

THE INFLUENCE OF VISCOSITY ON THE STRENGTH OF AN AXIALLY STRAINED PENDULAR LIQUID BRIDGE

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(First received 14 October 1988; accepted in revised form 2 October 1989)

Abstract—Pendular liquid bridges between two relatively moving solid particles (dynamic bridges) occur extensively throughout the powder technology industry, with a typical example being binder granulation. By means of a sensitive linear voltage differential force transducer, the influence of viscosity on the strength of an axially, periodically strained pendular bridge was investigated where *bridge strength* is defined as the maximum interparticle force developed during one complete sphere oscillation. The relative importance of the theoretically derived dimensionless groups governing bridge strength is experimentally established for systems exhibiting complete solid–liquid wettability. A simplified solution of dynamic bridge strength based on the superposition of lubrication theory and circular approximation is presented. For a small gap distance with sufficient bridge volume and in the limit of small Reynolds number, a good agreement between the experimental and present theoretical axial force response is observed, indicating the importance of the capillary number Ca in determining pendular bridge strength. The present quasi-static solution is zeroth-order in Ca and gap distance, and is expected to break down with increasing local inertial effects. Such inertial effects are governed by a modified Bond number. In the limit of low Ca , they lead to an increase in bridge strength due to an added mass effect, whereas in the limit of high Ca , they lead to a reduced, shifted force response due to an insufficient rate of vorticity propagation. Under industrially relevant conditions, the strength of the dynamic bridge was observed to exceed the static by over an order of magnitude. Since a significant number of powder processes such as granulation are not static in nature, viscous interactions between particles should be considered whenever a liquid interstitial phase is present.

INTRODUCTION

Pendular liquid bridges between two relatively moving solid particles occur extensively throughout the powder technology and related industries. Whether intentional or not, deposited moisture typically has a major influence on powder flow properties and mechanical processing ability ranging from the interruption of hopper flow or pneumatic conveying to the desirable extreme of particle size enlargement or granulation. If sufficient liquid is present between the particles to allow capillary regions to develop (Rumpf, 1962), the dynamic, interparticle viscous forces within the capillary region become akin to that experienced in the field of suspension rheology with such applications as slurry transport, filtration and sedimentation. The underlying theme in all such processes is that the ability to process the powder hinges on interparticle and particle–vessel forces—interactions which interstitial liquid can drastically influence.

As a typical example of an industrial application, powder granulation is a heavily utilized unit operation by the pharmaceutical, agricultural chemical, food stuffs, and metallurgical industries. A spraying system typically atomizes a binder and accompanying solvent onto a moving bed of powder (or powders) contained in a vessel such as a fluid bed, a rotating

drum or pan, or a high-speed mixer. As the solvent is evaporated and the associated array of powder particles undergo various relative displacements with respect to one another, the resulting pendular liquid bridges and capillary regions strengthen with time (Ennis, 1990) and eventually solidify, leaving behind homogeneous granules composed of the original powders. Clearly in such a process, the liquid bridge (and associated gas–liquid interface shape) can not be regarded as stationary, and hence the interparticle attractive force is expected to depend on binder viscosity in addition to surface tension and binder–solid wettability.

Static liquid bridges with constant physical properties and the related forces they induce have been studied extensively both theoretically and experimentally (Fisher, 1926; Mason and Clark, 1965; Boucher *et al.*, 1975; Schubert, 1977; Mehrota and Sastry, 1980). Surface tension and the resulting pendular interfacial shape induces an attractive force between two spherical particles due to a contact line force and a pressure deficiency resulting from the curvature of the interface. The related and physically more realistic phenomena of a dynamic bridge binding together two particles undergoing relative motion with respect to one another has received considerable less attention. Under industrially relevant conditions, the attractive force due to the dynamic bridge is often found to exceed the force due to a geometrically similar static pendular bridge by over an order of magnitude due to the viscous dissipation of energy

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induced by relative particle motion (Adams *et al.*, 1984; Mazzone *et al.*, 1987).

The present study is directed towards the establishment of the relative importance of the dimensionless groups governing the strength (or interparticle force) of dynamically strained pendular liquid bridges. The present paper is restricted to relative axial oscillatory two-particle motion and systems exhibiting nearly complete wetting and constant Newtonian behavior. First, the governing dimensionless groups are theoretically established based on the Navier–Stokes equations and associated boundary conditions and an approximate theoretical solution for the strength of an axially strained pendular liquid bridge is presented. The solution is approximate in the sense that the separate contributions of liquid viscosity and surface tension are solved individually and then superimposed. The viscous contribution is based on lubrication theory whereas the surface tension contribution is based on circular approximation. Second, an experimental confirmation of the relative influence of the dimensionless groups is presented, inferring that the capillary number Ca largely governs the strength of a dynamic pendular bridge. Most importantly, the experimental results are seen to scale down to finer particles. For low Ca , fluid inertia is seen to induce an added mass effect, and hence an increase in maximum axial bridge strength. In contrast, for high Ca , increases in Reynolds number lead to a phase shift in the experimental axial force response with a corresponding reduction in maximum axial bridge strength due to an insufficient rate of vorticity propagation.

THEORETICAL CONSIDERATIONS

Consider two spherical particles of radius a separated by a gap distance $2h_0$ of which the lower sphere oscillates in the axial direction with frequency ω and amplitude A at a velocity of $U'(t) = -2dh'/dt'$, where $h'(r)$ is the equation describing the spherical interface. As shown in Fig. 1, the two spheres are bound by a pendular liquid bridge of gas–liquid interfacial shape $g'(z)$, volume V' , viscosity μ , density ρ , kinematic viscosity ν , and surface tension γ .

We introduce a system of cylindrical coordinates made dimensionless with respect to sphere radius:

$$(r, \psi, z) = (r'/a, \psi', z'/a)$$

where primed variables refer to *dimensional* quantities and unprimed variables are *dimensionless*. In addition, pressure and velocity fields are given by

$$P = \frac{P'a}{\gamma}, \quad \mathbf{u} = \frac{\mathbf{u}'}{\omega A}$$

and

$$\nabla = a\nabla', \quad \nabla^2 = a^2\nabla'^2, \quad t = \omega t'$$

where pressure has been made dimensionless with respect to a characteristic capillary scale induced by surface tension.

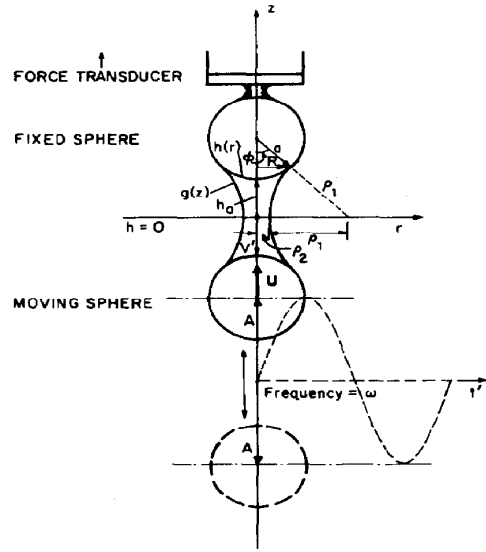


Fig. 1. Two spheres bound by a pendular bridge undergoing axial oscillatory motion.

Assuming quasi-static, axially symmetric flow thereby ignoring inertial effects due to an accelerating reference frame and azimuthal angle dependence, the Navier–Stokes and continuity equations become

$$Ca Re \left[\frac{1}{\alpha} \frac{\partial \mathbf{u}}{\partial t} + \mathbf{u} \cdot \nabla \mathbf{u} \right] = Ca \nabla^2 \mathbf{u} - \nabla P \quad (1)$$

$$\nabla \cdot \mathbf{n} = 0 \quad (2)$$

with the associated boundary conditions of

$$\mathbf{n} \cdot \mathbf{u} = \frac{1}{\alpha} g_t (1 + g_z^2)^{-1/2} \quad (3a)$$

$$\mathbf{t} \cdot (\mathbf{\sigma} - \mathbf{\sigma}_g) \cdot \mathbf{n} = 0 \quad (3b)$$

$$\mathbf{n} \cdot (\mathbf{\sigma} - \mathbf{\sigma}_g) \cdot \mathbf{n} = (\nabla \cdot \mathbf{n}) \quad (3c)$$

$$V = \pi \left\{ \int_0^z g(z)^2 dz - 2 \int_0^R r [Z(\phi) - h(r)] dr \right\} \quad (4)$$

$$u = 0, \quad v = 0 \quad \text{at} \quad z = +h(r, t) \quad (5a)$$

$$u = U(t), \quad v = 0 \quad \text{at} \quad z = -h(r, t) \quad (5b)$$

where (v, u) are the components of the fluid velocity $\mathbf{u}$ in the (r, z) -directions, U is the lower-sphere sinusoidal velocity, $\mathbf{n}$ and $\mathbf{t}$ are the normal and tangential unit vectors to the gas–liquid interface $g(z, t)$, the point $[R(\phi), Z(\phi)]$ represents the three-phase contact line of the pendular liquid bridge resulting from a filling angle of ϕ , and $\mathbf{\sigma}_g$ and $\mathbf{\sigma}$ are the total fluid stresses of the gas and liquid mediums, respectively. For the surrounding gas medium, the total stress $\mathbf{\sigma}_g$ is simply $P_g \mathbf{\delta}$.

One should note that solid and gas interfacial shapes are functions of both time and position. The

spherical solid interface is simply

$$h(r, t) = \varepsilon(t)/2 + 1 - (1 - r^2)^{1/2} \quad (6)$$

where $\varepsilon = 2h_0/a$ is the time-varying dimensionless gap distance. For the particle motion under consideration, the gap distance ε and lower-sphere velocity are given by

$$\varepsilon(t) = \varepsilon_0 + \alpha(1 + \cos t) \quad (7)$$

$$U(t) = -\frac{2}{\alpha}h_t = -\frac{1}{\alpha}\varepsilon_t = \sin t \quad (8)$$

where α is the dimensionless amplitude of oscillation, and ε_0 is the overall minimum in the time-varying gap distance ε or the closest point of approach during sphere oscillation. The gas-liquid interface shape $g(z, t)$ is clearly not known *a priori* since it is a function of the fluid pressure and velocity fields and must in general be determined implicitly from boundary conditions (3a)–(3c) and (4).

The no-slip boundary condition at the solid-liquid interface is represented by conditions (5a) and (5b). The continuity of tangential stress at the gas-liquid interface is given by condition (3b) whereas the jump in normal stress due to interfacial curvature is given by condition (3c) where, from differential geometry, the right-hand term of $\tilde{\nabla} \cdot \hat{n}$ can be recognized as the vector representation of twice the mean interfacial curvature H . The kinematic condition resulting from the motion of fluid particles situated at and defining the gas-liquid interface is given by condition (3a). Finally, the subsidiary condition of a given bridge volume V [eq. (4)] defines a unique liquid bridge shape $g(z)$ with the assumption of complete liquid-solid wetting (zero contact angle).

In summary, the fluid motion and resulting strength of the liquid bridge is governed by a Ca , Reynolds number Re , and dimensionless volume V given by

$$Ca = \frac{\mu\omega A}{\gamma}, \quad Re = \frac{\omega\omega A}{\nu}, \quad V = \frac{V'}{a^3}$$

as well as a dimensionless *minimum* gap distance ε_0 and amplitude α resulting specifically from the oscillatory particle motion being studied here and given by

$$\varepsilon_0 = \frac{2h_{0m}}{a}, \quad \alpha = \frac{A}{a}$$

and $2h_{0m}$ is the dimensional minimum gap distance. For the case of two spheres approaching at a constant relative velocity, liquid bridge strength is governed solely by the former three parameters.

A formal solution of eqs (1) and (2) with the associated boundary conditions of eqs (3)–(5) is a formidable task not attempted here. However, for the present experimental work, a solution based on the superposition of viscous and surface tension contributions to the strength of the liquid bridge is presented. For the present study, the contribution of surface tension is represented by the well-known toroidal or circular approximation (Fisher, 1926) of the strength

of a static pendular bridge whereas the viscous contribution to bridge strength is based on a lubrication analysis of two spheres in an infinite fluid medium. For such a problem, the dominant (or singular) contribution to the force acting on the lower moving sphere comes from the fluid in the immediate vicinity of the point of closest contact of the two spheres or pole ($r = 0$). Although such an analysis of the viscous contribution ignores the existence and influence of the gas-liquid interface on this inner region, it becomes justified in the limit of small gap widths, small Ca and sufficient bridge volume as is shown later.

Circular approximation

Surface tension governs the strength of the static bridge and contributes to the strength of the dynamic by its influence on the interfacial gas-liquid bridge shape $g(z, t)$ or boundary conditions (3a)–(3c) and eq. (4). In the limit of small Ca , the dynamic bridge strength approaches that of the static. This is easily shown formally by expanding the pressure and velocity fields as well as the gas-liquid interfacial shape in powers of the Ca or

$$P = \sum_{i=0}^{\infty} P_i Ca^i, \quad \tilde{u} = \sum_{i=0}^{\infty} \tilde{u}_i Ca^i, \quad g = \sum_{i=0}^{\infty} g_i Ca^i.$$

The zeroeth-order pressure field resulting from this asymptotic expansion is obtained from the Navier-Stokes eq. (1) and the normal stress balance (3c) which simplify to

$$\tilde{\nabla} P_0 = 0 \quad (9)$$

$$P_0 - P_g = \frac{g_{0,zz}}{(1 + g_{0,z}^2)^{3/2}} - \frac{1}{g_0(1 + g_{0,z}^2)^{1/2}} = 2H_0 \quad (10)$$

where H_0 is the zeroeth-order mean curvature of the interface. Therefore, one obtains the familiar Laplace-Young eq. (10) describing the zeroeth-order pressure field in the limit of $Ca \rightarrow 0$. As usual, applying the subsidiary condition (4) of a given bridge volume with eq. (10) establishes the shape of the static or zeroeth-order interface $g_0(z)$. In a similar straightforward manner, higher-order corrections to the interfacial shape $g(z, t)$ and pressure resulting from fluid motion can be obtained as outlined elsewhere [e.g. Park and Homsy (1984)].

Since we are largely concerned with the effect of viscosity on dynamic bridge strength for small gap widths, circular approximation will be adopted as an approximate solution to the strength of the static bridge, thereby avoiding well-known numerical solutions of eq. (10). The zeroeth-order pressure field then simplifies to

$$P_0 - P_g = 2H_0 = \left(\frac{1}{\rho_1} - \frac{1}{\rho_2} \right) \quad (11)$$

where the principle radii of curvature ρ_1 and ρ_2 are now given by

$$\rho_1 = \frac{1 + \varepsilon}{\cos \phi} - 1 \quad (12a)$$

$$\rho_2 = \sin \phi + \rho_1 (\sin \phi - 1) \quad (12b)$$

and ϕ is the filling angle resulting from a liquid bridge volume V and gap distance ε . Integrating over the wetted area of the sphere, we obtain the desired zeroth-order force acting on the upper (static) sphere

$$F'_{\text{stat0}}/\pi\gamma a = \sin^2 \phi \left[2 + \left(\frac{1}{\rho_1} - \frac{1}{\rho_2} \right) \right] \quad (13)$$

where the first term or factor of 2 results from the axial component of the contact line force acting along the contact line of length $2\pi R \sin \phi$. In obtaining the static force given by eq. (13), the imbalance in force between the upper and lower sphere resulting from the distortion of the interface about the meridian plane due to gravity is assumed negligible. For the small minimum gap distances and bridge volumes used during the present work (e.g. $\varepsilon = 0.0125$, $V = 0.128$), a relatively small discrepancy is exhibited between the exact numerical solution of the Laplace–Young equation and circular approximation [eq. (13)] as shown earlier by Mazzone *et al.* (1986).

Lubrication analysis

In obtaining eq. (9), it is assumed that radial and axial distances are of order a or sphere radius and the fluid pressure is of order γ/a . However, the combination of small gap distances and sphere motion can invalidate this assumption, particularly in the inner region of the bridge where the spheres nearly touch. In fact, in the limit of small gap widths, one may apply lubrication theory. Hence, viscosity contributes to the strength of a dynamic pendular bridge in two ways. First, it influences the dynamic gas–liquid interfacial shape $g(z, t)$ of the outer, non-lubrication region; although it can be significant, this effect as well as dynamic contact angle hysteresis is ignored in the present theoretical analysis. Second, viscosity governs the singular, inner contribution to dynamic bridge strength of the fluid in the proximity of the pole (assuming no inertial effects).

The appropriate radial and axial length scales are clearly time-varying due to the inherent unsteadiness of a problem with a time-varying gap distance $2h_0(t)$. In the limit of small gap distance, the axial coordinate is actually of order $2h_0$. Hence, using gap distance ε as a small parameter, one rescales or stretches the radial and axial coordinates to obtain

$$(\tilde{r}, \tilde{z}) = (r/\varepsilon^{1/2}, z/\varepsilon)$$

with pressure being rescaled by a viscous pressure scale or

$$P = \frac{P'a}{\mu\omega A}$$

In addition, the pressure and velocity fields are rescaled according to

$$\tilde{P} = \varepsilon^2 P, \quad \tilde{v} = \varepsilon^{1/2} v, \quad \tilde{u} = u.$$

Finally, the equation describing the solid–liquid interface shape [eq. (6)] as well as the pressure and velocity

are expanded in powers of ε or

$$\tilde{h} = \sum_{i=0}^{\infty} \tilde{h}_i \varepsilon^i, \quad \tilde{P} = \sum_{i=0}^{\infty} \tilde{P}_i \varepsilon^i, \quad \tilde{u} = \sum_{i=0}^{\infty} \tilde{u}_i \varepsilon^i.$$

Combining the above and retaining only zeroth-order terms, the governing eqs (1) and (2) yield a final expression for the force acting on the upper sphere of

$$F'_{\text{vis0}}/6\pi\mu a U = 1/4\varepsilon \quad (14)$$

where, again, primed variables refer to dimensional quantities.

Total force and higher-order effects

The total dynamic pendular bridge strength is obtained by the superposition of the surface tension contribution approximated by eq. (13) and the viscous contribution given by eq. (14). This superposition is justifiable in the limit of small Ca and Re , small gap distance ε , and sufficient bridge volumes. Small Ca and small $Ca Re$ implies that viscosity and fluid inertia do not effect the gas–liquid interface shape $g(z, t)$ and hence the surface tension contribution to the strength of the bridge. Conversely, small Ca and $Ca Re$ also implies that the boundary conditions applied at the interface [eqs (3a)–(3c)] in determining the fluid velocity field within the lubrication region may be relaxed to $\tilde{u} \rightarrow 0$ at infinity. Small gap distance ε and sufficient bridge volume justify a lubrication analysis of the viscous contribution. For the axial oscillation of the upper sphere under consideration, the viscous force is given by

$$F_{\text{vis}} = F'_{\text{vis}}/F_{\text{vc}} = \frac{\sin t}{4\varepsilon} \quad (15)$$

where F_{vc} is a characteristic Stokes force or

$$F_{\text{vc}} = 6\pi\mu\omega Aa. \quad (16)$$

The static, surface tension contribution [eq. (13)] was shown to be

$$F_{\text{stat}} = F'_{\text{stat}}/F_{\text{sc}} = \sin^2 \phi \left[2 + \left(\frac{1}{\rho_1} - \frac{1}{\rho_2} \right) \right] \quad (17)$$

where F_{sc} is the corresponding static characteristic force given by

$$F_{\text{sc}} = \pi a \gamma. \quad (18)$$

In combining the viscous and surface tension contributions, it is somewhat arbitrary as to which characteristic force should be selected. Since it is convenient to compare the strength of the static bridge to that of the dynamic, the capillary static scale given by eq. (18) is appropriate. Hence, the total force acting upon the upper sphere or *liquid bridge strength* is simply

$$F = \sin^2 \phi \left[2 + \left(\frac{1}{\rho_1} - \frac{1}{\rho_2} \right) \right] + 6Ca \frac{\sin t}{4\varepsilon}. \quad (19)$$

Therefore, this approximate relation governing the strength of a dynamic pendular bridge can be considered zeroth-order in ε and Ca . The apparent first-order dependence on Ca can be removed at high Ca

since the contribution due to surface tension, F_{stat} , can be neglected and the viscous force rescaled by the characteristic Stokes force. This relation [eq. (19)] is clearly approximate in the sense that the velocity field developed in the lubrication region cannot satisfy the kinematic boundary condition of the gas-liquid interface [eq. (3a)]—a condition which, in the strictest sense, cannot be relaxed with high Ca or inertial effects. Hence, a number of higher-order corrections will now be examined.

For quasi-static flow, the most apparent corrections to the Stokes flow relation given by eq. (15) are the additional inclusions of first-order inertial, zeroth-order shear stress, and first-order pressure terms. Equation (15) was obtained by solely integrating the zeroth-order pressure field over the wetted area of the sphere. For the case of a sphere approaching a plane wall in an infinite medium, Cox and Brenner (1967) obtained

$$F_{\text{vis}} = \frac{1}{5} \left(1 + \frac{1}{2} Re \right) \ln \frac{1}{\varepsilon} \quad (20)$$

where the total viscous force was then given by

$$F_{\text{vis}} = \frac{1}{\varepsilon} \left[1 + \frac{1}{5} \left(1 + \frac{1}{2} Re \right) \varepsilon \ln \frac{1}{\varepsilon} \right]. \quad (21)$$

Hence, the Stokes correction ($Re \rightarrow 0$) for the inclusion of shear stress and first-order pressure effects in calculating the repulsive force acting on the sphere is $O(\ln \varepsilon)$ whereas the inertial correction is $O(Re \ln \varepsilon)$. Inertial effects arise from both the $\hat{\mathbf{u}} \cdot \nabla \hat{\mathbf{u}}$ term of eq. (1) and the $\partial \mathbf{u} / \partial t$ term due to the inherently unsteady gap distance. However, since acceleration effects due to the sinusoidal motion have been neglected in the present analysis, both inertial corrections are convective in nature. Since the majority of experiments were performed at $Re \leq 1$ and $\varepsilon = 0.0125$, however, the maximum correction to the zeroth-order lubrication analysis leading to eq. (14) is less than 5%.

In obtaining eq. (21), we have neglected any inertial effects arising from either particle acceleration or an accelerating reference frame. In order to neglect such local inertial effects (Cox and Brenner, 1967), one must require that

$$|Re_a / Re| = a^2 \omega / \nu \ll [\max(1, |Re|)] \ln \frac{1}{\varepsilon} \quad (22)$$

where Re_a is an acceleration Reynolds number given by

$$Re_a = a^3 \omega^2 A / \nu^2.$$

The ratio Re_a / Re represents the ratio of viscous diffusion time to the natural time scale of the present problem, namely $1/\omega$. Neglecting local acceleration of the fluid represented by Re_a compared to the convective acceleration leading to eq. (21) implies that viscous diffusion is faster than a time scale of $1/\omega$. Such an assumption will be shown experimentally to break down for bridge materials in the limit of high frequencies.

Insufficient viscous diffusion time introduces the classical added mass and Basset force effects as shown by Stokes (1851) for the case of a single sphere oscillating in an unbounded fluid, or

$$F'_{\text{vis}} / 6\pi\mu\omega Aa = \text{Real} \left[\left(1 + \beta + \frac{1}{9} \beta^2 \right) e^{-i\omega t'} \right] \quad (23)$$

where β is a Reynolds parameter related to the viscous diffusion time given by

$$\beta^2 = -i \frac{a^2 \omega}{\nu} = -i Re_a / Re. \quad (24)$$

Basset (1888) similarly obtained for arbitrary time-dependent sphere motion:

$$F'_{\text{vis}} = 6\pi\mu U' a - 6\pi\mu a^2 (\pi\nu)^{-1/2} \int_0^{t'} \frac{dU'}{d\tau'} (t' - \tau')^{-1/2} d\tau' + \frac{2}{3} \pi \rho a^3 \frac{dU'}{d\tau'}. \quad (25)$$

The first term of eqs (23) and (25) is the steady Stokes force being $\pi/2$ out of phase with the sphere displacement and in phase with the sphere velocity, whereas the third term represents the added mass or acceleration contribution being in phase with the sphere displacement and $\pi/2$ out of phase with the steady Stokes force. (One should note the introduction of a minus sign in these relations since we are considering forces preventing sphere separation as positive.) Finally, the Basset force given by the middle term represents a memory integral of the acceleration and, for the single oscillating sphere case, is $\pi/4$ out of phase with both the displacement and velocity of the sphere. One expects similar competing effects with two close spheres vibrating axially with respect to one another and, indeed, such effects are exhibited by the experimental data to be presented.

An area of intense theoretical and experimental research (Dussan V., 1979; Hockings and Rivers, 1982; Cox, 1986) having a significant effect on the strength of an axially strained pendular bridge is that of dynamic liquid spreading on a solid surface and contact angle hysteresis. Two classes of spreading may be identified since both fluid viscosity and inertia impede the movement of the three-phase contact line $[R(\phi), Z(\phi)]$. The former case of viscous spreading controlled by Ca and having received the most attention has been shown to influence the three-phase macroscopic contact angle hysteresis (Johnson and Dettre, 1964; Huh and Mason, 1977). The latter case involving the influence of fluid inertia on the dynamics of liquid spreading, and hence the dynamic interfacial shape $g(z, t)$, has received considerable less attention. One would expect this effect to depend on Re [or more appropriately, $Ca Re$, in the light of the scaling leading to eq. (1)] and Ca . The group $Ca Re$ is, in fact, a Bond number representing the importance of fluid inertia in relation to capillary effects. These effects have been neglected in the present theoretical analysis.

A final area of concern is the influence of bridge volume on the strength of the dynamic pendular bridge. Although no exact solutions exist, Adams and Perchard (1984) have presented an approximate solution by integrating the viscous pressure field induced by sphere motion over the wetted area of the sphere [i.e. up to the three-phase contact line resulting from the given filling angle $[R(\phi), Z(\phi)]$], or

$$F_{vis}/6\pi\mu Ua = \frac{1}{4} \int_0^R r^3/h(r)^3 dr. \quad (26)$$

For a filling angle of ϕ , $R(\phi)$ is simply $\sin \phi$. This relation for the viscous contribution also differs from the previous expression given by eq. (14) by an extraneous factor of two since Adams and Perchard studied the symmetrical case of two spheres approaching at equal velocities. One should note, however, that such an analysis ignores the presence of the gas-liquid interface in computing the viscous force in the same sense as the analysis presented in obtaining eq. (14).

EXPERIMENTAL

The majority of the experimental work to be described herein is restricted to Newtonian liquid bridges possessing constant physical properties and exhibiting nearly complete solid wetting. Various oils were used to simulate industrially encountered binders with the aim of experimentally establishing the relative importance of the dimensionless groups governing bridge strength as well as confirming the present approximate theoretical solution. Industrially encountered pendular bridges typically possess the additional complexity of having time-varying physical properties and often exhibit viscous strengthening due to solvent evaporation, crystallization or polymeriz-

ation (Ennis, 1990). Binder strengthening time constants which characterize this viscous strengthening of the pendular bridge can conveniently be measured by the present experimental apparatus to be described.

Experimental apparatus

A schematic representation of the experimental apparatus used to measure the strength of a liquid bridge between two spherical particles is shown in Fig. 2 and is described in greater detail elsewhere (Ennis, 1990). A small stainless steel sphere of 2-mm radius is mounted to the piston of a sensitive linear variable differential force transducer of Prime Technologies design (FT5A). Minute displacements of the transducer piston due to the acting force cause an imbalance in magnetic reluctance resulting in a precalibrated output voltage (1 V/g). Since these displacements are typically less than 1% of the minimum sphere gap distance, transducer compliance is negligible. The second sphere is mounted to a retractable rod assembly whose vertical displacement is measured by a second linear variable differential transformer (LVDT). Both the transducer force response and LVDT signal are amplified and filtered with Validyne CD19 carrier demodulators which give up to 1 mV/V sensitivity. Data are then collected with an IBM personal computer equipped with a Data Translation 2805 interface board at a rate of 2000 data points per sphere oscillation cycle. Sphere alignment as well as minimum gap distance $2h_{om}$ and amplitude A adjustment are performed with the aid of a three-dimensional positioning support and a Nikon 6C projection microscope operating in shadow illumination mode and equipped with a two-dimensional vernier table. A motorized crank of variable radius

Experimental Dynamic Pendular Bridge Apparatus

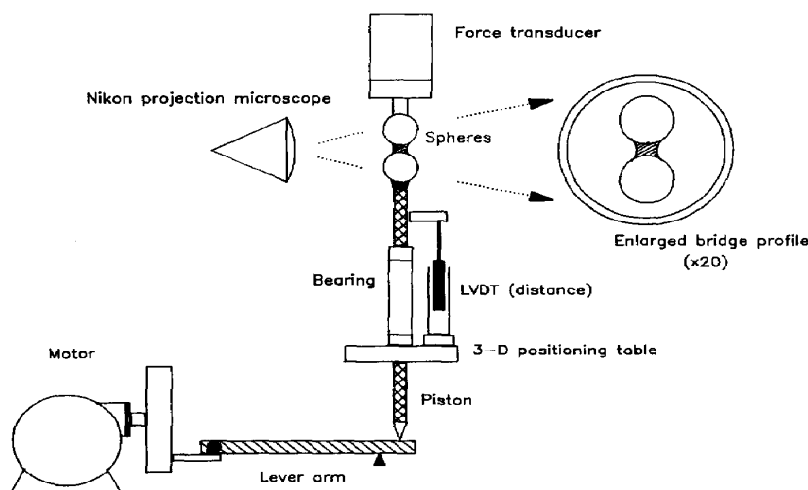


Fig. 2. Experimental liquid bridge apparatus for oscillatory particle motion.

and frequency supports a 42-cm lever arm which in turn provides the oscillatory vertical motion for the retracting rod and sphere assembly.

Since the lever arm has a lever ratio of approximately 20, oscillations of the lower sphere with amplitudes as small as one-quarter of a millimeter are easily obtained. The present system provides accurate control of sphere velocity at all points of sphere separation as opposed to the previous pneumatic arrangement which had an initial uncontrollable acceleration period (Mazzone *et al.*, 1987). Numerous oscillations can also be performed without bridge rupture which is a desirable feature for studying the time-dependent nature of binders.

After force transducer and LVDT calibration, 1 μ l of liquid bridge material is placed with the aid of a microliter syringe between the spheres. This amount of binder represents a moisture level on the order of 25 wt % which is within the range for industrial applications of granulation. A typical filling angle ϕ of approximately 38° was observed experimentally.

The frequency of oscillation ω of the bottom sphere was varied from 0.5 to 8 Hz while the minimum gap distance $2h_m$ between the spheres (Fig. 1) was varied from 0.025 to 0.10 mm. The amplitude of sphere oscillation was maintained at 0.25 mm. The chosen ranges of frequency, amplitude, and minimum separation are considered realistic values representing the dynamics of agglomerating particles based on an examination of high-speed photography of high-temperature agglomerating copper shot (Siegel, 1976) and video photography of the low-temperature granulation of glass ballotini.

Effect of Ca

A typical axial force response of the upper sphere is shown in Fig. 3 for a 10-S silicone oil and Ca of approximately 0.035. Starting from position A, the major contribution to liquid bridge strength is from surface tension alone. Viscous effects are negligible due to the large maximum gap distance and nearly zero sphere velocity. As the lower sphere approaches the upper static sphere, the liquid bridge strength increases to a small maximum at position B and then decays to an overall minimum at position C just before the point of minimum gap distance e_0 . The contribution of surface tension to dynamic bridge strength is due to the axial component of a contact line force and a capillary pressure deficiency. Hence, both components generally act to attract and prevent sphere separation for the usual case of complete solid-liquid wetting. In contrast, viscosity always acts to oppose sphere motion. Therefore, both viscosity and surface tension oppose sphere separation whereas only viscous effects oppose sphere approach. The maximum at position B is clearly a direct result of the opposing effects of viscosity and surface tension during sphere approach. The existence of such a peak will be a function of the Ca . Finally, as the spheres begin to separate shortly after reaching the minimum sphere gap distance e_0 , the liquid bridge strength rises to an overall maximum at position D. This maximum attractive force preventing sphere separation, F_{max} , occurring at position D is also clearly a function of Ca and, to a lesser degree, Re .

To confirm the importance of Ca as the major governing dimensionless group for systems exhibiting

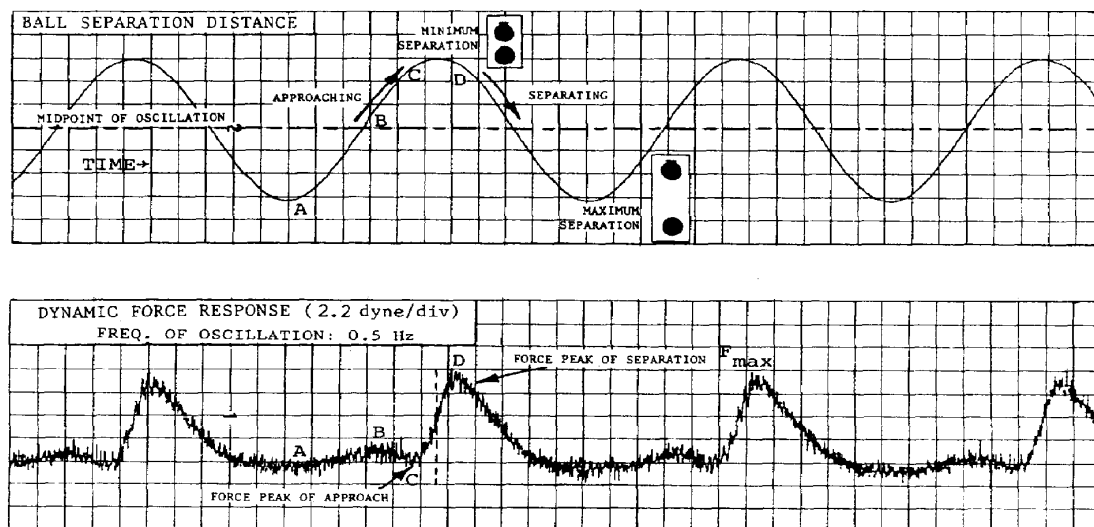


Fig. 3. Typical axial force response for $Ca = 0.035$ (silicone oil, $\nu = 10$ St, $\gamma = 19.4$ dyn/cm, $e_0 = 0.0125$, $\alpha = 0.125$, $V = 0.128$, $\omega = 0.5$ Hz).

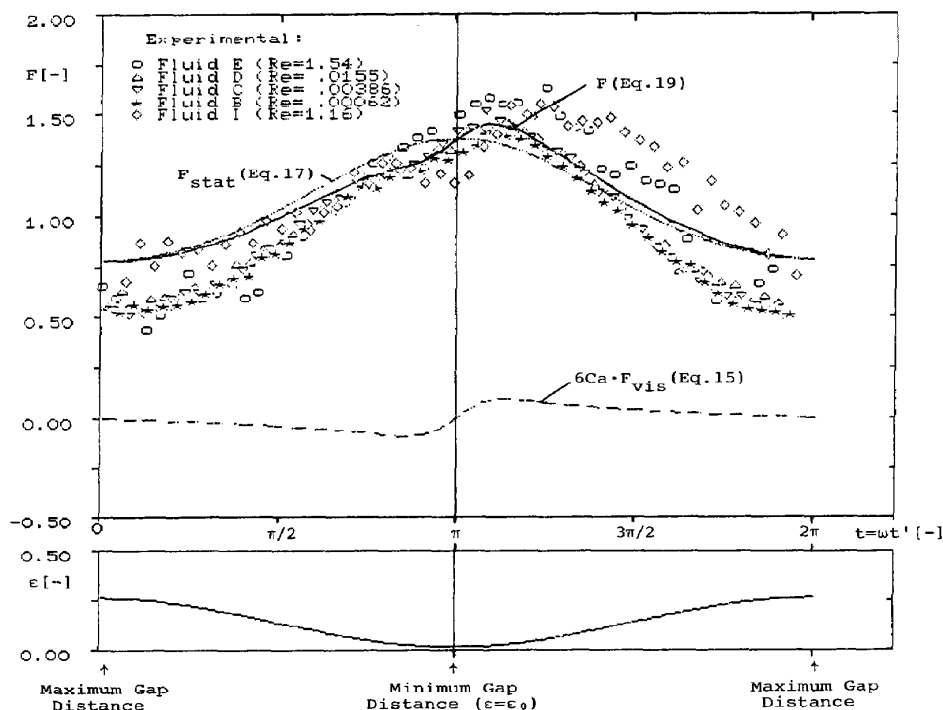
Table 1. Bridge materials and related properties (25°C)

Material	ID letter	Density (ρ) (g/cm ³)	Viscosity (μ) (P)	Surface tension (γ) (dyn/cm)
Silicone oil [†] —10 St	A	0.972	9.72	18.7
Silicone oil—5 St	B	0.971	4.86	18.8
Silicone oil—2 St	C	0.971	1.94	19.0
Silicone oil—1 St	D	0.970	0.97	18.9
Silicone oil—0.1 St	E	0.964	0.096	19.3
Silicone oil—125 St	F	0.980	122	23.2
Silicone oil—300 St	G	0.982	295	18.5
Silicone oil—1000 St	H	0.990	990	18.4
Dibutylphthalate [‡]	I	1.023	0.14	32.24
Immersion oil [§]	J	0.944	12.5	30.5

[†]Dow Corning (Silicone Fluid 200).[‡]Eastman Kodak Co., Inc.[§]R. P. Cargille Laboratories, Inc., Cedar Grove, NJ.

nearly complete solid-liquid wetting, three series of experiments were performed with a variety of bridge materials for Ca of 0.0035, 0.035 and 0.35. A summary of experimental bridge materials and their associated properties is given in Table 1. Newtonian liquid viscosities were measured by a combination of a Brookfield spindle viscometer and a Brookfield cone and plate viscometer equipped with a temperature-controlled cell. Surface tensions were measured by a combination of a Wilhelmy plate technique (Rosano Tensiometer) and a sessile drop gynecometer.

Figure 4 displays an axial force response with small viscous effects resulting from a Ca of 0.0035. The overall theoretical force response F given by eq. (19) is seen to closely follow the sinusoidal surface tension contribution given by circular approximation [eq. (17)]. The maximum force exhibited during sphere separation, $F_{\max}$, is of the order of a static bridge. The theoretical viscous contribution, which is an order of magnitude less than the corresponding static surface tension force, causes a slight negative and positive deviation from the surface tension contribution due to

Fig. 4. Axial force response with small viscous effects ($Ca = 0.0035$, $\epsilon_0 = 0.0125$, $\alpha = 0.125$, $V = 0.128$).

viscous repulsion during sphere approach and viscous attraction during separation. The experimental data follow a similar pattern and naturally deviate from the theoretical response at points of maximum sphere separation where circular approximation is no longer valid. Small deviations in the overall force response of the low-viscosity silicone oil and dibutylphthalate from the remaining data are likely due to the effects of imperfect solid wetting and fluid inertia with Re of the order of 1.

An overall axial force response exhibiting moderate viscous effects is shown in Fig. 5 for a Ca of 0.035. The theoretical contributions of surface tension and viscosity given by eqs (17) and (15), respectively, are now comparable in magnitude. Hence, there is a significant decrease in the overall attractive force during sphere approach due to viscous repulsion as observed for the total theoretical force response [eq. (19)], and exhibited by the experimental data.

Figure 6 displays an axial force response possessing extensive viscous effects occurring at a Ca of 0.35. The theoretical contribution of surface tension is now seen to be insignificant. Surface tension may still contribute significantly to the total force response with higher inertial effects, however, since it will effect the *exact* boundary conditions [eqs (3a)–(3c)] applied at the gas-liquid interface shape $g(z, t)$. The present approximate theoretical analysis does not account for this interaction of capillary and viscous lubrication

regions. A most important trend to note from Fig. 6 is the significant asymmetry and shift in the axial force response caused by an increase in Re . Only the most viscous fluids of 1000 and 300 St agree with the present approximate theory, having nearly symmetrical force peaks of approach and separation.

Figure 7 displays the effect of sphere radius on the axial force response for $Ca = 0.035$, $\varepsilon_0 = 0.0125$, $\alpha = 0.125$ and $V = 0.128$ for fluids of 5 and 10 St. Except for the smallest spheres of 0.53-mm radius, particle size is seen to have little effect on the dimensionless axial force response; deviations for the smallest spheres could easily be attributed to inaccurate adjustment of the minimum gap distance which is approximately 0.005 mm and approaching the limit of accuracy of the Nikon positioning vernier table. This would imply that the present theoretical analysis is sufficient to at least an order of magnitude in determining the strength of dynamically strained pendular bridges between the small particles of industrial interest and thus, in some respects, justifies the convenience of utilizing spheres of 2-mm radius for the majority of the experimental studies.

A summary of the effect of Ca on the maximum axial bridge strength $F_{\max}$ [calculated from eq. (19)] is shown in Fig. 8 for three minimum gap distances ε_0 . For low Ca ($< 10^{-3}$), the theoretical maximum bridge strength $F_{\max}$ approaches that of a static bridge whereas for high Ca ($> 10^0$), the maximum bridge

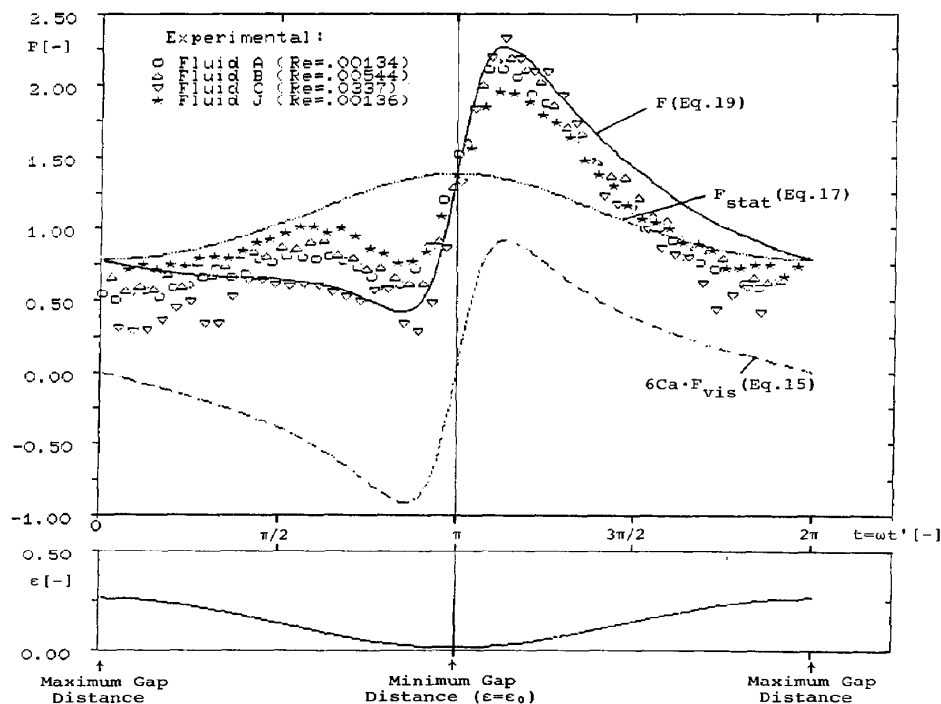


Fig. 5. Axial force response with moderate viscous effects ($Ca = 0.035$, $\varepsilon_0 = 0.0125$, $\alpha = 0.125$, $V = 0.128$).

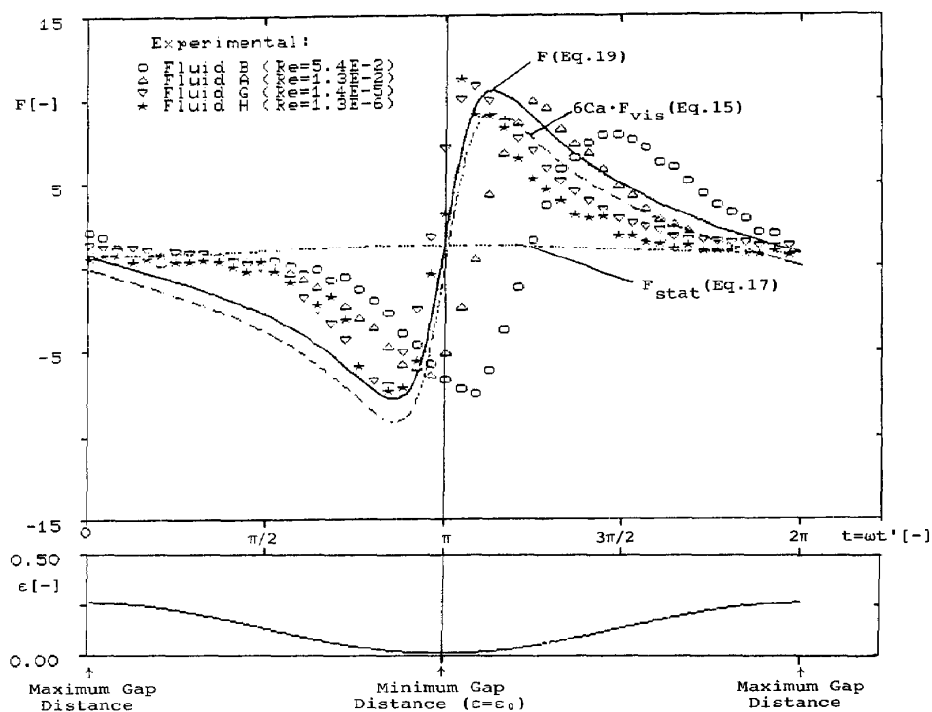


Fig. 6. Axial force response with extensive viscous effects ($Ca = 0.35$, $\epsilon_0 = 0.0125$, $\alpha = 0.125$, $V = 0.128$).

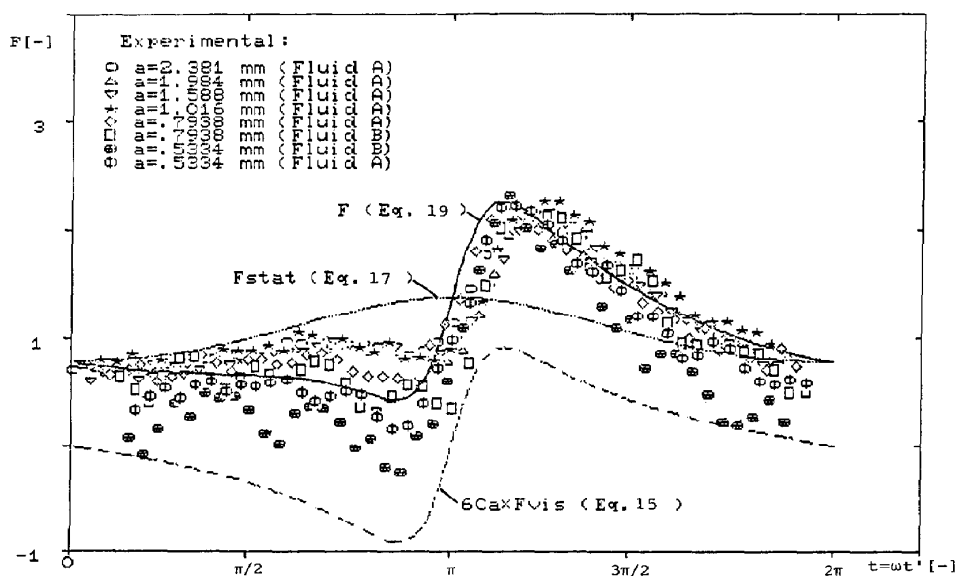


Fig. 7. Influence of particle radius on the dimensionless axial force response ($Ca = 0.035$, $\epsilon_0 = 0.0125$, $\alpha = 0.125$, $V = 0.128$).

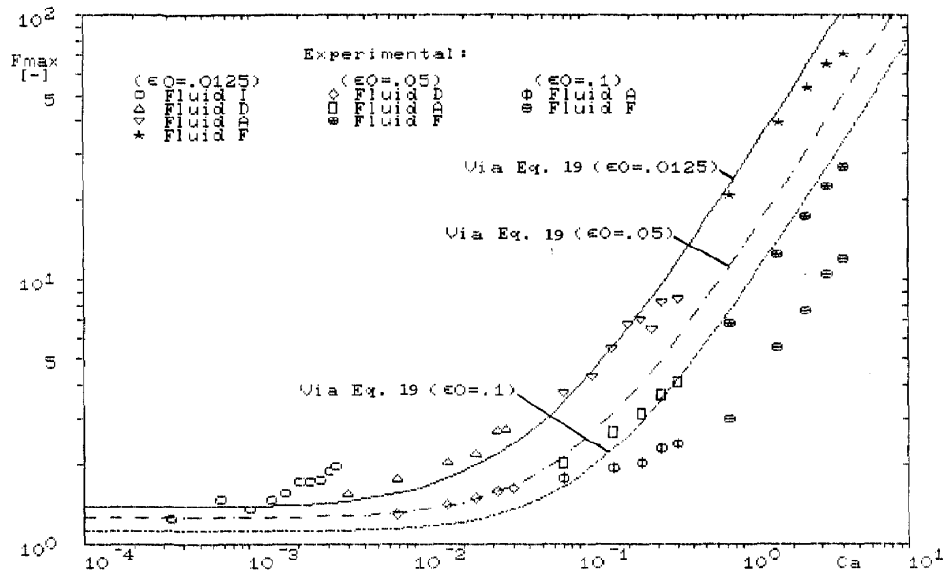


Fig. 8. Influence of Ca on maximum axial bridge strength $F_{\max}$ for various minimum gap distances ϵ_0 ($\alpha = 0.125$, $V = 0.128$).

strength approaches the viscous force induced by two particle relative axial motion in an infinite medium [eq. (15)] and is therefore independent of surface tension. Axial bridge strength (experimental and theoretical) is seen to be insensitive to Ca in the limit of both high and low Ca since the apparent dependence at high Ca can be removed by a rescaling with the viscous Stokes force [eq. (16)]. The boundaries of the industrially relevant, intermediate region are functions of ϵ_0 and α which govern the sphere motion. It is also seen from Fig. 8 that the present theoretical analysis significantly overestimates maximum bridge strength for the larger gap distances of 0.05 and 0.1. This is due to an increase in the relative contribution of inertial effects resulting from an invalidation of lubrication theory.

Inertial effects

For the minimum gap distance of 0.0125 used for the majority of experiments described, the theoretical maximum bridge strength resulting from eq. (19) agrees well with the experimentally measured maximum force for each of four fluids of Fig. 8 at their lower frequencies of oscillation or Ca . However, as sphere oscillation frequency ω or equivalently Ca is increased, experimental results for each fluid deviate from the theoretical curve due to increasing inertial effects.

It may be noted from the constant capillary number work presented in Figs 7–9 that no attempt was made to perform the experiments at constant Re . Hence, a convenient measure of the effect of fluid inertia on maximum bridge strength $F_{\max}$ at a constant capillary

number is provided as shown in Fig. 9 where the force is given as a function of the product $Ca Re$.

For the low- Ca region, positive deviations from the theoretical maximum are exhibited as Ca and, therefore also Re , is increased as shown for 1-St silicone oil (fluid I) in Fig. 8. A similar increase in maximum axial force with increasing Re can be seen in Figs 4 and 9. Such deviations most likely arise from the lubrication region where an added force is required to accelerate the fluid in the immediate vicinity of the singular pole and is, therefore, a local inertial effect. Convective inertial effects were shown to be small [eq. (20)] and can be ignored, particularly since they would act to decrease the required force during sphere separation. By an argument parallel to that given previously for two spheres oscillating in an infinite fluid [eq. (22)], one must require that

$$Bo = Ca Re / \alpha = \frac{\rho a^3 \omega^2 A}{\gamma a} \ll 1 \quad (27)$$

in order to neglect local acceleration effects in comparison with capillary pressure. Such a grouping of parameters can be considered as a modified Bond number Bo representing the ratio of displaced fluid inertial forces to capillary surface tension forces. It is interesting to note from Fig. 9 that for a constant Ca of 0.0035, positive deviations from the present quasi-static maximum theoretical force begin at $Ca Re \sim O(10^{-3})$ and hence $Bo \sim O(10^{-2})$. Letting $Bo = 10^{-2}$, one may solve eq. (27) for the critical frequency and, in turn, critical Ca at which fluid inertia becomes important for a nearly static pendular bridge. For the case of the 1-St oil (fluid I) of Fig. 8,

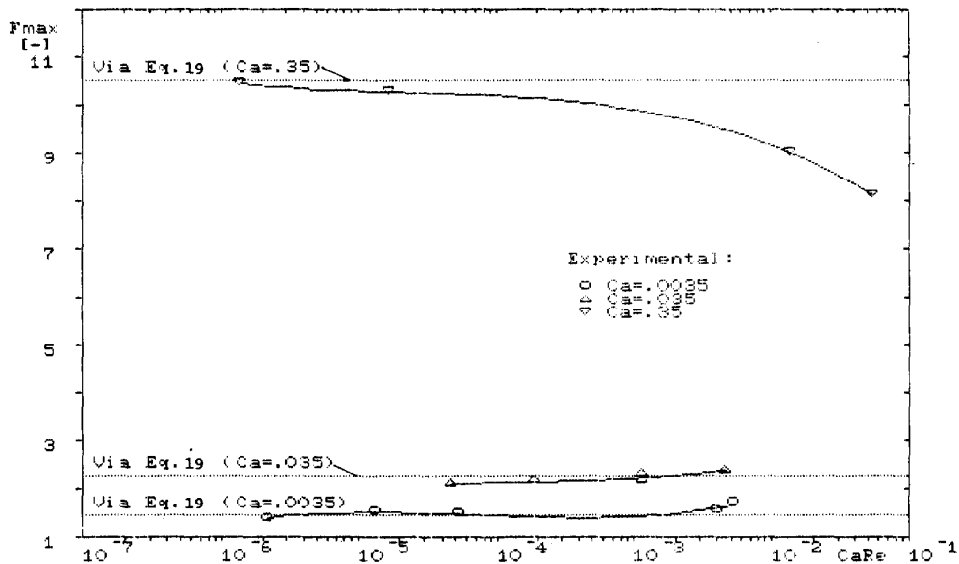


Fig. 9. Influence of fluid inertia on maximum axial bridge strength $F_{\max}$ ($\epsilon_0 = 0.0125$, $\alpha = 0.125$, $V = 0.128$).

a critical Ca of 0.002 is predicted which is in excellent agreement with initial experimental deviations.

The axial force responses for low- Ca bridges remain roughly symmetric about the point of minimum gap distance as shown in Fig. 4 despite increases in Re . This tends to further support the hypothesis of a local acceleration phenomenon since the added mass effect due to the fluid is expected to be in phase with decreasing sphere gap distance in light of eqs (23) and (25), and hence can only increase the observed force without introducing any phase lag. The Basset force may be regarded as a first-order correction to the inviscid, capillary flow field (Lawrence, 1986) which induces a positive phase lag due to a fading memory of maximum acceleration forces at the point of minimum gap distance. Such small increases in theoretical phase angle are exhibited in Fig. 4 with high Re (fluids E and I). Finally, for this low- Ca region, pressure remains of the order of a capillary scale, and hence the bridge shape does not differ considerably from that of a static bridge as shown in Fig. 10(a), further implying that the inner inviscid region and the bridge profile $g(z)$ do not influence one another.

Significant negative deviations from the theoretical maximum force are found for a Ca of 0.35 with increasing inertial effects as shown in Fig. 9. For the more viscous oils of 10 and 125 St (fluids A and F) as shown in Fig. 8, significant negative deviations are also exhibited as Ca and Re are increased. Finally, large phase shifts from the nearly oddly symmetric viscous theoretical response are observed as Re is increased at constant Ca as shown in Fig. 6. These effects can also be attributed to fluid inertia. To maintain the validity of the quasi-static analysis leading to

the viscous contribution of total bridge strength, one must require that the time required for vorticity propagation or viscous diffusion to the static measuring sphere surface from the moving sphere be small in comparison with the natural time scale of the problem, namely $1/\omega$. Increases in fluid inertia begin to invalidate this assumption, leading to the introduction of a minute time lag required for the full viscous force to develop. Hence, the maximum developed axial force occurs at a slightly larger gap distance and is, therefore, decreased. The same conclusion can be drawn from eq. (25) where, again, the Basset force arises as a first-order inertial correction to the viscous flow field. Contributions from the maximum fluid acceleration at minimum gap distance come at some phase lag after minimum separation and out of phase with velocity and the viscous force leading to the observed shift and decrease in the high- Ca force response as Re is increased.

By a similar calculation as was performed with the low- Ca bridge, the critical frequency or Ca at which inertial effects become important for a viscous, high- Ca bridge can be determined. At first glance, one may resort to eq. (22) based on two spheres oscillating in an infinite viscous fluid since the Navier-Stokes equations [eq. (1)] can be rescaled to remove Ca as explained earlier. Such a calculation based on vorticity propagation in an infinite *unbounded* fluid results in a critical frequency of 2 kHz for the 125-St oil (fluid F) of Fig. 8. Clearly, the presence of the gas-liquid interface cannot be ignored. This is to be expected for a high Ca since a rescaling of eqs (1)–(3) with a viscous pressure scale introduces the Ca into the normal stress balance along the gas-liquid interface $g(z, t)$. Repeating the calculation of critical frequency by eq. (27)

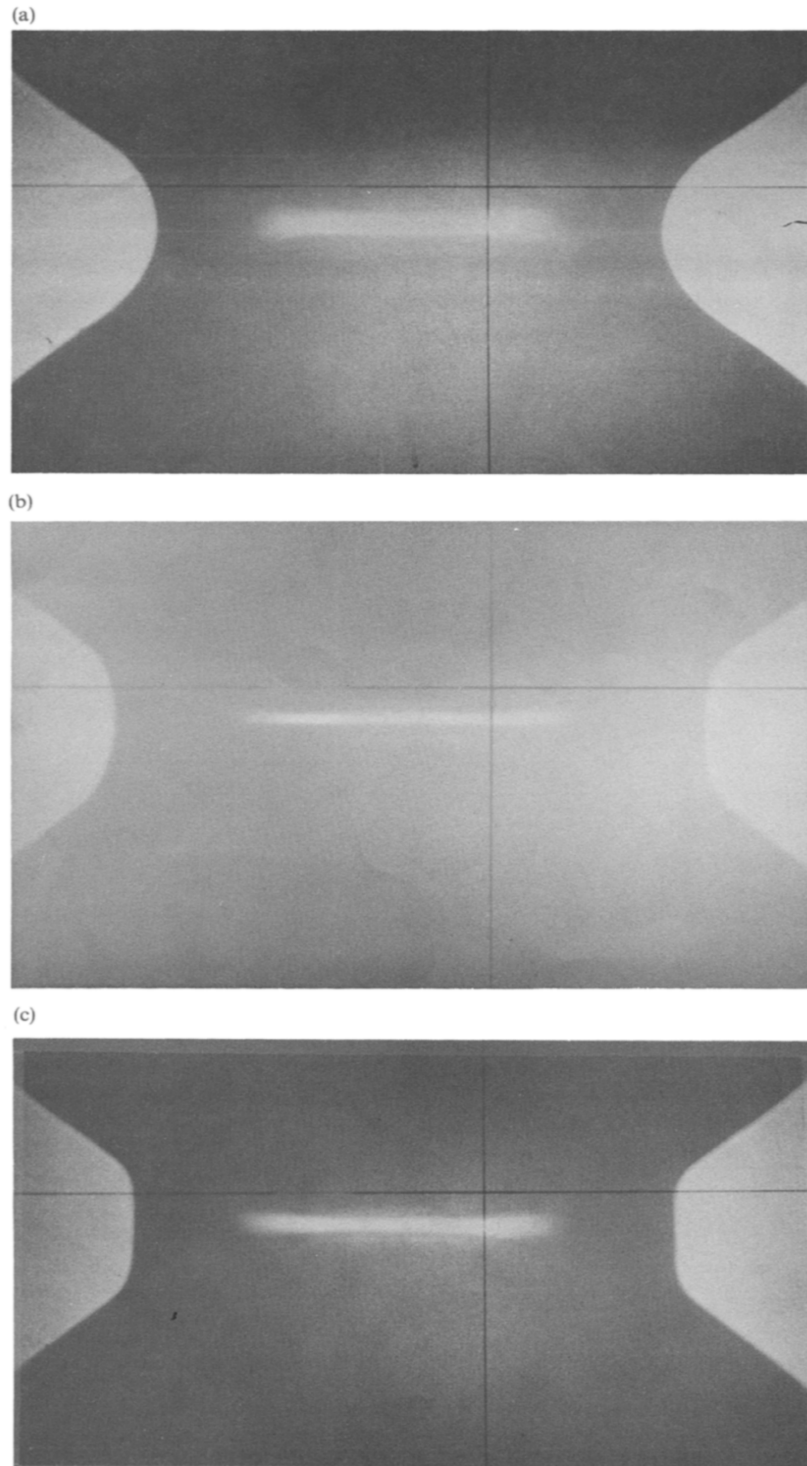


Fig. 10. (a) Influence of Ca on a typical bridge profile ($a = 2$ mm, $Ca = 0.0035$). (b) Influence of Ca on a typical bridge profile during sphere separation ($a = 2$ mm, $Ca = 0.35$). (c) Influence of Ca on a typical bridge profile during sphere approach ($a = 2$ mm, $Ca = 0.35$).

with $Bo = 0.01$, one obtains a critical Ca of 0.2 and 2 for the 10- and 125-St oils (fluids A and F), respectively, of Fig. 8, which again agrees well with experimentally observed deviations. The influence of high Ca on the gas-liquid interfacial profile is clearly observed in Fig. 10(b) and (c), where the profile tends to flatten during sphere approach. Such flattening resulting from the kinetics of surface spreading induced

by high Ca appears to significantly influence inertial interactions, and hence the total force response.

Effect of bridge volume

The effect of liquid bridge volume on the experimental maximum axial force F_{max} for the Ca studied in the present work as well as the effect of volume on static bridge strength is shown in Fig. 11. The

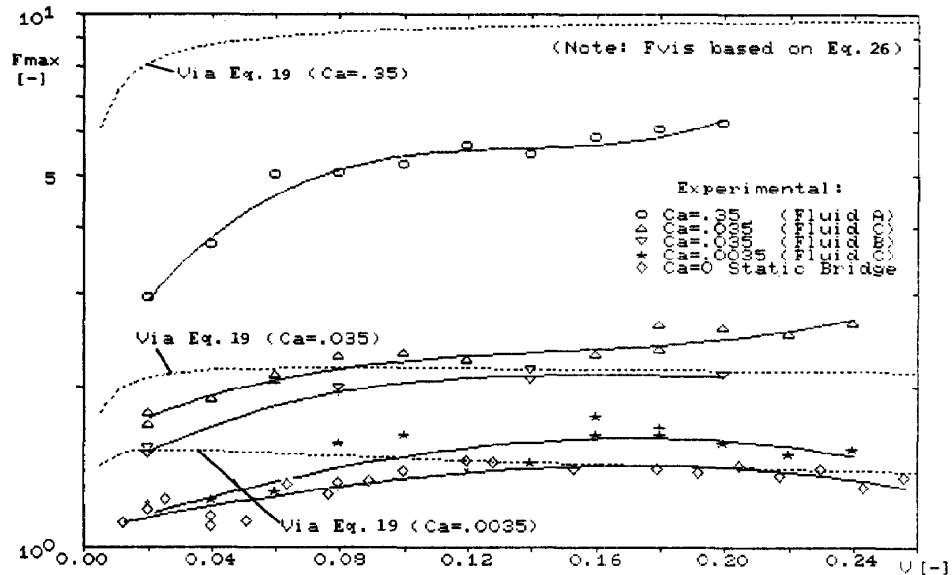


Fig. 11. Effect of bridge volume on maximum axial bridge strength ($\epsilon_0 = 0.0125$, $\alpha = 0.125$).

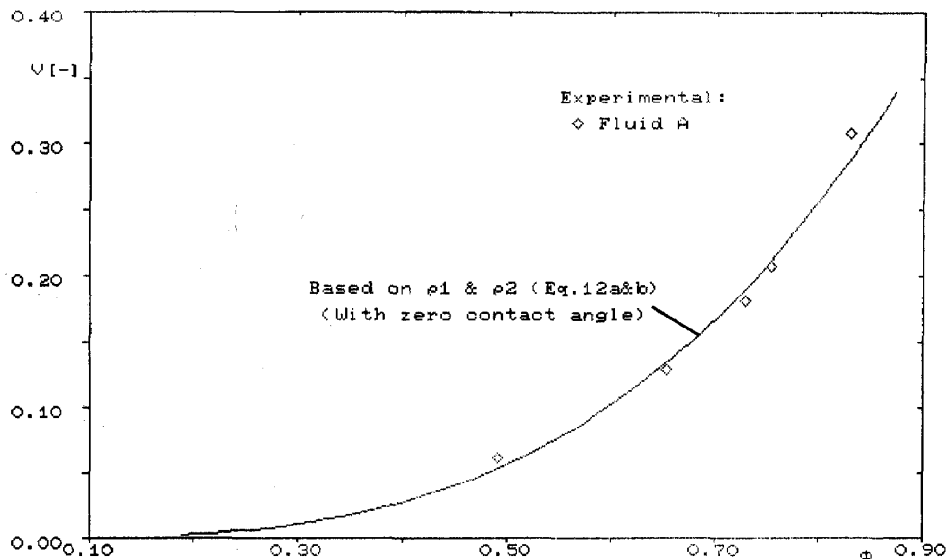


Fig. 12. Static bridge volume as a function of half-filling angle ϕ based on circular approximation ($\epsilon_0 = 0.0125$).

complete axial force response as a function of bridge volume is shown in Figs 13 and 14 for Ca of 0.0035 and 0.35, respectively. The maximum axial force is seen to be insensitive to bridge volume in the region of $V = 0.128$, the volume encountered in the previously presented work. This is to be expected for two reasons. First, the capillary or surface tension contribution is well known to be insensitive to volume since a decrease in mean curvature is balanced by an increase in wetted area as volume is increased. Second, the majority of the viscous contribution comes from the fluid in the immediate vicinity of the pole as described previously and is less sensitive to the surrounding fluid.

Shown for comparison in Fig. 11 are the corresponding theoretical curves for $Ca = 0.0035$, 0.035 and 0.35 where Adams and Perchard's result for the viscous force [eq. (26)], which integrates the lubrication solution over only the wetted area of the sphere, is taken as the viscous contribution to eq. (19). The theoretical filling angles based on circular approximation which are required for the viscous calculation [eq. (26)] are shown in Fig. 12 as a function of bridge volume for a minimum gap distance ε_0 of 0.0125. Good agreement is exhibited with the experimental filling angle of a 10-St oil which further justifies the use of circular approximation for the typically examined minimum gap distance of 0.0125.

For all Ca , the maximum axial force decreases as bridge volume is decreased below $V = 0.128$. For a nearly static bridge or small Ca this is due to the overriding effect of a decrease in wetted area over any changes in mean curvature. This is typical for the

minimum gap distances ε_0 of the order of 0.01 as shown in Mazzone *et al.* (1986). For higher Ca with more extensive viscous effects, the decrease in minimum bridge strength is due to a decrease of fluid in the vicinity of the gap or singular pole as well as an interaction between the bridge gas-liquid interface and the fluid at the pole which invalidates lubrication theory. Although all three theoretical curves display a drop in maximum bridge strength for very small volumes, two points should be noted. First, the theoretical bridge strength for a Ca of 0.0035 significantly overestimates experimental results and the exact solution of the Laplace-Young equation [eq. (10)] as expected for circular approximation (Mazzone *et al.*, 1986). Second, the viscous expression given by eq. (26) simply cuts off the contribution of lubrication theory outside the wetted area of the sphere and hence ignores any interaction between the interface and the inner fluid at the sphere gap. Therefore, the theoretical solutions at higher Ca again underestimate the drop in maximum bridge strength at small liquid volumes.

The effect of large bridge volumes is seen to depend on Ca . For small Ca with negligible viscous effects, the maximum bridge strength decreases due to a decrease in mean curvature with relatively little change in wetted area. Added mass effects may induce small positive deviations from the theoretical force response as shown in Fig. 13 for the 2-St oil (fluid C) at a volume $V = 0.16$. From the results of the previous section, a critical Ca of 0.004 is estimated for inertial effects which is close to the actual Ca of 0.0035. An increase in bridge volume to 0.22 tends to mitigate a possible inertial effect. The drop in axial force or

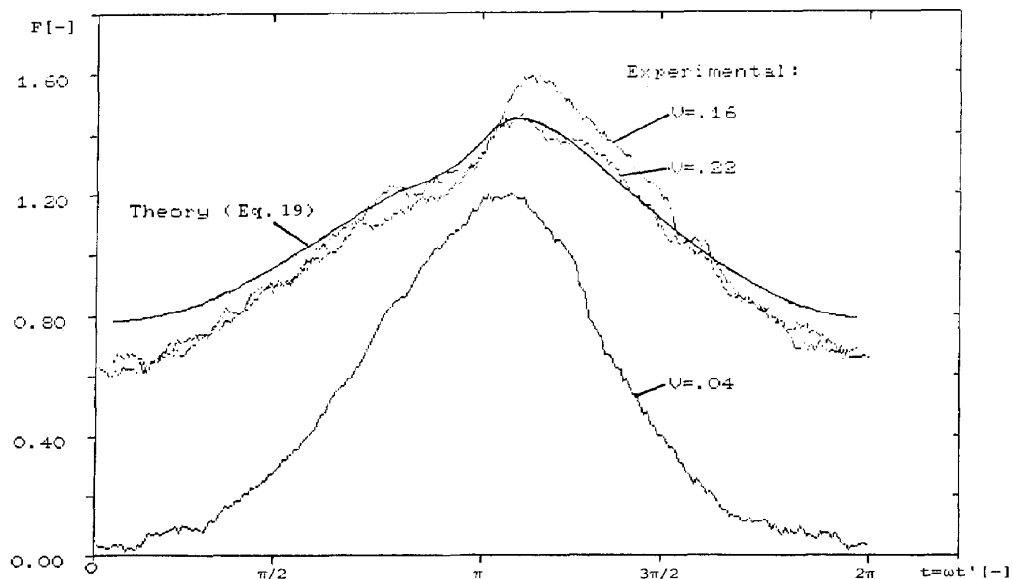


Fig. 13. Effect of bridge volume on a nearly static axial force response ($Ca = 0.0035$, $\varepsilon_0 = 0.0125$, $\alpha = 0.125$, fluid C).

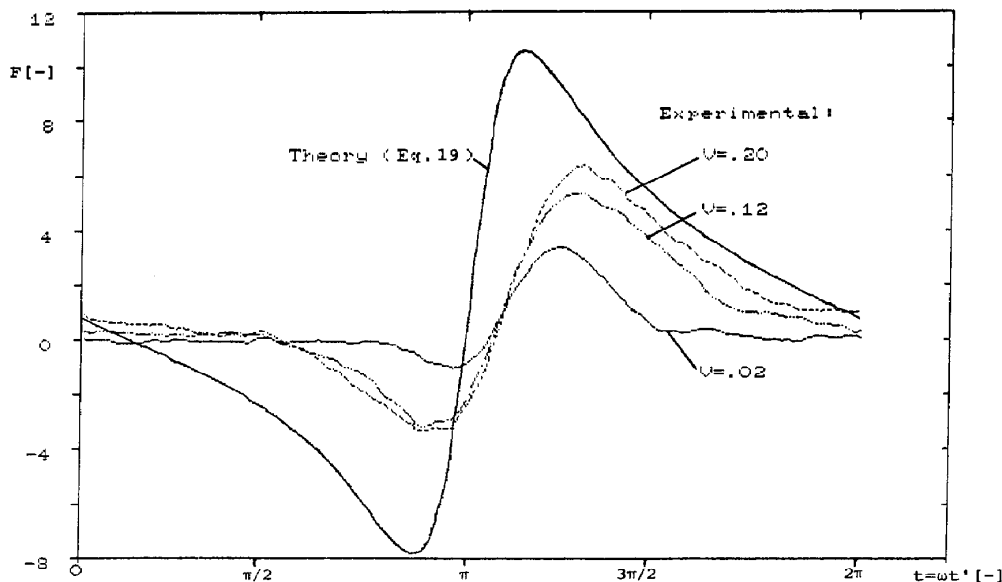


Fig. 14. Effect of bridge volume on a highly viscous axial force response ($Ca = 0.35$, $\varepsilon_0 = 0.0125$, $\alpha = 0.125$, fluid A).

bridge strength for the volume of 0.04 is due to bridge rupture at large gap distances (Fig. 13).

For large Ca with highly viscous effects, there is a slight increase in maximum bridge strength with increasing volume. These increases may again be related to inertial effects. First, one should note from the results of Fig. 8 that the 10-St oil clearly exhibits inertial effects in comparison with viscous diffusion at $Ca = 0.35$, leading to a decrease in bridge strength compared to theory as discussed previously. An increase in bridge volume will lessen the influence of the distorted, non-equilibrium interface on the lubrication region which may lead to an increase in experimental bridge strength toward the theoretical curve. Second, the experimental bridge strength for a 2-St fluid actually exceeds theoretical predictions at a Ca of 0.035 for larger bridge volumes. As shown previously for small Ca , this is likely due to an added mass effect as opposed to a phase lag, viscous diffusion effect. Finally, for a more viscous 5-St fluid, thereby lessening added mass effects, the maximum bridge strength for a Ca of 0.035 is seen to be slightly less than the corresponding bridge strength of the 2-St fluid. The bridge strength of the 5-St oil is also seen to approach the theoretical curve at higher bridge volumes, being unaffected by bridge volume.

CONCLUSIONS

The strength of dynamically strained pendular liquid bridges between relatively moving spherical particles can exceed the strength of their geometrically similar, static counterparts by an order of magnitude under industrially relevant conditions. This is largely due to the viscous dissipation of energy of the bridge

induced by relative particle motion and, hence, is governed by a Ca . For oscillatory Ca less than 10^{-3} , dynamic bridge strength is of the order of a static bridge and is insensitive to liquid viscosity and relative particle motion for small Re or inertial effects. In contrast, bridge strength is insensitive to surface tension and linearly related to Ca for Ca in excess of 10^0 (again assuming small Re). That is, bridge strength is only a function of viscosity at high Ca since the maximum axial bridge strength F_{max} can be rescaled by a viscous Stokes force eliminating the apparent dependence on Ca and surface tension.

These observations ignore inertial effects which can be significant. For small Ca , pendular bridge strength is due to the pressure deficiency resulting from the jump in normal stress induced by the curvature of the interface. A simple perturbation analysis indicates that the pressure field is constant and that inertial and viscous diffusion effects can be ignored on the zeroth order in Ca assuming small Re . Large inertial effects induce an increase in pendular bridge strength due to an added mass effect of the fluid arising from the local acceleration term of the Navier-Stokes relation. In fact, one may regard a low- Ca bridge as an inviscid bridge implying a similar superposition of ideal fluid theory and capillary pressure as was performed for the viscous bridge in the present work. Basset force interactions would be considered as first-order corrections to such an analysis inducing the observed experimental phase lag.

For arbitrary Ca and Re , no simplification of eq. (1) is possible except in the limit of small gap distances with sufficient bridge volume. This does not present a serious limitation in applicability since we are

largely concerned with forces induced at point contacts of nearly touching particles such as those occurring in industrial powder processes. On the zeroth order in gap distance and high Ca , pendular bridge strength is now due to a pressure force induced by the viscous diffusion term of eq. (1) where we are again assuming small Re in comparison to $(1/\varepsilon)$. With a large increase in inertial effects, the natural time scale of $1/\omega$ begins to approach the time scale of vorticity propagation invalidating a quasi-static analysis. In other words, the induced viscous force no longer develops instantaneously but instead requires some minute time lag which will increase with increasing Re . This characteristic time lag implies that the maximum pendular bridge strength occurring during sphere separation develops at a slightly larger gap distance and, hence, will be reduced. The resulting inertial phase shift and reduction in force is exhibited for all high- Ca experiments with correspondingly high Re .

In order to solidify a number of the present experimental observations regarding inertial effects on bridge strength, the problem of two nearly touching spheres oscillating axially with respect to one another in a viscous fluid must be undertaken {which has not been to the best of the authors' present knowledge [also see Lawrence (1986)]}. Inertial effects appear to be significantly influenced by bridge shape as observed for the high- Ca flattened profile and possible volume effects. Such topics are subjects of further investigation.

A theoretical analysis based on a simple superposition of circular approximation modeling zeroth-order Ca behavior and lubrication analysis modeling high- Ca viscous behavior adequately predicts the strength an axially strained pendular bridge over five orders of magnitude in Ca in the limit of small gap distance, sufficient bridge volume, and small inertial effects. Such an analysis has clearly been shown experimentally to break down for large inertial effects, large gap distance, and insufficient bridge volume where small bridge volumes bring about interactions between the lubrication region in the vicinity of the sphere gap and the interface unaccounted for by the present analysis.

The present work demonstrates the extreme influence liquid phase viscosity can have on the interparticle force between two relatively moving particles bound by a pendular bridge. Since a significant number of industrial applications involving powders are not static in nature, viscous interactions between particles should be considered whenever a liquid phase is present—particularly for finer powders where a high relative humidity can result in capillary condensation at contact points. As an additional example, previous models of the maximum force required to rupture a multi-particle, multi-bridge granule incorporate only the static, surface tension contribution of each individual pendular bridge (Rumpf, 1962; Schubert, 1977) and hence inadequately characterize the strength of a granule with a viscous interstitial binder.

Acknowledgements—Thanks are due to the International Fine Particle Research Institute for grant support. Helpful discussions with and the use of a surface tension gynecometer from Professor C. Maldarelli of The City College of CUNY are acknowledged with great appreciation. Lastly, the authors are indebted to Dr S. Spassov of Delft University for interesting conversations regarding inertial effects.

NOTATION

a	sphere radius, m
A	amplitude of sphere oscillation, m
Bo	Bond number [eq. (27)]
Ca	capillary number ($\mu\omega A/\gamma$)
F	total axial force [eq. (19)]
$F_{\max}$	maximum axial force ($F'_{\max}/F_{sc}$)
F_{sc}	characteristic surface tension force [eq. (18)]
F_{stat}	axial surface tension contribution to total force F [eq. (17)]
F_{vc}	characteristic viscous force [eq. (16)]
F_{vis}	axial viscous contribution to total force F [eq. (15)]
g	gas-liquid interfacial profile [$g(z, t) = g'/a$]
h	solid-liquid interfacial profile [$h(r, t) = h'/a$]
h_0	zeroth-order simplification of h (h'_0/a)
h_o	time-varying half-gap distance, m
h_{om}	minimum half-gap distance, m
H	mean interfacial curvature [eq. (11)]
$\hat{n}$	normal unit vector to gas-liquid interface $g(z, t)$
P	fluid pressure
P_g	atmospheric pressure
r	radial coordinate (r'/a)
R	radial contact position of liquid bridge (R'/a)
Re	Reynolds number ($a\omega A/\nu$)
Re_a	acceleration Reynolds number ($a^3\omega^2 A/\nu^2$)
t	time ($\omega t'$)
$\hat{t}$	tangential unit vector to gas-liquid interface $g(z, t)$
v	radial component of fluid velocity ($v'/\omega A$)
u	axial component of fluid velocity ($u'/\omega A$)
$\hat{u}$	fluid velocity ($\hat{u}'/\omega A$)
U	sinusoidal sphere velocity [eq. (8)]
V	bridge volume (V'/a^3)
z	axial coordinate (z'/a)
Z	axial contact position of liquid bridge (Z'/a)

Greek letters

α	dimensionless oscillation amplitude (A/a)
β	Reynolds number parameter
δ	identity matrix
γ	surface tension, N/m
ε	time-varying dimensionless gap distance ($2h_o/a$)
ε_0	minimum dimensionless gap distance ($2h_{om}/a$)
μ	Newtonian fluid viscosity, Pa s
ν	kinematic fluid viscosity, m ² /s
ρ	fluid density, kg/m ³
ρ_1, ρ_2	principal radii of curvature ($\rho'_1 a$ and $\rho'_2 a$)
$\bar{\sigma}$	total fluid stress tensor

$\bar{\sigma}_g$	total gas stress tensor ($P_g \bar{\delta}$)
ϕ	half-filling angle
ψ	azimuthal angle
ω	oscillation frequency, 1/s

Subscripts and superscripts (except as defined above)

i	summation indice
t	partial derivative with respect to time
z	partial derivative with respect to axial position
0	zeroth-order term
1	first-order term
'	dimensional variable
$\sim$	stretched lubrication variable

REFERENCES

- Adams, M. J. and Perchard, V., 1984, *Inst. chem. Engng Symp. Ser.* **91**, 12.
- Basset, A. B., 1888, *A Treatise on Hydrodynamics*, Vol. II. Deighton, Bell, Cambridge.
- Boucher, E. A., Evans, M. J. B. and McGary, S., 1982, *J. Colloid Interface Sci.* **89**, 154.
- Cox, R. G., 1986, *J. Fluid Mech.* **168**, 169.
- Cox, R. G., 1974, *Int. J. Multiphase Flow* **1**, 343.
- Cox, R. G. and Brenner, H., 1967, *Chem. Engng Sci.* **22**, 1753.
- Dussan V., E. B., 1979, *A. Rev. Fluid Mech.* **11**, 371.
- Ennis, B. J., 1990, PhD dissertation, The City College of the City University of New York, New York.
- Fisher, R. A., 1926, *J. agric. Sci.* **16**, 492.
- Hockings, L. M. and Rivers, A. D., 1982, *J. Fluid Mech.* **121**, 425.
- Huh, C. and Mason, S. G., 1977, *J. Fluid Mech.* **81**, 401.
- Lawrence, C., 1986, Ph.D. dissertation, The City College of the City University of New York.
- Mason, G. and Clark, W. C., 1965, *Chem. Engng Sci.* **20**, 859.
- Mazzone, D., 1985, Ph.D. dissertation, The City College of the City University of New York.
- Mazzone, D., Tardos, G. I. and Pfeffer, R., 1986, *J. Colloid Interface Sci.* **113**, 544.
- Mazzone, D., Pfeffer, R. and Tardos, G. I., 1987, *Powder Technol.* **51**, 71.
- Mehrota, V. P. and Sastry, K. V. S., 1980, *Powder Technol.* **25**, 203.
- Park, C. W. and Homsy, G. M., 1984, *J. Fluid Mech.* **139**, 291.
- Rumpf, H., 1962, in *Agglomeration* (Edited by W. A. Knepper), p. 379., Wiley, New York.
- Schubert, H., 1977, in *Agglomeration 77* (Edited by K. V. S. Sastry), p. 144. AIME.
- Siegell, J. H., 1976, Ph.D. dissertation, The City College of the City University of New York.
- Smith, P. G., 1980, Ph.D. thesis, University of London.
- Stokes, G. G., 1851, *Trans. Camb. phil. Soc.* **9**, 8.