Eur. Phys. J. E **20**, 365–368 (2006)
DOI 10.1140/epje/i2006-10031-3

THE EUROPEAN PHYSICAL JOURNAL E

Rapid Note

A thin-film model for corotational Jeffreys fluids under strong slip

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Received 19 April 2006 and Received in final form 2 August 2006 /
Published online: 11 August 2006 –

Abstract. We derive a thin-film model for viscoelastic liquids under strong slip which obey the stress tensor dynamics of corotational Jeffreys fluids.

PACS. 83.60.Bc Linear viscoelasticity – 47.50.-d Non-Newtonian fluid flows – 68.15.+e Liquid thin films

Dewetting liquid-polymer films on non-wetting substrates, such as silicone wafers grafted with a monolayer of brushes, play a prominent role in many nanotechnological applications. It is known that for these situations, polymer films on the scale of a few hundred nanometers typically show large slippage [1]. Furthermore, for highly entangled polymers, the assumption of a Newtonian fluid will cease to be valid. To understand the interplay of viscous and viscoelastic properties of liquid polymers on hydrophobically coated substrates, there is a need for refined theoretical methods that are able to capture and evolve the emerging morphologies and their longtime dynamics. Dimensionally reduced thin-film models have proved to be highly successful to enable quantitative predictions which are hard to obtain by studying the underlying free-boundary problem.

In this Rapid Note we make an important step in that direction by developing a new thin-film model that combines large slippage with viscoelastic properties. We show that, for the case of strong slip, we are able to derive a thin-film model from the general corotational Jeffreys model. Such a result has, to our knowledge, not been achieved before.

Our work is a continuation of reference [2], in which a family of thin-film models ranging from weak- to strong-slip regimes could be derived for Newtonian liquids, depending on the order of magnitude of the slip length (see also [3]). Modelling the viscoelastic properties of such polymers, generalized Maxwell and Jeffreys models have been widely used. In [4] we have previously discussed a weak-slip model combined with the linearized Jeffreys model to discuss effects of viscoelastic relaxation. More recently [5] we have shown that the strong-slip limit can also be recovered for the linear Jeffreys model. Here we show how to fully incorporate the non-linearities of the corotational Jeffreys model into our thin-film model.

We begin by presenting the underlying free-boundary problem for incompressible, viscous flow with velocity $\mathbf{u}$ and pressure p,

$$\boldsymbol{\nabla} \cdot \mathbf{u} = 0\,, \tag{1}$$

$$\rho \frac{\mathrm{d}\mathbf{u}}{\mathrm{d}t} = -\boldsymbol{\nabla} p + \boldsymbol{\nabla} \cdot \boldsymbol{\tau}, \tag{2}$$

where we assume that the traceless part of the symmetric stress tensor $\boldsymbol{\tau}$ obeys the corotational Jeffreys model with two internal time scales λ_1 and λ_2 [6],

$$\boldsymbol{\tau} + \lambda_1 \frac{D\boldsymbol{\tau}}{Dt} = \mu \left(\dot{\boldsymbol{\gamma}} + \lambda_2 \frac{D\dot{\boldsymbol{\gamma}}}{Dt} \right)\,. \tag{3}$$

Here, D/Dt denotes the Jaumann derivative which for arbitrary tensor fields Λ is given by

$$\frac{D\Lambda}{Dt} = \frac{\mathrm{d}\Lambda}{\mathrm{d}t} + \frac{1}{2}\left(\boldsymbol{\omega}\Lambda - \Lambda\boldsymbol{\omega}\right)\,, \tag{4}$$

where $\dot{\boldsymbol{\gamma}}$ and $\boldsymbol{\omega}$ denote the rate of strain tensor and the vorticity tensor, given by

$$\dot{\boldsymbol{\gamma}} = \nabla\mathbf{u} + (\nabla\mathbf{u})^{\dagger}, \qquad \boldsymbol{\omega} = \nabla\mathbf{u} - (\nabla\mathbf{u})^{\dagger}, \tag{5}$$

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respectively; $\mathrm{d}/\mathrm{d}t$ is the material derivative $\partial_t + \mathbf{u}\cdot\nabla$. We assume the viscosity μ as well as the relaxation parameters λ_1, λ_2 to be constant material parameters.

As boundary conditions we assume that the substrate is impermeable $\mathbf{u}\cdot\hat{\mathbf{e}}_z = 0$ at $z = 0$, and further the Navier-slip boundary condition

$$\mathbf{u}\cdot\hat{\mathbf{e}}_{||} = \frac{b}{\mu}\boldsymbol{\tau}\cdot\hat{\mathbf{e}}_{||}, \tag{6}$$

with a unit vector $\hat{\mathbf{e}}_{||}$ parallel to the substrate (*i.e.*, in x- or y-direction). Here, b is the slip length, which is considered a shear-rate–independent constant as in thin-film flow shear rates are in general low. A power law dependence of b on the shear rate has been observed for rather large shear rates only, see, *e.g.*, [7].

At the liquid surface the normal component of the stress is balanced by the Laplace pressure (with surface tension coefficient γ) and the disjoining pressure $V(z)$, while the tangential components of the stress tensor are zero. In the following we use the reduced pressure $p_R = p + V(z)$.

To simplify the algebra we restrict the calculations to the 2D case and denote the velocity field components by $\mathbf{u} = (u, w)$. We employ the strong-slip scaling, as in [2]. In this limit, the friction between the liquid and the substrate is too weak to maintain a non-zero xz-shear stress to lowest order, and lateral pressure gradients are balanced by the xx-component of the stress tensor. For the stress tensor we use the same scalings as in [4], which means we assume that corresponding components of the strain rate and stress tensor are of the same order. Hence, the dimensional stress tensor reads, in terms of the non-dimensional stress tensor components,

$$\frac{\mu}{T}\begin{pmatrix} \tau^{xx} & \dfrac{\tau^{xz}}{\varepsilon} \\ \dfrac{\tau^{xz}}{\varepsilon} & \tau^{zz} \end{pmatrix}, \tag{7}$$

where μ denotes the viscosity and T denotes the time scale, given by $T = L/U$. U denotes the characteristic velocity scale, such as the dewetting speed and L the relative scale of the lateral extension of the dewetting rim. If H denotes the relative height of the rim, then we let $\varepsilon = H/L \ll 1$.

The dimensionless corotational Jeffreys model can then be written as

$$\left(1+\lambda_1\frac{\mathrm{d}}{\mathrm{d}t}\right)\tau^{xx} - \lambda_1\left(\frac{\partial_z u}{\varepsilon^2} - \partial_x w\right)\tau^{xz} = 2\mu\left(1+\lambda_2\frac{\mathrm{d}}{\mathrm{d}t}\right)\partial_x u - \lambda_2\mu\left(\frac{(\partial_z u)^2}{\varepsilon^2} - \varepsilon^2(\partial_x w)^2\right), \tag{8}$$

$$\left(1+\lambda_1\frac{\mathrm{d}}{\mathrm{d}t}\right)\tau^{zz} + \lambda_1\left(\frac{\partial_z u}{\varepsilon^2} - \partial_x w\right)\tau^{xz} = 2\mu\left(1+\lambda_2\frac{\mathrm{d}}{\mathrm{d}t}\right)\partial_z w + \lambda_2\mu\left(\frac{(\partial_z u)^2}{\varepsilon^2} - \varepsilon^2(\partial_x w)^2\right), \tag{9}$$

$$\left(1+\lambda_1\frac{\mathrm{d}}{\mathrm{d}t}\right)\tau^{xz} + \frac{\lambda_1}{2}(\partial_z u - \varepsilon^2\partial_x w)(\tau^{xx} - \tau^{zz}) = \mu\left(1+\lambda_2\frac{\mathrm{d}}{\mathrm{d}t}\right)(\partial_z u + \varepsilon^2\partial_x w) + \lambda_2\mu\left(\partial_z u - \varepsilon^2\partial_x w\right)\partial_x u\,. \tag{10}$$

These equations are coupled to the non-dimensional governing equations, which read in the strong-slip case as

$$\partial_x u + \partial_z w = 0\,, \tag{11}$$

$$\varepsilon^2\mathrm{Re}^*\frac{\mathrm{d}u}{\mathrm{d}t} = -\varepsilon^2\partial_x p + \varepsilon^2\partial_x\tau^{xx} + \partial_z\tau^{xz}\,, \tag{12}$$

and

$$\varepsilon^2\mathrm{Re}^*\frac{\mathrm{d}w}{\mathrm{d}t} = -\partial_z p + \partial_x\tau^{xz} + \partial_z\tau^{zz}\,, \tag{13}$$

where $\mathrm{Re}^* - \mathrm{Re}/\varepsilon^2$ is the reduced Reynolds number.

The corresponding scaled boundary conditions at $z = h(x,t)$ are given by

$$-p_R + \frac{\varepsilon^2\partial_x h^2\tau^{xx} - 2\partial_x h\tau^{xz} + \tau^{zz}}{1+\varepsilon^2(\partial_x h)^2} = \frac{\partial_{xx}h}{(1+\varepsilon^2(\partial_x h)^2)^{3/2}} \tag{14}$$

and

$$\tau^{xz}\left(1-\varepsilon^2(\partial_x h)^2\right) - \varepsilon^2\partial_x h\,\overline{\tau} = 0\,. \tag{15}$$

In the last equation we have introduced the first normal stress difference $\overline{\tau}$ (commonly denoted by N_1 [6])

$$\overline{\tau} \equiv \tau^{xx} - \tau^{zz}\,. \tag{16}$$

Finally, the kinematic condition reads

$$\partial_t h = -\nabla\cdot\int_0^h \mathrm{d}z\ \mathbf{u}\,. \tag{17}$$

Note that $p_R = p + V(h)$, where $V(h)$ here denotes a disjoining pressure due to dispersion forces across the film.

At the substrate, $z = 0$, we have as usual the impermeability condition $w = 0$ and the slip condition $u = b\tau^{xz}$. In the strong-slip regime we have $b = \beta_s/\varepsilon^2$, β_s being the slip length parameter of order $\mathcal{O}(\varepsilon^0)$, see [2] for details.

For the derivation of the lubrication approximation we assume that the dynamic variables can be given in form of the asymptotic expansions

$$(u, w, h, p, \tau^{ij}) = (u_0, w_0, h, p_0, \tau_0^{ij}) + \varepsilon^2(u_1, w_1, h_1, p_1, \tau_1^{ij}) + O(\varepsilon^4)\,. \tag{18}$$

To leading order in ε we require from equations (8) and (9) that

$$\partial_z u_0 = 0 \qquad \text{or} \qquad \tau_0^{xz} = \frac{\lambda_2}{\lambda_1}\partial_z u_0\,. \tag{19}$$

The governing equations are, to leading order,

$$\partial_x u_0 + \partial_z w_0 = 0\,, \tag{20}$$

$$\partial_z\tau_0^{xz} = 0\,, \tag{21}$$

$$\partial_z p_0 = \partial_x\tau_0^{xz} + \partial_z\tau_0^{zz}\,, \tag{22}$$

and the leading-order boundary conditions at $z = h_0$ are

$$\tau_0^{xz} = 0\,, \tag{23}$$

$$p_{R0} - 2\left(\frac{\tau_0^{zz}}{2} - \partial_x h_0 \tau_0^{xz}\right) + \partial_{xx} h_0 = 0\,, \tag{24}$$

$$\partial_t h_0 - w_0 + u_0 \partial_x h_0 = 0\,. \tag{25}$$

Leading-order boundary conditions at the substrate are

$$w_0 = 0 \qquad \text{and} \qquad \tau_0^{xz} = 0\,. \tag{26}$$

We now integrate (21) with respect to z and use the boundary conditions (23) to find

$$\tau_0^{xz} = 0\,. \tag{27}$$

With the constitutive equations (19) and the boundary conditions (26) we are led to the plug flow condition $\partial_z u_0 = 0$.

The next-order equations for (8, 9) are then given by

$$\left(1 + \lambda_1 \frac{\mathrm{d}^0}{\mathrm{d}t}\right)\tau_0^{xx} = 2\left(1 + \lambda_2 \frac{\mathrm{d}^0}{\mathrm{d}t}\right)\partial_x u_0\,, \tag{28}$$

$$\left(1 + \lambda_1 \frac{\mathrm{d}^0}{\mathrm{d}t}\right)\tau_0^{zz} = 2\left(1 + \lambda_2 \frac{\mathrm{d}^0}{\mathrm{d}t}\right)\partial_z w_0\,, \tag{29}$$

where $\mathrm{d}^0/\mathrm{d}t = \partial_t + u_0\partial_x + w_0\partial_z$. Also, from (22), we find with the solution (27) and the boundary condition (24) for the pressure at the liquid surface $z = h_0$,

$$p_0 = \tau_0^{zz} - \partial_{xx} h_0 - V(h_0)\,. \tag{30}$$

The next-order ($O(\varepsilon^2)$) u-momentum equation is

$$\mathrm{Re}^* \frac{\mathrm{d}^0 u_0}{\mathrm{d}t} = -\partial_x p_0 + \partial_x \tau_0^{xx} + \partial_z \tau_1^{xz}\,.$$

Using (30) and denoting $u_0 = f(x,t)$ we obtain

$$\mathrm{Re}^* \left(\partial_t f + f\partial_x f\right) = \partial_x \overline{\tau_0} + \partial_x(\partial_{xx} h_0 + V) + \partial_z \tau_1^{xz}\,. \tag{31}$$

Integrating this last equation from 0 to h_0 we find, using the slip boundary conditions to the next order, $\tau_1^{xz} = f/\beta_s$,

$$h_0 \mathrm{Re}^* \left(\partial_t f + f\partial_x f\right) = \partial_x \int_0^{h_0} \mathrm{d}z\, \overline{\tau_0} \;-\; \overline{\tau_0}|_{z=h_0}\, \partial_x h_0 + h_0 \partial_x(\partial_{xx} h_0 + V) + \tau_1^{xz}|_{z=h_0} - \frac{f}{\beta_s}\,. \tag{32}$$

The next-order tangential stress boundary condition at $z = h_0$ is

$$\tau_1^{xz} = \overline{\tau_0}\, \partial_x h_0\,. \tag{33}$$

Hence

$$h_0 \mathrm{Re}^* \left(\partial_t f + f\partial_x f\right) = \partial_x \int_0^{h_0} \mathrm{d}z\, \overline{\tau_0} + h_0 \partial_x(\partial_{xx} h_0 + V) - \frac{f}{\beta_s}\,. \tag{34}$$

From equations (28) and (29) we obtain an equation for the difference of the diagonal terms of the stress tensor,

$$(1 + \lambda_1\partial_t + \lambda_1 f\partial_x - \lambda_1 z \partial_x f \partial_z)\overline{\tau_0} = 4\left(\partial_x f + \lambda_2 \partial_t \partial_x f + \lambda_2 f(\partial_x f)^2 - \lambda_2 z \partial_x f \partial_z \partial_x f\right)\,. \tag{35}$$

We now define a film average S of $\overline{\tau_0}$ as

$$S \equiv \frac{1}{4h_0}\int_0^{h_0} \mathrm{d}z\, \overline{\tau_0}\,. \tag{36}$$

We denote the r.h.s. of (35) by $G(x,t)$ and observe that the last term is zero. Integrating (35) with respect to z then yields

$$4h_0 S + \lambda_1 \partial_t(4h_0 S) + \lambda_1 f \partial_x(4h_0 S) + \lambda_1(4h_0 S)\partial_x f - \lambda_1 \overline{\tau_0}|_{z=h_0}\left(\partial_t h_0 + f\partial_x h_0 + h_0 \partial_x f\right) = h_0 G(x,t)\,. \tag{37}$$

Using the kinematic condition to leading order

$$\partial_t h_0 + \partial_x(f h_0) = 0 \tag{38}$$

and employing the definition of $G(x,t)$ then finally leads to

$$(1 + \lambda_1\partial_t + \lambda_1 f \partial_x)S = (\partial_x + \lambda_2 \partial_t \partial_x + \lambda_2 f \partial_{xx})f\,. \tag{39}$$

The lubrication model can finally be written as

$$h_0 \mathrm{Re}^* \left(\partial_t f + f\partial_x f\right) = \partial_x(4h_0 S) + h_0 \partial_x(\partial_{xx} h_0 + V) - \frac{f}{\beta_s} \tag{40}$$

together with (38) and (39). Note that only the advective non-linearities, but not the corotational non-linearities appear in (39). Indeed, these equations are identical to the strong-slip model derived in [5] but for the advective terms in the time derivatives in (39) and (40). This implies that the use of the upper or lower advective derivative instead of the Jaumann derivative (leading to Oldroyd's model A or B) leads to the same one-dimensional thin-film equation.

We are currently investigating the effect of viscoelastic relaxation in strongly slipping films on the morphology of dewetting rims by numerical solution of equations (38) to (40).

M.R. acknowledges support by the German Science Foundation (DFG) under grant RA1061/2-1, and A.M. and B.W. under grant MU1626/5-1, both in the priority programme SPP1164.

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