Shear-induced effects in confined non-Newtonian fluids under tension

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We study the influence of shear effects on the adhesive performance of a non-Newtonian fluid under tension, confined between two parallel flat plates. The upper plate is subjected to a pulling force, which is recorded during the separation process. We approach the problem analytically, and use a modified Darcy's law in the weak shear limit to derive the adhesive force and the separation energy. Our theoretical results demonstrate that, for relatively small separations, the adhesion strength is considerably reduced (enhanced) if the fluid is shear thinning (thickening). For larger plate separations, shear effects become negligible, and usual Newtonian behavior is observed. These findings are confirmed by a numerical solution of a more realistic version of the problem, which considers weak shear effects, plus the intrinsic elasticity of the lifting apparatus.

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An understanding of the mechanical response of confined thin films bounded by solids is a very important issue in several technological areas including adhesion, lubrication, fracture mechanics, wetting dynamics, and colloidal hydrodynamics [1–3]. On the academic side, the study of compliant adhesive layers is highly interdisciplinary, and its basic research involves a great variety of areas ranging from interfacial science and rheology to pattern formation and chemistry [4,5].

A quantitative study of the performance of an adhesive material can be done experimentally by measuring the energy required to separate two adhesively bonded surfaces. One effective way to examine relevant adhesive properties is provided by the so-called probe-tack test [6,7]. This test uses a probe that can be retracted uniaxially from an adhesive layer at a known rate while the applied force is recorded. A typical force-distance curve for a probe-tack test on a good adhesive exhibits a characteristic sharp peak in the measured force, followed by an abrupt drop, that defines a plateau at a lower value. Eventually, the force vanishes when the complete separation is achieved. From these curves two important quantities can be determined: (i) the peak adhesive force, which gives the maximum value of the applied force, and (ii) the separation energy (or, the work done during the entire separation process).

Most adhesives are based on weakly crossed-linked high-molecular weight polymers and present very complex rheological properties, many of them often not known in detail. However, adhesives may behave like fluids, so a fluid mechanics approach to understanding many adhesive problems seems reasonable. Very recently, some research groups tried to get more insight into the complicated rheological properties of conventional adhesives by studying the fundamentals of adhesion in viscous liquids, both Newtonian and non-Newtonian [8–11]. Though these fluids are not rigorously "true" adhesives, some of their behavior show interesting similarities with general properties reminiscent of conventional adhesive materials.

The work by Francis and Horn [8] applies lubrication theory to find an analytical solution of the force-distance curve for a *Newtonian* fluid in a sphere-plate geometry. By taking into account the compliance of the measurement apparatus in a probe-tack test, they have been able to reproduce the initial spike originally measured in adhesives. The same type of analysis has been employed in Refs. [9–11] for a plate-plate geometry, resulting in similar force-distance profiles. In addition to the characteristic force peak, Poivet *et al.* [9] have also detected a cavitation-induced force plateau when the confined fluid is highly viscous, and flows at high velocities. Derks *et al.* [11] used a *non-Newtonian*, yield stress fluid to model the viscoelastic properties of adhesives. They have found a very good agreement between their theoretical expression for the force-distance curves and their experimental results. The authors have also observed that the detailed shape of the contact area has a very modest influence on adhesive properties, both for Newtonian and yield stress fluids.

In contrast to most Newtonian fluids, non-Newtonian fluids differ widely in their hydrodynamic properties, with different fluids exhibiting a range of effects from elasticity and plasticity to shear thinning and shear thickening. Additional complexity comes from the fact that the majority of non-Newtonian fluids manifest several of these different properties simultaneously. For instance, polymer solutions may show both shear-thinning and normal stress effects [13,14], while other liquids have a yield stress, plus shear-thinning as well as elastic effects [15]. So, for force-distance curves obtained from measurements using non-Newtonian fluids, it is not trivial to identify precisely the origin of a given feature in the force-distance profile. An illustrative example of this fact can be found in the seminal work by Francis and Horn [8]. To investigate the deviation from Newtonian adhesion, they have performed experiments using non-Newtonian fluids. Their non-Newtonian force-distance curve is shifted from the Newtonian data, showing a reduced adhesion, notably for smaller probe/thin-film separation values. The authors in Ref. [8] pointed out a number of possible reasons for the observed adhesion reduction, ranging from viscoelasticity to shear-thinning effects, but no specific theoretical justification has been provided.

In this paper we systematically separate the influence that different non-Newtonian properties have on the fluid adhe-

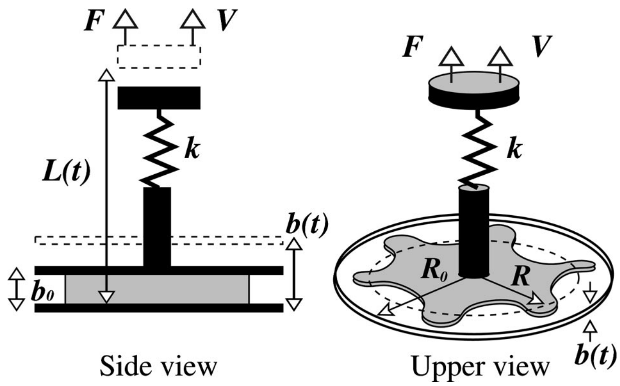

FIG. 1. Schematic representation of the plate-plate geometry.

sive performance. We study confined non-Newtonian fluids under tension, focusing on the isolated influence of shear thinning and shear thickening on the shape of the force-distance curves. Any other non-Newtonian effects are neglected. Within our approach, any deviation from Newtonian behavior may be attributed directly to shear-thinning (-thickening) effects. To avoid complicated rheological properties, which can quickly lead to intractable mathematics, we concentrate on the weak shear limit and study the problem analytically.

Consider a non-Newtonian, incompressible fluid of zero-shear viscosity η_0 located between two narrowly spaced circular, flat plates as depicted in Fig. 1. The outer fluid is Newtonian, and of negligible viscosity. As in Refs. [8–11] we assume that the lifting apparatus has spring constant denoted by k. One end of the apparatus moves at a specified constant velocity V, subjecting the upper plate to a pulling force F. The initial plate-plate distance is denoted by b_0. At a given time t the plate spacing is denoted by $b=b(t)$, while the deformation due to the stretching of the apparatus is $L-b$, where $L=b_0+Vt$. For the completely rigid apparatus case we have $b=L$ and $\dot{b}=V$, where $\dot{b}=db/dt$.

First, we calculate the pulling force F as a function of displacement L, deriving F analytically for the completely rigid case. Subsequently, we address the more realistic situation in which compliance is taken into consideration, by performing a numerical calculation. At the start of the adhesion measurement process, the plates begin to separate and as the outer fluid enters the system the non-Newtonian fluid must move radially inward to conserve volume. We follow Derks *et al.* [11] and derive F considering that the interface separating the fluids remains circular during the lifting process, with initial and final radii defined as R_0 and $R=R(t)$, respectively. Conservation of the non-Newtonian fluid volume leads to the relation $R^2b=R_0^2b_0$.

For the confined plate-plate geometry, a standard lubrication approximation applies [12], so that inertial and surface tension effects can be neglected. Under such circumstances, the relevant hydrodynamic equation for *Newtonian* fluids is Darcy's law

$$\nabla p=-\frac{12\eta}{b^2}\mathbf{u}, \tag{1}$$

where $\mathbf{u}=\mathbf{u}(r,\theta)$ and $p=p(r,\theta)$ are, respectively, the gap-averaged velocity and hydrodynamic pressure in the fluid, and (r,θ) denote polar coordinates. The viscosity of the fluid is represented by η.

To model the confined flow of *non-Newtonian* fluids under tension, we use a suitable form of the Darcy's law in the weak shear-thinning or -thickening limit. We follow Bonn *et al.* [13] and Constantin *et al.* [17], and consider a shear rate dependent viscosity

$$\eta(\omega^2)=\frac{\eta_0(1+\alpha\omega^2\tau^2)}{1+\omega^2\tau^2}, \tag{2}$$

where $\omega\approx u/b$ is the shear rate, $u=|\mathbf{u}|$, and τ denotes a characteristic relaxation time. The parameter α measures the shear dependence: $\alpha=1$ corresponds to the Newtonian fluids, and $\alpha<1$ ($\alpha>1$) gives the shear-thinning (-thickening) case.

By taking the weak non-Newtonian limit $(\omega\tau)^2\ll 1$ of Eq. (2), and substituting the resulting expression into Eq. (1), we obtain an alternative form of Darcy's law ideally suited to describe weak non-Newtonian effects in the plate-plate geometry,

$$\nabla p=-\frac{12\eta_0}{b^2}\left[1-\frac{\delta u^2\tau^2}{b^2}\right]\mathbf{u}. \tag{3}$$

Here $\delta=(1-\alpha)$ is a small parameter that expresses the non-Newtonian nature of the fluid under tension: $\delta=0$ corresponds to the Newtonian case, while $\delta>0$ ($\delta<0$) describes the shear-thinning (-thickening) case. An alternate route to our Eq. (3) is to start with viscosity depending on the square of the pressure gradient, as proposed by Kondic *et al.* [16], then approximate the pressure gradient with the velocity for small δ.

Volume conservation dictates a useful relation between the upper plate and the interface velocities $u(r)=-\dot{b}r/2b$. By substituting the latter into Eq. (3), we integrate our non-Newtonian Darcy's law to obtain the pressure field

$$p=\frac{3\eta_0\dot{b}}{b^3}(r^2-R^2)-\frac{3\eta_0\dot{b}^3\delta\tau^2}{8b^7}(r^4-R^4). \tag{4}$$

To conclude our derivation we integrate the pressure, Eq. (4), over the area occupied by the non-Newtonian fluid, and obtain the *dimensionless* pulling force

$$F=\frac{1}{b^5}-\delta\mathcal{A}\frac{1}{b^{10}}, \tag{5}$$

where $\mathcal{A}=(\tau^2V^2R_0^2b_0)/(6\zeta^5)$. In Eq. (5) lengths are rescaled by $\zeta=(3\pi\eta_0R_0^4b_0^2V/2k)^{1/6}$ and velocities by V. From now on we work with the dimensionless version of the equations. Taking the limit of vanishing δ, Eq. (5) reduces to the expression derived in Ref. [11] for Newtonian viscous fluids.

We analyze Eq. (5) in more detail, and try to explore the important aspects coming from the non-Newtonian contribution. Figure 2 depicts the force F as a function of the distance L. The black solid curve refers to the usual Newtonian case

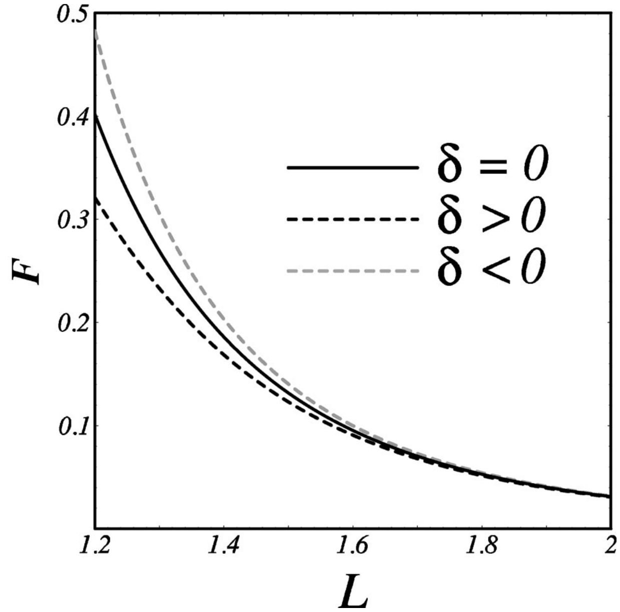

FIG. 2. Pulling force F as a function of L for the purely rigid case described by Eq. (5), for $\delta = 0.0, +0.1$, and -0.1.

$\delta = 0$, the black dashed curve describes the shear-thinning contribution for $\delta = +0.1$, and the gray dashed curve illustrates the shear-thickening behavior for $\delta = -0.1$. The parameter $\mathcal{A} = 5.0$.

By inspecting Fig. 2 we identify deviations from Newtonian adhesion in the force-distance curve profile, caused by weak shear effects. It is clear that the discrepancies between Newtonian and non-Newtonian behaviors are more significant for relatively small values of the displacement L, which coincides with the plate separation b in the completely rigid apparatus case. This fact is expected due to the $1/b^{10}$ dependence of the non-Newtonian term in Eq. (5). A more physical justification can be provided by analyzing how the shear rate ω depends on L. It can be shown that $\omega \sim L^{-3/2}$, so as L is increased, ω decreases, and the force is dominated by Newtonian effects for larger L. As a result, both non-Newtonian curves eventually coincide with the Newtonian one. However, relevant differences are clearly identified for smaller L: the shear-thinning curve lies below the Newtonian curve, while the shear-thickening curve lies above it. These results indicate that the adhesion performance is reduced (enhanced) for shear-thinning (-thickening) fluids. This reinforces, in a more precise and quantitative way, a proposition by Francis and Horn [8] that the reduced adhesion observed in their non-Newtonian experiments could be induced by shear-thinning effects. Our theoretical findings are also in agreement with the measurements performed by Poivet *et al.* [9] who detected a similar deviation in the force curve profiles, a behavior qualitatively attributed by them to shear thinning. As far as we know the experimental study of adhesion probe-tack tests involving *purely* shear-thickening fluids has not yet been investigated in the literature. It would be of interest to perform such experiments and verify the validity of our theoretical predictions (enhanced adhesion) for shear-thickening fluids.

The separation energy (work of separation) can be easily derived by integration of Eq. (5) as a function of displacement L, from initial position b_0 to infinity, yielding

$$W = \frac{1}{4b_0^4} - \frac{\delta \mathcal{A}}{9b_0^9}. \tag{6}$$

From Eq. (6) we clearly see that, for a given value of b_0 and $\mathcal{A}$, W is largest for the shear-thickening case. This fact could be guessed from Fig. 1, where it is evident that the area below the gray dashed curve is larger than the areas delimited by the other two curves and the L axis.

Typical force-distance curves increase sharply at initial stages of the plate separation process. For an ideal viscous Newtonian liquid, there is no apparent physical reason for the actual force to start at zero, increase quickly to a peak, and then decrease abruptly. However, this behavior can be described as a result of the elasticity of the apparatus [8,11]. We address this issue and calculate the complete form of the force-distance curves, considering the combined action of weak shear effects and the intrinsic rigidity of the lifting machine. To do it, we follow Francis and Horn [8] and assume that, during the entire separation process, there is a perfect balance between the hydrodynamic force and the spring restoring (dimensionless) force $L-b$ which results from the deflection of the apparatus. By using Eq. (5) and setting $V = db/dt$ we obtain a nonlinear first-order differential equation for $b = b(t)$,

$$\frac{1}{b^5}\frac{db}{dt} - \delta \mathcal{A} \frac{1}{b^{10}}\left(\frac{db}{dt}\right)^3 = L - b. \tag{7}$$

We solve Eq. (7) numerically for $b(t)$ and use the dimensionless relation $L = b_0 + t$ to write b as a function of L, and subsequently express F in terms of L.

Figure 3 presents the complete force-distance curves at

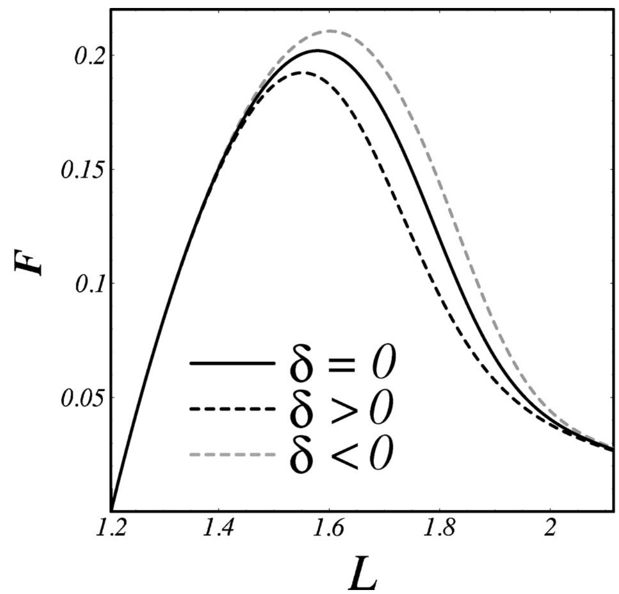

FIG. 3. Force-distance curves for the flexible apparatus case, obtained by numerically solving Eq. (7). The physical parameters are the same as those used in Fig. 2.

initial plate spacing $b_0 = 1.2$ obtained from the numerical solutions of Eq. (7). It contrasts the Newtonian ($\delta = 0$) black solid curve, with those calculated for $\delta = +0.1$ (black dashed curve) and $\delta = -0.1$ (gray dashed curve).

At the very beginning of the separation process, the elasticity of the apparatus dominates, regardless of the Newtonian or non-Newtonian nature of the fluid. At this stage the curves grow from zero when separation starts, and tend to amaximum when the apparatus is at maximum extension. Before a maximum peak is reached, the weak shear effects start to prevail, and the original single curve splits into three different curves. At this point the specific features of each curve start to be regulated by the non-Newtonian parameter δ. The peak adhesive force is the largest (lowest) for the shear-thickening (-thinning) case, and similarly to the purely rigid situation, the shear-thickening (-thinning) curve lies above (below) the Newtonian one. As expected, the area below the gray dashed curve is larger than the area under the black dashed and black solid curves, meaning that the weak shear-thickening effects do enhance the energy of separation. When a sufficiently large L is achieved, shear effects effectively vanish and purely viscous, Newtonian effects take over. Consequently, the three curves tend to meet again and collapse into a single curve for large L. We have also verified that the deviations from Newtonian behavior mentioned above become increasingly important for thinner layers (smaller b_0).

By employing a simple analytical approach we have been able to show that weak shear effects have an important role in determining force-distance profiles of probe-tack tests with non-Newtonian fluids. We have verified quantitatively that the adhesive performance of such fluids is enhanced (reduced) if they are shear thickening (thinning). It is hoped the contribution of this work will further advance the understanding of adhesion mechanisms, and motivate new theoretical and experimental investigations of the problem.

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