On the deformation and drag of a falling viscous drop at low Reynolds number

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The motion at low Reynolds number of a drop in a quiescent unbounded fluid is investigated theoretically by means of a singular-perturbation solution of the axisymmetric equations of motion. Special attention is paid to the deformation of the drop. It is shown that for small values of the Weber number We the drop will first deform exactly into an oblate spheroid and then, with a further increase in We, into a geometry approaching that of a spherical cap. These results are quite insensitive to the ratio of the viscosities of the two fluid phases. The first-order effect of the deformation on the drag of the drop is also included in the analysis.

1. Introduction

Interest in the motion of liquid drops and gas bubbles in a fluid medium has existed for many years and has resulted in a number of experimental and theoretical investigations of the laws governing this phenomenon which, from a practical point of view, is of considerable importance in various processes such as extraction and atomization. In most of these studies, the matter of drop deformation was found to require special attention because of the well-known influence which it exerts on the dynamics of such objects. Thus, although 'small' bubbles and drops are always found to be spherical, it has been observed experimentally (Haberman & Morton 1953) that an increase in their size is accompanied by a change in their shape from spherical to ellipsoidal to a spherical cap, with a corresponding effect on their terminal velocity, internal circulation, and stability. It is clear, therefore, that before the subject of drop and bubble dynamics can be placed on a firm basis, it is necessary that the factors which bring about deformation and the influence which this deformation can exert on the macroscopic parameters of the flow, such as the drag, be first quantitatively understood.

It is the purpose of this theoretical study to investigate one aspect of this general topic by restricting the analysis to the low-Reynolds-number range. This, as may be recalled, was attempted by Saito (1913) some years ago, but, in view of a fundamental error in Saito's work, a reinvestigation of this problem seemed desirable because of the valuable information its successful solution would provide.

We begin by considering the motion of a drop in an unbounded, quiescent fluid. Dimensionless variables will be employed throughout the analysis, and physical parameters pertaining to the interior of the drop will be distinguished from the corresponding exterior parameters by a caret. Also, a fixed spherical co-ordinate system will be used with its origin at the centre of mass of the drop.

Of particular importance in this investigation are the boundary conditions at the surface of this drop. These are readily available (Scriven 1960) if the fluids are immiscible, the surface tension is constant, the surface viscosity effects are negligible and axial symmetry is postulated. Thus, if $R = 1+\zeta(\mu)$ is the equation for the surface of the drop, with $\mu = \cos\theta$, it follows that, at $r = R(\mu)$,

$$\tau = \hat{\tau}, \tag{1a}$$

$$N = \hat{N} + \frac{1}{We}\left(\frac{1}{R_1}+\frac{1}{R_2}\right), \tag{1b}$$

$$u_t = \hat{u}_t, \tag{1c}$$

$$u_n = \hat{u}_n = 0, \tag{1d}$$

where τ represent the shear stress, N the normal stress, We the Weber number $(\rho a U^2/\sigma)$, u_t the tangential velocity component, u_n the normal velocity component, σ the interfacial tension, ρ the density of the exterior fluid, a the radius of the 'equivalent' spherical drop, U the terminal velocity of the drop, and R_1 and R_2 the two principal radii of curvature of the drop surface. The above have been made dimensionless by dividing all stresses by ρU^2, all velocities by U and all distances by a.

It is convenient now to express these boundary conditions in terms of the spherical co-ordinates r and μ and the corresponding spherical velocity components u_r and u_θ, with the requirement that $\max|\zeta(\mu)| \ll 1$ since the analysis will deal with drops which depart only slightly from a spherical shape. Then, if

$$\tan\alpha \equiv \frac{1}{R}\frac{dR}{d\theta} \to -(1-\mu^2)^{\frac{1}{2}}\frac{d\zeta}{d\mu} \quad \text{as} \quad \max|\zeta| \to 0,$$

one can show that

$$\tau = (\tau_{rr}-\tau_{\theta\theta})\sin\alpha\cos\alpha + \tau_{r\theta}(\cos^2\alpha - \sin^2\alpha) \to \tau_{r\theta} - (\tau_{rr}-\tau_{\theta\theta})(1-\mu^2)^{\frac{1}{2}}d\zeta/d\mu, \tag{2a}$$

$$N = \tau_{rr}\cos^2\alpha + \tau_{\theta\theta}\sin^2\alpha - 2\tau_{r\theta}\cos\alpha\sin\alpha \to \tau_{rr} + 2\tau_{r\theta}(1-\mu^2)^{\frac{1}{2}}\,d\zeta/d\mu, \tag{2b}$$

$$u_t = u_\theta\cos\alpha + u_r\sin\alpha \to u_\theta - u_r(1-\mu^2)^{\frac{1}{2}}\,d\zeta/d\mu, \tag{2c}$$

$$u_n = u_r\cos\alpha - u_\theta\sin\alpha \to u_r + u_\theta(1-\mu^2)^{\frac{1}{2}}\,d\zeta/d\mu, \tag{2d}$$

while (Landau & Lifschitz 1959), as $|\zeta| \to 0$,

$$\frac{1}{R_1}+\frac{1}{R_2} \to 2 - 2\zeta - \frac{d}{d\mu}\left((1-\mu^2)\frac{d\zeta}{d\mu}\right). \tag{3}$$

Here, τ_{rr}, $\tau_{r\theta}$, and $\tau_{\theta\theta}$ are the conventional components of the stress tensor in spherical co-ordinates (Pai 1956). In the present dimensionless notation, for example, and for incompressible fluids,

$$\tau_{rr} = -p + \frac{2}{Re}\frac{\partial u_r}{\partial r} \quad \text{and} \quad \hat{\tau}_{rr} = -\hat{p} + \frac{2\kappa}{Re}\frac{\partial \hat{u}_r}{\partial r},$$

where $Re = Ua/\nu$ is the Reynolds number and κ the ratio of the viscosity of the interior to that of exterior fluid.

In view of the axial symmetry of the flow, it is possible next to express the equations of motion in terms of the stream functions ψ and $\hat{\psi}$, so that

$$\frac{1}{Re}D^4\psi = \frac{1}{r^2}\frac{\partial(\psi, D^2\psi)}{\partial(r,\mu)} + \frac{2}{r^2}(D^2\psi)\left\{\frac{\mu}{1-\mu^2}\frac{\partial\psi}{\partial r} + \frac{1}{r}\frac{\partial\psi}{\partial\mu}\right\} \tag{4}$$

for the exterior region $R(\mu) < r < \infty$, and

$$\frac{\kappa}{\gamma Re}D^4\hat{\psi} = \frac{1}{r^2}\frac{\partial(\hat{\psi}, D^2\hat{\psi})}{\partial(r,\mu)} + \frac{2}{r^2}(D^2\hat{\psi})\left\{\frac{\mu}{1-\mu^2}\frac{\partial\hat{\psi}}{\partial r} + \frac{1}{r}\frac{\partial\hat{\psi}}{\partial\mu}\right\} \tag{5}$$

for the region inside the drop. Here

$$D^2 \equiv \frac{\partial^2}{\partial r^2} + \frac{1-\mu^2}{r^2}\frac{\partial^2}{\partial\mu^2},$$

$\partial(\psi, D^2\psi)/\partial(r,\mu)$ is the Jacobian of ψ and $D^2\psi$ with respect to r and μ, and γ is the ratio of the density of the interior to that of the exterior fluid. Equations (4) and (5) must now be solved subject to the boundary conditions given by equations (1)–(3), and to the requirement that

$$\psi \to \tfrac{1}{2}r^2(1-\mu^2) \quad \text{as} \quad r \to \infty, \tag{6}$$

which corresponds, at infinity, to the condition of free streaming relative to the centre of mass of the drop.

It should be remarked at this point, however, that equation (1*b*) is not a boundary condition in the strict sense of the word. It is instead an equation for determining $\zeta(\mu)$, and thereby the deformation of the drop, as a function of We, Re, κ and γ, which are the parameters of the system. Of course, since thermodynamic considerations easily lead to the conclusion that $\max|\zeta(\mu)| \to 0$ as $U \to 0$, one might expect $\zeta(\mu)$ to be linear in U for small terminal velocities. And yet, surprisingly enough, the deformation becomes proportioned to U^2, rather than to U, as $U \to 0$. This result, also noted by Saito (1913), will now be discussed in some detail.

2. The creeping flow solution

As is already well known, the solution of equations (4) and (5), with the inertia terms set identically equal to zero everywhere in the flow field, was arrived at some years ago by Hadamard (1911) and by Rybczynski (1911) under the assumption that the drop remained exactly spherical. This solution, denoted by ψ_0 and $\hat{\psi}_0$, for the regions outside and inside the drop respectively, describes the motion correctly only in the limit $Re \to 0$ and is given by

$$\psi_0 = \frac{1-\mu^2}{4}\left[2r^2 - \frac{3\kappa+2}{\kappa+1}r + \frac{\kappa}{\kappa+1}\frac{1}{r}\right] \tag{7a}$$

and

$$\hat{\psi}_0 = -\frac{(1-\mu^2)(r^2-r^4)}{4(\kappa+1)}. \tag{7b}$$

The corresponding pressure field is easily shown to be

$$p_0 = -\frac{ga}{U^2}\mu r - \frac{3\kappa+2}{\kappa+1}\frac{\mu}{2r^2 Re},$$

where g is the gravitational acceleration, if the pressure at $\mu = 0$ and $r \gg 1$ is arbitrarily set equal to zero. Similarly, for the region inside the drop,

$$\hat{p}_0 = -\frac{ga\gamma}{U^2}\mu r + \frac{5\kappa}{\kappa+1}\frac{\mu r}{Re} + \Pi,$$

where Π is an undetermined constant. Now, because of equations (1*b*), (2*b*) and (3)

$$\tau_{rr} - \hat{\tau}_{rr} = \frac{3\mu}{2Re}\frac{3\kappa+2}{\kappa+1} + \frac{ga\mu}{U^2}(1-\gamma) + \Pi = \frac{1}{We}\left\{2 - 2\zeta - \frac{d}{d\mu}\left((1-\mu^2)\frac{d\zeta}{d\mu}\right)\right\}, \tag{8}$$

from which ζ is to be determined subject to the conditions that

$$1 - \frac{1}{2}\int_{-1}^{1}(1+\zeta)^3 d\mu = 0 \quad \text{or} \quad \int_{-1}^{1}\zeta\, d\mu = 0 \quad \text{for} \quad \max|\zeta(\mu)| \ll 1, \tag{9}$$

since the characteristic length a has been set equal to the radius of the 'equivalent' spherical drop, and

$$\int_{-1}^{1}\zeta\mu\, d\mu = 0 \quad \text{for} \quad \max|\zeta(\mu)| \ll 1, \tag{9a}$$

since the origin of the co-ordinate system has been chosen to coincide with the centre of mass of the drop. From an overall force balance, however,

$$ga(1-\gamma)/U^2 = -3F_D/4\pi, \tag{10}$$

where F_D ($\equiv$ drag$/a^2\rho U^2$) is the dimensionless drag on the drop. In our case

$$F_D = \frac{2\pi}{Re}\frac{3\kappa+2}{\kappa+1} \tag{10a}$$

and hence
$$\tau_{rr} - \hat{\tau}_{rr} = \Pi = \frac{1}{We}\left\{2 - 2\zeta - \frac{d}{d\mu}\left((1-\mu^2)\frac{d\zeta}{d\mu}\right)\right\}.$$

From this it clearly follows that

$$\Pi = 2/We \quad \text{and} \quad \zeta(\mu) \equiv 0,$$

if $\zeta(\mu)$ is also to satisfy equations (9) and (9*a*). Thus, as observed by Saito (1913), the drop will remain spherical for all values of the Weber number, as long as the inertia terms of the equations of motion can be safely neglected in the flow field both inside and outside the drop.

3. Inertial effects

It is apparent from the above results that deformation is caused by inertia effects, and that these need to be included in our analysis. At first, it might seem logical to attack equations (4) and (5) by means of an iterative procedure in which the inertia terms would be obtained from the lower order approximations, but, since it is known that in problems of uniform streaming at infinity the creeping flow solution does not provide a uniform approximation to all the required properties of the flow, one should not expect such a straightforward perturbation expansion to generate a valid result. Thus, in view of the fact that Saito's (1913) analysis is based on precisely such a classical perturbation technique, it is not surprising that his solution is incorrect and that it does not satisfy the boundary condition of uniform streaming at infinity.

Fortunately, a correct way of overcoming this difficulty has been described recently by Proudman & Pearson (1957), who applied the method of singular perturbation expansions (see also Kaplun & Lagerstrom 1957) to the classical problem of improving the Stokes solution for the flow past a solid sphere, a problem which is clearly similar, in many respects, to the one at present under consideration. The same method of approach will therefore be adopted here, and the work of Proudman & Pearson will serve as a constant reference for many of the details of the analysis which follows.

An important point in the development of the singular perturbation expansion is concerned with the failure of the creeping flow solution, as given by equation ($7a$), to provide a uniform approximation to the flow as $r \to \infty$. Thus, when $r \gg 1$, it is necessary to replace ψ_0 by an appropriate 'outer' solution which, in this case, satisfies the familiar Oseen equation and also a matching requirement with the Stokes solution (Proudman & Pearson 1957). In particular, by referring to Proudman & Pearson and to equation ($7a$) we can easily show that, in the 'outer' region, the stream function—denoted here by Ψ—is given by

$$Re^2\Psi = \frac{\rho^2}{2}(1-\mu^2) - \frac{3\kappa+2}{2(\kappa+1)}Re\,(1+\mu)\,[1-e^{-\frac{1}{2}\rho(1-\mu)}] + O(Re^2), \tag{11}$$

where $\rho = Re\,r$.

It is now possible to proceed with the solution of the problem by letting

$$\psi = \psi_0 + Re\,\psi_1 + \ldots \quad \text{for} \quad r \geqslant 1+\zeta \tag{12a}$$

and

$$\hat{\psi} = \hat{\psi}_0 + Re\,\hat{\psi}_1 + \ldots \quad \text{for} \quad 0 \leqslant r \leqslant 1+\zeta, \tag{12b}$$

with the understanding that equation ($12a$) is to hold only throughout the 'Stokes' region and that an appropriate matching requirement between equations (11) and ($12a$) is also to be satisfied. Of course, the equations for ψ_1 and $\hat{\psi}_1$ follow directly from equations (4) and (5), respectively, if the inertia terms are approximated by means of the corresponding zero-order solutions ψ_0 and $\hat{\psi}_0$. Thus,

$$D^4\psi_1 = 3Q_2(\mu)\frac{3\kappa+2}{\kappa+1}\left\{\frac{1}{r^2} - \frac{1}{2r^3}\frac{3\kappa+2}{\kappa+1} + \frac{1}{2r^5}\frac{\kappa}{\kappa+1}\right\}, \tag{13}$$

where

$$Q_n(\mu) \equiv \int_{-1}^{\mu} P_n(\mu)\,d\mu,$$

in which $P_n(\mu)$ is the Legendre polynomial of degree n. Furthermore, since by coincidence $\hat{\psi}_0$ also satisfies the complete equation (5),

$$D^4\hat{\psi}_1 = 0. \tag{14}$$

A particular integral of equation (13) is

$$\frac{Q_2(\mu)}{8}\,\frac{3\kappa+2}{\kappa+1}\left\{r^2 - \frac{r}{2}\,\frac{3\kappa+2}{\kappa+1} - \frac{1}{2r}\,\frac{\kappa}{\kappa+1}\right\},$$

and the complete solution of equation (13) that vanishes both at $r = 1$ and, because of symmetry, at $\mu = \pm 1$, is, therefore,

$$\psi_1 = \frac{Q_2(\mu)}{8}\frac{3\kappa+2}{\kappa+1}\left\{r^2 - \frac{r}{2}\frac{3\kappa+2}{\kappa+1} + \frac{\kappa}{\kappa+1} - \frac{1}{2r}\frac{1}{\kappa+1}\right\} + \sum_{n=1}^{\infty}\{A_n(r^{n+3}-r^{-n}) + B_n(r^{n+1}-r^{-n}) + C_n(r^{-n+2}-r^{-n})\}\,Q_n(\mu).$$

This must now be matched to the 'outer' solution given by equation (11). It is easily seen, however, that, if only those terms up to and including $O(Re)$ are retained,

$$Re^2\Psi = -\rho^2 Q_1(\mu) - \frac{3\kappa+2}{2(\kappa+1)}Re\left\{-\rho Q_1(\mu) + \frac{\rho^2}{4}[Q_1(\mu)-Q_2(\mu)] + O(\rho^3)\right\},$$

while, in the (ρ, μ) variables,

$$Re^2(\psi_0 + Re\,\psi_1) = -\rho^2 Q_1 + \frac{3\kappa+2}{2(\kappa+1)}Re\left\{\rho Q_1(\mu) + \frac{\rho^2}{4}Q_2(\mu)\right\} + \sum_{n=1}^{\infty}\{A_n Re^{-n}\rho^{n+3} + B_n Re^{-n+2}\rho^{n+1}\}\,Q_n(\mu).$$

Hence the matching requirement is satisfied if

$$A_n = 0 \quad \text{for} \quad n \geqslant 1, \quad B_n = 0 \quad \text{for} \quad n \geqslant 2, \quad B_1 = -\frac{1}{8}\frac{3\kappa+2}{\kappa+1}.$$

Therefore

$$\psi_1 = -\frac{Q_1(\mu)}{8}\frac{3\kappa+2}{\kappa+1}\left(r^2 - \frac{1}{r}\right) + \frac{Q_2(\mu)}{8}\frac{3\kappa+2}{\kappa+1}\left\{r^2 - \frac{r}{2}\frac{3\kappa+2}{\kappa+1} + \frac{\kappa}{\kappa+1} - \frac{1}{2r}\frac{\kappa}{\kappa+1}\right\} + \sum_{n=1}^{\infty} C_n[r^{-n+2} - r^{-n}]\,Q_n(\mu). \quad (15)$$

Similarly, a solution of (14) which vanishes at $r = 1$ and at $\mu = \pm 1$, and which, for all $0 \leqslant r \leqslant 1$, yields finite velocities, is

$$\hat{\psi}_1 = \sum_{n=1}^{\infty} \hat{C}_n\,[r^{n+3} - r^{n+1}]\,Q_n(\mu). \quad (16)$$

The coefficients C_n and $\hat{C}_n$ can finally be determined by making use of equations ($1a$) and ($1c$) which yield

$$C_1 = \frac{1}{16}\left(\frac{3\kappa+2}{\kappa+1}\right)^2, \quad C_2 = -\frac{1}{80}\frac{\kappa(3\kappa+2)(5\kappa+6)}{(\kappa+1)^3},$$

$$\hat{C}_1 = -\frac{1}{16}\frac{3\kappa+2}{(\kappa+1)^2}, \quad \hat{C}_2 = \frac{(3\kappa+2)(4\kappa+5)}{80(\kappa+1)^3},$$

$$C_n = \hat{C}_n = 0 \quad \text{for} \quad n \geqslant 3,$$

so that

$$\psi_1 = -\frac{Q_1(\mu)}{8}\frac{3\kappa+2}{\kappa+1}\left[r^2 - \frac{3\kappa+2}{2(\kappa+1)}r + \frac{\kappa}{2(\kappa+1)}\frac{1}{r}\right] + \frac{Q_2(\mu)}{8}\frac{3\kappa+2}{\kappa+1} \times\left[r^2 - \frac{r}{2}\frac{3\kappa+2}{\kappa+1} + \frac{\kappa(5\kappa+4)}{10(\kappa+1)^2} - \frac{1}{2r}\frac{\kappa}{\kappa+1} + \frac{1}{10r^2}\frac{\kappa(5\kappa+6)}{(\kappa+1)^2}\right] \quad (17)$$

and

$$\hat{\psi}_1 = \frac{3\kappa+2}{16(\kappa+1)^2}\left\{(r^2-r^4)\,Q_1(\mu) - \frac{4\kappa+5}{5(\kappa+1)}(r^3-r^5)\,Q_2(\mu)\right\}. \quad (18)$$

4. The deformation of the drop

The solution just obtained can now be used to calculate the deformation of the drop for small values of the Weber number, *We*. First, however, it is necessary to determine the normal stresses at the surface of the drop. This may be accomplished in a straightforward, although somewhat tedious manner, by noting that, up to an additive constant, the pressure distribution at $r = 1$ may be derived quite readily by integrating with respect to μ the apporpriate equation of motion, evaluated at $r = 1$, containing the term $\partial p/\partial \mu$. Thus, if, for example, at $r = 1$,

$$p = p_0 + Re\, p_1 + \ldots$$

and

$$\tau_{rr} = \tau_{rr}^{(0)} + Re\, \tau_{rr}^{(1)} + \ldots,$$

it can be shown that

$$Re\, p_1 = \Pi_1 - \frac{1}{16}\frac{3\kappa+2}{(\kappa+1)^2}\left\{(3\kappa+2)\,P_1(\mu) - \frac{P_2(\mu)}{5(\kappa+1)}[15\kappa^2+27\kappa+10]\right\} + \frac{P_2(\mu)}{12(\kappa+1)^2}$$

and that

$$Re\, \tau_{rr}^{(1)} = \frac{1}{16}\frac{3\kappa+2}{(\kappa+1)^2}\left\{3(\kappa+2)\,P_1(\mu) - \frac{P_2(\mu)}{5(\kappa+1)}[15\kappa^2+43\kappa+30]\right\} - \frac{P_2(\mu)}{12(\kappa+1)^2} - \Pi_1, \tag{19}$$

where Π_1 is a constant. Similarly, for the region inside the drop and again for $r = 1$,

$$Re\, \hat{p}_1 = \hat{\Pi}_1 + \frac{\kappa}{16}\frac{3\kappa+2}{(\kappa+1)^2}\left\{10P_1(\mu) - \frac{7P_2(\mu)}{5(\kappa+1)}(4\kappa+5)\right\} + \frac{\gamma P_2(\mu)}{12(\kappa+1)^2}$$

while

$$Re\, \hat{\tau}_{rr}^{(1)} = -\frac{\kappa}{16}\frac{3\kappa+2}{(\kappa+1)^2}\left\{6P_1(\mu) - \frac{3P_2(\mu)}{5(\kappa+1)}(4\kappa+5)\right\} - \frac{\gamma P_2(\mu)}{12(\kappa+1)^2} - \hat{\Pi}_1. \tag{20}$$

Finally, since (Taylor 1962; Brenner & Cox 1963)

$$F_D = \frac{2\pi}{Re}\left(\frac{3\kappa+2}{\kappa+1}\right) + \frac{\pi}{4}\left(\frac{3\kappa+2}{\kappa+1}\right)^2 + \ldots,$$

it follows from equations (1b), (3), (8), (10), (19) and (20) that

$$\frac{1}{We}\left\{2\zeta + \frac{d}{d\mu}\left[(1-\mu^2)\frac{d\zeta}{d\mu}\right]\right\} = \frac{P_2(\mu)}{80}\frac{3\kappa+2}{(\kappa+1)^3}\{27\kappa^2+58\kappa+30\}$$
$$- \frac{\gamma-1}{12(\kappa+1)^2}P_2(\mu) + \Pi_1 - \hat{\Pi}_1$$

and, therefore, because of equations (9) and (9*a*),

$$\zeta = -\frac{We\, P_2(\mu)}{4(\kappa+1)^3}\left\{\left(\frac{81}{80}\kappa^3 + \frac{57}{20}\kappa^2 + \frac{103}{40}\kappa + \frac{3}{4}\right) - \frac{\gamma-1}{12}(\kappa+1)\right\}. \tag{21}$$

We can clearly see then, that, for small values of *We*, the drop will deform exactly into a spheroid. This result is in qualitative, but not quantitative, agreement with Saito's (1913) conclusion even though, as was mentioned earlier, Saito's analysis was based on an incorrect approach. In principle, the spheroid can, of course, be both oblate or prolate depending on whether the coefficient multiplying the function $P_2(\mu)$ is, respectively, negative or positive. Yet, because, according to equation (21), $\zeta/P_2(\mu) < 0$ even for drops of mercury in water ($\gamma = 13{\cdot}6$, $\kappa \sim 1$) one can safely conclude that in all cases of physical significance the drop will be deformed into an *oblate* rather than a prolate spheroid. As a matter of fact, it is interesting to note that the deformation is only slightly affected by the viscosity of the interior phase since, for very viscous drops ($\kappa \to \infty$),

$$\zeta = -0{\cdot}25\, We\, P_2(\mu),$$

whereas for the geometrically opposite limiting case of small gas bubbles ($\kappa \to 0, \gamma \to 0$)

$$\zeta = -0{\cdot}21\, We\, P_2(\mu).$$

5. The drag and higher-order deformation of a slightly deformed drop

The effect of the deformation on the drop can now be determined by a 'creeping flow' analysis of the appropriate equations of motion for the flow past a slightly deformed drop. Thus, if once again inertia effects are completely neglected, we can set

$$\psi_0 = -\frac{Q_1(\mu)}{2}\left[2r^2 - \frac{3\kappa+2}{\kappa+1}r + \frac{\kappa}{\kappa+1}\frac{1}{r}\right] + We \sum_{n=1}^{\infty} (B_n r^{2-n} + C_n r^{-n})\, Q_n(\mu) \quad (22)$$

and

$$\hat{\psi}_0 = \frac{Q_1(\mu)}{2(\kappa+1)}(r^2 - r^4) + We \sum_{n=1}^{\infty} (\hat{B}_n r^{n+3} + \hat{C}_n r^{n+1})\, Q_n(\mu), \quad (23)$$

where all the coefficients B_n, C_n, $\hat{B}_n$, $\hat{C}_n$ are, to a first approximation $O(1)$. These can in turn be determined from the boundary conditions, at

$$r = 1 - \lambda\, We\, P_2(\mu),$$

as given by (1) and (2). The straightforward but tedious computations, which will not be reported here, yield

$$\psi_0 = -\frac{Q_1(\mu)}{2}\left[2r^2 - \frac{3\kappa+2}{\kappa+1}r + \frac{\kappa}{\kappa+1}\frac{1}{r}\right] + \frac{\lambda\, We}{10(\kappa+1)^2}\left\{(3\kappa^2 - \kappa + 8)\,r - 3(\kappa^2 - \kappa + 2)\frac{1}{r}\right\}$$
$$\times Q_1(\mu) - \frac{6\lambda\, We}{5(\kappa+1)}\left\{\left(\frac{3\kappa}{2} + \frac{9}{7}\right)\frac{1}{r} - \left(\frac{3\kappa}{2} + \frac{2}{7}\right)\frac{1}{r^3}\right\} Q_3(\mu) \quad (24)$$

and

$$\hat{\psi}_0 = \frac{Q_1(\mu)}{2(\kappa+1)}(r^2 - r^4) + \frac{\lambda\, We}{5(\kappa+1)^2}\{2(2-\kappa)\,r^4 - 3(\kappa-1)\,r^2\}\, Q_1(\mu)$$
$$+ \frac{6\lambda\, We}{35(\kappa+1)}\{5r^6 - 12r^4\}\, Q_3(\mu), \quad (25)$$

in which

$$\lambda \equiv \frac{1}{4(\kappa+1)^3}\left\{(\tfrac{81}{80}\kappa^3 + \tfrac{57}{20}\kappa + \tfrac{103}{40}\kappa + \tfrac{3}{4}) - \frac{\gamma - 1}{12}(\kappa+1)\right\}. \quad (26)$$

The drag can now be obtained immediately from (24), since for creeping flow (Payne & Pell 1960)

$$\frac{\text{drag}}{a^2 \rho U^2} Re = 8\pi \lim_{r\to\infty}\left\{\frac{1}{r(1-\mu^2)}\left[\frac{r^2}{2}(1-\mu^2) - \psi_0\right]\right\},$$

which may in turn be combined with a recent result (Brenner & Cox 1963) to yield

$$\frac{F_D Re}{2\pi} = \frac{3\kappa+2}{\kappa+1} + \frac{Re}{8}\left(\frac{3\kappa+2}{\kappa+1}\right)^2 + \frac{1}{40}\left(\frac{3\kappa+2}{\kappa+1}\right)^3 Re^2 \ln Re + \frac{\lambda\, We}{5(\kappa+1)^2}(3\kappa^2-\kappa+8)+\ldots, \tag{27}$$

where, of course, the last term represents the effect of the deformation to $O(We)$. The terminal velocity of the drop may then be determined by combining (27) with (10).

The solution just derived may also be used to generate an expression for the deformation ζ up to $O(We^2/Re)$. Thus, it can be shown from equations (24) and (25) and the appropriate equations of motion, that, at $r = 1-\lambda\, We P_2$ and up to an additive constant,

$$\tau_{rr} = \frac{3\kappa+6}{2(\kappa+1)\,Re} P_1(\mu) + \frac{ga}{U^2}(1-\lambda\, We\, P_2)\,P_1 + \frac{\lambda\, We\, P_1}{10(\kappa+1)^2 Re}\{12+27\kappa-21\kappa^2\} + \frac{\lambda\, We\, P_3}{10(\kappa+1)\,Re}(9\kappa-\tfrac{426}{7}) + Re\,\tau_{rr}^{(1)}, \tag{28}$$

where $\tau_{rr}^{(1)}$ is given by (19). Similarly,

$$\hat{\tau}_{rr} = -\frac{3\kappa}{(\kappa+1)\,Re} P_1(\mu) + \frac{ga\gamma}{U^2}(1-\lambda\, We\, P_2)\,P_1 + \frac{\lambda\, We\, P_1}{10(\kappa+1)^2 Re}\{60\kappa-12\kappa^2\} + \frac{\lambda\, We\, P_3}{10(\kappa+1)\,Re}\frac{582\kappa}{7} + Re\,\hat{\tau}_{rr}^{(1)} - \Pi \tag{29}$$

and, therefore, because of equations (1*b*), (2*b*), (3)†, (10), (27), as well as our earlier results,

$$\zeta = -\lambda\, We\, P_2(\mu) - \frac{3\lambda(11\kappa+10)}{70(\kappa+1)}\frac{We^2}{Re}P_3(\mu)+\ldots, \tag{30}$$

with λ given by (26). Thus, according to (30) and in agreement with the experimental observations (Haberman & Morton 1953) the drop will first deform into an oblate spheroid and then, with a further increase in the Weber number, into a geometry approaching that of a spherical cap.

6. Concluding remarks

The principal results of this theoretical analysis are contained in equations (27) and (30) which depict quantitatively the nature of the drop deformation at low Reynolds numbers and the influence which this deformation can exert on the drag of the drop. These equations may now be recast into a more convenient form, in which only one of the dimensionless groups, the Reynolds number Re, contains U, by noting that $We = (\rho\nu^2/a\sigma)\,Re^2$. Thus, we can express the deformation by

$$\zeta = -\lambda\left(\frac{\rho\nu^2}{a\sigma}\right) Re^2 P_2(\mu) - \frac{3\lambda(11\kappa+10)}{70(\kappa+1)}\left(\frac{\rho\nu^2}{a\sigma}\right)^2 Re^3 P_3(\mu)+\ldots, \tag{30'}$$

† It should be noted that We is $O(Re^2)$. Consequently, the deformation ζ is also $O(Re^2)$. Thus, the linearized boundary conditions, (2) and (3), are still applicable to the present analysis since the ζ^2 terms would be $O(Re^4)$.

whereas, for the drag, we obtain

$$(F_D - F_{Dsph})\frac{Re}{2\pi} = \lambda\frac{3\kappa^2-\kappa+8}{5(\kappa+1)^2}\left(\frac{\rho\nu^2}{a\sigma}\right)Re^2+\ldots, \qquad (27')$$

where F_{Dsph} denotes the appropriate dimensionless drag of a spherical drop.

It should be kept in mind of course that the analysis just presented contains a number of limitations which we shall presently discuss. To begin with, we must require that $(\rho\nu^2/a\sigma)\,Re^2$ be small in view of our assumption that the drop is 'almost spherical'. Also, it is clear that our results are of significance only as long as both $Re < O(1)$ and $(\gamma/\kappa)\,Re < O(1)$, since the theoretical development was based on a perturbation expansion about the creeping flow solution of the equations of motion, both inside and outside the drop. And finally, since it can easily be shown that the term of $O(Re^2 \ln Re)$ which appears in the expansion for ψ in the case of a spherical drop (Proudman & Pearson 1957) does not contribute to the deformation—this term is after all only a multiple of the appropriate 'Stokes' solution, equation (7), which, as we have seen, satisfies automatically the requirement of normal stress continuity across the drop interface—it follows that (30) will contain a term of $O(Re\,We)$ which arises from the $O(Re^2)$ term in the expansion for ψ. Thus, (30′) should contain a term of $O(\rho\nu^2\,Re^2/a\sigma)$ which is of the same order of magnitude in Re as the second term in that equation. For the same reason, an additional term of $O(Re^2)$ should appear in (27)—but not in equation (27′) since this term has already been incorporated in F_{Dsph}. Thus, if we are to retain in equations (27), (30) and (30′) only those terms which are shown, we must impose the further restriction that $\rho\nu^2/a\sigma \gg 1$. This last condition is, however, difficult to achieve in general, owing to the small numerical values of ν for most fluids. Consequently, it appears that, although included for completeness, the last term in (27), (30) and (30′) would be of little significance in many cases of practical interest since each of these equations should contain in addition another term, of equal order of magnitude in Re to the one shown last.

A few remarks should finally be added concerning the possible influence of surface active agents on the results of this paper. This is necessary, because repeated experimental investigations (Griffith 1962) have firmly established that even small amounts of surfactants can have a profound influence on the terminal velocity and on the internal circulation of drops and bubbles.

The most likely explanation for this observation was originally provided by Frumkin & Levich (1947; see also Levich 1962). According to their theory, during the fall of a drop surfactant molecules are swept to the rear portion of its surface thus setting up a composition gradient, and therefore a surface tension gradient, which in turn opposes the motion of the liquid along the interface. On the basis of this model and by means of a theoretical analysis of the appropriate creeping flow equations, Frumkin & Levich were then able to derive an expression for the drag similar to equation (10*a*) with, however, $(\kappa + k/a)$ instead of κ, where k is a function of the amount and the type of surfactant present, and a is the radius of the sphere. The Frumkin & Levich solution also satisfies the normal stress balance at the surface of the drop, exactly as was the case with equations (7*a*) and (7*b*). Thus it would appear that the presence of surfactants should have no influence on the shape of the drop, which would still remain spherical, and that their only effect would be to increase the viscosity ratio κ by an amount equal to k/a.

These conclusions, which are based on the Frumkin & Levich theory, are of course applicable only as long as the inertia effects can be neglected. On the other hand, since the deformation of the drop, as given by equation (30), is so insensitive to the numerical value of κ, one would naturally expect, in view of what has been said above, that equation (30) would also remain relatively unaffected by the presence of surfactants. It would seem, therefore, that whereas the internal circulation of drops can indeed be profoundly influenced by the addition of surfactants to the system, the shape of a falling drop is determined primarily by the hydrodynamic forces and the static surface tension, and to a lesser extent by the presence of surface active molecules along the surface.

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REFERENCES

Brenner, H. & Cox, R. G. 1963 *J. Fluid Mech.* **17**, 561.
Frumkin, A. N. & Levich, V. G. 1947 *Zhur. Fiz. Khim.* **21**, 1183.
Griffith, R. M. 1962 *Chem. Engng Sci.* **17**, 1057.
Haberman, W. L. & Morton, R. K. 1953 *David Taylor Model Basin Rep.* no. 802.
Hadamard, J. 1911 *Compt. Rend.* **152**, 1735.
Kaplun, S. & Lagerstrom, P. A. 1957 *J. Math. Mech.* **6**, 585.
Landau, L. D. & Lifschitz, E. M. 1959 *Fluid Mechanics*. London: Pergamon Press.
Levich, V. G. 1962 *Physicochemical Hydrodynamics*, Ch. 8. New York: Prentice-Hall.
Pai, S. I. 1956 *Viscous Flow Theory. I. Laminar Flow.* New York: Van Nostrand.
Payne, L. E. & Pell, W. H. 1960 *J. Fluid Mech.* **7**, 529.
Proudman, I. & Pearson, J. R. A. 1957 *J. Fluid Mech.* **2**, 237.
Rybczynski, W. 1911 *Bull. Acad. Sci. de Cracovie*, A, p. 40.
Saito, S. 1913 *Science Rep. Tôhoku Imp. Univ., Sendai, Japan,* **2**, 179.
Scriven, L. E. 1960 *Chem. Engng Sci.* **12**, 98.
Taylor, T. D. 1962 Ph.D. Dissertation, Univ. of California, Berkeley.