problem, also used for model validation, is the determination of the deformation of an initially spherical capsule freely suspended in a simple shear flow.

4. Large deformation of an initially unstressed capsule in a simple shear flow

We consider the case of an initially unstressed capsule freely suspended in the simple shear flow (10). This configuration has been studied for different shear rates and initial capsule shapes when the membrane obeys a NH law [28,31,32,37,35,38,40] or a SK law [35,39]. The bending rigidity of the membrane has also been taken into account [41,33,34].

The numerical studies first allow one to determine the range of validity of the asymptotic approximation (Section 3.3) which is found to apply within at most 10% for deformations $D \leqslant 0.1$–0.15, as shown in Fig. 6. The smaller limit corresponds to values of ν_s near unity, i.e., to nearly area incompressible membranes. For larger values of deformation, three types of capsule behaviour can be identified depending on the sign of the principal tensions in the membrane. In particular, it is found that for any membrane constitutive law, there exists a shear rate interval $\varepsilon_L < \varepsilon < \varepsilon_H$ in which all the membrane is under elastic traction. The equilibrium solution in this range is then mechanically stable and the membrane model with no bending resistance is appropriate.

When $\varepsilon < \varepsilon_L$, the capsule reaches an equilibrium deformed state, but this equilibrium can be mechanically unstable owing to the presence of negative principal tensions that cause membrane buckling in the absence of bending resistance (Fig. 5a). The compression occurs in the equatorial area where buckling can also be observed experimentally [14].

For $\varepsilon > \varepsilon_L$, as the shear rate increases, the membrane deformation and subsequent area dilation also increase. The tensions in the membrane become all positive due to the contribution of the isotropic term in (2) or (3). The capsule then reaches a steady deformed shape (Fig. 5b) with the membrane continuously rotating around it (*tank-treading* rotation), as has been reported experimentally [3,4,42].

For large shear rates such that $\varepsilon > \varepsilon_H$, the capsule is highly elongated and exhibits high curvature tips that are bent off the main elongation direction by the flow vorticity. Negative tensions occur in the tip area (Fig. 5c). Such tips have also been observed experimentally for capsules with nylon membranes right before break-up, albeit for small values of the viscosity ratio [3].

The value of ε_L depends only slightly on the membrane constitutive law (because the deformation is moderate) and is insensitive to the numerical method [40]. As we go from a strain-softening (NH) to a strain-hardening law (SK),

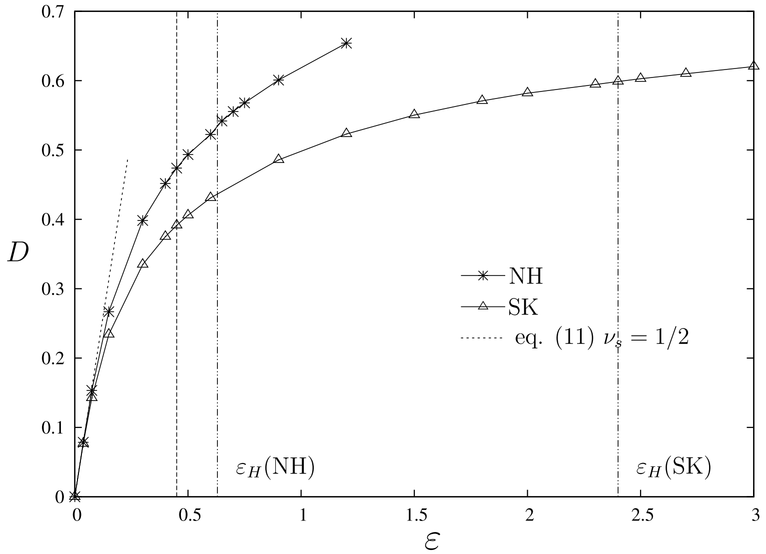

Fig. 6. Steady deformation in the shear plane as a function of membrane constitutive law. The small deformation approximation (11) is valid for $D \leqslant 0.15$. The value of ε_L (dashed line) is almost the same for the two laws. The values of ε_H (mixed dashed lines) depend on the law. A finite element representation of the membrane allows for compressive tensions in the membrane and for shear rates such that $\varepsilon > \varepsilon_H$.

ε_L decreases from 0.45 to 0.4. Conversely, as the value of ε_H occurs for large deformations, it depends on the membrane mechanical properties and increases as the strain hardening effects in the membrane increase (Fig. 6). However, the behaviour of the capsule undergoing negative tensions is strongly method dependent. For example, with a method based on local equilibrium at each node and membrane interpolation by means of cubic B-spline functions, the numerical model becomes unstable when negative tensions occur [35]. Conversely, with a numerical technique based on a variational form of (7) and a finite element method for the membrane, negative tensions are tolerated by the model and stable solutions are obtained with apparent folding at the equator ($\varepsilon < \varepsilon_L$) or at the tips ($\varepsilon > \varepsilon_H$) [40]. This may be explained by the fact that a finite element model introduces some *numerical* bending rigidity to the membrane that prevents overlapping of the nodes and that acts in a fashion that is similar to the mechanical bending rigidity. Such models, although not exact from the mechanics point of view, are nevertheless useful to model a capsule with a wall that is mostly under extension, and subjected to small compression in relatively small areas, only. Accounting for a finite bending stiffness of the membrane, may remove the folds (depending on the bending modulus) and prevent the low shear instability. It must be realised though, that shell models are much more complicated that simple membrane models.

An advantage of the numerical models is that they give access to quantities that are very difficult to measure experimentally such as the elastic tension level in the membrane. As shown in Fig. 7, the maximum steady state tension in the membrane T_{max}/G_s increases sharply with ε and with the strain hardening properties of the membrane. Such information, correlated with a failure criterion for the membrane, may allow to predict the occurrence of capsule burst under flow.

4.1. Effect of osmotic pressure

When the capsule is subjected to a positive osmotic pressure difference $p^{(0)}$ between the internal and external phases, the pre-stress can compensate the negative tensions that appear at low shear rates and membrane buckling can thus be avoided [9]. In all that follows, the capillary number ε is based on the *inflated* capsule radius a, rather than the *unstressed* capsule radius a_0, because it is a that is usually measured. Altogether, the global effect of pre-stress is to decrease the capsule deformation for a given shear rate and to increase the elastic tension in the membrane at a given deformation level. Furthermore, as shown in Fig. 8, the effects of pre-stress and of strain hardening add-up to decrease the membrane deformation for a given flow strength. Correspondingly, a capsule with a 10% pre-inflation and a SK membrane necessitates quite large values of ε to reach a 60% deformation. Finally, pre-stress decreases significantly

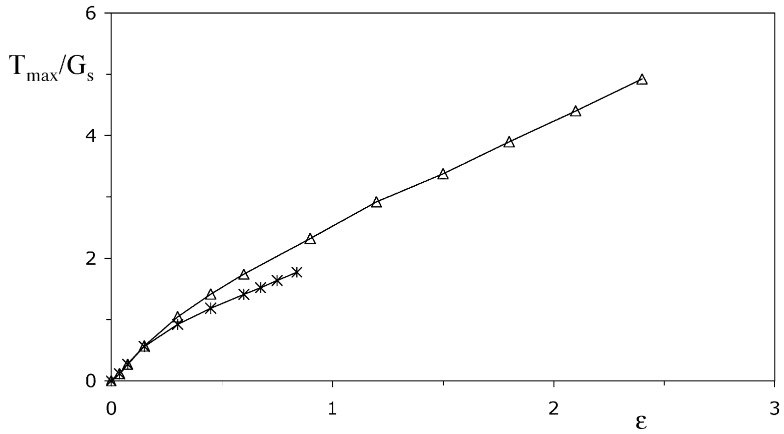

Fig. 7. Maximum steady elastic tension in the membrane as a function of shear strength. Same legend as in Fig. 6. Adapted from [35].

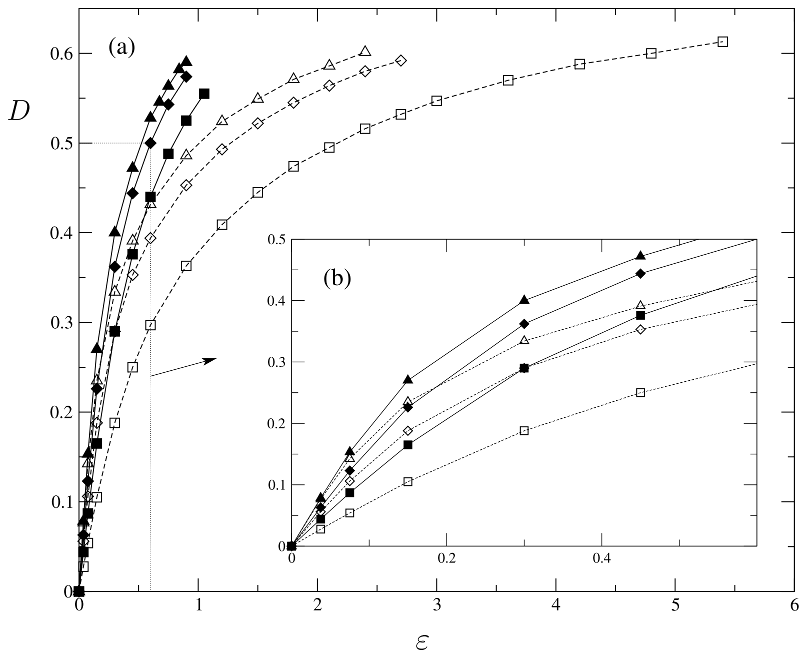

Fig. 8. Steady deformation in the shear plane as a function of pre-inflation and membrane constitutive law. △: $\alpha = 0$; ◇: $\alpha = 2.5\%$; □: $\alpha = 10\%$; filled symbols: NH law; open symbols: SK law ($C = 1$). The capillary number ε is based on the *inflated* capsule radius a. Reprinted with permission from Lac and Barthès-Biesel [9], © 2005 American Institute of Physics.

the value of ε_L that can even disappear. It increases only slightly the value of ε_H, because at large deformation, the effect of the capsule initial state becomes relatively unimportant.

5. Conclusion

Understanding and controlling the flow of a suspension of capsules represents an important challenge for many industrial processes. In particular, it is essential to ensure that no damage will occur when the particles are subjected to flow induced deformations. Most available theoretical results pertain to an isolated capsule freely suspended in shear flow. They show that the membrane constitutive law plays an essential role in determining the capsule behaviour in shear flow. Another important parameter is the osmotic pressure inside the capsule that tends to rigidify the membrane. It is clear too, that bending effects can be important, even for very thin membranes, when they are subjected to compression. This effect has not yet been studied in enough detail and it would be interesting to couple the roles of bending and of membrane constitutive law.

The models developed for small or large deformations of a single capsule can be used to conduct an inverse analysis of experiments and thus obtain information on the mechanics of the capsule membrane [3,4,14,42]. However, except for the pioneering work of Chang and Olbricht [3] there are very few experimental observations of flow induced burst, because the capsules were too resistant or the flow strengths not large enough. This is certainly a domain where more information is needed.

There are very few theoretical or experimental results regarding flow of a suspension of capsules, although this is a situation with many industrial or biological applications. The analysis of the hydrodynamic interaction between two capsules represents a first step in the direction of semi-dilute suspensions models [43,44]. There is certainly need for powerful numerical techniques that can study the hydrodynamic interactions of a collection of capsules and predict the apparent viscosity of the suspension as well as the normal stress effects that are expected to occur. There is also need for experimental observations of interacting capsules.

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