Apparatus-specific analysis of fluid adhesion measurements

B. A. Francis and R. G. Horn
Ian Wark Research Institute, University of South Australia, Mawson Lakes, SA 5095, Australia

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A thin film of viscous liquid between two solids acts as an adhesive due to the large force resisting separation of the solids. This effect is exploited in pressure-sensitive adhesive bonds. One method of investigating such bonds is to use a probe tack test in which a rigid probe is indented into a thin adhesive layer coating a rigid flat base. These experiments are characterized by two quantities: the total work of separation, that is, the work done in extracting the rigid punch from the adhesive film, and the peak adhesive force, otherwise known as the adhesive strength. Little effort, however, has been spent on understanding the connection between these quantities and the apparatus used to measure them. In this article we shall study the simplest case of fluid adhesion where a spherical probe and flat are bound by a high viscosity Newtonian polymer melt (polydimethylsiloxane or polybutene) and examine the role of apparatus-specific parameters in determining the measured adhesive strength and work of separation. We shall show how a dimensionless master-curve can be derived to capture the dependence of the adhesive strength on the testing regime. Specific attention is paid to the effect of system compliance on the adhesion, as introduced by the presence of a compliant load cell used to measure the adhesive force. A relationship linking the adhesive strength of a Newtonian film and the work of separation is also presented. © 2001 American Institute of Physics. [DOI: 10.1063/1.1351057]

I. INTRODUCTION

The study of adhesion generated by thin fluid films has often been overlooked in favor of the more surface-oriented adhesion mechanisms as discussed in thermodynamic adhesion studies. This is appropriate when considering simple linear elastic systems where adhesion results principally from surface forces, and where dissipation does not need to be considered directly by the surface energetics approach. Modifications to these simple theories have generally been in the form of semiempirical dissipation factors. In these cases, adhesion would generally be reduced by the introduction of any intermediate layer due to a reduction in the interfacial energies. However, the presence of a fluid does contribute significantly to the adhesion through energy dissipation in very thin, high viscosity films.

Pressure sensitive adhesives (PSA), typically 20–60 μm thick and made predominantly from high molecular weight polymer blends, are an example of a fluid adhesive film, though they often exist in the rubbery mechanical flow state rather than a fluid melt per se. Pressure sensitive adhesives can be studied using several techniques such as: (a) the experimentally simple though qualitatively difficult peel test, or (b) the fundamental, yet more abstracted probe tack test. While the former allows for direct experimental comparison with real world adhesives, and is thus an attractive option for manufacturers of PSA systems, the latter is equally valuable as it reduces the large number of uncontrollable parameters found in peel experiments. The tack test is also closer to a standard mechanical tensile test, where a sample is elongated while simultaneously measuring the resultant tensile force. Standard solid sample tensile tests require only minor corrections to remove the effects of compliance, equivalent to the removal of the Hookean elongation of the load cell. The effective elongation of the load cell may be similarly removed in the fluid adhesion test.

Several studies have attempted to explore the perturbation of the adhesion measurement caused by the compliance inherent in tape adhesion and contact adhesion experiments, however, little attention has been paid to the influence of the compliant load cell in the probe tack measurements. The current study examines the experimental and analytic considerations that apply to a model probe tack test measurement, with broader implications for other compliant tensile tests. To this end, a force balance approach is employed, where it is assumed that the restoring force of the load cell must at all times equal the integrated load supported by the film. This approach is significantly different from typical rheological studies involving squeeze film experiments under either constant load or constant speed conditions. To avoid the complexity of viscoelasticity, which can quickly lead to intractable mathematics, we have chosen to concentrate on Newtonian polymeric melts. Experimentally the adhesion is tested by the introduction of a bulk fluid between two rigid bodies (a sphere and flat). The adhesive force is measured as a function of time and displacement as the spherical body is separated from the flat. Under ideal conditions, this can be analyzed by using the Taylor equation for Newtonian fluids:

\[ F_H = 6\pi R \eta \left( \frac{dD}{dt} \frac{1}{D} \right). \]  

This equation gives the hydrodynamic force \( F_H \) on a sphere
of radius $R$ moving along a line perpendicular to a flat surface, when they are separated by a thin film of thickness $D(D \ll R)$ of liquid having viscosity $\eta$. This contrasts to more common parallel plate squeeze-film experiments found in the field of rheology. Film failure is a separate mechanism operative in real PSA tapes, however we shall leave discussion of cavitation and fibrillation to another paper.\(^{29}\)

The Taylor equation has been used elsewhere to examine hydrodynamic interactions on a molecular scale using the surface force apparatus (SFA) in conjunction with small organic molecules and short chain length polymer melts.\(^{19,21–23}\)

Other researchers have since extended this regime to include much higher molecular weight melts or solutions where limited viscoelastic effects are observed with polymer solutions under oscillatory strains or applied drive step functions.\(^{24,25}\)

The analysis presented here is equivalent to the SFA experiments using a constant speed drive. The Taylor theory has also been expanded to examine the effects of slippage of fluids along the rigid body surfaces\(^ {26}\) and the effects of finite compliance in the bounding solid bodies.\(^ {27}\)

Matthewson has applied a modification to the Taylor equation to examine the adhesion of spheres to surfaces coated with fluid films.\(^ {28}\)

In this article, after describing a generic experimental arrangement in which fluid adhesion is to be tested, a theoretical section outlines a method of reducing the results to simple master curves for the adhesive strength and work of separation. Experimental results are then presented which confirm the validity of the theoretical approach.

II. FORCE BALANCE IN A COMPLIANT MEASUREMENT SYSTEM

The elements of the system under consideration are shown in Fig. 1. The force between a solid sphere and a flat plate is measured by means of a load cell which has a stiffness $k$. One end of the load cell is driven by the testing machine at a constant speed $V$. The sphere is separated by a minimum distance $D$ from a rigid flat plate by a layer of liquid. Provided $D \ll R$ the hydrodynamic force acting between the sphere and the plate is dominated by the thin film of liquid between them, given by Eq. (1), with negligible contribution from the fluid away from the axis of symmetry. This justifies the replacement of the sphere by a more practical spherical cap or lens.

At any instant the force in the load cell, $F_S = k \Delta$, where $\Delta$ is the extension of the load cell, is equal to the hydrodynamic force given by Eq. (1). This quasistatic approximation neglects inertia of the solids, a valid assumption in the experiments considered here. The extension at any instant is given as $\Delta(t) = L(t) - D(t)$, where the zero of the drive function $L(t)$ is chosen so that $L = D$ when the force or the compliance is zero. The force balance is then

$$F_H(D,D) = F_S(D,t),$$

which produces a first order differential equation

$$6 \pi \eta R^2 \frac{D}{D(t)} = k[L(t) - D]$$

for a specified drive function $L(t)$. For simplicity we restrict the analysis to a linear drive:

$$L(t) = D_0 + Vt.$$  \(4\)

Some simple considerations allow an insight into how the force profile $F(t)$ is likely to develop. On commencement of the drive, the upper spherical surface remains almost stationary so $\Delta$, and therefore the restoring force $F_S$, must increase linearly with slope equal to $Vt$ or $Vt/k$, respectively. The increasing force is transmitted by the load cell to the sphere-fluid-flat system, gradually pulling the spherical surface from the plate. At long times, $D$ becomes large, the hydrodynamic force $(F_S \propto 1/D)$ becomes weak, the spring deflection is correspondingly small, and so the force is given by Eq. (1) with $D = L$ and $D \approx V$. Between short and long times there is a peak in the force. This peak is often called the adhesive strength of the adhesive. Some typical data presented in Fig. 2 illustrates the different phases of the force curve development, indicating the two distinct extreme regions, and how they relate to the shape of the force curve.

The force maximum is located by $F_S = kD = k(L - D) = 0$, i.e., $D = L = V$. The peak force must occur when the speed of separation equals the drive speed $V$. However, since $D(t) \neq L(t)$, the separation $D$ at which the peak occurs is not immediately apparent, nor is the magnitude of the peak force. Indeed, both the peak separation and peak force must be solved simultaneously. In the following section we derive the relationship between the peak force (adhesive strength) and the separation. We ignore the possibility that the thin fluid film can fail under tension, or that significant deformation of the bounding (rigid) solids could occur, both of which are possible in real-world adhesion tests.

III. THEORY

Combining Eqs. (3) and (4) gives

$$6 \pi \eta R^2 \frac{D(t)}{D(t)} = k[D_0 + Vt - D(t)].$$  \(5\)
We introduce a characteristic length $\beta$ which combines all of the experimental parameters apart from the initial film thickness:

$$\beta = \left[ \frac{6\pi \eta R^2 V}{k} \right]^{1/2}. \quad (6)$$

Equation (5) can be rewritten as

$$\frac{1}{V} \frac{dD}{dt} = \left\{ \frac{D(t)}{\beta} \right\} \left[ \frac{L(t)}{\beta} - \frac{D(t)}{\beta} \right]. \quad (7)$$

The variables are associated in dimensionless groups, which can then be substituted for the following dimensionless variables, displayed in a script font:

$$\mathcal{D} = \frac{D}{\beta}, \quad \mathcal{L} = \frac{L}{\beta}, \quad \mathcal{T} = \frac{Vt}{\beta}. \quad (8a)$$

with the dimensionless velocity given by

$$\mathcal{D} = \frac{d\mathcal{D}}{d\mathcal{T}} = \left( \frac{1}{V} \right) \frac{dD}{dt}. \quad (8b)$$

Similarly, the dimensionless force would be written as

$$\mathcal{F} = \frac{F}{k\beta}. \quad (8c)$$

Substituting for the above quantities into Eq. (7) then returns the very concise Bernoulli differential equation:

$$\frac{d\mathcal{D}}{d\mathcal{T}} = \mathcal{D}(\mathcal{L} - \mathcal{D}). \quad (9)$$

A further substitution $\gamma = (1/\mathcal{D})$ leads to the solution,

$$\gamma = e^{-(1/2)\mathcal{D}} \left[ \frac{e^{(1/2)\mathcal{D}}}{\mathcal{D}_0} - i \frac{\pi}{2} \left( \text{erf} \left( \frac{i\sqrt{2}}{\sqrt{2}} \right) - \text{erf} \left( \frac{i}{\sqrt{2}} \mathcal{D}_0 \right) \right) \right]. \quad (10)$$

This equation may be used to solve for the entire force-drive profile (examples of which are shown in Fig. 3), where the dimensionless drive position is also equal to $\mathcal{D}_0 + \mathcal{T}$. Since the time dependent force always equals the spring restoring force, the dimensionless force is given by

$$\mathcal{F} = \mathcal{L} - \mathcal{D}. \quad (11a)$$

hence

$$\mathcal{F} = \left( \mathcal{D}_0 + \mathcal{T} - \frac{1}{\beta} \right). \quad (11b)$$

If we now restrict the analysis to the force maxima (where, as noted previously, $D = V$ and so $\mathcal{D} = 1$) we can use Eqs. (10) and (11) to predict a priori the adhesive strength for a given fluid adhesion experiment. Substituting for the dimensionless separation speed back into Eq. (9) gives a very simple quadratic relationship between the drive position and the actual separation at the peak force point:

$$\mathcal{D}_p^2 - \mathcal{L}_p \mathcal{D}_p + 1 = 0, \quad (12)$$

where the subscript $p$ denotes values corresponding to the force peak. Rewriting for the peak drive position we obtain:

$$\mathcal{L}_p = \frac{1}{\mathcal{D}_p} + \mathcal{D}_p, \quad (13)$$

allowing us to determine at what point (and hence what time) the force will reach its zenith. From Eqs. (11a) and (13) it is seen that the peak force is given by

$$\mathcal{F}_p = \frac{1}{\mathcal{D}_p} \mathcal{Y}_p. \quad (14)$$

Substitution of Eq. (13) into Eq. (10) then gives the required relationship between $\mathcal{D}_p$ (and hence peak force) and $\mathcal{D}_0$. 

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**FIG. 2.** Representative experimental data obtained for a 100 Pa s PDMS fluid, 88 mm lens surface, and a drive speed of 10 $\mu$m/s. (a) Showing how the separation of the two surfaces develops with time. The dashed line represents the constant-speed drive function $L(t)$. (b) The corresponding force–drive curve. In this case the force (solid line) is shown together with the two extreme cases described in the text, the constant velocity Taylor force (dashed line) and the linear Hookes law force (solid line) given by the conditions $dD/dt = V$ and $D(t) = D_0$, respectively.

**FIG. 3.** Five dimensionless force curves, shown as a function of the dimensionless drive function, with different initial separations.
We have thus obtained an analytic solution for the dimensionless adhesive strength (peak force) for a generalized sphere-viscous fluid-flat adhesion experiment. We can use this result to create a master curve allowing the prediction of the adhesive strength from the initial conditions. While the resulting equation has a purely analytic basis, the solution is not in a convenient form. After presenting experimental data to test this result using a range of different experimental conditions, a more practical empirical form of this master-curve relationship will be presented.

IV. EXPERIMENTAL CONFIRMATION

A. Experimental details

The experimental apparatus is described elsewhere, and will therefore be discussed only briefly here. The apparatus consists of an optical quality convex glass lens as the upper surface, having a radius of curvature chosen between 20 and 200 mm, and a lower flat glass surface. The surfaces can be separated with speeds between 0.1 and 1000 μm/s and the drive position measured with micron resolution, with the concomitant force measured to ±0.3 mN resolution. The load cell used for the experiments reported here had a measured spring constant of 242 kN/m. All separations are measured relative to a solid–solid contact position established in the absence of fluid.

The fluids tested included a range of polydimethylsiloxane (PDMS) melts (denoted PDMS-I, II, and III) supplied by Dow Corning and United Chemicals (with viscosity measured as 60, 100, and 820 Pa s, respectively) and a polybutene melt (PB-I) supplied by Sigma-Aldrich (700 Pa s). All fluids were used without further preparation, and were rheologically characterized using a Rheometrics RS-5000 stress controlled rheometer. PDMS-I and II were found to be Newtonian over the entire accessible range of shear rates (up to a maximum of 10 s⁻¹) under steady shear conditions. PDMS-III did show slight viscoelastic behavior at higher shear rates, with a moderate Maxwellian time constant equal to 6 s.

B. Force curve analysis

Several sets of experimental data showing the development of force as a function of drive position (or equivalently, of time) are presented in Fig. 3. The corresponding theoretical fits are also shown (solid lines) which can be generated using either the solution presented above or a Runge–Kutta numerical method. The data, presented on dimensionless axes, are fitted with extremely good fidelity with no adjustable parameters. The analytic expression, Eq. (10), allows the force at any given time to be calculated without knowledge of the force history. In contrast, the Runge–Kutta routine requires a sequential development of the force curve.

In all of the experiments described, the initial separation is much larger than the lower resolution bound of ±0.5 μm. The only limitation to this theory is the continued requirement that the curvature of the gap between sphere and flat must be small compared to the separation. A ratio of $R/D \approx 200$ was found to give a reasonable limit to the applicability of the Taylor equation, that is, $D \leq 450 \mu m$ in the experiments presented. In all cases studied, the peak forces occurred well within this range.

C. Peak force analysis

We will now examine the magnitude of the peak force in more detail, and in particular the relationship between the underlying physical quantities and the magnitude of this measurement of adhesive strength. In order to do this we shall consider results for a range of fluids, each studied using a number of drive speeds, sphere radii, and importantly the initial separation. As described previously, we use the peak force as an easily quantifiable measure of the adhesive strength of the fluid film, with due appreciation that the concept of adhesive strength may not strictly hold for Newtonian fluids as compared to viscoelastic adhesive films. Presently, the concept of adhesive strength is as a relative term used to compare the adhesive ability as a function of film thickness.

Figure 4 shows the peak force as a function of the initial separation in microns. Decreasing the film thickness increases the effective film strength, as does increasing the drive speed, as expected from cursory inspection of the Taylor equation. The peak force–peak separation relationship may be linearized by plotting either the inverse of the force against separation, the force against the inverse of separation, or plotting log force against log separation. Dividing by a suitable constant (from the Taylor equation) the function can be used to measure the effective bulk Newtonian viscosity, as illustrated in Fig. 4(b). This cannot be used to characterize...
non-Newtonian fluids due to the nonuniform shear rates generated in the sphere-flat geometry.\textsuperscript{19,29,30}

D. System master-curve analysis

The direct application of the previous theoretical scaling is shown in Figs. 5(a) and 5(b). The peak data that is used to construct these figures is taken from systems having different fluid viscosity, sphere radii, and drive speeds. Under this dimensionless scaling the data collapses onto a single master-curve given exactly by the theoretical analysis. The scaling regime does not necessarily allow the effect of compliance to be singled out, since the physical parameters are intimately bound in the dimensionless analysis. It is important to note that in this respect the load cell compliance is as much a part of the measured quantity as the fluid viscosity.

Assuming all other parameters remain fixed, then decreasing the value of $k$ would result in larger values of $D_p$ and $F_p$. However, the actual peak force $F_p$ (scaled by $\sqrt{k}$) would be reduced, as expected for a more compliant system.

From Fig. 5 the dimensionless force can be well-approximated by the following empirical expression:

$$F_p = 1.2(0.27 + 0.9 D_0 + D_0^{1/3})^{-1}$$

or in dimensional variables,

$$F_p = 1.2k\left[0.27 + 0.9 \frac{D_0}{\beta} + \left(\frac{D_0}{\beta}\right)^{1/3}\right]^{-1}.$$  

We do not suggest that this empirical form contains any more insightful information other than being a convenient approximation, avoiding a numerical solution. This allows us to quickly predict the adhesive strength over a range of at least three orders of magnitude in the initial separation.

Equation (15a) can also be rewritten in a form that allows the initial separation to be calculated from a measured value of peak force. A reasonable empirical solution to this is

$$D_0 = \frac{1}{0.74 F_p} e^{-F_p/0.65}.$$  

The convenience derived from this form will become apparent in the next section.

E. Work of separation

In comparison to the PSA probe tack test, the total work of separation of these fluid films is defined as the integral of the force with respect to drive position or separation, where we denote this term $W_{\lambda}$. In practice there must be some limits to this integration, which we discuss below. This quantity must be distinguished from the more usual work of adhesion, which is strictly a reversible thermodynamic quantity associated with interfacial energies between materials. With the fluid film, the work of separation results from dissipation.

The difficulty in calculating this work of separation is that there is no obvious termination point for the integral because the force does not decay sufficiently rapidly. This is apparent in the work of separation as defined for the bulk fluid noncompliant experiment, obtained by integration of the Taylor equation:

$$W = 6 \pi \eta R^2 V \ln \left(\frac{D(t)}{D_0}\right)$$

which diverges as $D(t) \to \infty$. For the compliant experiment, integrating the force with respect to the drive function gives us the following relationship for adhesion:

$$\int_{L_0}^{L} F dL = \int_{D_0}^{D(t)} \mathcal{F}(T) dD + \frac{1}{2} \mathcal{F}^2(T),$$

where the right-hand side represents the partitioning between energy lost through viscous dissipation, $\mathcal{W}_d$, and that which is stored in the load cell, respectively. The magnitude of the integral increases unbounded with separation, and therefore to determine the adhesion, this integration must be taken either to some fixed end point, $D_f$ (taking it to infinity is not consistent with the limitations of the underlying Taylor equation), or to a film rupture point. Making the substitution for $\mathcal{F} = (1/D) dD/dT$ in the left-hand side of Eq. (18), we may rewrite the completed integration in terms of the viscous dissipation:

$$\mathcal{W}_d(D_f) = \ln \left(\frac{L_f}{L_0}\right) - \frac{1}{2} \mathcal{F}_f^2 \ln \left(\frac{D_f}{D_0}\right).$$

The last term may be neglected if the residual force is small at the end point, i.e., $\mathcal{F}_f \approx 0$, in which case $L_f \approx D_f$. Under these conditions, the work of separation is the same as in the noncompliant case, and is equivalent to the dimensional Eq. (17).

\begin{figure}[h]
\centering
\includegraphics[width=\textwidth]{figure5.png}
\caption{(a) dimensionless peak separation as a function of the dimensionless initial separation. (b) dimensionless force for the same range of fluids (105 and 700 Pa s PDMS-II and PB-I, respectively, at 1, 10, 50, and 100 \textmu m/s drive speed). The graph shows both the analytic solution to Eq. (10) (- - -) and an empirically fitted approximation (---) described by Eq. (15a). The two lines are indistinguishable on this scale. In (b) the force corresponding to the noncompliant case ($k \to \infty$) is also shown (solid line).}
\end{figure}
Making use of Eq. (16), the work of separation may be written as

\[ W_A = \frac{F_p}{0.65} + \ln(0.74F_pD_f). \]  

(20)

In this way it is possible to relate the total work of separation, \( W_A \), directly to the dimensionless adhesive strength \( F_p \). In Fig. 6 we see that the approximate solution from Eq. (20) gives a very close fit to experimental results, and also to numerical results generated directly from the Taylor equation in a compliant drive system using experimental values for \( D_0 \) and \( D_f \). To obtain the real work from \( W_A \), we simply use the scaling relationship:

\[ W_A \approx W_A k \beta^2. \]

(21)

We note (since \( \beta \approx k^{-1/2} \)) that \( W_A \) is independent of \( k \).

F. Non-Newtonian fluids

Although the behavior of Newtonian fluids remains the central question examined in this article, it is possible to investigate the deviation from this Newtonian adhesion. In the context of polymeric fluids, this may be achieved by either increasing the molecular weight of the polymers, or altering the structure to a more branched form, thus introducing viscoelasticity. PDMS is often used as a constant viscosity standard; however, it may also show considerable elasticity at sufficient molecular weights. As discussed previously, the PDMS-III sample showed moderate elasticity in oscillatory rheometry measurements, though essentially a constant viscosity in the steady shear mode. It could be suggested that any deviation from Newtonian behavior may be attributed directly to the presence of elasticity in the fluid flow response, in an otherwise Newtonian system. The following results replicate the previous experiments and are presented using the same scaled axes.

Figure 7 shows that the peak force data can once again be collapsed onto a single dimensionless master-curve, though one translated away from that corresponding to the Newtonian fluid. Unfortunately, the specific cause of this deviation cannot be distinguished from this result alone. As implied earlier, this reduced adhesion may derive entirely from the loading effects due to viscoelasticity, which are well-known.\(^{16,31,32} \) Alternatively, it is also known that high molecular weight polymers may exhibit slip over surfaces given sufficient shear stress at the surface.\(^{33,34} \) This has the effect of increasing the flow of fluid from between confining bodies, thus decreasing the hydrodynamic adhesion.\(^{26,30} \) It is also possible that this reduction could result from shear-thinning at the surfaces due to high shear rates localized along the surface,\(^{29} \) or due to elastohydrodynamic deformation.\(^{27} \) The latter can probably be discounted here, since no effect was observed in the polybutene fluid, PB-I, which has a similarly high viscosity.

We currently lack a suitable viscoelastic model for the hydrodynamics, and in particular one which allows multimodal analysis as would be required. Slippage is difficult to distinguish from the effects of bulk elasticity. The slippage of polymers has, however, been well-studied, for example, Migler et al.\(^{35,36} \) and by Inn and Wang\(^ {37} \) using similar PDMS systems. Migler et al. observed that slippage does occur under conditions similar to those found in this experiment, though for the particular case of pure and uniform shear flow. Analysis of the slippage of fluids in the sphere-flat geometry\(^ {26} \) has usually examined hydrophobic slip where shear stress dependence is ignored, though there have been exceptions.\(^ {30} \) Certainly it is unlikely that the constant slip
length models available would sufficiently model the transitions that occur between no-slip and slip boundary conditions. Further work will be required to address this question, which would require the ability to use different types of surfaces, and thus identify if the adhesion reduction is surface-dependent.

V. CONCLUSIONS

This article has developed a robust analysis of a simple fluid adhesion probe experiment, specifically using the sphere-flat geometry which has become popular in PSA characterization experiments. Our work clarifies the way in which adhesive strength in dissipative systems depends on the compliance of the measurement system. Within the limits of the lubrication approximation on which the Taylor equation (1) is based,19 we have found a useful correlation between work of separation and dimensionless adhesive strength. Other limits to our work would occur at very small values of the initial separation where the pressures become very high, resulting in possible cavitation and elastohydrodynamic deformation of the solids.

The present analysis is confined to Newtonian fluids and is not directly applicable to viscoelastic fluids such as those used for pressure-sensitive adhesives. Having said that, however, the important message remains that measurement of adhesive strength from a measurement of peak force would in general be dependent on system compliance and on the speed at which surfaces are separated. Comparisons of adhesive strength between different PSAs would only be appropriate if measurements were made in systems having equivalent compliance. Furthermore, we have found29 that compliance is an important contributing factor in determining the specific deadhesion mechanism in model PSA fluids, such as cavitation, meniscus instability, and fibrillation.

Our experiments on fluids having some viscoelasticity show that the peak force is shifted from the Newtonian value, but interestingly, though shifted, the data still collapses onto a master curve if the same scaling is used. Further work needs to be undertaken to isolate the cause of this translation, specifically, whether it occurs at the bulk fluid level, or is isolated at the solid surfaces as would be expected from a slip mechanism.

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APPENDIX

The preceding analysis has concentrated on the effect of compliance in the measurement of force between two rigid bodies, in a sphere-flat geometry. A different, though related question concerns the effect of compliance on the maximum negative pressure that is generated in any given separation process. This is particularly important when the fluid has a finite cohesive strength, and thus allows the possibility of fluid rupture. The specific examination of cavitation is left for a future paper,20 however, the following section will derive the relationship that allows the maximum pressure to be determined for any given experiment. In a similar fashion to the previous analysis, let us define a dimensionless maximum pressure as

$$P = \frac{2\pi R}{k} P_r(0),$$

where the maximum pressure $P_r(0)$ occurs on the axis of symmetry ($r=0$) and may be determined as a standard result of lubrication theory:

$$P_r(0) = \frac{3\eta R \dot{D}}{D^2}.$$  

A very simple relationship is obtained involving the instantaneous force:

$$P = \frac{1}{D} \mathcal{F}.$$  

Similar to the evolution of the force (Fig. 3), the pressure initially increases, reaches a maximum, and then decreases. The maximum depends on $D_0$ as shown in Fig. 8. We are presently unable to measure the pressure directly.

For the experiments presented in this article, the relationship may be adequately approximated by

$$P_{\text{max}} \approx \frac{0.6}{D_0}$$ for $D_0 < 0.1$.  

These solutions are derived with the explicit assumption that neither elastohydrodynamic deformation nor slippage occurs, as these would significantly alter the film pressure profiles.27 The analysis is limited to Newtonian fluids; fluid elasticity could alter the picture significantly, as recently discussed by Creton and Lakrout.38

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FIG. 8. Predicted maximum pressure as a function of initial separation (solid line) and a simple inverse approximation given by Eq. (A4) (dashed line).