Int. J. Multiphase Flow Vol. 15, No. 4, pp. 627–638, 1989
Printed in Great Britain.
0301-9322/89 \$3.00 + 0.00

THE LUBRICATION FORCE BETWEEN SPHERICAL DROPS, BUBBLES AND RIGID PARTICLES IN A VISCOUS FLUID

G. Barnocky and R. H. Davis†

Department of Chemical Engineering and Center for Low-gravity Fluid Mechanics and Transport Phenomena, University of Colorado, Boulder, CO 80309-0424, U.S.A.

(*Received* 7 *October* 1988; *in revised form* 12 *December* 1988)

Abstract—A theoretical prediction of the lubrication force resisting the close approach of two spherical drops of arbitrary radii and viscosities separated by a thin viscous fluid layer is presented. The hydrodynamic resistance is predicted to be weaker than that for two colliding rigid spheres in near contact due to the mobility of the drop interfaces. Solutions are also presented for the limiting cases of (a) a drop or bubble approaching a rigid sphere or flat plate, (b) a drop approaching a bubble or a flat free surface and (c) a rigid sphere approaching a flat free surface. When one of the interfaces is completely mobile, as for a bubble or a free surface, the ratio of the lubrication force to that for two rigid spheres with the same relative velocity and reduced radius is predicted to be less than one-fourth; when one interface is completely rigid or immobile, this ratio is predicted to be greater than one-fourth. The results are shown to be in good agreement with previous solutions based on the method of bispherical coordinates when the gap is much smaller than the radius of the smallest drop.

Key Words: drops, bubbles, lubrication, near-contact, hydrodynamic resistance, boundary integral techniques

1. INTRODUCTION

The dynamic interaction of viscous drops with other drops, bubbles or rigid spheres dispersed in an immiscible fluid is of fundamental importance to a variety of multiphase processes such as flotation, extraction, raindrop formation and enhanced oil recovery. In addition, the motion of a viscous drop toward or away from a solid boundary is important in the processing of immiscible materials involving a moving solidification front. Whether or not the drops or bubbles make contact and/or coalesce is substantially determined by the hydrodynamic resistance to near-contact, relative motion and by any attractive and repulsive forces which may be present. Several theoretical approaches have been taken previously to predict the hydrodynamic force on two spherical drops in relative motion, including bispherical coordinates (Haber *et al.* 1973; Wacholder & Weihs 1972; Rushton & Davies 1973, 1978), the method of reflections (Hetsroni & Haber 1978) and imaging techniques (Fuentes *et al.* 1988). Each of these solution methods yields an infinite series for the force between the drops that diverges when the distance between the drops becomes much smaller than their size.

In order to elucidate the nature of the hydrodynamic forces resisting the relative motion of dispersed drops close to one another, Davis *et al.* (1989) used lubrication theory to describe the flow in the narrow gap between two approaching drops and boundary integral theory to describe the tangential motion of the drop interfaces that arises due to the shear stress exerted by the fluid being squeezed out from between the drops. In their development, it is assumed that the two drops are of the same viscosity and that they have sufficiently high surface tension to remain spherical. The dimensionless lubrication force was shown to depend on a single parameter, $m \equiv \lambda^{-1}\sqrt{a/h_0}$, where λ is the ratio of the drop viscosity to that of the continuous phase, a is the reduced radius of the two drops and h_0 is the distance between the surfaces of the two drops at the axis of symmetry. This parameter describes the mobility of the interfaces; when $m \ll 1$, the drops behave as rigid spheres, whereas when $m \gg 1$, the drop interfaces are fully mobile.

†To whom all correspondence should be addressed.

The present paper directly builds on that of Davis *et al.* (1989) by considering spherical drops that, in general, have different viscosities; the extension to deformable drops will be examined later. Of special interest are the important limiting cases of a drop moving through a viscous fluid toward a planar rigid or free surface.

2. THEORETICAL DEVELOPMENT

Figure 1(a) shows two spherical drops of radii a_1 and a_2 and viscosities $\lambda_1\mu$ and $\lambda_2\mu$, respectively, approaching each other along their line-of-centers with relative velocity $W = V_1 - V_2$ (negative values of W describe drops that are receding from one another). In the limit as the radius of one drop goes to infinity, the problem becomes that of a drop approaching a planar interface. The drops are immersed in an immiscible fluid of viscosity μ, and the distance between the drops is assumed to be small relative to either drop radius. Our objective is to predict the lubrication force which resists the motion of the approaching drops as a function of their radii, separation, relative velocity and relative viscosities. In order to accomplish this goal, the pressure profile in the gap between the drops must be found, and this requires that the tangential velocity and stress profiles at each interface be determined. The flow inside the drops is governed by the boundary integral form of the Stokes equations, while the motion of the fluid in the gap is described by lubrication theory.

Lubrication flow in the gap

As in the case of two rigid spheres in near-contact motion, the dynamic pressure gradient, $\partial p/\partial r$, and radial velocity profile, $u(r, z)$, for the fluid in the gap are related by the fully-developed form of the radial momentum equation:

$$\frac{\partial p}{\partial r} = \mu \frac{\partial^2 u}{\partial z^2}, \qquad [1]$$

where r and z represent the radial and axial coordinates, respectively. Equation [1] is valid in the gap region provided that $h_0/a \ll 1$ and Re $h_0/a \ll 1$, where the Reynolds number is defined as $\mathrm{Re} \equiv \rho W a/\mu$, and ρ is the gap fluid density. Since the fluid pressure is constant across the gap, [1] may be integrated directly, subject to the interfacial boundary conditions on the surface of drop 1, $u(r, z_1) = u_1(r)$, and on the surface of drop 2, $u(r, r_2) = u_2(r)$, where $u_1(r)$ and $u_2(r)$ are the unknown velocities of the drop interfaces. This yields

$$u(r, z) = \frac{1}{2\mu}\frac{\partial p}{\partial r}(z - z_2)(z - z_1) - \frac{u_1}{h}(z - z_2) + \frac{u_2}{h}(z - z_1). \qquad [2]$$

The spherical drop surfaces may be approximated by paraboloids in the region of near-contact, so that the gap-thickness profile is given by

$$h(r) = z_2 - z_1 = h_0 + \frac{r^2}{2a}. \qquad [3]$$

Figure 1(b) illustrates that the velocity profile is, in general, asymmetric across the gap;

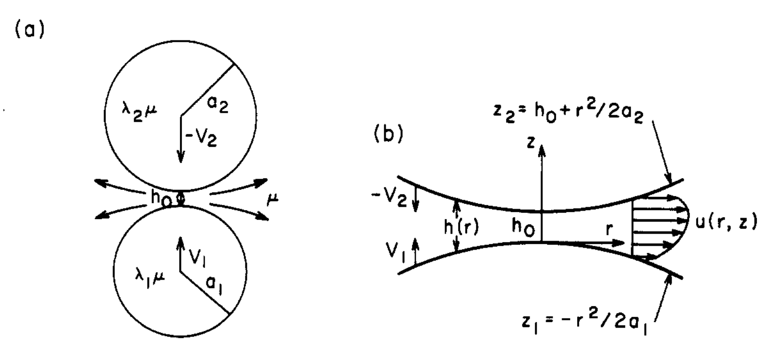

Figure 1. Schematic of two spherical drops moving toward one another along their line-of-centers: (a) side view; (b) close-up of near-contact region.

consequently, the tangential stresses on the two drop interfaces are predicted to differ when the drops have different viscosities.

By differentiating [2], the corresponding tangential stress on each drop interface in the near-contact region, $f_1(r)$ and $f_2(r)$, can be determined as a function of the pressure gradient and the two interfacial velocities:

$$f_1(r) = \mu \left.\frac{\partial u}{\partial z}\right|_{z=z_1} = -\frac{h}{2}\frac{\partial p}{\partial r} + \frac{\mu}{h}(u_2 - u_1) \qquad [4]$$

and

$$f_2(r) = -\mu \left.\frac{\partial u}{\partial z}\right|_{z=z_2} = -\frac{h}{2}\frac{\partial p}{\partial r} - \frac{\mu}{h}(u_2 - u_1). \qquad [5]$$

A macroscopic mass balance performed on the fluid being squeezed out of the gap relates the pressure gradient in [4] and [5] to the unknown interfacial velocities, $u_1(r)$ and $u_2(r)$:

$$\pi r^2 W = 2\pi r \int_{z_1}^{z_2} u(r,z)\,\mathrm{d}z = \pi r h\left(u_2 + u_1 - \frac{h^2 \partial p/\partial r}{6\mu}\right). \qquad [6]$$

Substituting [6] into [4] and [5] gives the relationship for the tangential stress on each drop interface as a function of the two unknown interfacial velocities:

$$f_1(r) = \frac{\mu}{h}\left(\frac{3rW}{h} - 2u_2 - 4u_1\right) \qquad [7]$$

and

$$f_2(r) = \frac{\mu}{h}\left(\frac{3rW}{h} - 2u_1 - 4u_2\right). \qquad [8]$$

Note that in the limit that the drop interfaces become immobile, $u_1 = u_2 = 0$, and so $f_1 = f_2 = 3\mu r W/h^2$, which is the well-known result from lubrication theory for the stress on two colliding solid spheres, first described by Reynolds (1886). In general, however, [7] and [8] contain four unknowns: f_1, f_2, u_1 and u_2 which must be determined in order to compute the hydrodynamic lubrication force on each drop. Consequently, in order to solve for these interfacial velocities and stresses, two more equations are necessary. These may be obtained by considering the flow inside each drop, as described in the next subsection.

The flow inside the drops

Since we restrict our attention to low Reynolds number flow inside each drop, we may make use of the boundary integral form of the Stokes equations (see Rallison & Acrivos 1978). This approach is especially convenient for the current application because it provides a direct relationship between the interfacial velocity and tangential stress for each drop interface. Following the development of Davis *et al.* (1989), the drop interfaces are treated as nearly flat in the vicinity of near-contact, and we restrict our attention to point stresses or Stokeslets acting on the interfaces. The proper forms of the boundary integral equations, which under these conditions relate the interfacial velocity for axisymmetric flow in a half-space to the interfacial stress distribution, become

$$u_1(r) = \frac{1}{\lambda_1 \mu}\int_0^\infty \phi(r', r) f_1(r')\,\mathrm{d}r' \qquad [9]$$

and

$$u_2(r) = \frac{1}{\lambda_2 \mu}\int_0^\infty \phi(r', r) f_2(r')\,\mathrm{d}r', \qquad [10]$$

where

$$\phi(r', r) = \frac{1}{2\pi}\frac{r'}{(r^2 + r'^2)^{1/2}}\int_0^\pi \frac{\cos\theta\,\mathrm{d}\theta}{(1 - k^2\cos\theta)^{1/2}} \qquad [11]$$

is an elliptic-type Green's function kernel and $k^2 = 2rr'/(r^2 + r'^2)$. Note that $\phi(r', r)$ has a logarithmic singularity at $r' = r$ of the form

$$\phi(r', r) \sim -\frac{1}{2\pi} \ln\left(\frac{|r' - r|}{r}\right) \quad \text{as } r' \to r. \qquad [12]$$

Equations [9] and [10] may be substituted into [7] and [8] to eliminate the interfacial velocities, with the result being two coupled Fredholm integral equations of the second kind which must be inverted for the two unknown interfacial stresses:

$$f_1(r) = \frac{\mu}{h}\left[\frac{3rW}{h} - \frac{2}{\lambda_2 \mu}\int_0^\infty \phi(r', r) f_2(r')\,\mathrm{d}r' - \frac{4}{\lambda_1 \mu}\int_0^\infty \phi(r', r) f_1(r')\,\mathrm{d}r'\right] \qquad [13]$$

and

$$f_2(r) = \frac{\mu}{h}\left[\frac{3rW}{h} - \frac{4}{\lambda_2 \mu}\int_0^\infty \phi(r', r) f_2(r')\,\mathrm{d}r' - \frac{2}{\lambda_1 \mu}\int_0^\infty \phi(r', r) f_1(r')\,\mathrm{d}r'\right]. \qquad [14]$$

Once [13] and [14] are solved for f_1 and f_2, the pressure gradient may be determined from

$$\frac{\partial p}{\partial r} = -\frac{f_1 + f_2}{h}, \qquad [15]$$

which was obtained simply by adding [4] and [5], and which satisfies a force balance on a differential element of the fluid in the gap. Finally, the total lubrication force, which is equal in magnitude but opposite in sign on each drop, is the well-known integral of the pressure over the surface area of the near-contact region of each drop. The force may be expressed explicitly in terms of the tangential interfacial stresses using integration by parts:

$$F = 2\pi \int_0^\infty p(r) r\,\mathrm{d}r = \pi \int_0^\infty \frac{(f_1 + f_2) r^2}{h}\,\mathrm{d}r. \qquad [16]$$

Strictly speaking, the upper limit of the integral in [16] should be in the range $\sqrt{ah_0} \ll r \ll a$, based on our previous assumptions of the narrowness of the lubrication region, but since $p \to 0$ for $r \gg \sqrt{ah_0}$, it is valid to replace this limit by $r = \infty$.

Scaling

For the flow inside the narrow gap between the drops, the axial length scale is h_0, the radial length scale is $\sqrt{ah_0}$, the axial velocity scale is W and the radial velocity scale is $W\sqrt{a/h_0}$. Using these scales to nondimensionalize the governing equations, the lubrication force on the drops is found to depend on two dimensionless parameters:

$$m_1 = \lambda_1^{-1}\sqrt{\frac{a}{h_0}} \qquad [17]$$

and

$$m_2 = \lambda_2^{-1}\sqrt{\frac{a}{h_0}}. \qquad [18]$$

The ratio $m_1/m_2 = \lambda_2/\lambda_1$ represents the relative viscosity between the two drops and may be treated as the second parameter in place of [18]. From the condition of continuity of tangential stress across each drop interface,

$$\mu \left.\frac{\partial u}{\partial z}\right|_{z_1^+} = \lambda_1 \mu \left.\frac{\partial u}{\partial z}\right|_{z_1^-} \qquad [19]$$

and

$$\mu \left.\frac{\partial u}{\partial z}\right|_{z_2^-} = \lambda_2 \mu \left.\frac{\partial u}{\partial z}\right|_{z_2^+}, \qquad [20]$$

and from the fact that the appropriate length scale in both the radial and axial directions for flow

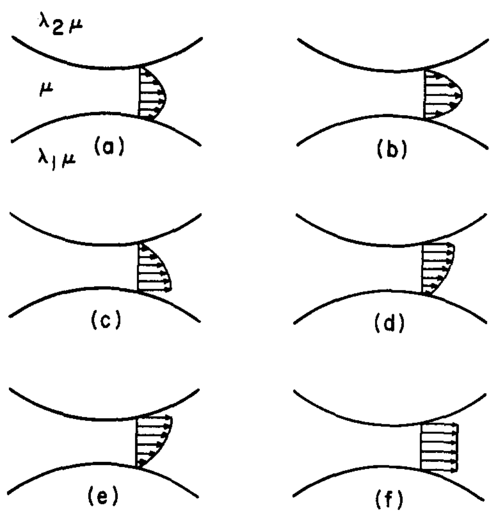

Figure 2. Sketches of the parabolic velocity profile in the gap between two spherical fluid drops moving toward one another with: (a) $m_2 \ll 1$, $m_1 = O(1)$; (b) $m_2 \ll 1$, $m_1 \ll 1$; (c) $m_2 \ll 1$, $m_1 \gg 1$; (d) $m_2 \gg 1$, $m_1 = O(1)$; (e) $m_2 \gg 1$, $m_1 \ll 1$; (f) $m_2 \gg 1$, $m_1 \gg 1$. Figure 1(b) shows the general case of $m_1 = O(1)$, $m_2 = O(1)$.

inside the drops is $\sqrt{ah_0}$, the parameters m_1 and m_2 are found to be measures of the mobility of the interfaces of drops 1 and 2, respectively. In particular, m_1 and m_2 give approximate measures of the ratios of the interfacial velocities for drops 1 and 2, respectively, to the average radial velocity in the gap between them. Depending on the values of m_1 and m_2, the parabolic flow profile in the gap between the drops may have several different shapes, as sketched in figures 2(a–f). Note that in the limit for which the interface of drop 2 becomes immobile ($m_2 \to 0$ and $\lambda_2/\lambda_1 \to \infty$), the problem becomes that of drop 1 approaching a rigid sphere or plate [figures 2(a–c)]. Likewise, in the opposite limit for which the interface of drop 2 becomes fully mobile ($m_2 \to \infty$ and $\lambda_2/\lambda_1 \to 0$), the problem becomes that of drop 1 approaching a bubble or a free surface [figures 2(d–f)]. Each of these limiting cases gives important results which are described in the next section.

3. RESULTS AND DISCUSSION

Drops of the same viscosity ($\lambda_1 = \lambda_2 = \lambda$, $m_1 = m_2 = m$)

For arbitrary values of the relative viscosities of the two drops, [13] and [14] are coupled and must be inverted numerically. However, Davis *et al.* (1989) developed both numerical and approximate solutions of the lubrication force on approaching drops for the important case when the drop viscosities are equal. Substituting $\lambda_1 = \lambda_2 = \lambda$ into [13] and [14] gives two identical Fredholm integral equations of the second kind of the form

$$f(r) = \frac{\mu}{h}\left[\frac{3rW}{h} - \frac{6}{\lambda\mu}\int_0^\infty \phi(r', r) f(r')\,\mathrm{d}r'\right], \qquad [21]$$

where $f(r) = f_1(r) = f_2(r)$. Davis *et al.* numerically inverted [21] for the interfacial stress and computed the lubrication force from [16] where $f_1(r) + f_2(r)$ was replaced by $2f(r)$. In addition, they showed that the numerical solution for the hydrodynamic force can be conveniently expressed as a Padé approximant of the form

$$\hat{F}(m) \equiv \frac{F}{6\pi\mu a^2 \dfrac{W}{h_0}} \approx \frac{1 + 0.38\,m}{1 + 1.69\,m + 0.43\,m^2}, \qquad [22]$$

where $\hat{F}(m)$ is the force acting on the drops made dimensionless with the lubrication force on two rigid spheres and $m = \lambda^{-1}\sqrt{a/h_0}$. They also stated that [22] is an accurate approximation to the numerical result to within 1–2% for all values of m. These results are used in the following subsections which describe the limiting cases of drops approaching rigid and free interfaces.

A drop or bubble near an immobile interface ($\lambda_2 \to \infty, m_2 \to 0$)

In the limit that drop 2 behaves like a rigid sphere or plate ($\lambda_2 = \infty$), [13] and [14] may be simplified to

$$f_1(r) = \frac{\mu}{h}\left[\frac{3rW}{h} - \frac{4}{\lambda_1\mu}\int_0^\infty \phi(r', r) f_1(r')\,dr'\right] \quad [23]$$

and

$$f_2(r) = \frac{\mu}{h}\left[\frac{3rW}{h} - \frac{2}{\lambda_1\mu}\int_0^\infty \phi(r', r) f_1(r')\,dr'\right]. \quad [24]$$

Equations [23] and [24] show that f_1 and f_2 are no longer coupled, and [23] is a Fredholm integral equation of the second kind which may be inverted numerically to obtain a solution for the interfacial stress on drop 1. Subsequently, once $f_1(r)$ is known, [24] may be integrated to obtain the stress on the rigid sphere or plate, $f_2(r)$, and the hydrodynamic force acting on the drop may then be computed from [16].

Since [23] differs from [21] only by a constant factor of 3/2 multiplying the integral term, the lubrication force, $F(m_1)$, on a drop of viscosity λ_1 approaching a rigid body, can be related to $\hat{F}$, the hydrodynamic force on two drops of the same viscosity, by simple algebraic manipulations of [23] and [24], with the result being

$$\frac{F}{6\pi\mu a^2 \dfrac{W}{h_0}} = \frac{3}{4}\hat{F}\left(\frac{2m_1}{3}\right) + \frac{1}{4}. \quad [25]$$

A key feature to note is that, when the drop is sufficiently close to the solid so that $\sqrt{a/h_0} \gg \lambda_1$, the drop interface is fully-mobile ($m_1 \ll 1$) and the force on the drop to leading-order is one-fourth of that on a rigid sphere approaching a solid body—independent of the viscosity of the drop. This is shown figure 3 where the lubrication force given by [25] is plotted vs the dimensionless gap between a spherical drop of radius a and a solid plane boundary for various values of the ratio of drop viscosity to continuous-phase viscosity. For comparison, the exact results using bispherical coordinates are also shown; these were obtained by using the analysis of Haber *et al.* (1973) and by keeping a large number of terms when the gap size was made small. In the limit as $h_0/a \to 0$, the force asymptotes to one-fourth of that for a solid sphere ($\lambda_1 = \infty$), regardless of whether the drop is a bubble ($\lambda_1 = 0$) or has a finite viscosity. This same conclusion was drawn by Beshkov

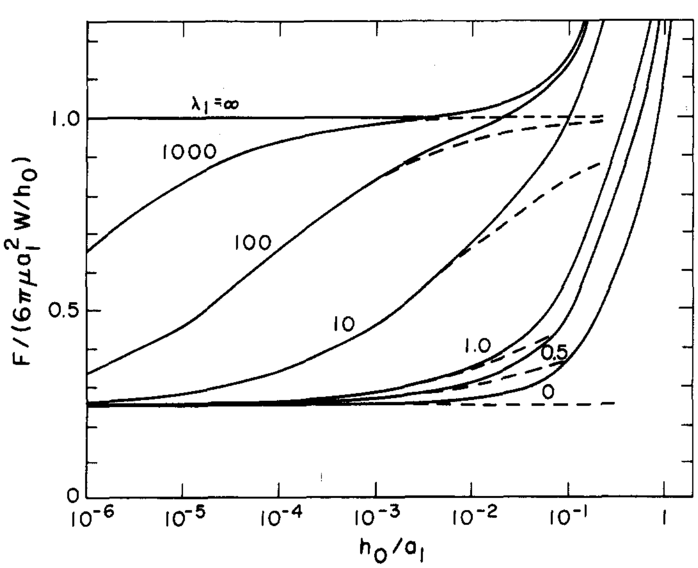

Figure 3. The dimensionless lubrication force as a function of the dimensionless separation for drop 1 approaching a rigid plate for several values of λ_1. The solid lines are from Haber *et al.* (1973) using bispherical coordinates; the dashed lines are our new near-contact solution.

et al. (1978) who examined the limiting form ($h_0/a \to 0$) of the series solution developed by Haber *et al.* (1973). Finally, it is seen in figure 3 that highly viscous drops ($\lambda_1 \gg 1$) behave as rigid spheres for sufficiently large separations ($h_0/a_1 \gg \lambda_1^{-2}$) but that they behave as bubbles with freely mobile interfaces for very small separations ($h_0/a_1 \ll \lambda_1^{-2}$).

Following Davis *et al.* (1989), regular asymptotic expansions may also be used to solve [23] and [24] and to illustrate the behavior of the lubrication force acting on a drop approaching a rigid body when the drop is much more viscous than the surrounding fluid. When the drop is very viscous ($\lambda_1 \gg \sqrt{a/h_0}$, $m_1 \ll 1$), the tangential velocity of the drop interface, u_1, is small, and both the drop and rigid interfaces strongly resist the flow of fluid from the gap, as shown in figure 2(b). Equations [23] and [24] may then be solved using asymptotic expansions in increasing powers of the mobility parameter, m_1. When the resulting expressions for $f_1(r)$ and $f_2(r)$ are substituted into [16], a series expansion for the lubrication force results:

$$\frac{F}{6\pi\mu a^2 \dfrac{W}{h_0}} = 1 - 0.65\, m_1 + 0.59\, m_1^2 - 0.55\, m_1^3 + 0.51\, m_1^4 - 0.48\, m_1^5 + O(m_1^6). \qquad [26]$$

Beshkov *et al.* (1978) give an approximate relationship identical to the first two terms of [26] except that the coefficient of -0.65 is replaced by -1.96, a difference of a factor of 3. At present, we have not determined the source of this discrepancy. Nevertheless, in the limit that $m_1 = 0$, both solutions give the well-known result for two colliding solid spheres as first developed by Reynolds (1886).

In the opposite limit when the viscosity of the drop is comparable to or smaller than that of the continuous phase ($\lambda_1 \ll \sqrt{a/h_0}$, $m_1 \gg 1$), the drop interface represents little resistance to flow in the gap, and the majority of the resistance results from the presence of the opposing rigid interface, as shown in figure 2(c). Under these conditions, [23] yields a Fredholm integral equation of the first kind that can be inverted to solve for $f_1(r)$. Davis *et al.* (1989) have numerically inverted an integral similar to this which differs only by a factor of 2/3. Using their result, the hydrodynamic force resisting the relative motion of the drop approaching a rigid body is given by

$$\frac{F}{6\pi\mu a^2 \dfrac{W}{h_0}} = \frac{1}{4} + \frac{0.657}{m_1}, \qquad [27]$$

clearly indicating that, in the limit as $m_1 \to \infty$, the hydrodynamic resistance which the drop experiences as it moves toward a rigid surface approaches one-fourth of that experienced by a rigid sphere under otherwise identical conditions. This result is independent of the drop viscosity, provided that the condition $\lambda_1 \ll \sqrt{a/h_0}$ is met. An approximate solution given by Beshkov *et al.* (1978) is identical to [27] except that the coefficient 0.657 is replaced by 0.981, a difference of a factor of $\sqrt{2}$.

Finally, for a drop of arbitrary viscosity approaching a rigid body, the Padé approximant [22] may be substituted into [25] to obtain a new Padé approximant

$$\frac{F}{6\pi\mu a^2 \dfrac{W}{h_0}} \approx \frac{1 + 0.47\, m_1 + 0.047\, m_1^2}{1 + 1.13\, m_1 + 0.19\, m_1^2}, \qquad [28]$$

which approximates the full numerical solution (dashed lines in figure 3) to within 1–2% for all values of m_1.

A drop near a fully mobile interface ($\lambda_2 \to 0, m_2 \to \infty$)

In the limit that drop 2 behaves like a bubble or free planar surface ($\lambda_2 = 0$), the tangential stress on the interface of drop 2 is negligible compared to that on the interface of drop 1. By utilizing the result that $f_2 \to 0$ in [7] and [8], it is easy to show that

$$f_1(r) = \frac{3\mu}{h}\left(\frac{rW}{2h} - u_1\right). \qquad [29]$$

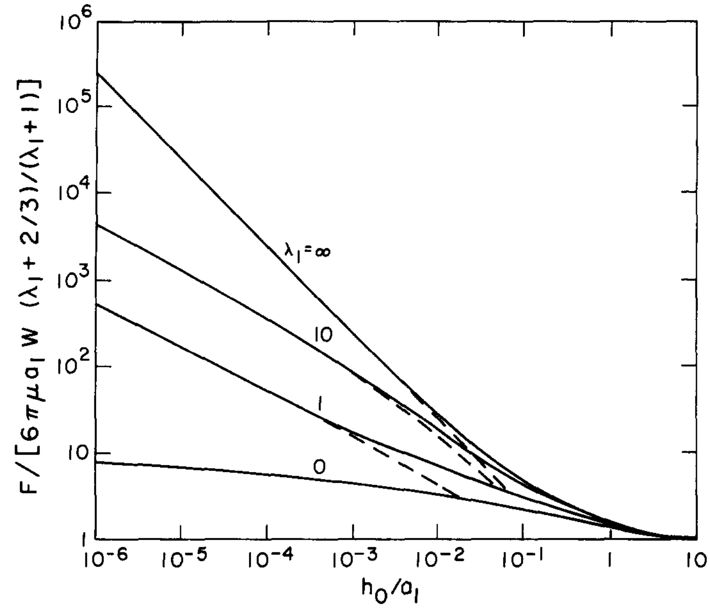

Figure 4. The dimensionless lubrication force as a function of the dimensionless separation for drop 1 approaching a free surface for several values of λ_1. The solid lines are from Haber *et al.* (1973) using bispherical coordinates; the dashed lines are our new near-contact solution.

Then, by substituting u_1 from [9] into [29], we obtain an integral equation solely in terms of $f_1(r)$:

$$f_1(r)=\frac{3\mu}{h}\left[\frac{rW}{2h}-\frac{1}{\lambda_1\mu}\int_0^{\infty}\phi(r',r)f_1(r')\,\mathrm{d}r'\right]. \qquad [30]$$

Equation [30] is a Fredholm integral equation of the second kind and must in general be inverted numerically to obtain a solution for the interfacial stress on drop 1. As in the limiting case for a drop approaching a rigid body, however, [30] can be related to the integral equation [21] which was previously solved by Davis *et al.* (1989), and the corresponding lubrication force acting on a drop approaching a free surface is given by

$$\frac{F}{6\pi\mu a^2\dfrac{W}{h_0}}=\frac{1}{4}\hat{F}\left(\frac{m_1}{2}\right), \qquad [31]$$

where $\hat{F}$ is the lubrication force resisting the approach of two drops of the same viscosity defined in [22]. Figure 4, a plot of the force made dimensionless with the Hadamard–Rybczynski formula for an isolated drop (see Batchelor 1967) as a function of the dimensionless separation, shows that these new lubrication results (---) are in good agreement with the bispherical coordinate solutions of Haber *et al.* (1973) (——) when the gap is sufficiently small relative to the size of the drop.

When the drop is very viscous ($\lambda_1 \gg \sqrt{a/h_0}$, $m_1 \ll 1$), the drop interface strongly resists the flow of fluid from the gap, as shown in figure 2(e). Again, using the asymptotic expressions of Davis *et al.* for the interfacial stress acting on two drops of the same viscosity, the hydrodynamic force may then be approximated by an asymptotic expansion in increasing powers of the mobility parameter, m_1:

$$\frac{F}{6\pi\mu a^2\dfrac{W}{h_0}}=\frac{1}{4}-0.164\,m_1+0.111\,m_1^2-0.077\,m_1^3+0.054\,m_1^4-0.038\,m_1^5+O(m_1^6). \qquad [32]$$

In the limit as $m_1 \to 0$, which corresponds to a rigid or extremely viscous sphere approaching a bubble or free interface, [32] shows that the force again becomes equal to one-fourth of that for two rigid spheres with the same reduced radius and relative velocity. Brenner (1961) previously showed that predictions of the drag on a rigid sphere approaching a planar free surface could be obtained from the solution for two equal rigid spheres moving toward one another using symmetry.

His series solution using the method of bipolar coordinates diverges, however, when the gap distance gets much smaller than the size of the sphere.

In the opposite limit, when the viscosity of the drop is comparable to or smaller than that of the continuous phase ($\lambda_1 \ll \sqrt{a/h_0}$, $m_1 \gg 1$), the drop interface represents little resistance to flow in the gap, as shown in figure 2(f). To leading order, the velocity profile is flat and has a magnitude of $u = rW/2h$ given by the mass balance [6]. The tangential stress and resulting hydrodynamic force resisting the relative motion of the drop under these conditions may be found from numerically inverting [30], which in this limit becomes a Fredholm integral equation of the first kind. Again, utilizing the solution developed by Davis *et al.* for the case of two drops of the same viscosity, this yields

$$\frac{F}{6\pi\mu a^2 \frac{W}{h_0}} = \frac{0.438}{m_1} \quad \text{or} \quad F = 8.26\lambda_1 \mu a W \sqrt{\frac{a}{h_0}}. \qquad [33]$$

A key result is that the lubrication force is proportional to the viscosity of the fluid inside the drop and is independent of the viscosity of the fluid in the gap. A similar result has been obtained by Beshkov *et al.* (1978) and by Ivanov & Traykov (1976) who argued that the dependency of the drag on the viscosity of the drop results from the fact that, in the near gap limit, most of the energy is being dissipated within the drop rather than within the gap. Moreover, the relative velocity under the action of a constant applied force decreases only in proportion to $h_0^{1/2}$, rather than in proportion to h_0 as in the rigid sphere case, and so coalescence can occur in a finite time without the requirement of an attractive force that increases in magnitude as the gap decreases. Finally, the resistance on a single drop approaching a bubble or a free surface in this limit is one-half of the value obtained by Davis *et al.* for the corresponding problem of a drop approaching another drop or half-space composed of the same fluid.

For a drop of arbitrary viscosity moving through an immiscible viscous fluid toward a bubble or a free surface [figure 2(d)], we can again make use of the Padé expression [22] to obtain an approximation to the full numerical solution. Substituting [22] into [31] yields a new Padé approximant of the form

$$\frac{F}{6\pi\mu a^2 \frac{W}{h_0}} \approx \frac{1}{4}\left(\frac{1 + 0.19\,m_1}{1 + 0.84\,m_1 + 0.11\,m_1^2}\right), \qquad [34]$$

which approximates the full numerical solution (dashed lines in figure 4) to within 1–2% for all values of m_1.

Drops of arbitrary viscosity

For arbitrary values of m_1 and m_2, [13] and [14] must be solved numerically using an iterative method. The procedure used was to initially choose $f_2(r) = 3\mu r W/h^2$, the tangential stress on a rigid sphere in close approach toward another rigid sphere, and then to compute $f_1(r)$ by numerically inverting [13]. Next, using the new $f_1(r)$, an improved $f_2(r)$ was computed by numerically inverting [14]. This simple, successive-substitution scheme converged rapidly for all values of m_1 and m_2 and required no artificial weighting.

The results of the numerical calculations are illustrated in figure 5, a plot of the dimensionless force on each sphere as a function of the mobility parameter m_1 for various viscosity ratios, $\lambda_2/\lambda_1 = m_1/m_2$. The limiting cases of $\lambda_2/\lambda_1 = \infty$ and $\lambda_2/\lambda_1 = 0$, corresponding to the force on a drop approaching a rigid body and a free surface, respectively, have been described in the two previous subsections. In addition, as $m_1 \to 0$, drop 1 behaves like a rigid sphere or plate; whereas when $m_1 \to \infty$, drop 1 behaves like a bubble or free surface. For $\lambda_2/\lambda_1 = \infty$, the dimensionless hydrodynamic force approaches a value of 1/4 as $m_1 \to \infty$; whereas for $\lambda_2/\lambda_1 = 0$, this force approaches a value of 1/4 as $m_1 \to 0$. The important case of $\lambda_2/\lambda_1 = 1$ corresponds to a drop approaching another drop or interface of the same fluid, the details of which have been discussed previously by Davis *et al.* (1989).

Figure 6 illustrates a comparison of our near-contact solution to a previous solution by Haber *et al.* (1973) who used the method of bispherical coordinates. The conditions illustrated in the plot

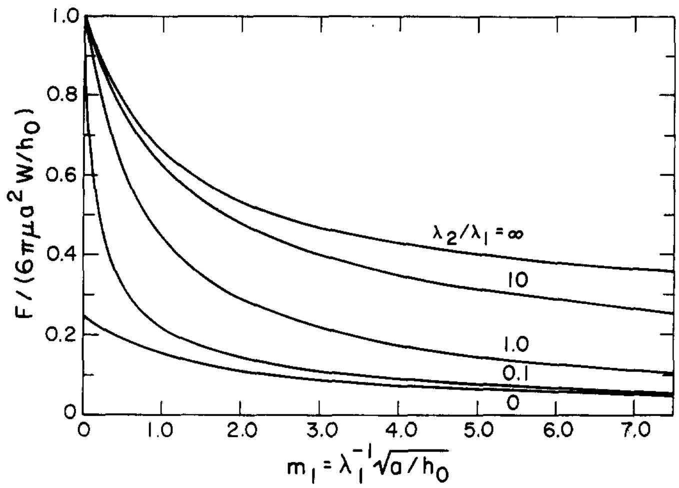

Figure 5. The dimensionless lubrication force as a function of the mobility parameter of drop 1 for several values of the viscosity ratio, λ_2/λ_1.

are for drops of the same size, $a_1 = a_2 = 2a$, moving at equal and opposite velocities, $V_1 = -V_2$. The hydrodynamic resistance force is made dimensionless using the Hadamard–Rybczynski formula for an isolated drop. For a very viscous drop approaching another viscous drop or solid body ($\lambda_1 \gg 1$, $\lambda_2 \gg 1$), an interesting feature of the curves shown in figure 6 is that the slope of these log–log plots changes from -1 to $-1/2$ as h_0/a decreases. This represents the transition from the drops exhibiting rigid sphere behavior for moderate separations to their interfaces becoming mobile at very small separations.

The new lubrication results agree with the exact results when h_0/a is sufficiently small. For very viscous drops in near contact (λ_1, $\lambda_2 \geqslant O(a/h_0)^{1/2}$), this is expected to be the case when $h_0/a \ll 1$. For drops of moderate viscosity or when one of the interfaces is fully mobile, one of two more stringent conditions, $\sqrt{h_0/a} \ll \lambda_1$ or $\sqrt{h_0/a} \ll \lambda_2$, is required. Finally, when both $\lambda_1 \leqslant O(h_0/a)^{1/2}$ and $\lambda_2 \leqslant O(h_0/a)^{1/2}$, such as may be the case for a gas bubble approaching another gas bubble or a free

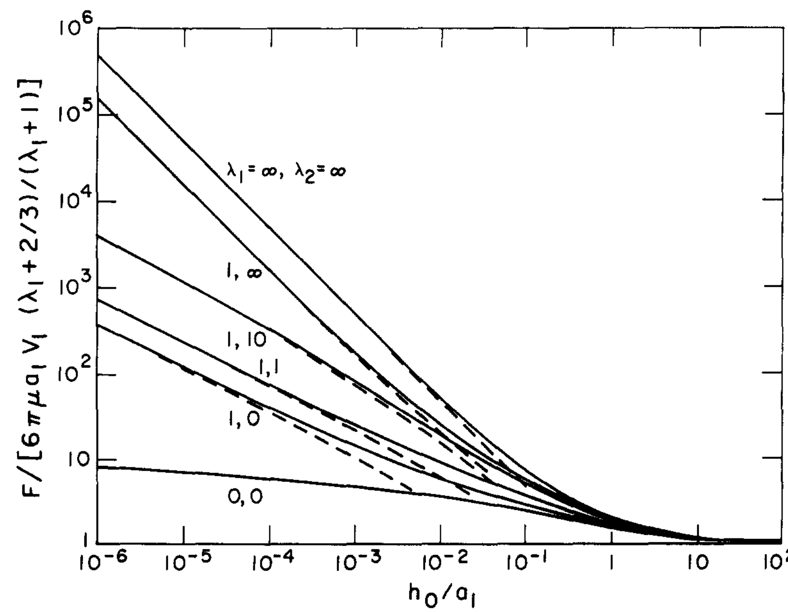

Figure 6. The dimensionless force as a function of the dimensionless separation for $a_1 = a_2 = 2a$ and $V_1 = -V_2$, for several values of λ_1 and λ_2. The solid lines are from Haber *et al.* (1973) using bispherical coordinates; the dashed lines are our new near-contact solution.

surface, the lubrication force does not dominate over the contribution to the force in the outer region, and the near-contact results described in this paper do not apply.

4. CONCLUDING REMARKS

This work presents quantitative predictions of the hydrodynamic lubrication force resisting the near-contact motion of two spherical drops of different viscosities moving along their line-of-centers toward one another through a third phase composed of a different fluid. Lubrication theory was used to model the flow of the fluid within the narrow gap between the drops, and the boundary integral formulation of the Stokes equations was used for the fluid flow within the drops. In the limit that the viscosity of one drop becomes large and its interface is immobile, the force on the drops is at least one-fourth of the lubrication force between two colliding rigid spheres and is proportional to the viscosity of the fluid in the gap. In addition, very viscous drops at moderate separations ($\lambda_1, \lambda_2 \gg \sqrt{a/h_0}$) experience a force resisting their motion that is inversely proportional to the minimum distance between the drop surfaces. Hence, a constant applied force is insufficient to push the drop into contact with a solid boundary in a finite time, and an additional mechanism—such as the action of van der Waals attractive forces—is required.

In contrast, significant tangential motion of the drop interfaces occurs due to the radial squeeze flow in the gap for drops with moderate relative viscosity at small separations. As the interface of one drop becomes fully mobile, the force is always less than one-fourth of that for two rigid spheres. Moreover, when the interface of one drop is fully mobile and the interface of the other drop is nearly so, the hydrodynamic resistance is proportional to the viscosity of the more viscous drop, and equals one-half the value acting on the drop moving toward an interface composed of the same fluid as the drop. Further, the force resisting the relative motion of the drops is inversely proportional to the square root of the minimum distance between the drop surfaces. This permits contact between two drops to occur in a finite time when they are subject to a constant force pushing them together, and thus has important implications in droplet coalescence.

In the analysis, inertia effects have been neglected relative to viscous effects. For the flow in the narrow gap between the drops, inertia effects are negligible when Re $(h_0/a) \ll 1$ for the nearly rigid and partially mobile cases, and when Re $\sqrt{h_0/a}/\lambda \ll 1$ for the fully mobile cases, where λ is the viscosity of the more viscous drop, $\text{Re} \equiv \rho Wa/\mu$ is the Reynolds number and ρ is the fluid density of the continuous phase. For the flow inside each drop, inertial effects are negligible when $\text{Re}(\rho_d/\rho) \ll 1$, where ρ_d is the density of the fluid comprising the drop.

In addition, it is assumed that the drops remain spherical. In order for any flattening of the drop interfaces to be small relative to the gap size, a normal stress balance on the interfaces requires that $(a/h_0)^2\,\text{Ca} \ll 1$ for the nearly rigid and partially mobile cases, and that $\lambda(a/h_0)^{3/2}\,\text{Ca} \ll 1$ for the fully mobile case, where λ is the viscosity of the more viscous drop, $\text{Ca} \equiv \mu W/\gamma$ is the capillary number and γ is the smaller of the two interfacial tensions. Each of these conditions is met when the drops are sufficiently small ($a \leqslant 10\,\mu$m, typically). However, when larger drops become close together, it is expected that the pressure which builds up to squeeze the fluid out from between the drop surfaces will also cause deformation of the interfaces near the axis of symmetry. The combination of lubrication theory and boundary integral theory developed in this work will still apply when deformation is important, but the normal stress balance must also be used to infer the gap thickness profile, $h(r, t)$.

Acknowledgements—This work was supported by NSF grant CBT-8451014, and by NASA grant NAGW-951 from the Microgravity Sciences and Applications Division of the Office of Space Sciences and Applications.

REFERENCES

BATCHELOR, G. K. 1967 *An Introduction to Fluid Dynamics*, p. 236. Cambridge Univ. Press, Cambs.

BESHKOV, V. N., RADOEV, B. P. & IVANOV, I. B. 1978 Slow motion of two droplets and a droplet towards a fluid interface. *Int. J. Multiphase Flow* **4,** 563–570.

BRENNER, H. 1961 The slow motion of a sphere through a viscous fluid towards a plane surface. *Chem. Engng Sci.* **16,** 242–251.

DAVIS, R. H., SCHONBERG, J. A. & RALLISON, J. M. 1989 The lubrication force between two viscous drops. *Phys. Fluids A* **1,** 77–81.

FUENTES, Y., KIM, S. & JEFFREY, D. J. 1988 Mobility functions for two unequal viscous drops in Stokes flow. Part 1. Axisymmetric motions. *Phys. Fluids* **31,** 2445–2455.

HABER, S., HETSRONI, G. & SOLAN, A. 1973 On the low Reynolds number motion of two droplets. *Int. J. Multiphase Flow* **1,** 57–71.

HETSRONI, G. & HABER, S. 1978 Low Reynolds number motion of two drops submerged in an unbounded arbitrary velocity field. *Int. J. Multiphase Flow* **4,** 1–17.

IVANOV, I. B. & TRAKOV, T. T. 1976 Hydrodynamics of thin liquid films. Rate of thinning of emulsion films from pure liquids. *Int. J. Multiphase Flow* **2,** 397–410.

RALLISON, J. M. & ACRIVOS, A. 1978 A numerical study of the deformation and burst of a viscous drop in an extensional flow. *J. Fluid Mech.* **89,** 191–200.

REYNOLDS, O. 1886 On the theory of lubrication and its application to Mr. Beauchamp Tower's experiments. *Phil. Trans.* **177,** 157.

RUSHTON, E. & DAVIES, G. A. 1973 The slow unsteady settling of two fluid spheres along their line of centres. *Appl. scient. Res.* **28,** 37–61.

RUSHTON, E. & DAVIES, G. A. 1978 The slow motion of two spherical particles along their line of centres. *Int. J. Multiphase Flow* **4,** 357–381.

WACHOLDER, E. & WEIHS, D. 1972 Slow motion of a fluid sphere in the vicinity of another sphere or a plane boundary. *Chem. Engng Sci.* **27,** 1817–1828.