TIME-DEPENDENT DROP DEFORMATION
IN A ROTATING HIGH VISCOSITY FLUID

By

H. A. STONE (*Division of Applied Sciences, Harvard University, Cambridge, MA*)

AND

J. W. M. BUSH (*Dept. of Applied Mathematics and
Theoretical Physics, University of Cambridge, England*)

Abstract. An exact solution is presented which describes the time-dependent deformation of a nearly spherical drop suspended on the rotation axis of a more dense rotating viscous fluid. The solution is demonstrated to be similar, though not identical, to that derived from the commonly invoked assumption that the external flow field is purely extensional.

1. Introduction. The spinning drop tensiometer is a device used to measure the interfacial tension between two fluids [12]. A drop of liquid less dense than the suspending fluid is placed in a horizontally oriented circular cylinder which is rotated about its symmetry axis. Centrifugal forces drive the lighter drop fluid along the rotation axis, while deformation of the drop is resisted by interfacial tension forces as well as viscous forces in the two fluid phases. The interfacial tension may be inferred from the final equilibrium shape of the drop, which necessarily corresponds to a prolate ellipsoidal form. Moreover, rheological information on the drop and suspending fluids may be deduced from the time-dependent distortion of the drop.

Previous studies analyzing the time-dependent features of this problem [5, 7] have assumed that since the drop is being stretched, the fluid motion may be analyzed as if the drop has been placed in an extensional flow. Hsu and Flumerfelt [5] indicated that this approximation is only valid for highly distorted drop shapes. Recently, Hu and Joseph [6] numerically solved the Navier-Stokes equations using a finite-element method to determine the time-dependent shape of the drop in a spinning drop device; the limit of highly stretched shapes was studied in some detail.

Here we present an exact solution for the fluid motion and time-dependent drop shape in the case that the drop remains nearly spherical. Wall effects are neglected, and fluid motions both inside and outside the drop are assumed to be dominated by viscous effects and so may be described as centrifugally-forced Stokes flows. The problem is solved by applying a vector solution method [e.g., 4] and is a suitable example problem for a graduate course on viscous flows [e.g., 2, 8].

2. Problem statement. Consider an unbounded Newtonian fluid with viscosity μ and density ρ rotating with constant angular velocity $\boldsymbol{\Omega}$. An initially spherical drop of Newtonian liquid with viscosity $\lambda\mu$, density $\rho - \Delta\rho$, and undeformed radius a, is placed on the rotation axis, where its position is stable provided $\Delta\rho > 0$. The interface between the drop and suspending fluids is characterized by a constant interfacial tension γ. The centrifugal pressure field dominates the gravitational pressure field provided the local gravitational acceleration g satisfies $g/(a\Omega^2) \ll 1$; gravitational effects are henceforth neglected. We proceed by calculating the centrifugally-driven time-dependent distortion of the drop.

It is convenient to describe the fluid motion relative to a coordinate system that rotates with angular velocity $\boldsymbol{\Omega}$ [e.g., 3]. The suspending and drop fluids are assumed to be sufficiently viscous that inertial and Coriolis forces are dynamically unimportant throughout the fluid domain. Balancing the characteristic centrifugal pressure $O(\Delta\rho\Omega^2 a^2)$ and the viscous stresses $O(\mu u/a)$ yields a characteristic velocity with magnitude $u = O(\Delta\rho\Omega^2 a^3/\mu)$. Coriolis forces may be neglected relative to viscous forces provided the Taylor number is sufficiently small, $\Omega a^2/\nu \ll 1$, and inertial effects may be likewise neglected provided the Reynolds number is sufficiently small, $ua/\nu = \Delta\rho\Omega^2 a^4/(\rho\nu^2) \ll 1$. In the low Taylor and Reynolds number limit that is considered here, the fluid motions inside and outside the drop are described by Stokes equations.

Choosing $a, \Delta\rho\Omega^2 a^3/\mu, \Delta\rho\Omega^2 a^2$, and $\lambda\Delta\rho\Omega^2 a^2$ as the characteristic length, velocity, pressure in the external fluid, and pressure in the drop, we may write the dimensionless governing equations as

$$\text{external fluid: } \nabla \cdot \boldsymbol{\sigma} = -\nabla p_d + \nabla^2 \mathbf{u} = \mathbf{0}, \quad \text{drop fluid: } \nabla \cdot \hat{\boldsymbol{\sigma}} = -\nabla \hat{p}_d + \nabla^2 \hat{\mathbf{u}} = \mathbf{0}, \quad (1)$$

$$\nabla \cdot \mathbf{u} = 0, \qquad\qquad\qquad\qquad \nabla \cdot \hat{\mathbf{u}} = 0.$$

Here we denote the velocity, dynamic pressure, and stress fields as $(\mathbf{u}, p_d, \boldsymbol{\sigma})$ and carets indicate the drop fluid variables. The dimensional dynamic pressure differs from the actual fluid pressure by incorporating the centrifugal term: $p_d = p + \rho(\boldsymbol{\Omega} \wedge \mathbf{r})^2$, where $\mathbf{r}$ denotes the position vector measured from the rotation axis. The centrifugal pressure term is the signature of the background rotation and appears explicitly in the boundary conditions.

The boundary conditions of continuity of velocity and tangential stress and the normal stress balance are

$$\mathbf{u} - \hat{\mathbf{u}} = \mathbf{0} \qquad\qquad \text{on } S, \qquad\qquad (2)$$

$$\mathbf{t} \cdot \mathbf{n} \cdot \boldsymbol{\sigma} - \lambda \mathbf{t} \cdot \mathbf{n} \cdot \hat{\boldsymbol{\sigma}} = 0 \qquad\qquad \text{on } S, \qquad\qquad (3)$$

and

$$\mathbf{n} \cdot (\mathbf{n} \cdot \boldsymbol{\sigma} - \lambda \mathbf{n} \cdot \hat{\boldsymbol{\sigma}}) = \frac{1}{\mathcal{B}}\nabla_s \cdot \mathbf{n} + \tfrac{1}{2}(\widehat{\boldsymbol{\Omega}} \wedge \mathbf{r})^2 \quad \text{on } S, \qquad\qquad (4)$$

where $\mathbf{n}$ and $\mathbf{t}$ denote the unit normal and tangent vectors along the deformed surface S, $\nabla_s \cdot \mathbf{n}$ represents the interface curvature and $\widehat{\boldsymbol{\Omega}} = \boldsymbol{\Omega}/\Omega$ is a unit vector aligned with

the rotation axis. The rotational Bond number, $\mathcal{B} = \nabla\rho\Omega^2 a^3/\gamma$, prescribes the relative magnitudes of the centrifugal stresses causing deformation and interfacial tension stresses resisting distortion. At large distances from the drop $|\mathbf{u}| \to 0$. The solution we describe here is valid for small Bond numbers, $\mathcal{B} \ll 1$.

To describe the nearly spherical shape distortions we write the drop shape as

$$r = 1 + \varepsilon f(\theta, \phi, t), \tag{5}$$

where $\varepsilon \ll 1$. We will see that the small parameter ε is in fact equal to the rotational Bond number. The normal velocity along the interface dictates the time-rate-of-change of the unknown shape function $f(\theta, \phi, t)$:

$$\mathbf{u} \cdot \mathbf{n} = \hat{\mathbf{u}} \cdot \mathbf{n} = \frac{\partial f}{\partial t}, \tag{6}$$

where a convective time scale, $\mu/(\Delta\rho\Omega^2 a^2)$, is chosen as the characteristic time. At long times (see Eq. 14) a quiescent steady state is established in which there is a balance between the centrifugal and interfacial stresses. The resulting steady drop shapes may be determined analytically as a function of $\mathcal{B}$ [11].

3. Solution. The centrifugal term $(\widehat{\mathbf{\Omega}} \wedge \mathbf{r})^2$ is the driving force responsible for fluid motion and drop deformation. Since Stokes equations are linear we expect the solution for the velocity field to be linear in the dyadic $\widehat{\mathbf{\Omega}}\widehat{\mathbf{\Omega}}$. We thus see a departure from the commonly applied mathematical model of a drop in a straining flow, in which the far-field is described by a rate of strain tensor $\mathbf{E}^\infty$, since $\operatorname{tr}\mathbf{E}^\infty = \nabla \cdot \mathbf{u}^\infty = 0$ but $\operatorname{tr}\widehat{\mathbf{\Omega}}\widehat{\mathbf{\Omega}} = 1$.

In order to construct a solution we follow the method outlined by Hinch [4] and discussed in detail by Nadim and Stone [9]. Since the procedure is similar to the analysis of a drop in a straining flow we only outline the important intermediate results. The velocity and pressure fields in either fluid are represented as

$$\mathbf{u}(\mathbf{r}, t) = \nabla\phi + \mathbf{r} \wedge \nabla\psi + \nabla(\mathbf{r} \cdot \mathbf{A}) - 2\mathbf{A}, \tag{7}$$

$$p_d(\mathbf{r}, t) = 2\nabla \cdot \mathbf{A} + \text{constant}, \tag{8}$$

where ϕ, ψ, and $\mathbf{A}$ are scalar and vector harmonic functions, respectively. Also, we neglect any contributions to $\mathbf{A}$ that are divergence-free. The appropriate harmonic functions linear in the driving force $\widehat{\mathbf{\Omega}}\widehat{\mathbf{\Omega}}$ are

$$\phi = \alpha\widehat{\mathbf{\Omega}}\widehat{\mathbf{\Omega}} : \left(\frac{\mathbf{I}}{r^3} - \frac{3\mathbf{rr}}{r^5}\right) + \frac{\xi}{r}, \qquad \hat{\phi} = \hat{\alpha}\widehat{\mathbf{\Omega}}\widehat{\mathbf{\Omega}} : (r^2\mathbf{I} - 3\mathbf{rr}), \tag{9}$$

$$\psi = 0, \qquad \hat{\psi} = 0, \tag{10}$$

$$\mathbf{A} = \beta\frac{\widehat{\mathbf{\Omega}}\widehat{\mathbf{\Omega}} \cdot \mathbf{r}}{r^3}, \qquad \widehat{\mathbf{A}} = 3\hat{\beta}[r^2\mathbf{r} + 2r^2\widehat{\mathbf{\Omega}} \cdot \mathbf{r}\widehat{\mathbf{\Omega}} - 5r^2(\widehat{\mathbf{\Omega}} \cdot \mathbf{r})^2\mathbf{r}] \cdot \hat{\xi}(\widehat{\mathbf{\Omega}} \cdot \mathbf{r})\widehat{\mathbf{\Omega}}, \tag{11}$$

where $\alpha, \beta, \xi, \hat{\alpha}, \hat{\beta}, \hat{\xi}$ are functions of time which are to be determined. $\hat{\xi}$ makes a time-dependent contribution to the pressure but does not directly contribute to the velocity field. A term proportional to $(\widehat{\mathbf{\Omega}} \cdot \widehat{\mathbf{\Omega}})\mathbf{r}$ may be added to $\widehat{\mathbf{A}}$, but only contributes to the

pressure a constant term which can then be absorbed into $\hat{\xi}$. We thus observe that since $\operatorname{tr}\widehat{\mathbf{\Omega}}\widehat{\mathbf{\Omega}} = 1$ there are six required harmonic functions as compared to four such functions in the case of a drop in a straining flow. The detailed velocity, pressure, and rate-of-strain fields are given in the appendix.

The drop shapes may be expanded in surface spherical harmonics [1, 9] and the leading-order deformation must be linear in $\widehat{\mathbf{\Omega}}\widehat{\mathbf{\Omega}}$. Hence, we seek

$$f(\theta, \phi, t) = \delta(t)\widehat{\mathbf{\Omega}}\widehat{\mathbf{\Omega}} : \left(\mathbf{I} - \frac{3\mathbf{r}\mathbf{r}}{r^2} \right), \tag{12}$$

where $\delta(t)$ is to be determined. It follows that for nearly spherical shapes

$$\nabla_s \cdot \mathbf{n} = 2[1 - 2\varepsilon\delta(1 - 3(\widehat{\mathbf{\Omega}} \cdot \mathbf{n}_0)^2)] + O(\varepsilon^2), \tag{13}$$

where $\mathbf{n}_0$ denotes the unit normal to the spherical surface. We may now determine the seven unknown functions by straightforward application of the boundary conditions. At leading order, velocities are $O(1)$ so that combining equations (4) and (13) we identify $\varepsilon = \mathcal{B}$. For completeness, the solutions for each of the coefficients are included in the appendix.

The quantity of most interest is the time-dependent shape of the drop, which we find to be

$$r = 1 + \frac{\mathcal{B}}{24}[1 - 3(\widehat{\mathbf{\Omega}} \cdot \mathbf{n}_0)^2] \left[\exp\left\{ -\frac{40(1 + \lambda)}{(3 + 2\lambda)(16 + 19\lambda)} \frac{t}{\mathcal{B}} \right\} - 1 \right]. \tag{14}$$

The drop attains an ellipsoidal steady shape with a relaxation time $\frac{(3+2\lambda)(16+19\lambda)}{40(1+\lambda)}\mu/\gamma$. This completes the solution. Because the results described here are limited to nearly spherical shapes, it is not possible to compare the predictions of Eq. (14) with the recent numerical calculations of Hu and Joseph [6], who focussed on the large distortion limit. We note that the corresponding analytical solution for the drop shape in an extensional flow has the similar, though not identical, form

$$r = 1 + \mathcal{C}\frac{(16 + 19\lambda)}{8(1 + \lambda)}\mathbf{E}^{\infty} : \mathbf{n}_0\mathbf{n}_0 \left[1 - \exp\left\{ -\frac{40(1 + \lambda)}{(3 + 2\lambda)(16 + 19\lambda)}t \right\} \right], \tag{15}$$

where $\mathcal{C} = \mu a|\mathbf{E}^{\infty}|/\gamma$ is the capillary number and time has been nondimensionalized using the surface tension relaxation time $\mu a/\gamma$ [10].

The time-dependent contributions (i.e., the exponential terms) to (14) and (15) are identical if time is scaled in both problems with $\mu a/\gamma$. However, since the rotating problem has a quiescent steady state, fluid viscosity does not influence the final shape and the magnitude of the distortion is thus independent of λ. As a final remark we note that in the rotating case the deforming drop creates a disturbance flow which decays as $O(1/r^2)$. Each end of the drop exerts a force $\Delta\rho\Omega^2 a^4$ on the fluid, and these equal and opposite forces contribute to produce a dipolar flow at large distances from the drop.

Appendix. Using the velocity and pressure representations given by (7)–(11) yields the detailed velocity, pressure, and rate-of-strain fields for the exterior fluid:

$$\mathbf{u}(\mathbf{r}, t) = 3\alpha(t) \left[-\frac{\mathbf{r}}{r^5} - \frac{2\widehat{\boldsymbol{\Omega}}\widehat{\boldsymbol{\Omega}} \cdot \mathbf{r}}{r^5} + \frac{5(\widehat{\boldsymbol{\Omega}} \cdot \mathbf{r})^2 \mathbf{r}}{r^7} \right] - \beta(t) \frac{3(\widehat{\boldsymbol{\Omega}} \cdot \mathbf{r})^2 \mathbf{r}}{r^5} - \xi \frac{\mathbf{r}}{r^3}, \tag{16}$$

$$p_d(\mathbf{r}, t) = p_0 + 2\beta(t) \left[\frac{1}{r^3} - \frac{3(\widehat{\boldsymbol{\Omega}} \cdot \mathbf{r})^2}{r^5} \right], \tag{17}$$

$$\mathbf{e}(\mathbf{r}, t) = 3\alpha(t) \left[-\frac{\mathbf{I}}{r^5} - \frac{2\widehat{\boldsymbol{\Omega}}\widehat{\boldsymbol{\Omega}}}{r^5} + \frac{5\mathbf{rr}}{r^7} + \frac{10\widehat{\boldsymbol{\Omega}} \cdot \mathbf{r}}{r^7}(\widehat{\boldsymbol{\Omega}}\mathbf{r} + \mathbf{r}\widehat{\boldsymbol{\Omega}}) + \frac{5(\widehat{\boldsymbol{\Omega}} \cdot \mathbf{r})^2 \mathbf{I}}{r^7} - \frac{35(\widehat{\boldsymbol{\Omega}} \cdot \mathbf{r})^2 \mathbf{rr}}{r^9} \right]$$
$$- 3\beta(t) \left[\frac{\widehat{\boldsymbol{\Omega}} \cdot \mathbf{r}}{r^5}(\widehat{\boldsymbol{\Omega}}\mathbf{r} + \mathbf{r}\widehat{\boldsymbol{\Omega}}) + \frac{(\widehat{\boldsymbol{\Omega}} \cdot \mathbf{r})^2 \mathbf{I}}{r^5} - \frac{5(\widehat{\boldsymbol{\Omega}} \cdot \mathbf{r})^2 \mathbf{rr}}{r^7} \right] - \xi \left[\frac{\mathbf{I}}{r^3} - \frac{3\mathbf{rr}}{r^5} \right]. \tag{18}$$

Inside the drop, the flow fields assume the form

$$\hat{\mathbf{u}}(\mathbf{r}, t) = 2\hat{\alpha}(t)[\mathbf{r} - 3\widehat{\boldsymbol{\Omega}} \cdot \mathbf{r}\widehat{\boldsymbol{\Omega}}] + 6\hat{\beta}(t)[r^2\mathbf{r} + 2(\widehat{\boldsymbol{\Omega}} \cdot \mathbf{r})^2 \mathbf{r} - 5r^2\mathbf{r} \cdot \widehat{\boldsymbol{\Omega}}\widehat{\boldsymbol{\Omega}}], \tag{19}$$

$$\hat{p}_d(\mathbf{r}, t) = \hat{p}_0 + 42\hat{\beta}(t)[r^2 - 3(\widehat{\boldsymbol{\Omega}} \cdot \mathbf{r})^2] + 2\hat{\xi}(t), \tag{20}$$

$$\hat{\mathbf{e}}(\mathbf{r}, t) = 2\hat{\alpha}(t)[\mathbf{I} - 3\widehat{\boldsymbol{\Omega}}\widehat{\boldsymbol{\Omega}}]$$
$$+ 6\hat{\beta}(t)[2\mathbf{rr} + r^2\mathbf{I} - 5r^2\widehat{\boldsymbol{\Omega}}\widehat{\boldsymbol{\Omega}} + 2(\widehat{\boldsymbol{\Omega}} \cdot \mathbf{r})^2 \mathbf{I} - 3\widehat{\boldsymbol{\Omega}} \cdot \mathbf{r}(\widehat{\boldsymbol{\Omega}}\mathbf{r} + \mathbf{r}\widehat{\boldsymbol{\Omega}})], \tag{21}$$

where p_0 and $\hat{p}_0$ are constant reference pressures.

Applying the boundary conditions determines the six coefficients in the above equations as well as $\delta(t)$, which appears in the shape function (12). Continuity of velocity yields the three relations

$$-3\alpha - \xi = 2\hat{\alpha} + 6\hat{\beta}, \tag{22}$$

$$15\alpha - 3\beta = 12\hat{\beta}, \tag{23}$$

$$-6\alpha = -6\hat{\alpha} - 30\hat{\beta}. \tag{24}$$

Continuity of tangential stress leads to

$$48\alpha - 6\beta - \lambda(-12\hat{\alpha} - 96\hat{\beta}) = 0. \tag{25}$$

The normal stress balance yields two equations:

$$-2\beta + 4\xi + 24\alpha - \lambda(4\hat{\alpha} - 6\hat{\beta} - 2\hat{\xi}) = 4\frac{\varepsilon}{\mathcal{B}}\delta + \tfrac{1}{2}, \tag{26}$$

$$-72\alpha + 18\beta - \lambda(-12\hat{\alpha} + 18\hat{\beta}) = -12\frac{\varepsilon}{\mathcal{B}}\delta - \tfrac{1}{2}, \tag{27}$$

in addition to the usual equilibrium condition $\hat{p}_0 - p_0 = 2/\mathcal{B}$. Finally, the kinematic condition $(\hat{\mathbf{u}} \cdot \mathbf{n} = \partial f/\partial t)$ requires

$$2\hat{\alpha} + 6\hat{\beta} = \varepsilon \frac{d\delta}{dt}. \tag{28}$$

We see that Eqs. (22) and (26) determine, respectively, ξ and $\hat{\xi}$. The remaining five equations decouple and are solved for $\alpha, \beta, \hat{\alpha}, \hat{\beta}$, and δ. In particular, we find

$$\alpha(t) = -\frac{(2+3\lambda)}{6(16+19\lambda)(3+2\lambda)} \exp\left\{-\frac{40(1+\lambda)}{(16+19\lambda)(3+2\lambda)}\frac{t}{\mathcal{B}}\right\}, \tag{29}$$

$$\beta(t) = -\frac{1}{6(3+2\lambda)} \exp\left\{-\frac{40(1+\lambda)}{(16+19\lambda)(3+2\lambda)}\frac{t}{\mathcal{B}}\right\}, \tag{30}$$

$$\hat{\alpha}(t) = -\frac{(19+16\lambda)}{12(16+19\lambda)(3+2\lambda)} \exp\left\{-\frac{40(1+\lambda)}{(16+19\lambda)(3+2\lambda)}\frac{t}{\mathcal{B}}\right\}, \tag{31}$$

$$\hat{\beta}(t) = \frac{1}{12(16+19\lambda)} \exp\left\{-\frac{40(1+\lambda)}{(16+19\lambda)(3+2\lambda)}\frac{t}{\mathcal{B}}\right\}, \tag{32}$$

$$\delta(t) = \frac{1}{24}\left[\exp\left\{-\frac{40(1+\lambda)}{(16+19\lambda)(3+2\lambda)}\frac{t}{\mathcal{B}}\right\} - 1\right]. \tag{33}$$

REFERENCES

[1] R. G. Cox, *The deformation of a drop in a general time-dependent fluid flow*, J. Fluid Mech. **37**, 601–623 (1969)

[2] D. A. Edwards, H. Brenner, and D. T. Wasan, *Interfacial Transport Processes and Rheology*, Butterworth-Heinemann, Boston, 1991

[3] H. P. Greenspan, *The Theory of Rotating Fluids*, Cambridge University Press, Cambridge, 1968

[4] E. J. Hinch, *Hydrodynamics at low Reynolds numbers*, in A Brief and Elementary Introduction: Disorder and Mixing (Eds. E. Guyon, J. P. Nadal and Y. Pomeau), NATO ASI Series, Series E, 152: 43–55 (1988)

[5] J. C. Hsu and R. W. Flumerfelt, *Rheological applications of a drop elongation experiment*, Trans. Soc. Rheol. **19**, 523–540 (1975)

[6] H. H. Hu and D. D. Joseph, *Evolution of a liquid drop in a spinning drop tensiometer*, J. Colloid and Interface Sci. **162**, 331–339 (1994)

[7] D. D. Joseph, M. S. Arney, G. Gillberg, H. Hu, D. Hultman, C. Verdier, and T. M. Vinagre, *A spinning drop tensioextensometer*, J. Rheology **36**, 621–662 (1992)

[8] L. G. Leal, *Laminar Flow and Convective Transport Processes*, Butterworth-Heinemann, Boston, 1990

[9] A. Nadim and H. A. Stone, *The motion of small particles and droplets in quadratic flows*, Stud. Appl. Math. **85**, 53–73 (1991)

[10] J. M. Rallison, *The deformation of small viscous drops and bubbles in shear flows*, Ann. Rev. Fluid Mech. **16**, 45–66 (1984)

[11] D. K. Rosenthal, *The shape and stability of a bubble at the axis of a rotating liquid*, J. Fluid Mech. **12**, 358–366 (1962)

[12] B. Vonnegut, *Rotating bubble method for the determination of surface and interfacial tension*, Rev. Sci. Instr. **13**, 6–9 (1942)