

of  $B$ , as well as  $b_3$  can be taken as the free parameters. That the solution (2.3) is real valued follows from the results obtained earlier in [1]. The dependence of  $\varphi$  on time can then be found by inverting an elliptic integral.

The method of investigation used in this paper indicates that the precession (2.3) is unique under the conditions (2.2). For  $C = 0$ ,  $B = 0$  and  $\lambda = 0$ , we obtain a precession of general form in the classical problem of the motion of a rigid body corresponding to a solution [2], which, despite the Hess conditions for the distribution of the mass of the body, does not occur as a special case in the Hess solution.

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## THE EFFECT OF THE SURFACE TENSION GRADIENT ON THE MOTION OF A SPHERICAL AND DEFORMED DROP†

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It is shown that, when the Stokes equations are used, a drop which is falling through a viscous fluid can only maintain a strictly spherical shape when there are specific distributions of the surface tension. Deviations from these distributions will cause some deformation of the drop. These results are obtained using a more general solution of the Stokes equations compared with the solutions which were considered earlier [1].

THE MOTION of a spherical drop in a viscous fluid has been studied both theoretically and experimentally. It has been pointed out [2] that agreement between the experimental and theoretical results can be attained if account is taken of the effect of surfactants and the changes on the surface of a drop associated with them. Moreover, the surface tension distribution on the drop may manifest itself in the shape of its surface.

The deformation of a drop which falls through a viscous fluid has been treated in the Oseen approximation, taking into account inertial effects, by the method of matched asymptotic expansions [1, 3]. It was concluded [1] that deformations of the surface cannot occur and the drop will remain spherical within the framework of the inertia-less Stokes equations when the surface tension on the surface of the drop is constant and there is no change in the rate of flow around the spherical drop.

Let us consider the flow around a drop of radius  $R$  by another fluid with a velocity  $U$  at a large distance from the drop. This flow relative to the drop arises as a result of its falling through the fluid under the action of gravitational and Archimedean forces. The surface tension  $\sigma$  varies along the surface of the drop  $\sigma(\theta)$ . There are various reasons for this change in the surface tension: the existence of surfactants, a non-uniform temperature field, etc.

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The motion of the fluid inside and outside the drop is denoted by the subscripts  $i$  and  $e$ , respectively. At low Reynolds numbers ( $Re \ll 1$ ), the steady motion of the internal and external fluid is described in the Stokes approximation by the system of equations:

$$\Delta \mathbf{v} = \mu^{-1} \nabla P, \quad \mathbf{v} \cdot \mathbf{v} = 0 \quad (1)$$

where  $P$  is a generalized function of the pressure, which includes the external forces ( $P = P - \rho g z$ ) and  $\mu$  is the viscosity of the fluid. Since the flow around a sphere is axisymmetrical, the problem is solved using a stream function  $\psi$  which is defined as follows

$$u = -\frac{1}{r^2 \sin \theta} \frac{\partial \psi}{\partial \theta}, \quad v = \frac{1}{r \sin \theta} \frac{\partial \psi}{\partial r} \quad (2)$$

where  $u$  and  $v$  are the radial and transverse components of the velocity. The boundary conditions at infinity and on the surface of the drop  $S$  have the form

$$r \rightarrow \infty \quad \psi_e \rightarrow 1/2 U r^2 \sin^2 \theta \quad (3)$$

$$\text{At } \psi_i = 0, \quad \psi_e = 0, \quad \frac{\partial \psi_i}{\partial r} = \frac{\partial \psi_e}{\partial r} \quad (4)$$

$$\tau_e - \tau_i = -\frac{1}{R} \frac{d\sigma}{d\theta} \quad (5)$$

$$N_e - N_i = 2\sigma/R \quad (6)$$

Here,  $\tau_e$ ,  $N_e$ ,  $\tau_i$  and  $N_i$  are the shear and normal stresses on the surface of the drop, respectively. The general solution of the equations for the stream function is represented in the form of infinite series in Gegenbauer polynomials [4]:

$$\psi_e = \sum_{n=2}^{\infty} (A_n r^n + B_n r^{-n+1} + C_n r^{n+2} + D_n r^{-n+3}) J_n(\zeta) \quad (7)$$

$$\psi_i = \sum_{n=2}^{\infty} (a_n r^n + b_n r^{-n+1} + c_n r^{n+2} + d_n r^{-n+3}) J_n(\zeta) \quad (8)$$

where  $\zeta = \cos \theta$ . On satisfying the boundary conditions (4) and (5) and taking account of the boundary conditions of the stream functions at infinity (3) and the property of the finiteness of the velocities within the drop, we establish that all the coefficients in (7) and (8) for the stream functions are expressed in terms of  $B_2$ ,  $B_n$ ,  $n \geq 3$ , which depend on the gradient of the surface tension by virtue of condition (5)

$$B_2 = \frac{R^3}{\mu_e + \mu_i} \left( \frac{U \mu_i}{2} - \frac{1}{4} \int_0^\pi \sin^2 \theta \frac{d\sigma}{d\theta} d\theta \right) \quad (9)$$

$$B_n = -\frac{n(n-1)}{4} \frac{R^{n+1}}{\mu_e + \mu_i} \int_0^\pi J_n(\zeta) \frac{d\sigma}{d\theta} d\theta \quad (10)$$

Knowing the external flow around the drop we can determine the force which acts on the drop due to the surrounding fluid, which will be in the direction of the flow velocity and will be equal to

$$F = -2\pi \mu_e U R \frac{2\mu_e + 3\mu_i}{\mu_e + \mu_i} + \pi R \frac{\mu_e}{\mu_e + \mu_i} \int_0^\pi \sin^2 \theta \frac{d\sigma}{d\theta} d\theta \quad (11)$$

where the first term represents the force obtained by Hadamard and Rybczynski and the second term is associated with the existence of a surface-tension gradient. The latter increases the drag since, from (5),  $\partial\sigma/\partial\theta < 0$ . The minus sign in the expression for the force indicates that it acts in the opposite direction to the motion of the sphere. The drag force acting on the drop is balanced by a driving force

$$F = -4/3 \pi R^3 k, \quad k = g(\rho_i - \rho_e) \quad (12)$$

From this condition the velocity  $U$  is determined

$$U = \frac{2(\mu_e + \mu_i)}{2\mu_e + 3\mu_i} \frac{kR^2}{3\mu_e} + \frac{1}{4\mu_e + 6\mu_i} \int_0^\pi \sin^2 \theta \frac{d\sigma}{d\theta} d\theta \quad (13)$$

Expression (13) is identical with the expression presented in [5] for the velocity of a drop with an arbitrary surface tension distribution depending on the presence of surfactants.

In earlier investigations associated with the motion of a drop with a constant or variable surface tension [5], a condition on the normal stresses acting on the surface of the drop was usually not considered. However, when there is a surface tension gradient, it is important that a boundary condition on the normal stresses should be satisfied since it provides the possibility of determining the distribution of the surface tension under which the drop remains spherical. When the boundary conditions on the shear and normal stresses are jointly taken into account, one obtains

$$B_2 = -UR^3 + \frac{kR^5}{3\mu_e}, \quad B_n = 0 \quad n \geq 3 \quad (14)$$

By representing the surface tension  $\sigma$  in the form of a series in Legendre polynomials with unknown coefficients  $\alpha_m$

$$\sigma = \sum_{m=0}^{\infty} \alpha_m P_m(\xi) \quad (15)$$

and taking into account the relationship between Legendre polynomials and Gegenbauer polynomials, we obtain

$$\frac{d\sigma}{d\theta} = \sum_{m=1}^{\infty} \alpha_m (-\sin \theta) m(m+1) \frac{J_{m+1}(\xi)}{1-\xi^2} \quad (16)$$

By comparing (9), (10) and (14) and taking into account (16) and the orthogonality conditions for Gegenbauer polynomials, we obtain a system of equations for finding all the  $\alpha_m$ :

$$\sum \alpha_m m(m+1) \int_{-1}^{+1} J_2(\xi) \frac{J_{m+1}(\xi)}{1-\xi^2} d\xi = \frac{2}{3} kR^2 \frac{\mu_e + \mu_i}{\mu_e} - U(2\mu_e + 3\mu_i) \quad (17)$$

$$\sum \alpha_m m(m+1) \int_{-1}^{+1} J_n(\xi) \frac{J_{m+1}(\xi)}{1-\xi^2} d\xi = 0 \quad n \geq 3$$

the solution of which yields

$$\alpha_1 = kR^2 \frac{\mu_e + \mu_i}{\mu_e} - U \left( \frac{9}{2} \mu_i + 3\mu_e \right), \quad \alpha_m = 0, \quad m \geq 2 \quad (18)$$

Consequently, in the general case, the surface tension on the surface of a spherical drop must be expressed by the formula

$$\sigma = \alpha_0 + \alpha_1 \cos \theta \quad (19)$$

where  $\alpha_0$  is the surface tension on the large circle of the drop ( $\theta = \pi/2$ ). Relationship (19) is a necessary condition for the drop to preserve its spherical shape in the case of a variable surface tension.

If the variability of the surface tension is caused by the existence, for example, of a non-uniform temperature field  $\sigma = \sigma(T)$  which satisfies the Laplace equation  $\nabla^2 T = 0$  (for low Peclet numbers) it is possible to find the temperature distribution along the surface of the spherical drop. It will have the form [6]

$$T(R, \theta) = T(R, \pi/2) + \lambda \cos \theta \quad (20)$$

where the coefficient  $\lambda$  is proportional to the radius of the drop and the temperature gradient at infinity and depends on the ratio of the thermal conductivity of the drop and the medium in which the drop is moving.

If  $\sigma$  depends on the temperature such that it may be assumed that  $\partial\sigma/\partial T = \text{const}$ , then  $\sigma(\theta) = (\partial\sigma/\partial T) \lambda \cos \theta + \sigma(\pi/2)$ . Consequently, in the case under consideration, the surface tension has a cosinusoidal

distribution over the surface of the drop and the drop retains its spherical form, as follows from the general considerations which lead to formula (19).

It may be noted from Eq. (13) that the terminal setting velocity of the drop as a function of the quantity  $\partial\sigma/\partial\theta$  may be changed from the maximum velocity determined by the Hadamard–Rybczynski formula

$$U' = \frac{2}{3} kR^2 \frac{\mu_c + \mu_i}{\mu_c (3\mu_i + 2\mu_c)} \tag{21}$$

to the minimum velocity, which is identical with the velocity of a solid sphere

$$U = \frac{2}{9} kR^2 / \mu_c \tag{22}$$

In this case, the surface tension distribution on the surface of the sphere (19) varies from a constant value  $\alpha_0$  up to the value

$$\sigma = \alpha_0 + \frac{1}{3} kR^2 \cos \theta \tag{23}$$

Here, we do not consider the case when, for example, owing to certain causes the direction of rotation of the vortex which develops within the drop changes into the opposite direction on account of the occurrence of a temperature gradient.

When a fluid drop moves at the velocity of a solid sphere (22) and with the corresponding surface tension distribution (23), the external flow of the fluid does not give rise to motion of the fluid within the drop. The stream function of the internal motion vanishes. The shear stresses of the external fluid which act on the sphere are balanced by the gradient of the surface tension. It can be seen from expression (23) that drops of large radii must have an appreciable surface-tension gradient in order that they can move at the velocity of a solid sphere. Drops of small radii can fall at such a limiting least terminal settling velocity even when there are exceedingly small changes in the surface tension.

Hence, by specifying the surface tension in the form  $\sigma = \alpha_0 + \alpha_1 P_1(\zeta)$  with a fixed value of the coefficient  $\alpha_1$  lying between 0 and  $\frac{1}{3}kR^2$ , the magnitude of the terminal settling velocity of a spherical drop is thereby fixed and it will lie between the Hadamard–Rybczynski velocity and the Stokes velocity.

The deformations of the drop are determined in the case when the surface tension has a distribution that differs from cosinusoidal and is expressed by the following dependence:

$$\alpha = \alpha_0 + (\alpha_1 + \alpha') P_1(\cos \theta) + \alpha_2 P_2(\cos \theta) \tag{24}$$

The shape of the drop is described by the equation

$$r = R(1 + \omega) \quad \left( \omega = \sum_{m=0}^{\infty} \beta_m P_m(\zeta) \right) \tag{25}$$

The boundary conditions on the surface of a deformed drop (25) have the same form (4)–(6) if, in the more general equalities

$$\begin{aligned} v_{nr} = 0, \quad v_{ni} = 0, \quad v_{\tau e} = v_{\tau i}, \quad p_{nne} - p_{nni} &= (1/R_1 + 1/R_2) \sigma \\ p_{n\tau e} - p_{n\tau i} &= -\partial\sigma/\partial s \end{aligned}$$

one neglects terms which are proportional to the small quantities  $\omega$ ,  $\omega'$  and  $\omega''$  when writing down the equation of the meridional curve for the deformed surface in the form of (25). The use of conditions (4) makes it possible to express the coefficients  $a_n$ ,  $c_n$  and  $D_n$ , occurring in the stream functions (7) and (8), in terms of  $B_n$ . The system of equations, composed of (5) and (6), the conditions for the conservation of the volume of the drop under deformation

$$\frac{4}{3} \pi R^3 + 2\pi R^3 \int_0^\pi \omega \sin \theta' d\theta = \frac{4}{3} \pi R^3 \tag{26}$$

and the condition for the balance of the forces acting on the drop

$$4\pi\mu_c \left[ -UR - B_2 R^{-2} + (2B_2 R^{-2} - UR) \left( \beta_0 + \frac{2}{5} \beta_2 - \frac{1}{35} \beta_4 \right) \right] = -\frac{4}{3} \pi R^3 k \tag{27}$$

serve to find the unknown coefficients  $\beta_m$  and  $B_n$ . Confining ourselves to the coefficients  $\beta_0 - \beta_4$ ,  $B_2$  and  $B_3$ , they are sought in the following form:

$$B_2 = B_{20} + B_{21}, \quad \beta_i = \beta_{i1}, \quad i = 0, 1, 2, 3, 4 \tag{28}$$

$B_{20}$  is a known quantity which occurs in the stream function which describes the flow around a spherical drop (14). From Eq. (26),  $\beta_0 = 0$  and it is found from Eqs (5) and (6), by using the relationship between Gegenbauer and Legendre polynomials and equating the coefficients accompanying identical polynomials, that  $\beta_1 = \beta_2 = \beta_4 = 0$  while the quantity  $\beta_3$  differs from zero:

$$\beta_3 = -\frac{14}{3} \frac{\alpha_2}{\alpha_0} \frac{4+\bar{\mu}}{M} \quad (29)$$

$$M = 10(1+\bar{\mu}) \left[ \text{Ca}(36\bar{\mu}-18) + \text{Bo} \left( 1 - \frac{23}{3} \bar{\mu} \right) \right] +$$

$$+ (2+3\bar{\mu}) [ \text{Ca}(90+69\bar{\mu}) - \text{Bo}(28+8\bar{\mu}) ]$$

$$\bar{\mu} = \frac{\mu_i}{\mu_e} \quad \text{Ca} = \frac{U\mu_e}{\alpha_0} \quad \text{Bo} = \frac{g(\rho_i - \rho_e)R^2}{\alpha_0}$$

where Ca is the capillary number, Bo is the Bond number and  $B_3$  is determined from the equation

$$B_3 \frac{R^{-4}}{\alpha_0 \mu_e} = \frac{\alpha_2}{\alpha_0} \frac{2}{2+3\bar{\mu}} - \frac{3}{7} \beta_3 \frac{\text{Ca}(18-36\bar{\mu}) + \text{Bo} \left( -1 + \frac{23}{3} \bar{\mu} \right)}{2+3\bar{\mu}} \quad (30)$$

The resulting shape of the drop is close to the shape of a so-called spherical cap

$$r = R \left[ 1 + \frac{1}{2} \beta_3 \cos \theta (5 \cos^2 \theta - 3) \right] \quad (31)$$

The drop is flattened in its frontal part and prolate in its rear part. Such a shape for falling drops has been observed [7, 8]. In this paper it has been obtained as a result of the assumption that the surface tension along the surface of the drop varies according to the law (24), which is more complex than a sinusoidal law, and when the internal and external flows are described using Stokes equations.

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