Slow Flow of a Non-Newtonian Fluid Past a Droplet

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The problem of slow flow past a droplet is considered where both materials may be represented as fluids of grade 3 and where the outer fluid is of infinite extent. Both non-Newtonian and inertial effects are included in the analysis. A double perturbation technique and the method of matched asymptotic expansions are employed to obtain solutions to the equation of motion.

Solutions, in the form of Legendre polynomial series, are obtained for the stream function (both outside and inside the droplet), for the drag force exerted on the droplet, and for the shape of the droplet.

The results obtained are in complete agreement with those obtained by other workers for the flow of a Newtonian fluid past a Newtonian droplet and for the flow of a fluid of grade 3 past a solid sphere. Droplet shape predictions are in qualitative agreement with experimentally observed shapes.

Flow of a Newtonian fluid past a Newtonian droplet with constant surface tension and negligible inertial effects was solved in 1911 by Hadamard (*1*) and, independently, by Rybczynski (*2*) [see also Levich (*3*, Chapter 8)]. The experimental usefulness of this solution is limited by its failure to include inertial effects and to account for a nonuniform surface tension caused by trace quantities of surfactants which are almost always present in such experiments.

Taylor and Acrivos (*4*) applied the method of matched asymptotic expansions (*5* to *8*) to this problem in order to account for inertial effects. They obtained several terms in the resulting expansions for the stream function, drag force and droplet shape. Wellek, Agrawal, and Skelland (*9*) have verified experimentally that their shape results are valid for Reynolds number less than 20.

Wasserman and Slattery (*10*) evaluated the effects of trace quantities of surface-active agents on the motion of a slowly moving droplet or gas bubble. Mass transfer of surfactant molecules from the unbounded fluid to the droplet's surface was assumed to be controlled by molecular and convective diffusion in the unbounded fluid. Their perturbation solution indicates that the terminal velocity of the droplet and the drag force which the surrounding fluid exerts on the droplet can be very sensitive to slight changes of surfactant concentration in the unbounded fluid even though the droplet does not lose its spherical shape.

In what follows we wish to extend the solution of Taylor and Acrivos (*4*) to cases where both the continuous and discontinuous phases are incompressible viscoelastic fluids. [Viscoelastic is used here in the sense that the materials obey neither of the classical linear relations, Newton's law of viscosity and Hooke's law of elasticity. Subclasses of such materials are fluids which show a finite relaxation time and fluids which exhibit normal stresses in viscometric flows (*11*, p. 47). The term viscoelastic is often used in the literature in referring to one of these subclasses.] Our primary objective is to predict the shape of these droplets. The experiments of Wellek, Agrawal, and Skelland (*9*) with Newtonian systems give us some reason to hope that such predictions will be reasonable descriptions of experimental data. The calculations of Wasserman and Slattery (*10*) suggest that we should not expect from this study an accurate representation for the drag force on a droplet.

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MODEL FOR MATERIAL BEHAVIOR

Since it is generally believed at the present time that a wide variety of materials may be described as Noll simple fluids (*12*, p. 60), we wish to base the description of our viscoelastic fluids upon this model. Unfortunately, the generality of the theory of Noll simple fluids precludes its use in all but a few restricted classes of flows (*11*, *13*, *14*).

Coleman and Noll (*15*) have shown that a Noll simple fluid with a fading memory may be approximated by a Rivlin-Ericksen fluid of grade n. [By fading memory we mean qualitatively that deformations which occurred in the distant past should be less important in determining the present stress than those which occurred in the recent past (*12*, p. 101). The assumption here is that real fluids with which we commonly work have a fading memory.] Truesdell (*16*; *12*, p. 495) comes to essentially the same conclusions for a fluid with sufficiently small characteristic time s_o. Putting Truesdell's arguments in a dimensionless form serves to consolidate these results (*17*). For the flow to be considered here we find that, with a sufficiently small value of the Weissenberg number $\lambda = s_o U/a$, the Noll simple fluid with fading memory may be approximated by a fluid of grade n. (We use λ rather than N_{Wi} for the Weissenberg number, since it occurs so frequently in what follows.) Here a, the radius of a sphere having the same volume as our droplet, denotes a characteristic length of our geometry; U, the magnitude of the velocity of the undisturbed stream as $r \longrightarrow \infty$, is a magnitude of velocity characteristic of our flow. For sufficiently small values of U/a, a fluid of grade n should be an excellent description of real fluid behavior in flow past a droplet.

The simplest Rivlin-Ericksen fluid of grade n which exhibits both normal stresses and a shear-dependent viscosity in a viscometric flow (*11*) is a fluid of grade 3 (*12*, p. 494; *16*; *18*):

$$\boldsymbol{\tau} = \phi_1 \mathbf{A}^{(1)} + \phi_2 \mathbf{A}^{(2)} + \phi_3 [\mathbf{A}^{(1)}]^2 + \phi_4 \mathbf{A}^{(3)} + \phi_5 [\mathbf{A}^{(1)} \cdot \mathbf{A}^{(2)} + \mathbf{A}^{(2)} \cdot \mathbf{A}^{(1)}] + \phi_6 \, tr\, [\mathbf{A}^{(1)}]^2 \mathbf{A}^{(1)} \quad (1)$$

Here $\boldsymbol{\tau}$ is the extra stress tensor, where the ϕ_i are constants under isothermic conditions, and the Rivlin-Ericksen tensors $\mathbf{A}^{(i)}$ are defined as (*19*, sections 10 and 16):

$$\mathbf{A}^{(1)} = 2\mathbf{d} = \nabla \mathbf{v} + (\nabla \mathbf{v})^T \quad (2)$$

$$\mathbf{A}^{(n)} = (\nabla \mathbf{A}^{(n-1)}) \cdot \mathbf{v} + (\nabla \mathbf{v})^T \cdot \mathbf{A}^{(n-1)} + \mathbf{A}^{(n-1)} \cdot \nabla \mathbf{v} \quad n > 1 \quad (3)$$

It is this model which we shall use to describe the behavior of our viscoelastic fluids.

DEFINING EQUATIONS

Consider the slow, steady state flow of a fluid of grade 3, the behavior of which is described by Equation (1) past a droplet of a second fluid of grade 3. Both fluids are assumed to be incompressible and mutually immiscible and the outer fluid is taken to be of infinite extent. The interface is taken as being a Newtonian surface fluid with constant surface tension and negligible surface viscosity (*20* to *22*). Denoting the properties of the interior fluid by quantities with carats, one obtains, under the above conditions, the equations of motion and continuity for this problem:†

$$\rho (\nabla \mathbf{v}) \cdot \mathbf{v} = -\nabla P + \nabla \cdot \boldsymbol{\tau} \quad (4)$$

$$\nabla \cdot \mathbf{v} = 0 \quad (5)$$

Here the force per unit mass $\mathbf{f}$ is assumed to be representable by the gradient of a scalar φ

$$\mathbf{f} = -\nabla \varphi \quad (6)$$

and has been included in the generalized pressure

$$P = p + \rho \varphi \quad (7)$$

Let us define as dimensionless variables

$$r^* = \frac{r}{a}, \quad \mathbf{v}^* = \frac{\mathbf{v}}{U}, \quad \boldsymbol{\tau}^* = \frac{a \boldsymbol{\tau}}{\phi_1 U}$$

$$P^* = \frac{aP}{\phi_1 U}, \quad \mathbf{A}^{(n)*} = \left(\frac{a}{U}\right)^n \mathbf{A}^{(n)} \quad (8)$$

We define a to be the radius of a spherical droplet of the same volume and U to be the magnitude of the velocity of the undisturbed stream as $r \longrightarrow \infty$. Dimensionless variables are used exclusively below; the asterisks are discarded for simplicity. The equations of motion become

$$N_{Re} (\nabla \mathbf{v}) \cdot \mathbf{v} = \nabla \cdot \boldsymbol{\tau} - \nabla P \quad (9)$$

while the equations of continuity remain unchanged in form. The Reynolds numbers are here defined in terms of the zero shear viscosities, ϕ_1 and $\hat{\phi}_1$, and the droplet equivalent radius.

The boundary conditions for this problem are

$$\text{at } r = R: \quad \mathbf{v} \cdot \mathbf{n} = \hat{\mathbf{v}} \cdot \mathbf{n} = 0 \quad (10a)$$

$$\text{at } r = R: \quad \mathbf{v} \cdot \boldsymbol{\nu} = \hat{\mathbf{v}} \cdot \boldsymbol{\nu} \quad (10b)$$

$$\text{at } r = R: \quad \mathbf{t} \cdot \mathbf{n} = \kappa \hat{\mathbf{t}} \cdot \mathbf{n} + \frac{N_{Re}}{N_{We}} \left[\frac{1}{R_1} + \frac{1}{R_2}\right] \mathbf{n} \quad (10c)$$

$$\text{at } r = R: \quad \mathbf{t} \cdot \boldsymbol{\nu} = \kappa \hat{\mathbf{t}} \cdot \boldsymbol{\nu} \quad (10d)$$

$$\text{as } r \longrightarrow 0: \quad \hat{\mathbf{v}} \text{ remains finite} \quad (10e)$$

$$\text{as } r \longrightarrow \infty: \quad v_z \longrightarrow 1, \quad v_x \longrightarrow 0, \quad v_y \longrightarrow 0 \quad (10f)$$

where $R = R(\theta)$ denotes the surface of the droplet. Here $\mathbf{n}$ and ν are, respectively, unit vectors normal and tangential to the droplet surface, R_1 and R_2 are the principal radii of curvature of the surface, κ is the ratio of the zero shear viscosity of the interior fluid to that of the exterior

$$\kappa = \hat{\phi}_1 / \phi_1 \quad (11)$$

and N_{We} is the Weber number

$$N_{We} = \frac{\rho a U^2}{\sigma} \quad (12)$$

†For simplicity, equations that are identical in form for both the interior and exterior fluids will be written only in terms of the exterior fluid. Thus it is understood that equations identical to Equations (1) and (2), for example, but with each variable replaced by its corresponding interior variable, exist and must be taken into account in the analysis.

METHOD OF SOLUTION

In spherical polar coordinates with the origin taken near the center of the droplet (the precise position of the origin will be defined later) and the line $\theta = 0$ in the downstream direction, a dimensionless stream function is defined such that equations of continuity, after axial symmetry is assumed, are identically satisfied:

$$v_r = \frac{1}{r^2} \frac{\partial \psi}{\partial \mu}, \quad v_\theta = \frac{1}{r(1-\mu^2)^{1/2}} \frac{\partial \psi}{\partial r} \quad (13)$$

where

$$\mu = \cos \theta \quad (14)$$

Utilizing the definitions of the stream functions and eliminating the generalized pressure P between the two nonzero components of each equation of motion [this is equivalent to using the third component of the vorticity equation as given by Goldstein (*23*, p. 115)]

$$D_r^4 \psi - f(\mathbf{T}) = N_{Re} \left[\frac{1}{r^2} \frac{\partial(\psi, D_r^2 \psi)}{\partial(r, \mu)} + \frac{2}{r^2} (D_r^2 \psi)(L_r \psi)\right] \quad (15)$$

where $\mathbf{T}$, the non-Newtonian portion of the extra stress tensor, is defined by

$$\mathbf{T} = \boldsymbol{\tau} - \mathbf{A}^{(1)} \quad (16)$$

The operators D_r^2 and L_r are those given by Goldstein (*23*, p. 115):

$$D_r^2 = \frac{\partial^2}{\partial r^2} + \frac{(1-\mu^2)}{r^2} \frac{\partial^2}{\partial \mu^2} \quad (17a)$$

$$L_r = \frac{\mu}{1-\mu^2} \frac{\partial}{\partial r} + \frac{1}{r} \frac{\partial}{\partial \mu} \quad (17b)$$

The function $f(\mathbf{T})$ is defined as

$$f(\mathbf{T}) = -\frac{\partial}{\partial r} \left\{ r^2 (1-\mu^2)^{1/2} \frac{\partial (T_{r\theta}/r)}{\partial r} + \frac{\partial[(1-\mu^2) T_{\theta\theta}]}{\partial \mu} + 4(1-\mu^2)^{1/2} T_{r\theta} + \mu T_{\theta\theta} + \mu T_{\phi\phi} \right\} + (1-\mu^2) \frac{\partial}{\partial \mu} \left\{ \frac{\partial T_{rr}}{\partial r} + \frac{1}{r} \frac{\partial[(1-\mu^2)^{1/2} T_{r\theta}]}{\partial \mu} + \frac{2}{r} T_{rr} - \frac{1}{r} T_{\theta\theta} - \frac{1}{r} T_{\phi\phi} \right\} \quad (18)$$

The method of matched asymptotic expansions is to be applied to the region outside the droplet. Near the droplet the stream function is assumed to have an expansion

$$\psi \sim \sum_{i=0}^{\infty} \varepsilon_i (N_{Re}) \psi_i (r, \mu) \quad (19)$$

where

$$\varepsilon_o (N_{Re}) = 1, \quad \lim_{N_{Re} \to 0} \left(\frac{\varepsilon_{n+1}}{\varepsilon_n}\right) = 0 \quad (20)$$

The n^{th} partial sum of Equation (19) is required to satisfy, to order ε_n, the equation of motion outside the droplet and the boundary conditions at the surface of the droplet. This statement may be illustrated in the following manner. All of the pertinent equations and boundary conditions may be put in the form

$$f(\psi) = 0 \tag{21}$$

The n^{th} partial sum of Equation (19) satisfies an equation of this form to order ε_n, if

$$\lim_{N_{Re}\to 0}\left[\frac{f\left(\sum_{i=0}^{n} \varepsilon_i(N_{Re})\psi_i(r,\mu)\right)}{\varepsilon_n}\right] = 0 \tag{22}$$

But such an expansion will not satisfy the uniform stream condition at infinity (*4*). To remedy this, "outer" variables are introduced:

$$\rho = rN_{Re}, \quad \Psi = N_{Re}^2 \psi \tag{23}$$

It follows that

$$\mathbf{v}^o = N_{Re}\,\mathbf{v}, \quad \mathbf{T}^o = \frac{1}{N_{Re}^2}\mathbf{T},$$

$$D_\rho^2 = \frac{1}{N_{Re}^2} D_r^2, \quad L_\rho = \frac{1}{N_{Re}} L_r \tag{24}$$

where the superscript o denotes an outer variable and D_ρ^2 and L_ρ are those operators defined by Equations (17) with r replaced by ρ. In terms of these variables, the equation of motion becomes

$$D_\rho^4\Psi = \frac{1}{\rho^2}\frac{\partial(\Psi, D_\rho^2\Psi)}{\partial(\rho,\mu)} + \frac{2}{\rho^2}(D_\rho^2\Psi)(L_\rho\Psi) + N_{Re}f_\rho(\mathbf{T}^o) \tag{25}$$

where f_ρ is given by Equation (18) with r replaced by ρ. This form of the equation of motion removes the explicit dependence on Reynolds number, except in the non-Newtonian terms; an asymptotic expansion based on this equation should be valid in the region far from the droplet. An expansion

$$\Psi \sim \sum_{i=0}^{\infty} \varepsilon_i(N_{Re})\Psi_i(\rho,\mu) \tag{26}$$

is defined whose n^{th} partial sum is required to satisfy, to order ε_n, Equation (25) and the uniform stream condition at infinity. The n^{th} partial sums of the expansions (19) and (26) are required to match to order ε_n in some region of space. More complete discussions of the method of matched asymptotic expansions are given in several sources (*5, 7, 8, 24, 25*).

An expansion similar to (19) is now defined for the region inside the droplet:

$$\hat{\psi} \sim \sum_{i=0}^{\infty} \varepsilon_i(N_{Re})\hat{\psi}_i(r,\mu) \tag{27}$$

Only one expansion is required for this region, since no singularities in the equation are expected.

The expansions (19), (26), and (27) imply similar expansions for velocity, stress, and all other flow quantities. Remembering that $\varepsilon_o \equiv 1$, we substitute these expansions into the appropriate governing equations and we take the limit to order unity in order to yield the equations for the initial approximation:

$$D_r^4\hat{\psi}_o = f(\hat{\mathbf{T}}_o) \tag{28}$$

$$D_r^4\psi_o = f(\mathbf{T}_o) \tag{29}$$

and

$$D_\rho^4\Psi_o = \frac{1}{\rho^2}\left\{\frac{\partial(\Psi_o, D_\rho^2\Psi_o)}{\partial(\rho,\mu)}\right\} + \frac{2}{\rho^2}(D_\rho^2\Psi_o)(L_\rho\Psi_o) \tag{30}$$

Due to the complexity of the terms of the form $f(\mathbf{T}_o)$, it is now necessary to define a further expansion of the stream function. Combining and rearranging Equations (1) and (16), we find that

$$\mathbf{T} = \lambda[\mathbf{A}^{(1)}\cdot\mathbf{A}^{(1)} + \beta_1\mathbf{A}^{(2)}] + \lambda^2[\beta_2\mathbf{A}^{(1)}\,tr\,(\mathbf{A}^{(1)})^2 + \beta_3\mathbf{A}^{(3)} + \beta_4(\mathbf{A}^{(1)}\cdot\mathbf{A}^{(2)} + \mathbf{A}^{(2)}\cdot\mathbf{A}^{(1)})] \tag{31}$$

Here we choose the characteristic time of the fluid $s_o = (\phi_3/\phi_1)$ and write the Weissenberg number λ as (*17*)

$$\lambda = \frac{\phi_3 U}{\phi_1 a} = \frac{\phi_3 N_{Re}}{a^2\rho} \tag{32}$$

For simplicity, we also define

$$\beta_1 = \frac{\phi_2}{\phi_3}, \quad \beta_2 = \frac{\phi_1\phi_6}{\phi_3^2}, \quad \beta_3 = \frac{\phi_1\phi_4}{\phi_3^2}, \quad \beta_4 = \frac{\phi_1\phi_5}{\phi_3^2} \tag{33}$$

Available data from the literature (see Appendix) indicate the magnitude of these coefficients for two fluids. The form of Equation (31) suggests that the stream function, both inside and outside the droplet, be further expanded in the form

$$\psi_i \sim \sum_{n=0}^{\infty} \lambda^n \psi_{in} \tag{34}$$

In order for this expansion to have a wide range of applicability, it is necessary for any given fluid that the Weissenberg number λ be very small. This is not an additional restriction on our development, however, since we stated in our discussion of Equation (1) that λ should be small in order to approximate the behavior of a Noll simple fluid by the behavior of a fluid of grade 3. It is not necessary to define an expansion of this nature for the outer region, since Equation (30) may be solved in its present form. Substituting expansions of the form of Equation (34) into Equations (28) and (29) and equating powers of λ, we have

$$D_r^4\hat{\psi}_{oo} = 0 \tag{35a}$$

$$D_r^4\hat{\psi}_{om} = f(\hat{\mathbf{T}}_{om}) \quad m > 1 \tag{35b}$$

$$D_r^4\psi_{oo} = 0 \tag{36a}$$

and

$$D_r^4\psi_{om} = f(\mathbf{T}_{om}) \quad m > 1 \tag{36b}$$

It follows from Equations (31) and (34) that terms of the form $\mathbf{T}_{om}$ depend only on previously obtained solutions, that is, $\psi_{oo}, \cdots, \psi_{o(m-1)}$. The governing equations are greatly simplified by this expansion.

The location of the bounding surface still remains a problem. Boundary conditions (*10a*) to (*10d*) are to be applied at the surface of the droplet, while the position in space of the droplet surface is initially unknown. It is possible to carry the unknown shape through the calculations and, remembering that the volume of the droplet must remain constant, solve for the shape of the droplet. We justify below taking a considerably simpler approach; the boundary conditions are applied at a specified surface and the true shape is calculated by an iterative procedure.

Having assumed axial symmetry, we represent the surface of the droplet by

$$R = 1 + \zeta(\mu) \tag{37}$$

For a small deformation $\zeta(\mu)$, Landau and Lifschitz (*26*) have obtained, correct to the first order in ζ

$$\frac{1}{R_1}+\frac{1}{R_2}=2-2\zeta-\frac{d}{d\mu}\left[(1-\mu^2)\frac{d\zeta}{d\mu}\right] \quad \zeta<<1 \tag{38}$$

Boundary condition (10*c*) then becomes

$$\mathbf{t}\cdot\mathbf{n}=\kappa\,\hat{\mathbf{t}}\cdot\mathbf{n}+\frac{N_{Re}}{N_{We}}\left\{2-2\zeta-\frac{d}{d\mu}\left[(1-\mu^2)\frac{d\zeta}{d\mu}\right]\right\}\mathbf{n} \quad \zeta<<1 \tag{39}$$

and the constant volume condition is, to the first order in ζ

$$\int_{-1}^{1}\zeta d\mu=0 \tag{40}$$

The origin of the coordinate system is now specified to be coincident with the center of mass of the droplet:

$$\int_{-1}^{1}\zeta\mu d\mu=0 \tag{41}$$

Hadamard and Rybczynski find that the assumption of a spherical shape is consistent with neglecting both inertial and non-Newtonian effects. It is reasonable to expect that the inclusion of these effects, on the scale of this problem, will produce only small deviations from spherical and that use of Equations (38) and (39) is justified.

The procedure used to obtain a solution is then as follows. The droplet is initially assumed to be spherical. This assumption overspecifies the problem and the normal stress boundary condition (39) is temporarily discarded. After the governing equations are solved subject to the remaining boundary conditions and the assumption of spherical shape, stresses are calculated and substituted into Equation (39), which is solved for the deformation. This procedure is repeated using the newly calculated shape as the initial assumption until the required accuracy is obtained. Further details and a mathematical justification of this method have been given elsewhere (*25*).

INITIAL APPROXIMATION

The governing equations for the initial approximation are given by Equations (30), (35*a*), and (36*a*), subject to the boundary conditions (10*a*), (10*b*), (10*d*) to (10*f*), and the matching condition. This is precisely the Hadamard-Rybczynski problem as discussed by Taylor and Acrivos (*4*). For this problem the outer solution is simply the uniform stream

$$\Psi_o=\frac{1}{2}\rho^2(1-\mu^2) \tag{42}$$

while the inner solution and the solution for the interior of the droplet are the Hadamard-Rybczynski solutions

$$\psi_{oo}=\frac{1-\mu^2}{4}\left\{2r^2-\frac{3\kappa+2}{\kappa+1}r+\frac{\kappa}{\kappa+1}\frac{1}{r}\right\} \tag{43}$$

and

$$\hat{\psi}_{oo}=\frac{(1-\mu^2)(r^2-r^4)}{4(\kappa+1)} \tag{44}$$

FIRST NON-NEWTONIAN PERTURBATION

No new outer solution need be obtained for this perturbation. The solution (42) is the full outer solution of zero order in Reynolds number and has been fully matched with ψ_{oo}. Further terms of the form ψ_{oj} in the inner expansion must not contribute to the outer solution.

From Equations (18), (31), and (34) to (36) and the Hadamard-Rybczynski solutions (43) and (44), the governing equations for the next perturbation are

$$D_r^4\psi_{01}=-\frac{3\mu(1-\mu^2)(1+\beta_1)(3\kappa+2)}{(\kappa+1)^2}\left(\frac{3\kappa+2}{r^5}-\frac{6\kappa}{r^7}\right) \tag{45}$$

and

$$D_r^4\hat{\psi}_{01}=0 \tag{46}$$

The solution of the homogeneous equation

$$D_r^4\psi=0 \tag{47}$$

that vanishes at $r=1$ and $\mu=\pm1$ is

$$\psi=\sum_{n=1}^{\infty}[A_n(r^{-n+2}-r^{-n})+B_n(r^{n+1}-r^{-n})+C_n(r^{n+3}-r^{-n})]\,Q_n(\mu) \tag{48}$$

where

$$Q_n(\mu)=\int_{-1}^{\mu}P_n(\mu)d\mu \tag{49}$$

and $P_n(\mu)$ is the Legendre polynomial of degree n. A particular integral of Equation (45) is

$$\psi_{01}=-\frac{(3\kappa+2)(1+\beta_1)}{4(\kappa+1)^2}\times\left[\frac{3\kappa+2}{r}+\frac{\kappa}{r^3}+\frac{a}{r^2}+b+cr^3+dr^5\right]Q_2(\mu) \tag{50}$$

and hence the general solutions of Equations (45) and (46) are

$$\psi_{01}=\frac{-(3\kappa+2)(1+\beta_1)}{4(\kappa+1)^2}\times\left[\frac{(3\kappa+2)}{r}+\frac{\kappa}{r^3}+\frac{a}{r^2}+b+cr^3+dr^5\right]Q_2(\mu)+\sum_{n=1}^{\infty}[A_n(r^{-n+1}-r^{-n})+B_n(r^{n+1}-r^{-n})+C_n(r^{n+3}-r^{-n})]\,Q_n(\mu) \tag{51}$$

and

$$\hat{\psi}_{01}=\sum_{n=1}^{\infty}[\hat{A}_n(r^{-n+1}-r^{-n})+\hat{B}_n(r^{n+1}-r^{-n})+\hat{C}_n(r^{n+3}-r^{-n})]\,Q_n(\mu) \tag{52}$$

The boundary and matching conditions having been satisfied, these solutions become

$$\psi_{01}=\frac{(3\kappa+2)(1+\beta_1)}{20(\kappa+1)^2}\left[\frac{5\kappa^2+10\kappa+6}{\kappa+1}\left(1-\frac{1}{r^2}\right)-5(3\kappa+2)\left(\frac{1}{r}-\frac{1}{r^2}\right)+10\kappa\left(\frac{1}{r^2}-\frac{1}{r^3}\right)\right]Q_2(\mu)-\frac{3\kappa}{10(\kappa+1)^3}\left[(1-\beta_1)-\frac{\hat{\lambda}}{\lambda}(1-\hat{\beta}_1)\right]\times\left(1-\frac{1}{r^2}\right)Q_2(\mu) \tag{53}$$

$$\hat{\psi}_{01}=\frac{(3\kappa+2)\lambda(1+\beta_1)}{20(\kappa+1)^3\hat{\lambda}}-\frac{3\kappa\lambda}{10(\kappa+1)^3\hat{\lambda}}\times\left[(1-\beta_1)-\frac{\hat{\lambda}}{\lambda}(1-\beta_1)\right](r^5-r^3)Q_2(\mu) \tag{54}$$

SECOND NON-NEWTONIAN PERTURBATION

The solution for this perturbation closely parallels the solution for the first non-Newtonian perturbation. The governing equations for this perturbation are obtained from Equations (35) and (36) and the previously obtained solution:

$$D_r^4\psi_{02} = \sum_{i=6}^{13} [m_i + n_i\mu^2](1-\mu^2)r^{-i} \tag{55}$$

and

$$D_r^4\hat{\psi}_{02} = \hat{C}(1-\mu^2)r^2 \tag{56}$$

Here

$$\hat{C} = \frac{-1}{(\kappa+1)^4}\left\{\frac{7(1+\beta_1)(1+\hat{\beta}_1)(3\kappa+2)\lambda}{20(\kappa+1)\hat{\lambda}} \times [(\kappa+1)^2\mu_1 - 2] + 63\hat{\beta}_2 - 12\hat{\beta}_3 + 63\hat{\beta}_4\right\} \tag{57}$$

$$\mu_1 = \frac{12\kappa}{(1+\beta_1)(3\kappa+2)(\kappa+1)^2}\left[(1-\beta_1) - \frac{\hat{\lambda}}{\lambda}(1-\hat{\beta}_1)\right] \tag{58}$$

and the coefficients m_i and n_i are given in Table 1.

It is not difficult to show that particular integrals satisfying Equations (55) and (56) are

$$\psi_{02} = \sum_{i=2}^{9} m_i^* r^{-i} Q_1(\mu) + \left[\frac{4n_7}{315} r^{-3}\ln r + \sum_{i=2}^{9} n_i^* r^{-i}\right] Q_3(\mu) \tag{59}$$

and

$$\hat{\psi}_{02} = \frac{\hat{C}}{140} r^6 Q_1(\mu) \tag{60}$$

where the m_i^* and n_i^* are given in Table 2.

After adding the solutions to the homogeneous equations and satisfying the boundary and matching conditions, we obtain solutions for the second perturbation:

$$\psi_{02} = \sum_{n=-1}^{9} a_n r^{-n} Q_1(\mu) + \left[\frac{4n_7}{315} r^{-3}\ln r + \sum_{n=1}^{9} b_n r^{-n}\right] Q_3(\mu) \tag{61}$$

and

$$\hat{\psi}_{02} = [\hat{a}_1(r^2 - r^4) - \hat{a}_2(r^4 - r^6)]\, Q_1(\mu) + \hat{b}(r^4 - r^6)\, Q_3(\mu) \tag{62}$$

where

$$a_{-1} = \delta_1 + \frac{1}{6(\kappa+1)}\sum_{n=2}^{9}[n^2 + 3n(\kappa+1) - (3\kappa+4)]\, m_n^* - \frac{\hat{\lambda}^2\kappa\hat{C}}{105(\kappa+1)\lambda^2} \tag{63a}$$

$$a_0 = 0 \tag{63b}$$

$$a_1 = -a_{-1} - \sum_{n=2}^{9} m_n^* \tag{63c}$$

$$a_n = m_n^* \qquad 9 \ge n \ge 2 \tag{63d}$$

$$b_1 = \delta_2 + \frac{1}{14(\kappa+1)}\sum_{n=2}^{9}[n^2 + n(3+7\kappa) - 3(6+7\kappa)]\, n_n^* \tag{63e}$$

$$b_3 = n_3^* - b_1 - \sum_{n=2}^{9} n_n^* \tag{63f}$$

$$b_n = n_n^* \qquad n \ne 1, 3 \tag{63g}$$

$$\hat{a}_1 = \frac{\hat{C}(7\kappa+3)}{420(\kappa+1)} - \frac{\lambda^2}{6(\kappa+1)\hat{\lambda}^2}\sum_{n=2}^{9}(n^2-1)\, m_n^* - \frac{\lambda^2}{\hat{\lambda}^2}\delta_1 \tag{63h}$$

$$\hat{a}_2 = \frac{\hat{C}}{140} \tag{63i}$$

$$\hat{b} = \frac{-\lambda^2}{14(\kappa+1)\hat{\lambda}^2}\sum_{n=2}^{9}[n^2 - 4n + 3]\, n_n^* - \frac{\lambda^2}{\hat{\lambda}^2}\delta_2 \tag{63j}$$

$$\delta_1 = \left[\frac{2\kappa(3\kappa+2)}{75(\kappa+1)^5}(1+\beta_1)\left(1-\frac{\hat{\lambda}}{\lambda}\right) + \frac{7\kappa(3\kappa+2)}{150(\kappa+1)^5}(1+\beta_1)\beta_1\left(1-\frac{\hat{\beta}_1\hat{\lambda}}{\beta_1\lambda}\right) - \frac{3\kappa}{5(\kappa+1)^4}\beta_2\left(3\kappa^2+1-\frac{4\hat{\beta}_2\hat{\lambda}^2}{\beta_2\lambda^2}\right) + \frac{3\kappa}{10(\kappa+1)^4}\beta_3\left(1-\frac{\hat{\beta}_3\hat{\lambda}^2}{\beta_3\lambda^2}\right) - \frac{3\kappa}{5(\kappa+1)^4}\beta_4\left(3\kappa^2+1-\frac{4\hat{\beta}_4\hat{\lambda}^2}{\beta_4\lambda^2}\right) - \frac{(3\kappa+2)\mu_1}{600(\kappa+1)^2}\left(3\kappa-5-8\kappa\frac{\hat{\lambda}}{\lambda}\right) - \frac{(3\kappa+2)\mu_1}{600(\kappa+1)^2}\beta_1(1+\beta_1)\left(9\kappa-5-14\kappa\frac{\hat{\beta}_1\hat{\lambda}}{\beta_1\lambda}\right)\right] \tag{63k}$$

and

$$\delta_2 = \left[\frac{8\kappa(3\kappa+2)}{175(\kappa+1)^5}(1+\beta_1)\left(1-\frac{\hat{\lambda}}{\lambda}\right) - \frac{27\kappa(3\kappa+2)}{350(\kappa+1)^5}\beta_1(1+\beta_1)\left(1-\frac{\hat{\beta}_1\hat{\lambda}}{\beta_1\lambda}\right) - \frac{9\kappa}{35(\kappa+1)^4}\beta_2\left(3\kappa^2-4+\frac{\hat{\beta}_2\hat{\lambda}^2}{\beta_2\lambda^2}\right) - \frac{12\kappa}{35(\kappa+1)^4}\beta_3\left(1-\frac{\hat{\beta}_3\hat{\lambda}^2}{\beta_3\lambda^2}\right) + \frac{3\kappa}{35(\kappa+1)^4}\beta_4\left(9\kappa^2-7-\frac{2\hat{\beta}_4\hat{\lambda}^2}{\beta_4\lambda^2}\right) - \frac{2(9+7\kappa)}{2205(\kappa+1)}n_7 - \frac{(3\kappa+2)\mu_1}{350(\kappa+1)^2}(1+\beta_1)\left(3\kappa-5-8\kappa\frac{\hat{\lambda}}{\lambda}\right) + \frac{3(3\kappa+2)\mu_1}{700(\kappa+1)^2}\beta_1(1+\beta_1)\left(4\kappa-5-9\kappa\frac{\hat{\beta}_1\hat{\lambda}}{\beta_1\lambda}\right)\right] \tag{63m}$$

TABLE 1. SECOND NON-NEWTONIAN PERTURBATION-EQUATION COEFFICIENTS

I	II	III	IV	V	VI
m_6	$\frac{6(3\kappa+2)}{(\kappa+1)^3}\left[10\kappa^2+20\kappa+12-\mu_1(\kappa+1)^2\right]$	$\frac{6(3\kappa+2)}{(\kappa+1)^3}\left[30\kappa^2+60\kappa+32-\mu_1(\kappa+1)^2\right]$	0	$-6\left(\frac{3\kappa+2}{\kappa+1}\right)^2$	$-4\left(\frac{3\kappa+2}{\kappa+1}\right)^2$
m_7	$-100\left(\frac{3\kappa+2}{\kappa+1}\right)^2$	$-130\left(\frac{3\kappa+2}{\kappa+1}\right)^2$	0	$2\left(\frac{3\kappa+2}{\kappa+1}\right)^3$	$\left(\frac{3\kappa+2}{\kappa+1}\right)^3$
m_8	$\frac{-6(6\kappa+5)}{(\kappa+1)^3}\left[10\kappa^2+20\kappa+12-\mu_1(\kappa+1)^2\right] + \frac{300(3\kappa+2)(2\kappa+1)}{(\kappa+1)^2}$	$\frac{-6(6\kappa+5)}{(\kappa+1)^3}\left[10\kappa^2+20\kappa+12-\mu_1(\kappa+1)^2\right] + \frac{300(3\kappa+2)(2\kappa+1)}{(\kappa+1)^2} - \frac{720\kappa}{\kappa+1}$	0	$48\,\frac{\kappa(3\kappa+2)}{(\kappa+1)^2}$	$24\,\frac{\kappa(3\kappa+2)}{(\kappa+1)^2}$
m_9	$-270\,\frac{\kappa(3\kappa+2)}{(\kappa+1)^2}$	$-120\,\frac{\kappa(3\kappa+2)}{(\kappa+1)^2}$	$\frac{\kappa(3\kappa+2)^2}{(\kappa+1)^3}$	$\frac{-9\kappa(3\kappa+2)^2}{(\kappa+1)^3}$	$\frac{\kappa(3\kappa+2)^2}{(\kappa+1)^3}$
m_{10}	0	0	0	$-375\left(\frac{\kappa}{\kappa+1}\right)^2$	0
m_{11}	$90\left(\frac{\kappa}{\kappa+1}\right)^2$	$270\left(\frac{\kappa}{\kappa+1}\right)$	$-18\,\frac{\kappa^2(3\kappa+2)}{(\kappa+1)^3}$	$132\,\frac{\kappa^2(3\kappa+2)}{(\kappa+1)^3}$	$-114\,\frac{\kappa^2(3\kappa+2)}{(\kappa+1)^2}$
m_{12}	0	0	0	0	0
m_{13}	0	0	$-114\left(\frac{\kappa}{\kappa+1}\right)^3$	$-84\left(\frac{\kappa}{\kappa+1}\right)^3$	$-684\left(\frac{\kappa}{\kappa+1}\right)^3$
n_6	$\frac{-48(3\kappa+2)}{(\kappa+1)^3}\left[10\kappa^2+20\kappa+12-\mu_1(\kappa+1)^2\right]$	$\frac{-48(3\kappa+2)}{(\kappa+1)^3}\left[10\kappa^2+20\kappa+12-\mu_1(\kappa+1)^2\right] - 960\left(\frac{3\kappa+2}{\kappa+1}\right)$	0	$54\left(\frac{3\kappa+2}{\kappa+1}\right)^2$	$32\left(\frac{3\kappa+2}{\kappa+1}\right)^2$
n_7	$720\left(\frac{3\kappa+2}{\kappa+1}\right)^2$	$1170\left(\frac{3\kappa+2}{\kappa+1}\right)^2$	$-6\left(\frac{3\kappa+2}{\kappa+1}\right)^3$	$-36\left(\frac{3\kappa+2}{\kappa+1}\right)^3$	$-51\left(\frac{3\kappa+2}{\kappa+1}\right)^3$
n_8	$\frac{30(12\kappa+5)}{(\kappa+1)^3}\left[10\kappa^2+20\kappa+12-\mu_1(\kappa+1)^2\right] - 1500\,\frac{(3\kappa+2)(2\kappa+1)}{(\kappa+1)^2}$	$\frac{30(12\kappa+5)}{(\kappa+1)^3}\left[10\kappa^2+20\kappa+12-\mu_1(\kappa+1)^2\right] - 1500\,\frac{(3\kappa+2)(2\kappa+1)}{(\kappa+1)^2} + 7200\,\frac{\kappa}{\kappa+1}$	0	$-180\,\frac{\kappa(3\kappa+2)}{(\kappa+1)^2}$	$-240\,\frac{\kappa(3\kappa+2)}{(\kappa+1)^2}$
n_9	$\frac{-990\kappa(3\kappa+2)}{(\kappa+1)^2}$	$\frac{-4800\kappa(3\kappa+2)}{(\kappa+1)^2}$	$\frac{55\kappa(3\kappa+2)^2}{(\kappa+1)^3}$	$\frac{189\kappa(3\kappa+2)^2}{(\kappa+1)^3}$	$\frac{457\kappa(3\kappa+2)^2}{(\kappa+1)^3}$
n_{10}	0	0	0	$-225\left(\frac{\kappa}{\kappa+1}\right)^2$	0
n_{11}	$3510\left(\frac{\kappa}{\kappa+1}\right)^2$	$6570\left(\frac{\kappa}{\kappa+1}\right)^2$	$-42\,\frac{\kappa^2(3\kappa+2)}{(\kappa+1)^3}$	$\frac{-120\kappa^2(3\kappa+2)}{(\kappa+1)^3}$	$\frac{-354\kappa^2(3\kappa+2)}{(\kappa+1)^3}$
n_{12}	0	0	0	0	0
n_{13}	0	0	$-90\left(\frac{\kappa}{\kappa+1}\right)$	$-90\left(\frac{\kappa}{\kappa+1}\right)^3$	$-540\left(\frac{\kappa}{\kappa+1}\right)^3$

$$\mathrm{I} = \frac{(1+\beta_1)(3\kappa+2)}{40(\kappa+1)}\,\mathrm{II} + \frac{\beta_1(1+\beta_1)(3\kappa+2)}{40(\kappa+1)}\,\mathrm{III} + \frac{9\beta_2}{2}\,\mathrm{IV} + \frac{3\beta_3}{4}\,\mathrm{V} + \frac{3\beta_4}{4}\,\mathrm{VI}$$

FIRST INERTIAL PERTURBATION

The first inertial perturbation for flow past a spherical droplet has been worked out by Taylor and Acrivos (4). The solutions for the stream function are

$$\psi_{10} = \frac{-(3\kappa+2)}{8(\kappa+1)}\left[r^2 - \frac{(3\kappa+2)}{2(\kappa+1)}\,r + \frac{\kappa}{2(\kappa+1)}\,\frac{1}{r}\right]Q_1(\mu) + \frac{(3\kappa+2)}{8(\kappa+1)}\left[r^2 - \frac{(3\kappa+2)}{2(\kappa+1)}\,r + \frac{\kappa(5\kappa+4)}{10(\kappa+1)^2} - \frac{\kappa}{2(\kappa+1)}\,\frac{1}{r} + \frac{\kappa(5\kappa+6)}{10(\kappa+1)^2}\,\frac{1}{r^2}\right]Q_2(\mu) \qquad (64)$$

$$\hat{\psi}_{10} = \frac{(3\kappa+2)}{16(\kappa+1)^2} \times \left[(r^2 - r^4)\,Q_1(\mu) - \frac{4\kappa+5}{5(\kappa+1)}\,(r^3 - r^5)\,Q_2(\mu)\right] \qquad (65)$$

$$\Psi_1 = \frac{-(3\kappa+2)}{2(\kappa+1)}\,(1+\mu)\,[1 - e^{-\frac{1}{2}\rho(1-\mu)}] \qquad (66)$$

and

$$\varepsilon_1(N_{Re}) = N_{Re} \qquad (67)$$

No further solutions for flow past an assumed spherical shape will be calculated. The restriction is here imposed that

$$N_{Re} \le \lambda^2 << \lambda \qquad (68)$$

Since λ has already been assumed to be much less than unity, from Equation (32) this is equivalent to

$$\frac{1}{N_{Re}} >> \frac{\phi_3}{\rho a^2} >> \left(\frac{\phi_3}{\rho a^2}\right)^2 N_{Re} \ge 1 \qquad (69)$$

For a given fluid, these conditions may easily be achieved experimentally by a proper choice of U and a. The imposition of this restriction insures that none of the neglected terms will be of the order of magnitude of the included terms.

DRAG FORCE

The dimensionless drag force exerted by the exterior fluid on the droplet may be calculated from the formula

$$F_D = 2\pi \int_{-1}^{1} [\mu\tau_{rr} - \mu P + (1-\mu^2)^{\frac{1}{2}}\,\tau_{r\mu}]_{r=1}\, d\mu \qquad (70)$$

The generalized pressure at $r = 1$ may be determined, up to an additive constant, by integration of the μ-component of the equation of motion evaluated at $r = 1$. The drag force calculated by this procedure is

$$F_D = \frac{2\pi(3\kappa+2)}{(\kappa+1)} + \frac{\pi N_{Re}(3\kappa+2)^2}{4(\kappa+1)^2} + \lambda^2\left\{\frac{-2\pi}{3}\sum_{n=1}^{9}(n-1)(n+2)(n+4)\,a_n + \frac{\pi(3\kappa+2)}{75(\kappa+1)^4}(1+\beta_1)\,[3\mu_1(3\kappa-2)(\kappa+1)^2 + 2(15\kappa^2 - 19\kappa - 19)] + \frac{\pi(3\kappa+2)}{75(\kappa+1)^4}\,\beta_1(1+\beta_1)\,[3\mu_1(3\kappa-2)(\kappa+1)^2 + 2(15\kappa^2 - 34\kappa - 34)] + \frac{12\pi}{5(\kappa+1)^3}\,\beta_2(33\kappa^3 - 3\kappa^2 + 7\kappa + 6) - \frac{2\pi}{5(\kappa+1)^3}\,\beta_3(7\kappa - 10) + \frac{2\pi}{5(\kappa+1)^3}\,\beta_4(198\kappa^3 - 18\kappa^2 + 45\kappa + 38)\right\} \qquad (71)$$

The first non-Newtonian perturbation, ψ_{01}, is such that all terms inside the integral in (70) are of odd order in μ. The quantity ψ_{01} does not contribute to the drag force and the non-Newtonian parameter λ does not appear in the solution for the drag force until terms of the second power in λ are reached.

TABLE 2. SECOND NON-NEWTONIAN PERTURBATION-SOLUTION COEFFICIENTS

i	m_i^*	n_i^*
2	$-\frac{1}{36}(m_6 + \frac{n_6}{5})$	$\frac{n_6}{30}$
3	$-\frac{1}{140}(m_7 + \frac{n_7}{5})$	0
4	$-\frac{1}{360}(m_8 + \frac{n_8}{5})$	$-\frac{n_8}{150}$
5	$-\frac{1}{756}(m_9 + \frac{n_9}{5})$	$-\frac{n_9}{495}$
6	$-\frac{1}{1400}(m_{10} + \frac{n_{10}}{5})$	$-\frac{n_{10}}{1125}$
7	$-\frac{1}{2376}(m_{11} + \frac{n_{11}}{5})$	$-\frac{n_{11}}{2145}$
8	$-\frac{1}{3780}(m_{12} + \frac{n_{12}}{5})$	$-\frac{n_{12}}{3675}$
9	$-\frac{1}{5720}(m_{13} + \frac{n_{13}}{5})$	$-\frac{n_{13}}{5850}$

DEFORMATION

The boundary condition (39) may be rewritten in terms of the extra stress tensor, the generalized pressure, and the body force potential:

$$2 - 2\zeta - \frac{d\zeta}{d\mu}\left[(1-\mu^2)\frac{d\zeta}{d\mu}\right] = \frac{N_{We}}{N_{Re}}\left[\tau_{rr} - \kappa\hat{\tau}_{rr} - P + \kappa\hat{P} + \frac{a}{U}\,\varphi\left(\frac{\rho}{\phi_1} - \kappa\,\frac{\hat{\rho}}{\hat{\phi}_1}\right)\right]_{r=1} \qquad (72)$$

The body force potential (dimensional) is specified to be of the form

$$\varphi = g a r \mu \qquad (73)$$

An overall force balance on the droplet then yields

$$F_D = \frac{4\pi a^2}{3\phi_1 U}\, g(\hat{\rho} - \rho) \tag{74}$$

and Equation (72) becomes

$$2 - 2\zeta - \frac{d}{d\mu}\left[(1-\mu^2)\frac{d\zeta}{d\mu}\right] = \frac{N_{We}}{N_{Re}}\left[\tau_{rr} - \kappa\hat{\tau}_{rr} - P + \kappa\hat{P} - \frac{3\mu}{4\pi}F_D\right]_{r=1} \tag{75}$$

The solution of this equation subject to conditions (40) and (41) is

$$\begin{aligned}
\zeta = {} & \frac{-N_{We}}{960(\kappa+1)^3}\left[243\kappa^3 + 684\kappa^2 + 638\kappa + 200 - 20\frac{\hat{\rho}}{\rho}(\kappa+1)\right]P_2(\mu) \\
& + \frac{\lambda N_{We}}{80N_{Re}(\kappa+1)^3}\left\{19\kappa^2 + 30\kappa + 16 + \beta_1(15\kappa^3 + 42\kappa^2 + 44\kappa + 24) - \frac{\hat{\lambda}}{\lambda}\left[23\kappa^2 + 17\kappa - 2\hat{\beta}_1(14\kappa^2 + 11\kappa)\right]\right\}P_2(\mu) \\
& + \frac{\lambda^2 N_{We}}{N_{Re}}\left\{\frac{11}{9450}n_7 - \frac{1}{120}\sum_{n=1}^{9}(n+2)(n^2+n-36)b_n + \frac{\kappa\hat{\lambda}^2}{5\lambda^2}\hat{b}\right. \\
& + \frac{(3\kappa+2)}{1500(\kappa+1)^4}(1+\beta_1)\left[30\kappa^2 - 37\kappa - 58 + \frac{9}{2}\mu_1(3\kappa+2)(\kappa+1)^2 + 5\kappa\frac{\hat{\lambda}}{\lambda} - \frac{75}{2}\mu_1\kappa(\kappa+1)^2\frac{\hat{\lambda}}{\lambda}\right] \\
& + \frac{(3\kappa+2)}{750(\kappa+1)^4}\beta_1(1+\beta_1)\left[15\kappa^2 - 5\kappa + 16 + \frac{9}{2}\mu_1(5\kappa-3)(\kappa+1)^2 - 110\kappa\frac{\hat{\beta}_1\hat{\lambda}}{\beta_1\lambda} + 55\mu_1\kappa(\kappa+1)^2\frac{\hat{\beta}_1\hat{\lambda}}{\beta_1\lambda}\right] \\
& - \frac{3}{100(\kappa+1)^3}\beta_2\left[27\kappa^3 + 42\kappa^2 - 20\kappa - 24\right] + \frac{1}{100(\kappa+1)^3}\beta_3\left[603\kappa + 20 - 8\kappa\frac{\hat{\beta}_3\hat{\lambda}^2}{\beta_3\lambda^2}\right] \\
& \left. - \frac{1}{100(\kappa+1)^3}\beta_4\left[81\kappa^3 + 126\kappa^2 - 51\kappa - 56 - 10\kappa\frac{\hat{\beta}_4\hat{\lambda}^2}{\beta_4\lambda^2}\right]\right\}P_3(\mu)
\end{aligned} \tag{76}$$

FLOW PAST DEFORMED SHAPE

We desire to determine, by an analysis of the flow past the deformed shape, those additional terms that are of the order of magnitude of the terms already included in the solution. With these limited goals in mind, it is not necessary to use the complete deformed shape given by Equation (76), which is of the form

$$\zeta = N_{We}\zeta_{10}P_2(\mu) + \frac{\lambda N_{We}}{N_{Re}}\zeta_{01}P_2(\mu) + \frac{\lambda^2 N_{We}}{N_{Re}}\zeta_{02}P_3(\mu) \tag{77}$$

It has been shown elsewhere (25) that if the problem is restated and solved with this shape as the initial assumed shape, the newly found deformation will be of the form

$$\zeta' = \zeta + \left[\frac{N_{We}^2}{N_{Re}}\zeta_{10}f_1(\mu) + \frac{\lambda N_{We}^2}{N_{Re}^2}\zeta_{01}f_2(\mu) + \frac{\lambda^2 N_{We}^2}{N_{Re}^2}\zeta_{02}f_3(\mu)\right] \tag{78}$$

From Equation (68) we see that the only new deformation term which may not be shown to be negligible with respect to ζ is $(\lambda N_{We}^2/N_{Re}^2)\zeta_{01}f_2(\mu)$. But this deformation may be obtained by considering the "creeping flow" past a droplet of spheroidal surface shape given by

$$R = 1 + \frac{\lambda N_{We}}{N_{Re}}\zeta_{01}P_2(\mu) \tag{79}$$

Such a problem has been solved by Taylor and Acrivos (4). Adapting their results to this case, we obtain for the additional terms due to the deformation:

$$\psi' = \frac{-\lambda N_{We}\zeta_{01}}{10N_{Re}(\kappa+1)^2}\times\left[(3\kappa^2 - \kappa + 8)r - 3(\kappa^2 - \kappa + 2)\frac{1}{r}\right]Q_1(\mu) + \frac{6\lambda N_{We}\zeta_{01}}{5N_{Re}(\kappa+1)}\times\left[\left(\frac{3\kappa}{2} + \frac{9}{7}\right)\frac{1}{r} - \left(\frac{3\kappa}{2} + \frac{2}{7}\right)\frac{1}{r^3}\right]Q_3(\mu) \tag{80a}$$

$$\hat{\psi}' = \frac{-\lambda N_{We}\zeta_{01}}{5N_{Re}(\kappa+1)^2}\left[2(2-\kappa)r^4 + 3(\kappa-1)r^2\right]Q_1(\mu) - \frac{6\lambda N_{We}\zeta_{01}}{35N_{Re}(\kappa+1)}(5r^6 - 12r^4)Q_3(\mu) \tag{80b}$$

$$F_D' = \frac{-2\pi\lambda N_{We}\zeta_{01}}{5N_{Re}(\kappa+1)^2}(3\kappa^2 - \kappa + 8) \tag{80c}$$

$$\zeta' = \frac{3(11\kappa + 10)\lambda N_{We}^2\zeta_{01}}{70N_{Re}^2(\kappa+1)}P_3(\mu) \tag{80d}$$

DISCUSSION

The principal results of this investigation, the drag force and the deformation, may be written in the form

$$F_D = B_0 + N_{Re}B_1 + \lambda^2 B_2 + \frac{\lambda N_{We}}{N_{Re}}B_3 \tag{81}$$

and

$$\zeta = N_{We}C_1P_2(\mu) + \frac{\lambda N_{We}}{N_{Re}}C_2P_2(\mu) + \frac{\lambda^2 N_{We}}{N_{Re}}C_3P_3(\mu) + \frac{\lambda N_{We}^2}{N_{Re}^2}C_4P_3(\mu) \tag{82}$$

where the constants B_n and C_n are functions of κ, $\hat{\rho}/\rho$, $\hat{\tau}/\tau$, $\hat{\beta}_i/\beta_i$, and the β_i and may be determined from Equations (71), (76), (80c), and (80d). The coefficients B_n and C_n are readily calculable once the physical characteristics of the system are known.

These solutions, as well as those for the stream functions, reduce to the Newtonian case in the limit as the Weissenberg number λ approaches zero. For a Newtonian fluid moving past a Newtonian droplet, these results agree with those of Taylor and Acrivos (4). For a fluid of grade 3 flowing past a solid sphere, these results are in agreement with those obtained by Giesekus (27).

Qualitative comparisons may be made between these results and some of the experimental data in the literature. For the solid sphere case, these results predict that the first non-Newtonian correction to the Stokes drag (dimensional) will be proportional to U^3/a. Tanner (*28*) and Caswell (*29*) attempted to confirm this experimentally. Both researchers used steel balls and polyisobutylene solutions; Tanner employed carbon tetrachloride as the solvent and Caswell decalin. They found general agreement with the theory for very slow flows, although they consider their results to be of a preliminary nature.

Griffith (*30*) indicates that surfactants, even in small concentrations, produce sizable deviations from a drag force predicted on the assumption of their absence. Wasserman and Slattery (*10*) confirm this theoretically. Unfortunately, no experimental studies to date have reported quantitative data on the effects of surfactants upon drag data. Since surfactants are nearly impossible to eliminate from an experiment, we will not discuss experimental drag measurements for a droplet here. We limit our comments to experimental observations of droplet deformation, since theoretical investigations (*3*, Chapter 8; *10*) suggest that surfactants have only a negligible effect on the shape of a droplet.

Several investigators (*9*, *31* to *35*) have observed, and qualitatively described, shapes of droplets falling through a second fluid. For a non-Newtonian bulk fluid, these investigators have observed that as the size, and hence the velocity, of the droplet was increased, the shape progressed from spherical to prolate spheroidal to ovate with the large end leading and finally to a teardrop shape with an extended rear. Although quantitative data are lacking, the deformation solution (82) may serve to explain this sequence of shapes. If the first two terms on the right-hand side of Equation (82) combine to yield a positive number multiplying $P_2(\mu)$ and the last two terms yield a positive number multiplying $P_3(\mu)$, the sequence of predicted shapes would be from spherical to prolate spheroidal to ovate with the large end leading. Figure 1, while not meant to be an exact representation of shapes for any particular problem, illustrates how the equations may yield prolate spheroidal and approximately ovate shapes.

Taylor and Acrivos have shown that the coefficient C_1 in Equation (82) is always negative. For the case of a water droplet we compare Equations (76) and (82) and, assuming that $\beta_1 < 0$, as indicated by the experimental data in the Appendix, find that C_2 is always positive and of the same order of magnitude as C_1. From Equation (68) we have

$$\lambda >> N_{Re} \quad (83)$$

and thus the first two terms on the right in our deformation solution (82) do combine to yield a positive number multiplying $P_2(\mu)$.

We also see that C_4 will always be a positive number as it is merely a multiple of C_2. Nothing quite so general may be said about C_3. However, several sample calculations have been made and the results are given in Table 3. The first two lines of Table 3 correspond to the data of Markovitz (see Appendix) for solutions of polyisobutylene in cetane at concentrations of 3.9 and 6.9%. The next two lines are the same except for the choice of β_1. Here we

TABLE 3. DROPLET SHAPE-SAMPLE CALCULATIONS

ϕ_1, poise	β_1	C_3	C_4
4.8	−0.42	−0.0214	0.139
60	−0.28	−0.0232	0.120
4.8	−0.50	−0.0205	0.150
60	−0.50	−0.0205	0.150
4.8	−0.58	−0.0184	0.160
60	−0.72	−0.0133	0.180

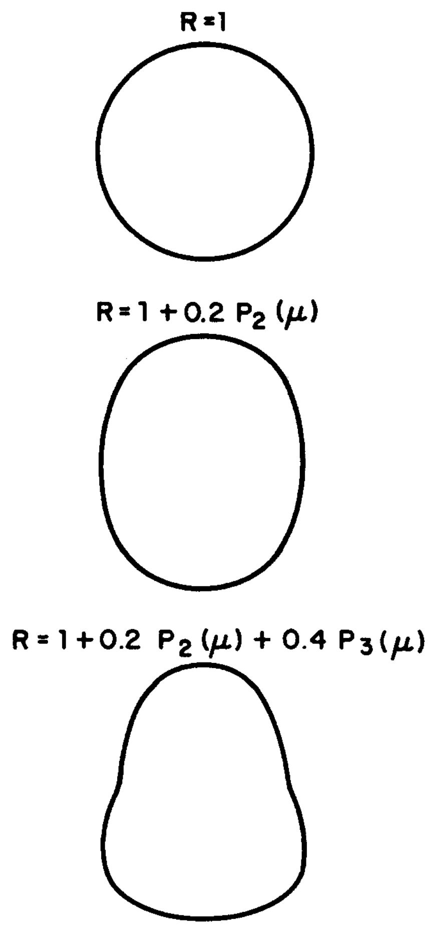

Fig. 1. Predicted droplet shapes.

have taken the value of β_1 that is dictated by the Weissenberg hypothesis. The last two lines of the table use values of β_1 which would correspond to a second normal stress difference of the same magnitude but of opposite sign from Markovitz's data. All of these calculations are based on a Newtonian droplet of viscosity 0.01 poise.

This wide range of properties produces a surprisingly narrow variation in the calculated values of C_3 and C_4. If one assumes that the density of the bulk phase is equal to that of pure cetane and that interfacial tension is 70 dynes/cm., then the last two terms in Equation (82) will combine to yield a positive term multiplying $P_3(\mu)$ for all cases considered in Table 3 except for an extremely small droplet. None of the values of C_3 and C_4 in Table 3 will produce a negative coefficient of $P_3(\mu)$ unless the droplet equivalent radius is less than 0.08 cm.

Thus we see that for the case of a Newtonian droplet falling through a non-Newtonian fluid, the same sequence of droplet shapes is predicted for a wide range of bulk fluid properties. This sequence is in agreement with the shapes that have been reported in the literature.

ACKNOWLEDGMENT

The support provided the authors by the National Science Foundation (M. G. Wagner, an N.S.F. Cooperative Graduate Fellowship awarded through Northwestern University; J. C. Slattery, GK-450) is gratefully acknowledged. The assistance provided by Professor Bruce Caswell in reconciling the results of this investigation with those of previous authors has been invaluable.

NOTATION

a = equivalent radius of droplet, that is, the radius of a spherical droplet of the same volume, cm.

$\mathbf{A}^{(i)}$ = Rivlin-Ericksen tensor of order i, Equations (6), (7)
B_i = coefficients in drag force equation, Equation (81)
$\hat{C}$ = coefficient in the equation for the second non-Newtonian perturbation, Equation (56)
C_i = coefficients in deformation solution, Equation (82)
$\mathbf{d}$ = rate of deformation tensor
D^2 = differential operator, Equation (17a)
$\mathbf{f}$ = body force vector, Equation (3)
F_D = drag force
g = body force constant, cm./sec.^{2}, Equation (78)
L = differential operator, Equation (17b)
m_i = coefficients in the equation for the second non-Newtonian perturbation, Equation (55))
m_i^* = coefficients in the solution for the second non-Newtonian perturbation, Equation (59)
$\mathbf{n}$ = unit normal vector
n_i = coefficients in the equation for the second non-Newtonian perturbation, Equation (55)
n_i^* = coefficients in the solution for the second non-Newtonian perturbation, Equation (59)
N_{Re} = Reynolds number
N_{We} = Weber number, Equation (12)
N_{Wi} = Weissenberg number
p = pressure, dynes/sq. cm.
P = generalized pressure, Equation (4)
P_n = Legendre polynomial of degree n
Q_n = integral of Legendre polynomial of degree n, Equation (54)
r = spherical coordinate
R = radius at the surface, measured from the centroid of the droplet
R_1, R_2 = principal radii of curvature of droplet surface
s_o = characteristic time of the fluid
$\mathbf{t}$ = stress tensor
$\mathbf{T}$ = non-Newtonian stress tensor, Equation (16)
$\mathbf{T}_{ij}$ = i^{th} inertial and j^{th} non-Newtonian term in the double expansion for the non-Newtonian stress tensor
U = magnitude of the free stream velocity, cm./sec.
$\mathbf{v}$ = velocity vector

Greek Letters

β_i = constants in the Rivlin-Ericksen equation, Equation (31)
ε_i = perturbation parameter
ζ = deformation, Equation (37)
ζ_{ij} = i^{th} inertial and j^{th} non-Newtonian term in double expansion of the deformation
θ = spherical coordinate
κ = ratio of zero shear viscosities, Equation (11)
λ = Weissenberg number (*47*) as defined by Equation (32)
μ = (cos θ), Equation (14)
ν = unit tangent vector
ρ = density, g./cc., outer radial variable
σ = surface tension, dynes/cm.
τ = extra stress tensor
ϕ_i = coefficients in the Rivlin-Ericksen equation, Equation (5)
φ = body force potential, sq. cm./sec.^{2}, Equation (3)
ψ = stream function
ψ_i = i^{th} term in inertial perturbation expansion of ψ, Equation (19)
ψ_{ij} = j^{th} term in non-Newtonian perturbation expansion of ψ_i, Equation (34)
Ψ = outer stream function
Ψ_i = i^{th} term in inertial perturbation expansion of Ψ, Equation (26)

Superscripts

$\hat{}$ = variable referring to interior of droplet
$'$ = additional term due to deformation
o = outer variable

LITERATURE CITED

1. Hadamard, J., *Compt. Rend.*, **152**, 1735 (1911).
2. Rybczynski, W., *Bull. Acad. Sci. Cracovie*, **A40**, (1911).
3. Levich, V. G., "Physicochemical Hydrodynamics," Chapt. 8, Prentice-Hall, Englewood Cliffs, N.J. (1962).
4. Taylor, T. D., and A. Acrivos, *Chem. Eng. Sci.*, **19**, 466 (1964).
5. Kaplun, S., and P. A. Lagerstrom, *J. Math. Mech.*, **6**, 585 (1957).
6. Kaplun, S., *ibid.*, **6**, 595 (1957).
7. Proudman, I., and J. R. A. Pearson, *J. Fluid Mech.*, **2**, 237 (1957).
8. Van Dyke, M., "Perturbation Methods in Fluid Dynamics," Academic Press, New York (1964).
9. Wellek, R. M., A. K. Agrawal, and A. H. P. Skelland, *AIChE J.*, **12**, 854 (1966).
10. Wasserman, M. L., and J. C. Slattery, *AIChE J.* (to be published).
11. Coleman, B. D., H. Markovitz, and W. Noll, "Viscometric Flows of Non-Newtonian Fluids," Springer-Verlag, New York (1966).
12. Truesdell, C., and W. Noll, "Handbuch der Physik," S. Flügge, ed., Vol. 3/3, Springer-Verlag, Berlin (1965).
13. Coleman, B. D., and W. Noll, *Phys. Fluids*, **5**, 840 (1962).
14. Slattery, J. C., *ibid.*, **7**, 1913 (1964).
15. Coleman, B. D., and W. Noll, *Arch. Rat. Mech. Anal.*, **6**, 355 (1960).
16. Truesdell, C., *Phys. Fluids*, **7**, 1134 (1964).
17. Slattery, J. C., *AIChE J.*, **14**, 517 (1968).
18. Coleman, B. D., and W. Noll, *Ann. New York Acad. Sci.*, **89**, 672 (1961).
19. Rivlin, R. S., and J. L. Ericksen, *J. Rat. Mech. Anal.*, **4**, 323 (1955).
20. Scriven, L. E., *Chem. Eng. Sci.*, **12**, 98 (1960).
21. Slattery, J. C., *ibid.*, **19**, 379 (1964).
22. ———, *Ind. Eng. Chem. Fundamentals*, **6**, 108 (1967).
23. Goldstein, S., "Modern Developments in Fluid Dynamics," Oxford Univ. Press, London, New York (1938).
24. Erdelyi, A., *Atti. Acad. Sci. Torino Cl. Sci. Fis. Mat. Nat.*, **95**, 651 (1961).
25. Wagner, M. G., Ph.D. thesis, Northwestern Univ., Evanston, Ill. (1967).
26. Landau, L. D., and E. M. Lifschitz, "Fluid Mechanics," Addison-Wesley, Reading, Mass. (1959).
27. Giesekus, H., *Rheol. Acta*, **3**, 59 (1963).
28. Tanner, R. I., *Quart. J. Mech. Appl. Math.*, **14**, 47 (1961).
29. Caswell, B., Ph.D. thesis, Stanford Univ., Palo Alto, Calif. (1963).
30. Griffith, R. M., *Chem. Eng. Sci.*, **17**, 1057 (1962).
31. Astarita, G., and G. Apuzzo, *AIChE J.*, **11**, 815 (1965).
32. Fararoui, A., and R. C. Kintner, *Trans. Soc. Rheol.*, **5**, 369 (1961).
33. Garner, F. H., K. D. Mathur, and V. G. Jenson, *Nature*, **180**, 331 (1957).
34. Mhatre, M. V., and R. C. Kintner, *Ind. Eng. Chem.*, **51**, 865 (1959).
35. Warshay, H., E. Bogusz, M. Johnson, and R. C. Kintner, *Can. J. Chem. Eng.*, **37**, 29 (1959).
36. Philippoff, W., *J. Appl. Phys.*, **28**, 984 (1956).
37. Markovitz, H., unpublished data, quoted in Truesdell, C., *Phys. Fluids*, **7**, 1134 (1964).

APPENDIX

The data of Philippoff (*36*) for a 15% solution of polyisobutylene in decalin at 30°C. yield:

$$\phi_1 = 9{,}320 \text{ poise}$$

$$\phi_2 = -22{,}500 \text{ dynes/(sq. cm.)(sec.}^2)$$

$$\phi_3 = 45{,}000 \text{ dynes/(sq. cm.)(sec.}^2)$$

$$\phi_5 + \phi_6 = 22{,}000 \text{ dynes/(sq. cm.)(sec.}^3)$$

where ϕ_2 was obtained on the assumption that $\phi_2 = -\frac{1}{2}\phi_3$. Thus

$$\beta_1 = -0.5, \quad \beta_2 + \beta_4 = 0.1$$

Markovitz (*37*) reports the following data for solutions of polyisobutylene in cetane:

Concentration, %	ϕ_1, poise	ϕ_2, g./cm.	ϕ_3, g./cm.	β_1
3.9	4.8	−0.050	0.12	−0.42
5.4	18.5	−0.30	1.1	−0.27
6.9	60	−2.1	7.6	−0.28

Manuscript received February 5, 1968; revision received April 14, 1969; paper accepted July 7, 1969. Paper presented at AIChE New York meeting.