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MOTIONS OF A FLUID DROP IN LINEAR AND QUADRATIC FLOWS

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Abstract—A theoretical analysis is presented of the flow field near a spherical fluid drop immersed in an incompressible Newtonian fluid which, at large distances from the drop, is undergoing an undisturbed flow. The undisturbed flows considered here are relevant to studies of drop motions near a phase boundary, and to some aspects of the coalescence of liquid drops. Exact solutions in closed form have been found using the harmonic function expansion in spherical coordinates. Calculation of the hydrodynamic force on the drop leads to a generalization of Faxen's law to a *fluid* particle in an *arbitrary* undisturbed creeping-flow. The solutions are then expressed in terms of the fundamental singularity solutions for Stokes flow in anticipation of future analysis of the drop coalescence. In addition, the deformed shapes are determined for a fluid drop freely suspended in an axisymmetric Poiseuillian flow.

INTRODUCTION

The present study is concerned with the dynamics of a droplet immersed in an immiscible fluid which, at large distances from the drop, undergoes an undisturbed flow. A number of different problems are of potential interest, corresponding to various types of application. Specifically, the translation of a fluid drop in a quiescent fluid near a phase boundary is relevant to coalescence of liquid drops. Drop motions in a general flow field are relevant to studies of suspension rheology, erythrocyte motion in capillary blood flow, and to some aspects of gel permeation chromatography, [1]. Another area of potential applications is to the formation of emulsions where one fluid phase is to be dispersed throughout a second, and in particular to the determination of the emulsification mechanisms in colloid mills and to the design of efficient mixing devices, [2,3].

When a fluid drop is suspended in a second fluid that is caused to shear, the flow-induced stress tends to deform the drop, and the interfacial tension between the phases resists this deformation. If the local shear rate is sufficiently large compared to the interfacial restoring force, the drop bursts into two or more fragments. Even when the drop does not burst, the distortion produced by a given flow is of interest in understanding the rheological behavior of flowing emulsions. Emulsions are known to exhibit such non-Newtonian characteristics as shear-dependent viscosity, viscoelasticity, and normal stress differences in rectilinear flow, even when the concentration of the dispersed phases is small. From a knowledge of the deformation of the drops forming the dispersed phase and of the disturbance flow in their vicinity, a constitutive equation can be developed, at least in principle, for the emulsion.

The problem has received considerable attention in the fluid mechanics literature over the past fifty years since Taylor's celebrate work on the viscosity of a fluid containing small drops of another fluid [4,5]. From a theoretical point of view, the drop motion problem is extremely difficult. The equation of motion must be solved for the flow both inside and outside the drop, with boundary conditions applied on its surface. However, the shape of the drop is not known, *a priori*, but must be determined as part of the solution. To date, three distinct methods have been commonly employed in studying drop deformation; namely, (1) a domain perturbation technique (i.e., asymptotic analysis) for slightly deformed drops [6-10], (2) a slender-body theory for highly elongated drops [11-14], and (3) a numerical analysis (i.e., boundary-integral method) for selected intermediate cases [15,16]. A plethora of studies, however, has been concerned with the *linear* undisturbed flow. Our particular contribution lies in a systematic investigation of the effect of flow parameters in the *quadratic* imposed-flow. The undisturbed flow considered here are the quadratic paraboloidal and stagnation flows which are essential for the analysis of drop motion near a phase boundary, [17]. The paraboloidal flow with a typical representation corresponds to Hagen-Poiseuille flow, and the solution can be used to determine the motion of a fluid drop through a tube of elliptic cross-section.

The present paper represents an initial study whose purpose is the generalization of previous theoretical work to the case of quadratic undisturbed flow. The analysis is formally carried out by the eigenfunction expansion for Stokes equations in spherical coordinates under the conditions where the drop deformation remains small. The theory determines the drop deformation and the general motion of a freely suspended drop in the prescribed mean flow. Then, the solutions are expressed in terms of the fundamental singularity solutions for Stokes flow for the purpose of future analysis of the drop coalescence near a phase boundary. The novel feature in the analysis is that the types of fundamental singularities needed to represent the solution have the same form (i.e., orientation) as for a *solid*, no-slip sphere except for magnitudes of the necessary singularities that depend on the viscosity ratio. Among the most interesting results is a generalization of Faxen's law to a *fluid* particle. According to the generalized Faxen's law, the translational velocity of a sphere freely suspended in an *arbitrary* undisturbed flow changes from the surface average (Faxen's) velocity to the local velocity of the primary flow at the drop center in the transition from a solid, no-slip sphere to an inviscid gas bubble.

PROBLEM STATEMENT

We consider a neutrally buoyant spherical drop suspended in an incompressible Newtonian fluid which is undergoing an undisturbed flow $\mathbf{U}^{\infty}(\mathbf{x})$ at infinity, as indicated in Figure 1. The interface between the two immiscible fluids 1 and 2 is assumed to be clean, mobile, and characterized completely by constant interfacial tension γ. The analysis which we consider is predicated on the neglect of inertia effects in the fluid both outside and inside the drop. Let a be the characteristic drop radius, and u_c the scaling of the undisturbed flow. Furthermore, define the Reynolds number, $\mathrm{Re}=au_c/\nu_2$, where ν_2 is the kinematic viscosity of fluid 2 outside the drop. Viewing the problem as a fixed laboratory observer, and requring

$$\mathrm{Re} \ll O(1), \tag{1}$$

the governing equations are approximated by the familiar Stokes equation plus the continuity equation in each fluid, i.e., in dimensionless form,

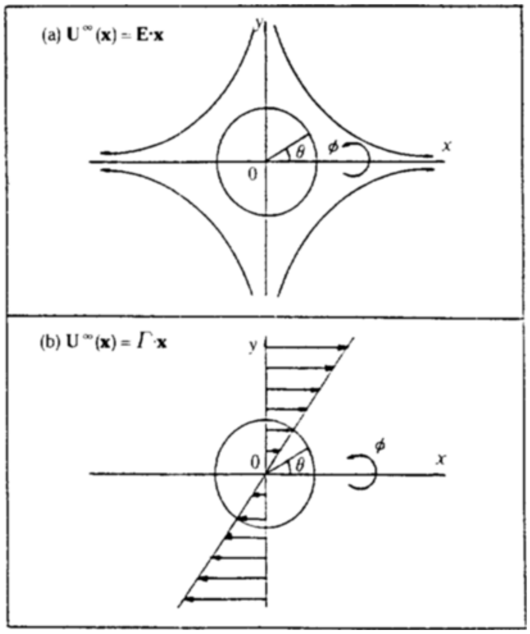


Fig. 1. Schematic sketch for (a) a uniaxial extensional flow and (b) a linear shear flow. The instantaneous coordinate of the drop center is x=O.

$$\nabla\cdot\tilde{\sigma}=-\nabla\tilde{p}+\varkappa\nabla^2\tilde{\mathbf{u}}=0,\ \nabla\cdot\tilde{\mathbf{u}}=0 \quad \text{for fluid 1} \tag{2}$$

$$\nabla\cdot\sigma=-\nabla p+\nabla^2\mathbf{u}=0,\ \nabla\cdot\mathbf{u}=0 \quad \text{for fluid 2} \tag{3}$$

in which σ (or $\tilde{\sigma}$) is the dimensionless stress tensor with the characteristic stress taken as

$$p_c=\frac{\mu_2 u_c}{a}$$

and $\varkappa$ is the viscosity ratio, i.e., $\varkappa=\mu_1/\mu_2$. The boundary condition far from the drop is

$$\mathbf{u}\to\mathbf{U}^{\infty}(\mathbf{x}),\quad p\to p^{\infty}(\mathbf{x}) \quad \text{as } |\mathbf{x}|\to\infty \tag{4}$$

in a laboratory frame of reference. On the interface separating fluids 1 and 2, $\mathbf{x}\in S$, we require

$$[|\mathbf{u}|]_s=0 \tag{5}$$

$$[|\mathbf{n}\cdot\sigma|]_s=\frac{1}{Ca}(\nabla\cdot\mathbf{n})\,\mathbf{n} \tag{6}$$

$$\mathbf{n}\cdot\mathbf{u}=\mathbf{n}\cdot\tilde{\mathbf{u}}=\frac{1}{|\nabla S|}\frac{\partial f}{\partial t} \tag{7}$$

where the symbol $[|\cdot|]_S$ denotes the jump in the bracketed quantity across S. In these equations, the drop surface S is conveniently specified using a spherical coordinate system, defined as $S: r-1-f(\theta,\phi,t)=0$. The vector $\mathbf{n}$ is the unit normal into fluid 2 at the interface, i.e., $\mathbf{n} = \nabla S/|\nabla S|$ and $\nabla \cdot \mathbf{n}$ is the surface curvature.

Equation (6) is the surface stress condition, and contains both continuity of tangential stress, and the normal stress balance between viscous and pressure stresses and capillary force. The parameter $Ca(=\mu u_c/\gamma)$ is the capillary number, i.e., the ratio of the deforming viscous force to the restoring surface tension force. Equation (7) is the kinematic condition which relates the normal velocity components at the drop surface to the rate of change of the drop shape. The equations and boundary conditions (2)-(7) are sufficient to completely determine the velocity and pressure fields in fluids 1 and 2, as well as the drop shape.

SOLUTION METHODOLOGY

Now, we have seen that the problem represented by (2)-(7) is both nonlinear, and unsteady due to the boundary conditions (6) and (7). Thus, the solutions for motion of a drop will depend on the prior history of the imposed flow it has experienced. Although the nonlinear drop deformation problem cannot be solved exactly (except by numerical methods), it can be solved approximately by an asymptotic method when the drop deformation remains small. The obvious physical requirement for this condition to be satisfied is that

$$Ca \ll O(1). \tag{8}$$

It is important to recognize that the capillary number Ca can be viewed as the ratio of the surface tension relaxation time scale, $\mu a/\gamma$, relative to the advection time scale, a/u_c, of the imposed flow. When the condition (8) is satisfied, the drop deformation will not only be in a *quasi-steady* (i.e., $\frac{\partial f}{\partial t}=0$), but the magnitude of the deformation will also be asymptotically small.

Since for a nearly spherical drop shape the boundary conditions can be extrapolated onto a sphere, the flow fields inside and outside the drop can be determined as a regular perturbation expansion, and hence the evolution of the distortion can be predicted unitl such time as it ceases to be small. The leading order approximation [i.e., for $f(\theta, \phi)$] thus represents the motion of a *spherical* drop immersed in the prescribed flow. When the velocity and stress fields have been determined from the leading order approximation equations, the normal stress condition (6) can be used to determine a first correction to the drop shape.

The most frequently used technique for the leading order problem is the use of eigensolutions of Laplace's equation in spherical coordinates. Lamb [18] derived a general solution of the creeping motion equations in a series of solid spherical harmonics. Specializing the general solution to separate domains involving the regions inside and outside the drop, we must have

$$p(\mathbf{x}) = p^\infty(\mathbf{x}) + \sum_{n=1}^{\infty} \frac{p_n}{r^{2n+1}}, \quad r = |\mathbf{x}| \tag{9}$$

$$\mathbf{u}(\mathbf{x}) = \mathbf{U}^\infty(\mathbf{x}) + \sum_{n=1}^{\infty} \left[\frac{\nabla\chi_n \times \mathbf{r}}{r^{2n+1}} + \nabla\frac{\phi_n}{r^{2n+1}} - \frac{n-2}{2n(2n-1)} r^2 \nabla \frac{p_n}{r^{2n+1}} + \frac{n+1}{n(2n-1)} \mathbf{r} \frac{p_n}{r^{2n+1}}\right] \tag{10}$$

for $r>1$, and

$$\tilde{p}(\mathbf{x}) = \sum_{n=1}^{\infty} \tilde{p}_n \tag{11}$$

$$\tilde{\mathbf{u}}(\mathbf{x}) = \sum_{n=1}^{\infty} \left[\nabla\tilde{\chi}_n \times \mathbf{r} + \nabla\tilde{\phi}_n + \frac{(n+3)r^2}{2\kappa(n+1)(2n+3)} \nabla\tilde{p}_n - \frac{n\mathbf{r}\tilde{p}_n}{\kappa(n+1)(2n+3)}\right] \tag{12}$$

for $r<1$. Here, p_n, χ_n, ϕ_n, $\tilde{p}_n$, $\tilde{\chi}_n$ and $\tilde{\phi}_n$ are the solid spherical harmonics of order n. It should be noted that the general solutions (9)-(12) automatically satisfy the governing differential equations (2) and (3), as well as the condition (4) of vanishing disturbances in the far field. The various spherical harmonics are to be determined from the boundary conditions (5) and (6) at the drop surface, i.e., continuity of tangential velocity and stress and zero normal velocity. All that is required for doing this is a specification of the undisturbed flow velocity $\mathbf{U}^\infty(\mathbf{x})$ and pressure $p^\infty(\mathbf{x})$ in terms of spherical harmonics plus a solution of the algebraic relationships that result from applying the boundary conditions (5) and (6) at the spherical drop surface.

In the analysis which follows, we shall use the general solution (9)-(12) to examine the case of a drop which moves through various undisturbed Stokes flows in an *unbounded* domain. The solution for the flow *outside* the drop will then be expressed in terms of the fundamental solutions of the creeping motion equations, and these results used in the forthcoming part of the present series to study drop motions near a phase boundary, as a simple model of 'coalescence'. In addition, it will be shown that a generalization of Faxen's law can be obtained to calculate the resistance of a drop suspended in an *arbitrary* undisturbed Stokes flow. Finally, the deformed shapes will be determined for a fluid drop freely suspended in an axisymmetric paraboloidal (Poiseuillian) flow.

UNIFORM STREAMING AND LINEAR FLOWS

Let us then begin by considering the case of a fluid drop immersed in a uniform streaming, linear shear or uniaxial extentional flow of an unbounded fluid, as depicted in Figure 1. Although a number of the linear-flow cases for a spherical drop in an unbounded domain have previously been solved elsewhere, by other methods, the solutions as their expression in terms of a superposition of fundamental singularities are a necessary preliminary to the use of reflections procedure for studying drop motions near a boundary [17,19].

For the case of a uniform streaming flow $\mathbf{U}^{\infty}(\mathbf{x}) = \mathbf{e}_x$, in an infinite fluid domain with no external boundaries, an exact solution for a fluid drop is the Hadamard-Rybczynski solution [20]. The velocity field outside the fluid drop in this solution can be represented by superposition of the fundamental solutions for a point force (i.e., Stokeslet) and a potential dipole, both applied at the drop center:

$$\text{Stokeslet}: \frac{3}{4}\frac{2/3+\kappa}{1+\kappa}\mathbf{u}_S(\mathbf{x};\mathbf{e}_x) \tag{13a}$$

$$\text{Potential Dipole}: -\frac{1}{4}\frac{\kappa}{1+\kappa}\mathbf{u}_D(\mathbf{x};\mathbf{e}_x) \tag{13b}$$

where κ is the viscosity ratio of the fluid drop relative to the suspending fluid. The fundamental solutions $\mathbf{u}_S$ and $\mathbf{u}_D$ for a Stokeslet $\boldsymbol{\alpha}$ and a potential dipole β are given by

$$\mathbf{u}_S(\mathbf{x};\boldsymbol{\alpha}) = \frac{\boldsymbol{\alpha}}{r} + \frac{(\boldsymbol{\alpha}\cdot\mathbf{x})\,\mathbf{x}}{r^3} \tag{14a}$$

and

$$\mathbf{u}_D(\mathbf{x};\beta) = -\frac{\beta}{r^3} + \frac{3(\beta\cdot\mathbf{x})\,\mathbf{x}}{r^5}. \tag{14b}$$

The most interesting feature of solution (13a,b) is that it is a superposition of precisely the same singularities as are needed for a rigid, *solid* sphere in the same flow. Indeed, as $\kappa\rightarrow\infty$, equations (13a,b) reduce to the velocity field for the case of a rigid, no-slip sphere, and is identical with the flow generated by the singularities, $\alpha = \frac{3}{4}\mathbf{e}_x$ and $\beta = -\frac{1}{4}\mathbf{e}_x$ at the origin. Thus, in spite of the fact that the boundary conditions at the drop surface are quite different from the solid sphere-i.e., continuity of tangential velocity and stress and zero normal velocity are required instead of the no-slip condition-it is only magnitude of the necessary singularities that changes rather than the type of singularities in the transition from a solid to fluid sphere.

It is also straightforward to solve for the motion of a fluid drop in an unbounded domain that is undergoing various linear undisturbed flow. We begin by considering the simplest case of an extensional flow

$$\mathbf{U}^{\infty}(\mathbf{x}) = \mathbf{E}\cdot\mathbf{x}$$

where the strain rate tensor $\mathbf{E} = \{E_{ij}\}$ is defined by $E_{ij} = 3\delta_{i1}\delta_{j1} - \delta_{ij}$. Note that $\mathbf{E}$ has been nondimensionalized with respect to the mean strain rate E (i.e., $u_c = Ea$). Expanding $\mathbf{U}^{\infty}(\mathbf{x})$ and $p^{\infty}(\mathbf{x})$ in terms of spherical harmonics, it can be easily shown that the exact solution for the velocity field *exterior* to a drop is equivalent to that generated by a stresslet and a potential quadrupole at the drop center, of the form:

$$\text{Stresslet}: -\frac{5}{2}\frac{2/5+\kappa}{1+\kappa}\mathbf{u}_{SS}(\mathbf{x};\mathbf{e}_x,\mathbf{e}_x) \tag{15a}$$

$$\text{Potential Quadrupole}: -\frac{1}{2}\frac{\kappa}{1+\kappa}\mathbf{u}_{PQ}(\mathbf{x};\mathbf{e}_x,\mathbf{e}_x) \tag{15b}$$

where the fundamental solutions, $\mathbf{u}_{SS}$ and $\mathbf{u}_{PQ}$, for a stresslet (γ,δ) and a potential quadrupole (ν,ζ) are given by

$$\mathbf{u}_{SS}(\mathbf{x};\gamma,\delta) = -\left[\frac{\gamma\cdot\delta}{r^3} - \frac{3(\gamma\cdot\mathbf{x})(\delta\cdot\mathbf{x})}{r^5}\right]\mathbf{x} \tag{16a}$$

and

$$\mathbf{u}_{PQ}(\mathbf{x};\nu,\zeta) = \zeta\cdot\nabla\mathbf{u}_D(\mathbf{x};\nu). \tag{16b}$$

Again the remarkable fact is that the singularities required to satisfy boundary conditions at the surface of a drop are the same as required for a no-slip sphere. It is only the magnitude of the coefficients that depends on the viscosity ratio, i.e., κ. Of course, the ratio of stresslet to potential quadrupole strength is not the same as for a solid sphere, except in the limit $\kappa\rightarrow\infty$ when the present solution for the velocity field exterior to the sphere reduces to

$$\mathbf{u}(\mathbf{x}) = \mathbf{U}^{\infty}(\mathbf{x}) - \frac{5}{2}\mathbf{u}_{SS}(\mathbf{x};\mathbf{e}_x,\mathbf{e}_x) - \frac{1}{2}\mathbf{u}_{PQ}(\mathbf{x};\mathbf{e}_x,\mathbf{e}_x)$$

which is identical with Chwang and Wu's result for the case of a solid sphere [21].

Another linear flow problem that we need for study of drop motion near a phase boundary is the steady simple shear flow past a neutrally buoyant drop [19]. In this problem, the fluid velocity at infinity, nondimensionalized with respect to $u_c = \Gamma a$ (Γ: shear rate), is

$$\mathbf{U}^{\infty}(\mathbf{x}) = y\mathbf{e}_x.$$

Since a simple shear flow can be represented as a superposition of a plane extensional flow and a rigid body rotation, we can easily determine a complete solution by superposition of the preceding results of (15a,b) and the rigid body rotation. The singularities required for construction of the solution *exterior* to the drop, apart from the rigid body rotation and primary flow, are a stresslet and a potential quadrupole of the form:

$$\text{Stresslet}: -\frac{5}{6}\frac{2/5+\varkappa}{1+\varkappa}\mathbf{u}_{SS}(\mathbf{x};\mathbf{e}_x,\mathbf{e}_y) \quad (17a)$$

$$\text{Potential Quadrupole}: -\frac{1}{6}\frac{\varkappa}{1+\varkappa}\mathbf{u}_{PQ}(\mathbf{x};\mathbf{e}_x,\mathbf{e}_y). \quad (17b)$$

The present solution, (17a,b), is identical to that obtained by Taylor [4] who investigated the viscosity of a fluid containing small drops of another fluid. Again, it is noteworthy that the same fundamental singularities apply for the fluid drop, as for the solid sphere, though their ratio of magnitudes reduces to the no-slip limit only for $\varkappa \to \infty$.

QUADRATIC PARABOLOIDAL AND STAGNATION FLOWS

We now consider various quadratic flows that will be necessary for solution of the problem of drop motion near a phase boundary The first case is a flow with a paraboloidal velocity profile.

$$\mathbf{U}^\infty(\mathbf{x}) = (\xi y^2 + z^2)\mathbf{e}_x,\ p^\infty(\mathbf{x}) = 2(\xi+1)x. \quad (18a, b)$$

The spherical drop is again assumed to be centered at the origin, see Figure 2 (in this case $u_c = Ka^2$, $p_c = \mu Ka$, with proportionality constant K). The form of the paraboloidal flow depends upon the value of the parameter ξ. When $\xi = 0$, the paraboloidal flow degenerates into a 2-dimensional Poiseuille flow. For $\xi > 0$, it can be interpreted as the pressure-driven flow through a tube of elliptic cross-section. The case $\xi < 0$ is primarily of interest as a local component of a more complicated flow.

Let us first consider the simple case of an axisymmetric paraboloidal flow with $\xi = 1$. In this case, the solution must be independent of the azimuthal angle ϕ. Thus, the only nonzero spherical harmonics in the general solution are those with rank zero, and, in addition, $\chi_n = 0$. The remaining spherical harmonics can be determined from the boundary conditions (5) and (6) at the drop surface in combination with the prescribed flow field at infinity that is incorporated into the general solutions, (9)-(12).

The velocity field *exterior* to the drop, corresponding to the exact solution for $\xi = 1$, can again be expressed by a superposition of the fundamental solutions for Stokes flow. The required form, apart from the primary flow, is:

$$\text{Stokeslet}: -\frac{1}{2}\frac{\varkappa}{1+\varkappa}\mathbf{u}_S(\mathbf{x};\mathbf{e}_x) \quad (19a)$$

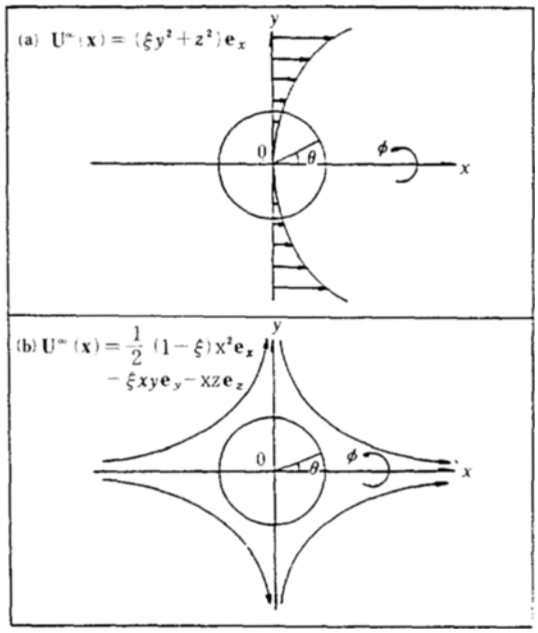


Fig. 2. Schematic sketch for (a) a quadratic paraboloidal flow, and (b) a quadratic stagnation flow.

$$\text{Potential Dipole}: -\frac{1}{12}\frac{2-5\varkappa}{1+\varkappa}\mathbf{u}_D(\mathbf{x};\mathbf{e}_x) \quad (19b)$$

$$\text{Stokes Quadrupole}: \frac{1}{12}\frac{2+7\varkappa}{1+\varkappa}\frac{\partial^2}{\partial x^2}\mathbf{u}_S(\mathbf{x};\mathbf{e}_x) \quad (19c)$$

$$\text{Potential Octupole}: -\frac{1}{24}\frac{\varkappa}{1+\varkappa}\frac{\partial^2}{\partial x^2}\mathbf{u}_D(\mathbf{x};\mathbf{e}_x). \quad (19d)$$

As suggested by the variable velocity gradient of the primary flow we also require an axial Stokes quadrupole $\frac{\partial^2}{\partial x^2}\mathbf{u}_S(\mathbf{x};\mathbf{e}_x)$ and a potential octupole $\frac{\partial^2}{\partial x^2}\mathbf{u}_D(\mathbf{x};\mathbf{e}_x)$ that is associated with the Stokes quadrupole to balance the power-law variations of the solution in r. It is again noteworthy that the singularities required in (19) for a drop are identical to those determined by Chwang and Wu [21] for rigid, solid sphere, with the coefficients reducing to the solid sphere values for $\varkappa \to \infty$. The drag on the drop comes solely from the contribution of the Stokeslet:

$$\mathbf{F} = 4\pi\frac{\varkappa}{1+\varkappa}\mathbf{e}_x \quad (20)$$

(the dimensional drag is $\mathbf{F}$ multiplied by μKa^3). As expected, the drag is an increasing function of the viscosity ratio $\varkappa$. Indeed, the drag becomes zero for an inviscid gas bubble (i.e., $\varkappa = 0$).

To construct an exact solution for the more general paraboloidal flow, (18) with $\xi \neq 1$, we need to determine a solution either for

$$\mathbf{U}^\infty(\mathbf{x}) = y^2\mathbf{e}_x \tag{21a}$$

or for

$$\mathbf{U}^\infty(\mathbf{x}) = z^2\mathbf{e}_x. \tag{21b}$$

Any general paraboloidal flow, (18), with $\xi \neq 1$, can then be constructed by superposition owing to the linearity of the problem. Moreover, the solution for (21a) [or (21b)] can be easily obtained by decomposing the exact solution (19) just obtained for $\mathbf{U}^\infty(\mathbf{x}) = (y^2 + z^2)\mathbf{e}_x$ into two *symmetric* parts corresponding to the 2-dimensional paraboloidal flows of (21a,b), see Yang& Leal [22]. The total hydrodynamic force acting on the spherical drop in the primary flow, (18), for arbitrary ξ can be obtained by superposition:

$$\mathbf{F} = 2\pi(1+\zeta)\frac{\kappa}{1+\kappa}\mathbf{e}_x. \tag{22}$$

Te torque $\mathbf{T}$ is obviously zero for arbitrary ξ.

Finally, we consider a quadratic stagnation flow with a velocity profile

$$\mathbf{U}^\infty(\mathbf{x}) = \frac{1}{2}(1+\xi)x^2\mathbf{e}_x - \xi xy\mathbf{e}_y - xz\mathbf{e}_z \tag{23a}$$

which obviously satisfies the creeping motion equations if the pressure associated with it is

$$p^\infty(\mathbf{x}) = (1+\xi)x. \tag{23b}$$

The stagnation plane is $x = 0$, as depicted in Figure 2. Although this type of quadratic flow may be of some intrinsic interest as a local component of a more complicated flow in an *unbounded* domain, it is primarily of interest in determining the motion of a particle near a phase boundary.

Let us consider, first, the simple case of the axisymmetric stagnation flow, (23) with $\xi = 1$. In this case, the exact solution for the flow fields exterior and interior to the drop involves the nonzero spherical harmonics, p_n and ϕ_n with $n = 1$, 3 and rank zero, in the general solution (9)-(12). Not surprisingly, in view of previous examples, the flow field exterior to the spherical drop can be expressed in terms of fundamental solutions of Stokes' equations. In particular, the exterior velocity associated with the spherical harmonics p_n and ϕ_n can be represented by a Stokeslet (required to produce a drag), a potential dipole (associated with the Stokeslet to account for the body-thickness effect), an axial Stokes quadrupole [as suggested by the variable velocity gradient of $\mathbf{U}^\infty(\mathbf{x})$] and a potential octupole (associated with the Stokes quadrupole):

$$\text{Stokeslet}: -\frac{1}{4}\frac{\kappa}{1+\kappa}\mathbf{u}_S(\mathbf{x};\mathbf{e}_x) \tag{24a}$$

$$\text{Potential Dipole}: -\frac{1}{12}\frac{2+\kappa}{1+\kappa}\mathbf{u}_D(\mathbf{x};\mathbf{e}_x) \tag{24b}$$

$$\text{Stokes Quadrupole}: -\frac{1}{12}\frac{2+7\kappa}{1+\kappa}\frac{\partial^2}{\partial x^2}\mathbf{u}_S(\mathbf{x};\mathbf{e}_x) \tag{24c}$$

$$\text{Potential Octupole}: \frac{1}{12}\frac{\kappa}{1+\kappa}\frac{\partial^2}{\partial x^2}\mathbf{u}_D(\mathbf{x};\mathbf{e}_x). \tag{24d}$$

The total hydrodynamic force on the drop is evaluated from the Stokeslet contribution:

$$\boldsymbol{F} = 2\pi\frac{\kappa}{1+\kappa}\mathbf{e}_x \tag{25}$$

which reduces in the limiting case of a solid sphere (i.e., $\kappa \to \infty$) to $\lim_{\kappa\to\infty}\mathbf{F} = 2\pi\mathbf{e}_x$.

Now, we consider the more general quadratic flow, (23) with $\xi \neq 1$. The solution exterior to the spherical drop is analogous to that for $\mathbf{U}^\infty(\mathbf{x}) = (\xi y^2 + z^2)\mathbf{e}_x$ in the previous example. In view of the linearity of the problem, it is sufficient to solve for the primary flow

$$\mathbf{U}^\infty(\mathbf{x}) = \frac{1}{2}x^2\mathbf{e}_x - xy\mathbf{e}_y \tag{26a}$$

or

$$\mathbf{U}^\infty(\mathbf{x}) = \frac{1}{2}x^2\mathbf{e}_x - xz\mathbf{e}_z \tag{26b}$$

in order to construct the exact solution for $\mathbf{U}^\infty(\mathbf{x})$ given by (23) with arbitrary ξ. However, if we note that the primary flow, (23) with $\xi = 1$, consists of two symmetric components of the type (26a,b), then decomposing the solution (24) into the two parts we can easily determine the velocity field for each component flow.

The resulting hydrodynamic force acting on the drop immersed in the primary flow, with $\mathbf{U}^\infty(\mathbf{x})$ given by (23), is thus

$$\mathbf{F} = \pi(1+\xi)\frac{\kappa}{1+\kappa}\mathbf{e}_x. \tag{27}$$

When $\kappa \to \infty$, Equation (27) reduces to the drag for the case of a solid, no-slip sphere, and is identical with the result of Chwang [23] for $\xi = 0$. It is noteworthy that a freely suspended drop at the stagnation point of the primary flow will translate with velocity $\mathbf{U} = \frac{1+\xi}{2}\frac{\kappa}{2+3\kappa}\mathbf{e}_x$ without applying a negative force $-\pi(1+\xi)\frac{\kappa}{1+\kappa}\mathbf{e}_x$. The induced translational velocity becomes zero as $\kappa \to 0$, so that an inviscid gas bubble will stay at the stagnation point at all times.

GENERALIZED FAXEN's LAW

Let us then turn to the general problem of a spherical drop immersed in an *arbitrary* undisturbed flow field $\{\mathbf{U}^\infty(\mathbf{x}), p^\infty(\mathbf{x})\}$ which itself satisfies the creeping motion equations. Following the preceding analysis, this problem may be solved directly by a specification of the various unknown spherical harmonics from the boundary conditions at the sphere surface, i.e., the continuity of tangential velocity and stress and zero-normal velocity. However, if we wish only to calculate the hydrodynamic force on a sphere (solid or fluid), and not the velocity field itself, it is possible to do so by evaluating only a small number of spherical harmonics as a consequence of the integral theorem for the spherical harmonics [20].

A general expression for the hydrodynamic force exerted on a particle of arbitrary shape can be derived by integrating the surface force $\mathbf{n}\cdot\sigma$ over a circumscribed sphere in the fluid:

$$\mathbf{F} = -4\pi\nabla p_1. \tag{28}$$

Thus, the hydrodynamic force on any spherical particle can be evaluated by determining the spherical harmonic p_1 from the boundary conditions at the sphere surface. Adopting the general method outlined by Brenner [24], we can determine directly this harmonic p_1 by utilizing the orthogonal properties and mean-value theorem of the spherical harmonics and the vector identities of $\nabla^4\mathbf{U}^\infty = \nabla^6\mathbf{U}^\infty = \mathbf{O}$ etc, in the creeping flow. The result for a spherical drop is simply given by

$$p_1 = -\left[\frac{3}{2}\frac{2/3+\kappa}{1+\kappa}\mathbf{U}^\infty(\mathbf{O}) + \frac{1}{4}\frac{\kappa}{1+\kappa}\nabla^2\mathbf{U}^\infty(\mathbf{O})\right]\cdot\mathbf{x}. \tag{29}$$

The linearity of the problem enables us to determine the resistance of a drop which moves with translational velocity $\mathbf{U}$ in an undisturbed flow. Combining (13a) with (29) we have

$$\mathbf{F} = 6\pi\frac{2/3+\kappa}{1+\kappa}\{\mathbf{U}^\infty(\mathbf{O}) - \mathbf{U}\} + \pi\frac{\kappa}{1+\kappa}\nabla^2\mathbf{U}^\infty(\mathbf{O}). \tag{30}$$

This result is a generalization of Faxen's law to a spherical drop immersed in an *arbitrary* undisturbed flow. It is a simple matter to reproduce Faxen's law by taking limit $\kappa\to\infty$ in the solution (30), i.e., $\mathbf{F} = 6\pi\{\mathbf{U}^\infty(\mathbf{O}) - \mathbf{U}\} + \pi\nabla^2\mathbf{U}^\infty(\mathbf{O})$ as $\kappa\to\infty$.

From the generalized Faxen's law, (30), we can evaluate the translational velocity $\mathbf{U}$ of a *freely* suspended neutrally buoyant drop in an *arbitrary* mean flow $\mathbf{U}^\infty(\mathbf{x})$:

$$\mathbf{U} = \langle\mathbf{U}^\infty\rangle_S - \frac{1}{3}\frac{1}{2+3\kappa}\nabla^2\mathbf{U}^\infty(\mathbf{O}) \tag{31a}$$

where the surface average of the primary flow $\langle\mathbf{U}^\infty\rangle_S$ is given by

$$\langle\mathbf{U}^\infty\rangle_S = \mathbf{U}^\infty(\mathbf{O}) + \frac{1}{6}\nabla^2\mathbf{U}^\infty(\mathbf{O}). \tag{31b}$$

It is of interest to note that the translational velocity (31a) is different from the surface average Faxen's velocity of the primary flow, which would be the result for a solid, no-slip sphere according to Faxen's law. Indeed, as $\kappa\to 0$, the translational velocity becomes the same as the local velocity of the primary flow at the drop center, $\mathbf{U} = \mathbf{U}^\infty(\mathbf{O})$. As an example of application, we determine the trajectory for a spherical drop freely suspended in an off-centered paraboloidal flow, $\mathbf{U}^\infty(\mathbf{x}) = \{\xi(y-y_0)^2 + (z-z_0)^2\}\mathbf{e}_x$, that is equivalent to a centered one (i.e., with $y_0 = z_0 = 0$) superimposed on a uniform streaming flow plus linear shear flows. The result is

$$\mathbf{U} = \left[\xi y_0^2 + z_0^2 - \frac{1+\xi}{3}\frac{\kappa}{2/3+\kappa}\right]\mathbf{e}_x. \tag{32}$$

The trajectory equation (32) are relevant to the problem of a spherical drop freely suspended at an arbitrary point in Poiseuillian flow through a cylindrical tube of elliptic cross-section.

DROP DEFORMATION

When a drop moves through a viscous fluid, the fluid in the neighborhood of the drop is disturbed. The disturbance generates a stress system which can be resolved into tangential and normal stresses acting at the drop surface. The tangential stress is assumed to be transmitted undiminished across the interface and thus establishes a system of velocity gradient in the vicinity of the interface. The normal stress, on the other hand, is discontinuous at the interface, and generates normal stress differences across the interface which can be balanced by capillary forces through interface deformation. The leading-order solutions obtained in the preceding sections for a *spherical* drop satisfy the conditions of continuity of the tangential velocity and stress at the undeformed interface, as well as zero-normal velocity. However, they do produce an imbalance in the normal stress components across the drop surface. Thus, to calculate a first correction to the drop shape, it is necessary to solve the differential equations (6) with the normal stress difference $[|\sigma\cdot\mathbf{n}|]_S$ evaluated from the leading-order solution.

As a simple illustration, consider the problem of a fluid drop freely suspended in a circular cylindrical tube through which a viscous fluid is moving axially. We suppose that the cylinder radius, R, is much larger than that of the drop (i.e., $\frac{1}{R}\ll 1$) so that the hydro-

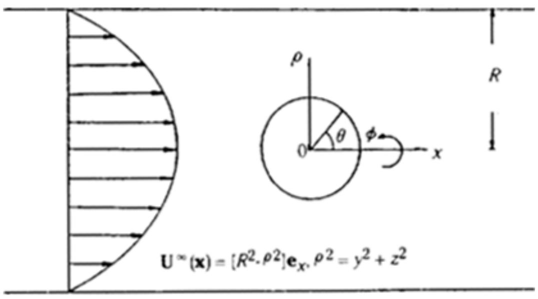


Fig. 3. Schematic sketch of a drop freely suspended in a Poiseuille flow.

dynamic wall-effect may be negligible. The cylinder axis is taken to be the x-axis, at which the drop center is situated, see Figure 3. At large distances from the drop the undisturbed flow is a quadratic paraboloidal (i.e., Poiseuillian) flow. Thus we set

$$\mathbf{U}^\infty(\mathbf{x}) = [R^2 - \rho^2]\mathbf{e}_x,\ \rho^2 = y^2 + z^2. \tag{33}$$

According to the generalized Faxen's law, the drop will move with a constant velocity

$$\mathbf{U} = [R^2 - \frac{2\kappa}{2+3\kappa}]\mathbf{e}_x$$

parallel to the axis, whereas the superficial flow of fluid occurs in the same direction with a mean velocity of $R^2/2\ \mathbf{e}_x$. Note that the velocity is nondimensionalized with respect to $u_c = Ka^2$, and K is a Poiseuille-flow parameter.

The normal stress difference $[|\sigma\cdot\mathbf{n}\cdot\mathbf{n}|]_S$ across the drop surface S can be evaluated by a superposition of the leading-order solutions for a uniform streaming and quadratic paraboloidal flows and expressed in terms of the Legendre polynomial of third order, P_3:

$$[|\sigma\cdot\mathbf{n}\cdot\mathbf{n}|]_S = p^{in} - p^\infty + \frac{1}{2}\frac{10+11\kappa}{1+\kappa}P_3(\eta),\ \eta = \cos\theta \tag{34}$$

where θ is the spherical polar angle measured from the x-axis, that is the axis of symmetry. In (34), p^{in} denotes the pressure at the interface inside the drop phase and p^∞ is the reference pressure far from the drop. This pressure difference, p^{in}-p^∞, is precisely balanced by interfacial tension for the drop in its undisturbed spherical shape (S: r-1 = 0), i.e., p^{in}-p^∞ $= \frac{2}{Ca}$. It is thus obvious that the first correction to the drop shape, $f(\theta, \phi)$, is independent of the azimuthal angle ϕ owing to the axisymmetry of the problem. The differential equation for the shape function $f(\theta)$ follows directly by substitution of (34) into (6), noting that $\mathbf{n} = \nabla S / |\nabla S|$, so that

$$\frac{d}{d\eta}\{(1-\eta^2)\frac{df}{d\eta}\} + 2f = -\frac{Ca}{2}\frac{10+11\kappa}{1+\kappa}P_3(\eta) \tag{35}$$

where $Ca = \frac{\mu K a^2}{\gamma}$. The equation (35) can be solved in terms of the Legendre polynomials subject to the conditions $\int_{-1}^{1} f(\eta)d\eta = 0$ since the characteristic length a has been set equal to the radius of the 'equivalent' spherical drop, and $\int_{-1}^{1} \eta f(\eta)d\eta = 0$ since the origin of the coordinate system has been chosen to coincide with the center of mass of the drop. The resulting solution for the drop shape $S(r,\theta)$ is

$$S(r, \theta) : r - 1 - f(\theta) = 0 \tag{36a}$$

where

$$f(\theta) = \frac{Ca}{2}\frac{1+1.1\kappa}{1+\kappa}P_3(\cos\theta). \tag{36b}$$

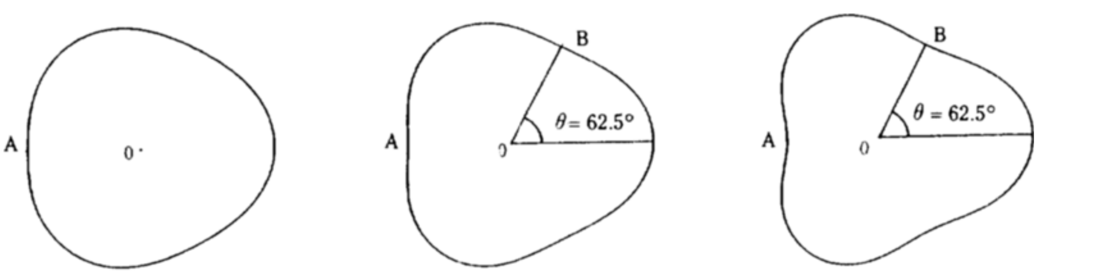
The computation shows that there exist three distinct cases depending on the capillary number, i.e.

Case Ⅰ : $Ca < Ca_I = 0.2857\,\frac{1+\kappa}{1+1.1\kappa}$,

Case Ⅱ : $Ca_I < Ca \le Ca_{II} = 0.3412\,\frac{1+\kappa}{1+1.1\kappa}$,

Case Ⅲ : $Ca_{II} < Ca$

that exhibit different deformation behaviors. The results are shown in Figure 4. In the case I, the surface curvature $\nabla\cdot\mathbf{n}$ is positive everywhere at the drop surface. When the capillary number has a critical value Ca_I the curvature is zero at the front stagnation point



Case I: $Ca = 0.7\ Ca_I$ Case II: $Ca = Ca_{II}$ Case III: $Ca = 1.5\ Ca_{II}$

Fig. 4. Deformed drop shapes corresponding to the three distinct cases I, II and III.

($\theta = 180°$), which is identified by letter A. On the other hand, for case II, the interface in the neighborhood of the stagnation point A becomes dented (i.e., $\nabla \cdot \mathbf{n} < 0$) into the drop (and $\nabla \cdot \mathbf{n} > 0$ elsewhere). As we can see in Figure 4, the waist appears at point B ($\theta = 62.5°$) if the capillary number is larger than the second critical value Ca_{II}. Although the present analysis cannot provide any quantitative informations for *very large* deformation, it can be expected from the result that the drop will deform without limit eventually leading to breakup as the capillary number increases further after the waist appears.

This completes our detailed study of solutions for a fluid drop in an *unbounded* fluid, immersed in a precribed mean flow at infinity. Calculation of the hydrodynamic force exerted on the drop leads to a Faxen-type law for a spherical *fluid* particle in an *arbitrary* undisturbed Stokes flow. These solutions play an important role in determining the general motion of a drop in the vicinity of a phase boundary. The most interesting, and important, feature of the unbounded domain solutions is that the disturbance flow in the fluid can always be expressed as a superposition of the same fundamental singularities at the drop center, as are required for motion of a solid, no-slip sphere in the same flow. Only the relative strengths of these singularities depend upon the viscosity ratio.

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