Mechanics of couple-stress fluid coatings

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Abstract

We outline here, the formal development of a theory of viscoelastic surface fluids with bending resistance - their kinematics, dynamics, and rheology. It is relevant to the mechanics of fluid drops and jets coated by a thin layer of immiscible fluid with rather general rheology. This approach unifies the hydrodynamics of two-dimensional fluids with the mechanics of an elastic shell in the spirit of a Cosserat continuum.

Introduction

Recently, Waxman[1,2] has developed a formal theory of viscoelastic surface fluids in which bending resistance was incorporated in a purely phenomenological way. Motivation for this two-dimensional continuum theory stems from a variety of applications: interfacial stability, emulsion rheology, red blood cell deformability, and coated drop and jet mechanics. The mechanics of Newtonian surface fluids, accounting for the evolving surface geometry, was first considered by Scriven[3] (as detailed by Aris[4]). Extension to viscoelastic surface rheologies[2] and the inclusion of bending resistance[1] in the formalism then followed.

Bending rigidity arises from the finite thickness structure of the fluid coating, e.g. surface tension at the multiple interfaces of a compound drop or jet, a layer of normally oriented rod-like molecules such as those which form the lipid bilayer membrane of biological cells, and electrically charged or polarized monolayers at a fluid interface. Whatever the molecular origins of the bending rigidity may be, the associated bending moments (or couple-stresses) may be included in the mechanics of the surface phase in a purely phenomenological way. However, it would clearly be of interest to see if averaging techniques could indeed reduce the mechanics of finite thickness fluid coatings to that of couple-stress surface fluids. Such averaging methods underlie the development and success of elastic shell theory.[5] The direct approach which we have adopted is motivated by the notion of a Cosserat surface which has been exploited by the shell theorists for some time now.[6]

We view our model continuum as a two-dimensional viscoelastic fluid, isotropic in the surface, and associate with each material point on this surface a 'director' (viz. an arrow) oriented along the local normal with its center of mass located at the surface. Changes in surface shape imply a reorientation of these directors which manifests itself dynamically in two ways: reorientation corresponds to curvature changes which generate bending moments, in addition the rate of reorientation corresponds to an internal angular momentum of the surface phase over and above any surface vorticity. We shall see that the director dynamics enters into the surface equations of motion through an asymmetric surface stress tensor and a transverse shearing stress.

There are three distinct facets to the formulation of surface continuum mechanics and we shall try to outline here the important ideas and results associated with each: the kinematics of evolving surface geometries, the conservation laws governing the mechanics of surface continua, and the rheological equations of state governing the surface stress and moment tensors. Further details may be found elsewhere.[1,2]

Evolving surface geometries

As the surface phase is generally located at the interface between two bulk fluids, motions in the bulk lead to a distortion of the interface and hence, an evolution of the surface phase geometry. In order to discuss the mechanics of surface continua we must be able to track the surface as it moves through space. In addition, since various key geometrical quantities (e.g. metric and curvature tensors) enter into the dynamical equations, it is useful to derive evolution equations for these quantities. But first we must establish a coordinate system on the surface. Following Scriven[3], we construct a set of 'fixed surface coordinates' u^α ($\alpha=1,2$) which label geometric points on the surface. As the surface evolves, these fixed coordinates move through space along the local normal to the surface; they are unaffected by any flow of the surface material tangential to the surface. Associated with these fixed coordinates are local tangent vectors $\mathbf{g}_\alpha$ which then define a metric tensor for the surface $a_{\alpha\beta}=\mathbf{g}_\alpha\cdot\mathbf{g}_\beta$. A base vector triad at each point on the surface consists of these tangent vectors $\mathbf{g}_\alpha$ and the local unit normal $\mathbf{n}$ to the surface. Thus, for example, the velocity $\mathbf{v}$ of the surface phase through space may be decomposed according to $\mathbf{v}=v^\alpha\mathbf{g}_\alpha+v^{(n)}\mathbf{n}$.

Here, v^α are the contravariant components of surface velocity and $v^{(n)}$ (a scalar) is the component of velocity in the direction of the local normal.

Let the surface be embedded in an inertial space described by general coordinates x^i (i=1,2,3) with corresponding base vectors $\underline{g}_i$. The surface location is expressed by a relation between the fixed surface coordinates and the space coordinates: $x^i = x^i(u^\alpha, t)$. It can be shown that the evolution of the surface geometry through space is governed by the equation

$$\frac{\partial x^i}{\partial t} = v^{(n)} n^i, \tag{1}$$

where the local normal to the surface has been decomposed as $\underline{n} = n^i \underline{g}_i$. That is, the fixed surface coordinates move through space in the direction of the local normal and do so at a rate given by the normal component of velocity of the surface phase. One may also obtain evolution equations for the metric ($a_{\alpha\beta}$) and curvature ($b_{\alpha\beta}$) tensors associated with the fixed surface coordinate system.

$$\frac{\partial a_{\alpha\beta}}{\partial t} = -2 v^{(n)} b_{\alpha\beta} \tag{2}$$

$$\frac{\partial b_{\alpha\beta}}{\partial t} = v^{(n)}{}_{,\alpha\beta} - v^{(n)} b^\sigma_\alpha b_{\sigma\beta} \tag{3}$$

Equation (2) expresses the fact that a normal velocity distribution over a curved surface leads to a stretching of the fixed surface coordinates (viz. the radial expansion of a spherical surface). Equation (3) describes geometric shape changes via an evolving curvature. The first term, being the second covariant derivative of the normal velocity over the surface, leads to new geometric forms (a first derivative would only express a tilting of the surface). The second term in (3) incorporates the effects of a changing surface metric in the shape changes (e.g. an expanding sphere has a changing radius of curvature though it remains spherical). Simple evolution equations may also be derived for the tangent and normal vectors as well as the Christoffel symbols of the fixed surface coordinate system. As may be seen from equations (1) - (3), the evolution of the surface geometry may be decoupled from the tangential flow of the surface phase in so far as it depends only on the normal component of velocity. However, the normal velocity is implicitly coupled to the tangential flow through the equations of motion governing the surface phase.

Surface equations of motion

The surface equations of conservation of mass, momentum, and angular momentum may all be derived in the fixed surface coordinate system through the use of the Reynolds transport theorem generalized to surface flows. If we may neglect mass exchange between the surface phase and the neighboring bulk fluids on the timescales of interest, then the conservation of surface mass leads to the following continuity equation for the surface mass density γ:

$$\frac{\partial \gamma}{\partial t} + (\gamma v^\alpha)_{,\alpha} - 2H v^{(n)} \gamma = 0. \tag{4}$$

The first two terms in (4) resemble those found in the continuity equation for bulk fluids. The third term is associated with the stretching of the fixed surface coordinates (cf. eq.2) $H = \frac{1}{2} b^\alpha_\alpha$ being the local mean curvature of the surface. If we bring this term to the right-hand side of (4) it appears as a 'mass sink' ($H < 0$ for a sphere) in that it represents a fixed amount of mass being spread over an ever increasing surface area (for $H < 0$ and $v^{(n)} > 0$).

The conservation of linear surface momentum leads us to the following equations of motion for the surface continuum. They may be thought of as dynamical boundary conditions which couple the adjacent bulk phases.

$$\gamma A^\alpha = (f^\alpha + \gamma G^\alpha) + (T^{\beta\alpha}{}_{,\beta} - g^\beta b^\alpha_\beta) \tag{5a}$$

$$\gamma A^{(n)} = (f^{(n)} + \gamma G^{(n)}) + (T^{\alpha\beta} b_{\alpha\beta} - g^\alpha{}_{,\alpha}) \tag{5b}$$

The right-hand sides of (5) closely resemble the equilibrium shell equations.[5] They represent the net tangential and normal forces acting on an element of surface. The surface stress tensor $T^{\alpha\beta}$ and transverse shear stress g^α will be discussed further below; they manifest themselves in local stress vectors (forces per unit length) acting on a curve bounding a small element of surface, $T^{\alpha\beta}$ leading to an in-plane stress, g^α corresponding to a stress normal to the surface along the bounding curve. These are usually termed internal stresses. External stresses act on a small element of surface and arise from body forces (e.g. gravitational and electrostatic forces) represented in (5) by $\underline{G} = G^\alpha \underline{g}_\alpha + G^{(n)} \underline{n}$, and from the neighboring bulk fluids exerting normal and tangential stresses on the surface phase embodied in $\underline{f} = f^\alpha \underline{g}_\alpha + f^{(n)} \underline{n}$. Expressions for f^α and $f^{(n)}$ are given elsewhere.[1,3,4] On the left-hand sides

of (5) we have the tangential and normal components of surface acceleration given by

$$A^{\alpha} = \frac{\partial v^{\alpha}}{\partial t} + v^{\beta} v^{\alpha}{}_{,\beta} - 2 v^{(n)} v^{\beta} b^{\alpha}_{\beta} - v^{(n)} v^{(n)}{}_{,\beta} a^{\alpha\beta} \tag{6a}$$

$$A^{(n)} = \frac{\partial v^{(n)}}{\partial t} + 2 v^{\beta} v^{(n)}{}_{,\beta} + v^{\alpha} v^{\beta} b_{\alpha\beta} . \tag{6b}$$

In addition to the intrinsic derivatives of velocity in equations (6), there are terms resembling centripetal and Coriolis accelerations. They arise from the time varying base vectors associated with the evolving surface coordinate system, i.e. evolving surface is a non-inertial reference frame. In equation (5b), we see how tan stresses conspire with the curvature to generate a normal force (a generalization of place condition). Similarly, the transverse shear in (5a) generates a tangential f a the curvature.

Considerations of angular momentum conservation lead to expressions for the transverse shear stress q^{α} and the antisymmetric part of the surface stress tensor $T^{[\alpha\beta]}$. (We decompose $T^{\alpha\beta}$ into a sum of symmetric and antisymmetric parts, $T^{\alpha\beta} = T^{(\alpha\beta)} + T^{[\alpha\beta]}$.) We find

$$q^{\alpha} = M^{\sigma\alpha}{}_{,\sigma} + \varepsilon^{\alpha\sigma}(H_{\sigma} - \gamma \mathcal{L}_{\sigma}), \tag{7}$$

$$T^{[\alpha\beta]} = \frac{1}{2}\left(b^{\alpha}_{\sigma} M^{\sigma\beta} - b^{\beta}_{\sigma} M^{\sigma\alpha}\right) - \frac{1}{2}\varepsilon^{\alpha\beta}\gamma H^{(n)}, \tag{8}$$

where $M^{\sigma\alpha}$ is the surface moment (or couple-stress) tensor. In equations (7) and (8), $\varepsilon^{\alpha\beta}$ is the contravariant alternating tensor of the surface, H_{σ} and $H^{(n)}$ are components of any externally imposed torque on the surface phase (e g. magnetic couples), and $\mathcal{L}_{\sigma}$ represents the tangential components of internal angular momentum associated with the tumbling motion of the directors. A complicated expression may be derived for $\mathcal{L}_{\sigma}$, but what is important is that it is determined entirely by the velocity field of the surface phase and the surface geometry (along with a presumed moment of inertia). Thus, it introduces no new unknowns into the equations of motion. In arriving at (8) we have assumed that each director spins about its local normal at a rate equal to one-half the local surface vorticity. That is, they are viscously coupled to their surface phase environments and hence, there is no normal component of internal angular momentum. We may use (7) and (8) to eliminate q^{α} and $T^{[\alpha\beta]}$ from the equations of motion (5). These equations simplify enormously for slow flows where we may neglect all terms associated with the inertia of the surface phase. It remains for us to give expressions for the symmetric surface stress and moment tensors $T^{(\alpha\beta)}$ and $M^{\alpha\beta}$.

Surface rheology

We concern ourselves here with surface fluids which are isotropic in the surface, and summarize the rheological laws discussed in detail by Waxman.[1,2] Allowing the surface phase to support an in-plane 'hydrostatic stress' in the absence of any motion, we write

$$T^{(\alpha\beta)} = -\Pi a^{\alpha\beta} + \mathcal{J}^{\alpha\beta}. \tag{9}$$

Here, Π is an isotropic surface pressure (or minus the net surface tension). It is related to the density, temperature, and chemistry of the surface phase and neighboring bulk phases via a thermodynamic equation of state. For incompressible surface continua, Π becomes a dynamic variable to be solved for along with the surface velocity field. The symmetric tensor $\mathcal{J}^{\alpha\beta}$ embodies the viscous and elastic components of stress. (An explicit dependence of Π on γ already represents an area elasticity.)

The Newtonian surface fluid is the simplest example of a viscous surface phase. It is described by a linear relation between stress and rate-of-strain;

$$\mathcal{J}^{\alpha\beta} = C^{\alpha\beta\gamma\delta} S_{\gamma\delta} \tag{10a}$$

$$C^{\alpha\beta\gamma\delta} = \kappa a^{\alpha\beta} a^{\gamma\delta} + \epsilon\left(a^{\alpha\gamma} a^{\beta\delta} + a^{\alpha\delta} a^{\beta\gamma} - a^{\alpha\beta} a^{\gamma\delta}\right) \tag{10b}$$

with κ and ϵ being coefficients of surface dilational and shear viscosity, respectively. The surface rate-of-strain is given by

$$S_{\gamma\delta} = \frac{1}{2}\left(v_{\gamma,\delta} + v_{\delta,\gamma}\right) - v^{(n)} b_{\gamma\delta}. \tag{11}$$

The first term represents the rate-of-deformation due to gradients in the tangential flow, the second term represents the geometric straining associated with the evolving surface

metric (cf. eq.2).

A simple viscoelastic surface fluid which exhibits both stress relaxation in a finite time and delayed elasticity is the 'corotational Jeffreys surface fluid' described by

$$\mathcal{T}^{\alpha\beta} + \lambda \frac{\partial_s \mathcal{T}^{\alpha\beta}}{\partial t} = C^{\alpha\beta\gamma\delta} \left\{ S_{\gamma\delta} + \omega \frac{\partial_s S_{\gamma\delta}}{\partial t} \right\} \tag{12}$$

Here, $C^{\alpha\beta\gamma\delta}$ is of the form (10b), and $\lambda > \omega > 0$ are stress relaxation and strain retardation time constants. The time-derivative operator in (12) represents a rate-of-change as seen from a frame which is translating and corotating (but not codeforming) with an element of surface material. It is the surface analogue of the Jaumann time-derivative. Its derivation and properties are discussed in detail in the work of Waxman.[2] Equation (12) is a quasi-linear rheological law of the rate-type; nonlinear modifications of (12) have also been discussed.[2]

A simple law for the bending moment tensor is motivated by Hookean elasticity. It describes a surface capable of storing potential energy in bending and relates the moment tensor to a measure of bending strain in a linear fashion;

$$M^{\alpha\beta} = C^{\alpha\beta\gamma\delta} K_{\gamma\delta}. \tag{13}$$

Again, $C^{\alpha\beta\gamma\delta}$ is of the form (10b) with κ and ϵ representing independent (positive) moduli of bending rigidity. An appropriate choice of bending strain for surface fluids is[2]

$$K_{\gamma\delta} = -(b_{\gamma\delta} - B_{\gamma\delta}) \quad , \quad \text{where} \tag{14a}$$

$$\frac{\partial_s B_{\gamma\delta}}{\partial t} = 0 \, , \quad B_{\gamma\delta} = \overset{\circ}{b}_{\gamma\delta} \quad \text{at} \quad t = 0. \tag{14b}$$

That is, $K_{\gamma\delta}$ measures the deviation of curvature from a comparison curvature $B_{\gamma\delta}$ which represents an initial reference curvature $\overset{\circ}{b}_{\gamma\delta}$ evolved forward in time in a corotational way. Viscoelastic moment relations of the rate-type may be constructed from (13) through the use of the surface corotational time-derivative operator.

Conclusions

It is hoped that the dynamical formulation outlined here for couple-stress surface fluids will provide a useful approximation to the dynamics of thin fluid coatings in evolving geometries. Application to the mechanics of cell membranes is anticipated in the near future.

References

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