# Stability of a Viscous Compound Fluid Drop

A compound drop composed of viscous Newtonian core and shell fluids is disturbed away from its motionless spherical state. The physical characteristics of the drop determine the decay rates of any disturbance. This damping process is accompanied by out-of-phase displacements of the two interfaces. Special limiting cases of a thin and thick shell are investigated.

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## SCOPE

A compound liquid drop, composed of a viscous Newtonian fluid surrounded by a shell of another viscous Newtonian fluid is studied here. Its equilibrium motionless state is a sphere in which the internal interface is concentric with the outer boundary when gravitational effects are small. A small disturbance will deform the drop slightly, but it eventually returns to its initial state. When the inertial and gravitational effects are negligible the disturbances are damped due to the dominant viscous dissipation of energy and the surface tension forces. The rate of damping and the relationship between the small displacements of the two interfaces is determined. Their dependence on the physical properties of the drop is also discussed.

Compound drops arise in technological areas, for example the production of fusion target pellets, and in processing techniques. In a laboratory, the construction of a layer around a drop does not always yield a monolayer, but sometimes a thin multilayered shell. In this case, previous studies on the dynamics of fluid interfaces (Miller and Scriven, 1968; Ramabhadran et al., 1976) may not be appropriate. Also, if the shell fluid is sufficiently thin, this layer can be considered as a thin coating to a droplet in an emulsion or as a model of a viscous membrane surrounding biological cells (Landman, 1983; Landman and Greenspan, 1982).

The dynamic behavior of a compound drop to small disturbances is treated in a classical way using a linearized stability analysis. The conservation equations governing the separate fluids are linearized and then their eigenvalues and eigenvectors are determined from a normal mode analysis.

## CONCLUSIONS AND SIGNIFICANCE

A quadratic equation to determine the rate of damping of small disturbances to a compound liquid drop composed of two viscous Newtonian fluids has been obtained and analyzed. The smaller decay rate determines the time scale for the drop to return to its initial motionless spherical state. This damping process is accompanied by out-of-phase displacements of the two interfaces in the $P_2$ (cos$\theta$) shape; that is, the drop relinquishes its concentric configuration. In constrast, Saffren, Elleman and Rhim (1982) found that for an inviscid compound drop, the higher frequency oscillations were in-phase, while the lower ones were out-of-phase, as defined in the discussion section. The dependence of the fluid properties and the thickness of the shell to the decay rates has been determined. In particular, if $\sigma_i/\sigma_o$ or $\mu_2/\mu_1$ is increased, the time scale of return to the initial state is diminished. For thin shells, those often required in processing techniques, the decay rate is of the order $[(R_o/R_i) - 1]^2$ and the relative displacement amplitudes of the outer and inner interface is determined by the surface tension ratio. For sufficiently thick shells, the boundaries effectively uncouple and the drop acts as if the core fluid did not exist.

## INTRODUCTION

The dynamics of droplets and their stability to small disturbances has been of much interest in recent years. In this paper the dynamics of a compound drop composed of two viscous Newtonian fluids is discussed. The purely oscillatory motion of an inviscid compound drop has been studied by Saffren, Elleman and Rhim (1982). Our work complements theirs, since it treats the viscous rather than the inviscid limit of the equations governing the motion of a compound drop. However, if the shell thickness is small, the viscous boundary layer effects should be important in determining the stability behavior of these drops. The work of Saffren et al. ignores these effects. If inertial effects and viscous effects were of comparable strength, damped oscillations would be evident.

Lamb (1932), Reid (1960), and Chandrasekhar (1961) have analyzed the stability of a simple viscous and inviscid drop immersed in another fluid. If the inertial effects are important the damping is also accompanied by oscillations. Miller and Scriven (1968) address many of these problems and some limiting cases (which also follow from this work).

The work of Patzer and Homsy (1975) incorporates a disjoining pressure term in the fluid shell. This additional term can give rise to an exchange of stability which indicates a possible rupture of the droplet. However, the disjoining pressure is only significant when the ratio of the thickness of the shell to drop radius is $O(10^{-4})$. This is many orders of magnitude smaller than the compound drops used for example in the manufacture of laser fusion pellets discussed by Kim et al. (1982) and Lee et al. (1982).

The following section 2 presents a discussion of the dynamics and stability of the compound, but the full mathematical analysis can be found in the Appendix. Special limiting cases of a thin shell are investigated following the discussion. Finally, a section discussing the general behavior of the damping process and its de-

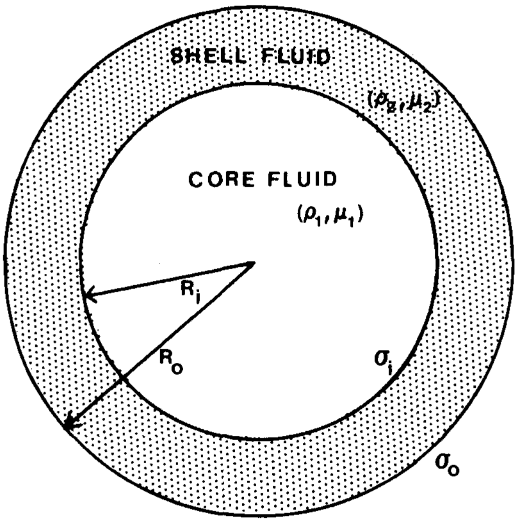



**Figure 1. Initial configuration of the two-fluid drop: Densities $\rho_1, \rho_2$; dynamic viscosities $\mu_1, \mu_2$; surface tensions $\sigma_i, \sigma_o$.**

pendence on the physical characteristics of the drop is presented.

## DISCUSSION

The initial configuration of the compound drop is assumed to be spherical, radius $R_o$. The core fluid is spherical, radius $R_i$, and is surrounded by a uniform shell, thickness $R_o - R_i$, of another fluid. The two fluids, which do not mix, are assumed to be very viscous. This drop is immersed in another fluid and is in a state of rest. It will be assumed that gravitational forces are small in comparison to the surface tension forces at the two interfaces, and that the host fluid has low density and is much less viscous than the fluids composing the drop, so that any dynamical effects from this external medium are negligible. The initial configuration is illustrated in Figure 1.

Any slight disturbance to the droplet causes it to deform and the fluids to be set in motion. Energy will be dissipated by the viscosity of the fluids and the drop will eventually return to its initial quiescent shape. The nature of this process is investigated here; both the decay rates and the relationship between the displacements of the two interfaces will be discussed.

The equations which govern the incompressible, viscous fluids are the usual conservation of mass and momentum equations, described by the Navier-Stokes equations. The inertial terms will be neglected since both fluids are supposed to be very viscous. The boundary conditions to be satisfied are given by the normal and tangential stress balance, which involve pressure, viscous stresses, and surface tension forces. The velocity of the two fluids at their common interface must be continuous and the radial velocity of each interface must equal the radial velocity of the bounding fluid (the kinematic conditions). Finally, the viscosity of the two fluids prohibits any slip at the interior interface.

Since only small disturbances of the initial motionless state (with constant pressures related to the surface tension forces) are considered, the equations governing the two-fluid system can be linearized. These equations and the nature of their solution are given in the Appendix. Here we describe the final system to be analyzed.

In terms of spherical coordinates, where $r$ is the spherical radius, $\theta$ the polar angle, and $\phi$ the azimuthal angle, the surface of the droplet slightly distributed away from a sphere radius $R_o$ is given by

$$r = R_o + y(\theta,\phi,t), \quad |y| \ll R_o, \tag{1}$$

and the interface between the core and shell fluids is displaced from radius $R_i$ as

$$r = R_i + x(\theta,\phi,t) \quad |x| \ll R_i. \tag{2}$$

These displacements can be expanded in spherical harmonics:

$$y = \sum_{n=2}^{\infty} e^{\lambda_n t} Y_n P_n(\cos\theta) + \sum_{n=2}^{\infty}\sum_{m=1}^{n} e^{\lambda_n t} Y_{nm} e^{im\phi} P_n^{(m)}(\cos\theta),$$
$$x = \sum_{n=2}^{\infty} e^{\lambda_n t} X_n P_n(\cos\theta) + \sum_{n=2}^{\infty}\sum_{m=1}^{n} e^{\lambda_n t} X_{nm} e^{im\phi} P_n^{(m)}(\cos\theta), \tag{3}$$

where $P_n^{(m)}$ are the associated Legendre polynomials of degree $n$. The $\lambda_n$ depends on $n$ but not on $m$ as is usual (Landman and Greenspan, 1982; Miller and Scriven, 1968), so that with no loss of generality the $m$ from the notation is dropped here; let $Y_n, X_n$ denote the arbitrary constants in the expansions. (These start with $n = 2$, because the volume of the drop remains fixed as does the position of its center of gravity.)

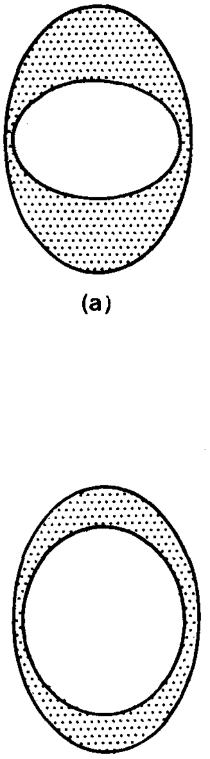
In the Appendix, it is shown that a linear system of equations in $Y_n$ and $X_n$ must be solved. This involves the $\lambda_n$ terms due to the time derivatives in the kinematic conditions. The equations can be written in the following form; here $\mu_1$ and $\mu_2$ are the dynamic viscosities of the core and shell fluid respectively, $\sigma_i$ is the interfacial tension at their common boundary, and $\sigma_o$ is the surface tension at the external boundary of the compound drop:

$$\begin{bmatrix} c\dfrac{J}{M} - \lambda_n & \beta c M^{n-1} K \\ c M^{n-1} K & \beta c L - \lambda_n \end{bmatrix} \begin{bmatrix} Y_n \\ X_n \end{bmatrix} = \begin{bmatrix} 0 \\ 0 \end{bmatrix}. \tag{4}$$

where $n \geq 2$ and

$$\beta = \sigma_i/\sigma_o, \quad \delta = \mu_1/\mu_2 \quad M = R_o/R_i,$$
$$c = \frac{\sigma_o}{\mu_2 R_i}\frac{n(n+2)(2n+1)}{2\Delta},$$
$$J = -M^{4n}[2n^2 + 1 + 2\delta(n^2-1) - M^{2n+1}(1-\delta)$$
$$(2n+1)(n^2-1)]P + M^{2n-3}(1-\delta)$$
$$(2n+1)Q + 2M^{-2}(1-\delta)^2(n^2-1)(2n^2+4n+3),$$
$$P = 2n(n+2) + \delta(2n^2+4n+3),$$
$$Q = 2(n^4 + 2n^3 - n^2 - 2n + 3)$$
$$+ \delta(n^2-1)(2n^2+4n+3),$$
$$K = -M^{2n+1}n(n+2)(2n-1)P$$
$$+ M^{2n-1}(2n+3)(n^2-1)[2(n^2-1) + \delta(2n^2+1)]$$
$$-2(1-\delta)(2n+3)(n^2-1)^2$$
$$+ 2M^{-2}(1-\delta)n^2(n+2)^2(2n-1), \tag{5}$$
$$L = -2M^{4n}(1+\delta)(n^2-1)(2n^2+4n+3)$$
$$-M^{2n+1}(2n+1)(n^4+2n^3-n^2-2n+3)[2+\delta(2n+1)]$$
$$+ 2M^{2n-1}\delta n(n+2)(2n-1)(2n+3)(n^2-1)$$
$$+ M^{2n-3}n(n+2)(2n+1)(n^2-1)[2-\delta(2n+1)]$$
$$+ 2M^{-2}(1-\delta)n(n+2)(2n^2+1),$$
$$\Delta = M^{4n}(2n^2+4n+3)[2n^2+1+2\delta(n^2-1)]P$$
$$-M^{2n+1}(1-\delta)(2n+1)^2(n^4+2n^3-n^2-2n+3)P$$
$$+ 2M^{2n-1}(1-\delta)n(n+2)(2n-1)(2n+3)(n^2-1)P$$
$$-M^{2n-3}(1-\delta)n(n+2)(2n+1)^2 Q$$
$$+ 2M^{-2}(1-\delta)^2 n(n+2)(2n^2+1)(2n^2+4n+3).$$

There are nontrivial solutions for the flow and pressure in the disturbed droplet only if there exists a nontrivial solution to Eq. 4; consequently the determinant of coefficients must be zero. This gives



(a)

(b)

**Figure 2. Configurations of the compound drop in the $P_2$ (cos$\theta$) mode: (a) out-of-phase case corresponding to $\lambda_+$; (b) in-phase case corresponding to $\lambda_-$.**

$$\lambda_n^2 - c\left(\frac{J}{M} + \beta L\right)\lambda_n + \beta c^2\left(\frac{J}{M}L - M^{2n-2}K^2\right) = 0. \quad (6)$$

A messy algebraic calculation shows that $\Delta$, and hence $c$, are positive and that $J$, $K$, and $L$ are negative for all relevant values of the parameters (namely $\beta$, $\delta > 0$, $M > 1$, $n \geq 2$). Also all the coefficients in Eq. 6 are positive, so that the two solutions $\lambda_\pm$ (dropping the subscript $n$) are negative:

$$\lambda_\pm = \frac{c}{2}\left\{\frac{J}{M} + \beta L \pm \left[\left(\frac{J}{M} - \beta L\right)^2 + 4\beta M^{2n-2}K^2\right]^{1/2}\right\}. \quad (7)$$

The corresponding eigenvectors, which determine the relative amplitudes of $Y_n$ and $X_n$, are $[Y_n/X_n]_\pm$ where the ratio of the entries is

$$\left(\frac{Y_n}{X_n}\right)_\pm = \frac{\left(\frac{J}{M} - \beta L\right) \pm \left[\left(\frac{J}{M} - \beta L\right)^2 + 4\beta M^{2n-2}K^2\right]^{1/2}}{2M^{n-1}K}. \quad (8)$$

The positive square root here corresponds to the $\lambda_+$ eigenvalue. Consequently, regardless of the sign of $(J/M) - \beta L$, since $K < 0$, the slower decay rate $\lambda_+$, gives rise to a negative relative boundary displacement. In this case the interfaces are called "out-of-phase." In contrast the relative boundary displacement of the faster decaying solution, corresponding to $\lambda_-$, is positive, and is called "in-phase." Hence the drop relinquishes its concentric configuration in the $\lambda_+$ case. Using a variety of parameter settings it can be verified numerically that the decay rates increase with $n$ so that drop will usually decay through the $n = 2$ mode. These configurations are illustrated in Figure 2. However, a higher mode may be observed if the initial conditions are carefully arranged for its excitation.

In general, numerical methods are required to investigate the variation of $\lambda_\pm$ with the fluid properties (given by $\beta,\delta$) and ratio of the drop radius to core radius (given by $M$). For $M$ of moderate size [$M \sim 0(1)$], it was found that $\lambda_- \leq 0(10\lambda_+)$. Thus, only the out-of-phase displacements would be observed in any laboratory situation. In other words, the $e^{\lambda_+ t}$ solution will be the important one in determining how the compound droplet regains its initial state. However, if the ratio $(Y_n/X_n)_+$ is very large, only the outer displacement will damp like $e^{\lambda_+ t}$. (See the thick shell case in the following section.)

## EXAMPLES AND LIMITING CASES

Expressions for several limiting cases can be deduced from the work above.

*Simple Drop.* For comparison, the case of a simple, very viscous drop composed only of shell fluid immersed in a much less viscous medium is considered. This is analogous to setting the core radius $R_i$ to zero and a similar procedure gives the decay rate of the boundary $r = R_o + \Sigma_{n=2}^{\infty} e^{\lambda_n t} Y_n P_n$ (cos$\theta$), (ignoring the $\phi$ dependence):

$$\lambda_n = -\frac{\sigma_o}{\mu_2 R_o}\frac{n(n+2)(2n+1)}{2(2n^2+4n+3)}. \quad (9)$$

These decay rates increase with $n$ so that the droplet will decay through the $n = 2$ mode as $-1.053\sigma_o/\mu_2 R_o$. Equation 9 is identical to the Miller and Scriven (1968) result for a droplet of high viscosity.

*Thin Shell.* When there is only a very thin layer of shell fluid, we set $M = R_o/R_i = 1 + h$, where $h \ll 1$. Then if the coefficients in Eq. 6 are expanded in a Taylor series (in $h$) about $M = 1$, it can be shown that the coefficient of $\lambda_n$ is

$$\frac{-\sigma_o(1+\beta)n(n+2)(2n+1)}{\mu_2 R_i \delta 2(2n^2+4n+3)} + 0(h), \quad (10)$$

while the constant term is $0(h^2)$. Hence the slower decay rate is determined by matching the linear and constant terms in Eq. 6, giving

$$\lambda_+ = \beta c \frac{\left(\frac{J}{M}L - M^{2n-2}K^2\right)}{\frac{J}{M} + \beta L} = 0(h^2). \quad (11)$$

These increase with $n$, so that the $n = 2$ mode determines the rate of decay. For moderate values of $\delta$, when $n = 2$,

$$\lambda_+ \sim -\frac{\sigma_o\beta}{\mu_2 R_i(1+\beta)\delta}(4\cdot 8)h^2 = -\frac{\sigma_i\sigma_o}{\mu_1 R_i(\sigma_i + \sigma_o)}(4\cdot 8)h^2. \quad (12)$$

When $\delta \simeq 10$ or larger, $|\lambda_+|$ is slightly larger than this, and when $\delta \simeq 0.1$ or smaller, it is slightly smaller.

The other eigenvalue $\lambda_-$ is found by matching the quadratic and linear terms of Eq. 6. This gives

$$\lambda_- \sim -\frac{(\sigma_i + \sigma_o)}{\mu_1 R_i}\frac{n(n+2)(2n+1)}{2(2n^2+4n+3)} + 0(h). \quad (13)$$

Notice that to highest order $\lambda_-$ is equal to the decay rate for a viscous drop, radius $R_i$, composed entirely of the core fluid (viscosity $\mu_1$), if its surface tension is replaced by $\sigma_i + \sigma_o$ (see the simple drop case above).

Excluding $0(h)$ terms the solutions are asymptotic to

$$e^{\lambda_+ t}\begin{bmatrix}\beta\\-1\end{bmatrix}, \quad e^{\lambda_- t}\begin{bmatrix}1\\1\end{bmatrix}. \quad (14)$$

*Thick Shell.* In this case, let $M \to \infty$. Then the dominant terms in $\Delta$, $J$, $K$, and $L$ must be studied. It can be easily verified that

$$\lambda_\pm \sim \frac{\sigma_o}{\mu_2 R_i}\frac{n(n+2)(2n+1)}{4\Delta}\left\{\left(\frac{J}{M} + \beta L\right) \pm \left|\frac{J}{M} - \beta L\right|\right\} \quad (15)$$

where

$$J \sim -M^{4n}[2n^2 + 1 + 2\delta(n^2 - 1)]P$$
$$K \sim -M^{2n+1}n(n + 2)P$$
$$L \sim -2M^{4n}(1 + \delta)(n^2 - 1)(2n^2 + 4n + 3) \quad (16)$$
$$\Delta \sim M^{4n}(2n^2 + 4n + 3)[2n^2 + 1 + 2\delta(n^2 - 1)]P.$$

Equation 15 is obtained from Eq. 7 using the fact that $4\beta M^{2n-2}K^2 \ll (J/M - \beta L)^2$ as $M \to \infty$. Consequently,

$$\lambda_+ \sim -\frac{\sigma_o}{\mu_2 R_i M}\frac{n(n + 2)(2n + 1)}{2(2n^2 + 4n + 3)} = -\frac{\sigma_o}{\mu_2 R_o}\frac{n(n + 2)(2n + 1)}{2(2n^2 + 4n + 3)} \quad (17)$$

and

$$\lambda_- \sim -\frac{\beta\sigma_o(1 + \delta)n(n + 2)(2n + 1)(n^2 - 1)}{\mu_2 R_i[2n^2 + 1 + 2\delta(n^2 - 1)][2n(n + 2) + \delta(2n^2 + 4n + 3)]} = -\frac{\sigma_i(\mu_1 + \mu_2)n(n + 2)(2n + 1)(n^2 - 1)}{R_i[2\mu_1(n^2 - 1) + \mu_2(2n^2 + 1)][\mu_1(2n^2 + 4n + 3) + 2\mu_2 n(n + 2)]}. \quad (18)$$

Further, the two linearly independent solutions $e^{\lambda t}[Y_n/X_n]$ are asymptotic to

$$e^{\lambda_+ t}\begin{bmatrix}1\\0\end{bmatrix}, \quad e^{\lambda_- t}\begin{bmatrix}0\\1\end{bmatrix}, \quad (19)$$

as $M \to \infty$. Therefore the boundary displacements become uncoupled as the outer radius becomes very large compared to the inner radius. In fact the asymptotic form of $\lambda_+$ represents the decay rate exhibited by disturbances of a viscous simple drop of shell fluid, radius $R_0$. The drop acts as if the small inner core did not exist. It is not difficult to show that the asymptotic expression for $\lambda_-$ is the decay rate of disturbances of a viscous simple droplet of core fluid, radius $R_i$, surrounded by a viscous host fluid made of the shell fluid, which extends to infinity; that is, the inner core acts as if the thick shell (large radius compared to the inner radius) extends to infinity. (This expression for $\lambda_-$ is also given in Miller and Scriven, 1968).

Note also that if $\mu_1 \gg \mu_2$, $\lambda_-$ reduces to the decay rate obtained for a highly viscous ($\mu_1$) simple drop immersed in a fluid of negligible viscosity and density. Similarly the other limit, $\mu_2 \gg \mu_1$ gives the decay rate for a cavity in a high viscosity fluid (Miller and Scriven, 1968).

## GENERAL BEHAVIOR

As mentioned in the discussion section above, in order to investigate the variation in $\lambda_n$ for different fluid properties, the solutions to the quadratic equation must be evaluated numerically. Some general results will be discussed here.

Since $\lambda_+$ evaluated at $n = 2$ is the slowest decay rate, it determines the time scale for decay. The eigenvector corresponding to this tells us that the displacements of the exterior and inner surfaces are always of opposite sign. Only as $M \to \infty$ do the inner and outer interfaces essentially act independently of each other (see the preceding thick shell case).

In Figure 3, the vertical axis is

$$\Lambda = \lambda_+ \Big/ \left(-\frac{\sigma_o}{\mu_2 R_o}\frac{20}{19}\right). \quad (20)$$

Here the numerator is the smallest decay rate (evaluated for $n = 2$) of a compound drop with prescribed values of the $\delta$ and $\beta$. The denominator is the smallest decay rate for a simple drop composed of the shell fluid, of radius $R_0$ (that is $R_i = 0$); it is found by setting $n = 2$ in Eq. 9. Figure 3 illustrates the variation of their ratio with $M = R_o/R_i$. Some remarks regarding this figure follow.

When $\mu_1 = \mu_2(\delta = 1)$ and $\sigma_i = 0$, then in the viscous approximation the two fluids of the drop are identical, and in effect there is only one boundary and one decay rate, giving $\Lambda = 1$. For all the other curves, when $M$ is close to 1, the ratio $\Lambda$ is very small, given by $O((M - 1)^2)$, as seen by Eq. 12. This is the thin shell approximation. As the thickness of the shell increases, the ratio increases, and it asymptotes to 1. This confirms the behavior given by Eq. 17 in the limit $M \gg 1$.

The ratio of the viscosity and surface tension coefficients influence the ratio $\Lambda$ and therefore $\lambda_+$ in the following way. If $\delta$ is fixed, increasing $\beta$ increases the decay rate. If $\beta$ is fixed, decreasing $\delta$ increases the decay rate. So for example, increasing the viscosity of the shell fluid is a stabilizing effect—the motion of the droplet is damped more quickly because of the increased energy dissipitation.

Similar qualitative behavior is exhibited for any $n$, and the decay rates increase with $n$. As mentioned previously, the $n = 2$ mode dominates the stability of the compound drop. However, a higher mode may be observed if the initial conditions are carefully and specifically arranged for its excitation.

Figure 4 gives a measure of the ratio of the outer and inner displacements corresponding to the decay rates in Figure 3. It can be seen that these displacements are of opposite sign, giving rise to the out-of-phase configuration discussed earlier and illustrated in Figure 2b. For thin shells ($M \to 1$), this ratio is just $-\beta$ and so is determined by the surface tension forces. As the shell thickness increases, the viscous forces play a role. As $M$ becomes sufficiently large, the ratio becomes large, which means that the eigenvector approaches [1,0], as in Eq. 19. This corresponds to the effective uncoupling of the two boundaries. (The curve corresponding to $\delta = \beta = 0.01$ also increases, but does so only for $M \gg 10$). Again, fixing $\delta$ and increasing $\beta$ increases the ratio of the displacement amplitudes. Decreasing $\delta$, for fixed $\beta$, has a similar effect.

This study was carried out on the assumption that the viscosities of the two fluids were very large, and also much greater than that of the surrounding medium. If the inertial terms for the drop and the external medium are included in the analysis, the decay rates will no longer be real. They would involve a small imaginary part,

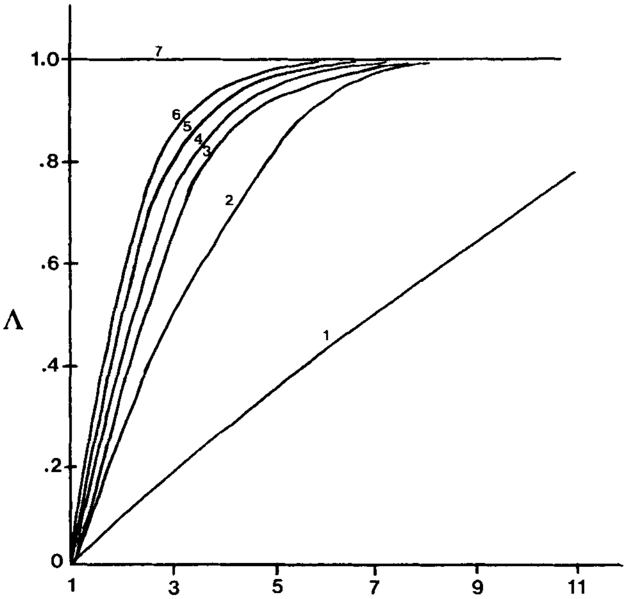



**Figure 3. $\Lambda$ vs. $M$ for different fluid properties: (1) $\delta = \beta = 0.1$; (2) $\delta = 10$, $\beta = 2$; (3) $\delta = \beta = 1$; (4) $\delta = \beta = 10$; (5) $\delta = 1, \beta = 10$; (6) $\delta = 0.1, \beta = 10$; (7) $\delta = 1, \beta = 0$.**

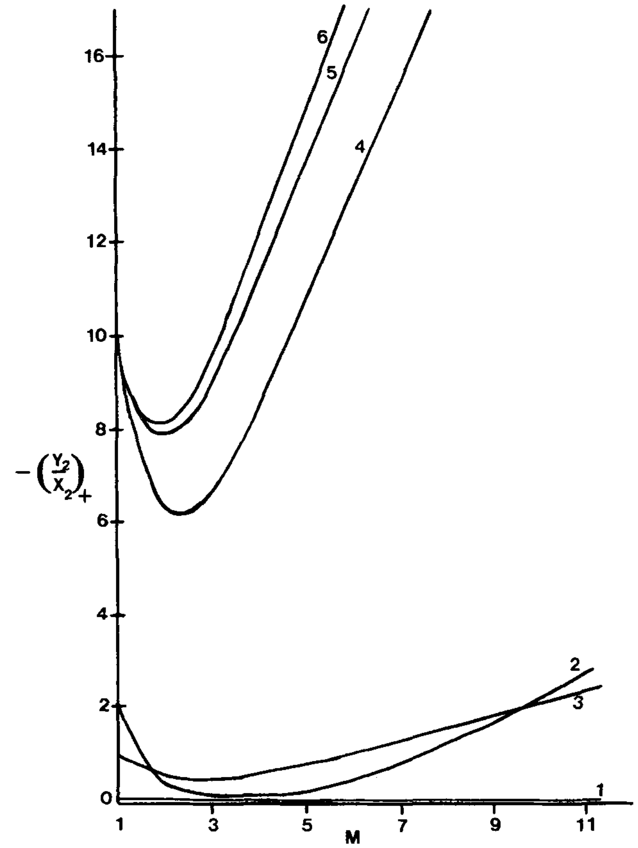



**Figure 4.** $-(Y_2/X_2)_+$ vs. $M$ for different fluid properties: (1) $\delta = \beta = 0.1$; (2) $\delta = 10, \beta = 2$; (3) $\delta = \beta = 1$; (4) $\delta = \beta = 10$; (5) $\delta = 1, \beta = 10$; (6) $\delta = 0.1, \beta = 10$.

so that the drop would return to its initial motionless state through a series of oscillations with decreasing amplitude. The case of a simple drop immersed in another fluid has been discussed by Miller and Scriven (1968), and various cases exhibiting damped oscillations arise. Again, for high viscosity fluids, there are no oscillations, only an aperiodic return to the spherical shape.


## ACKNOWLEDGMENT

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## NOTATION

$c$ = algebraic expression used in decay factor evaluation
$h$ = relative thickness of the shell fluid = $M - 1$
$J,K,L$ = algebraic expression in decay factor evaluation
$M$ = ratio of the initial radii $R_o/R_i$
$n$ = modal index of Legendre polynomial $P_n$ $(\cos\theta)$
$P$ = algebraic expression used in decay factor evaluation
$P_n, P_n^{(m)}$ = Legendre polynomials
$Q$ = algebraic expression used in decay factor evaluation
$r$ = radial coordinate
$R_i$ = initial radius of core fluid
$R_o$ = initial radius of compound drop
$x$ = inner radial displacement
$X_n, X_{nm}$ = amplitude of displacement to core radius
$y$ = outer radial displacement
$Y_n, Y_{nm}$ = amplitude of displacement to outer radius

### Greek Letters

$\beta$ = surface tension ratio $\sigma_i/\sigma_o$
$\delta$ = viscosity coefficient ratio $\mu_1/\mu_2$
$\Delta$ = algebraic expression used in decay factor evaluation
$\lambda_n, \lambda_+, \lambda_-$ = decay factors
$\mu_1$ = dynamic viscosity of core fluid
$\mu_2$ = dynamic viscosity of shell fluid
$\rho_1, \rho_2$ = density of core and shell fluids, respectively
$\sigma_i$ = surface tension coefficient at common interface between core and shell fluid
$\sigma_o$ = surface tension coefficient at exterior boundary of compound drop
$\theta, \phi$ = angular coordinates

## APPENDIX

The fluid problem concerns viscous, low Reynolds number flow of the two incompressible, Newtonian fluids composing the compound drop. It is assumed that the effects of gravity and the much less viscous external medium are negligible. Initially the two fluids are at rest and the two interfaces are concentric and spherical, radius $R_i$ and $R_o (R_o > R_i)$. Let $\rho_1$, $\mu_1$ and $\rho_2$, $\mu_2$ be the density and dynamic viscosity of the core and shell fluids respectively; let $\sigma_i$ be the interfacial tension between the two fluids, while $\sigma_o$ is the surface tension of the shell fluid with the surrounding medium. The fluid pressures in the core and shell fluids in this state of rest are

$$\bar{p} = \bar{\mathcal{P}} + \frac{2\sigma_i}{R_i}, \quad \bar{\mathcal{P}} = \frac{2\sigma_o}{R_o}. \tag{A1}$$

The equations of motion which describe the motion of these fluids are highly nonlinear. However, if the drop is deformed slightly only small deviations about the initial state of equilibrium need to be studied. This results in a linearization of the equations.

In terms of spherical coordinates, $r$ and $\theta$, the small deviations in the radial and latitudinal velocity components and fluid pressure in the core fluid are $u(r,\theta,t), v(r,\theta,t), p(r,\theta,t)$, and for the shell fluid $U(r,\theta,t), V(r,\theta,t), \mathcal{P}(r,\theta,t)$ respectively. The $\phi$-dependence and longitudinal velocity components are ignored here since the final system describing the evolution behavior of the perturbations is the same for the axisymmetric and asymmetric cases (Landman and Greenspan, 1982; Miller and Scriven, 1968).

The linearized equations governing the core and shell fluids are as follows.

*Core Fluid:* $r < R_i$

Mass Conservation:

$$\frac{1}{r^2}(r^2 u)_r + \frac{1}{r\sin\theta}(v\sin\theta)_\theta = 0. \tag{A2}$$

Momentum Conservation:

$$\rho_1 u_t = -p_r + \mu_1 \frac{1}{r}\nabla^2(ru), \tag{A3a}$$

$$\rho_1 v_t = -\frac{1}{r}p_\theta + \mu_1\left(\nabla^2 v + \frac{2}{r^2}u_\theta - \frac{v}{r^2\sin^2\theta}\right). \tag{A3b}$$

*Shell Fluid:* $R_i < r < R_o$

Mass Conservation:

$$\frac{1}{r^2}(r^2 U)_r + \frac{1}{r\sin\theta}(V\sin\theta)_\theta = 0. \tag{A4}$$

Momentum Conservation:

$$\rho_2 U_t = -\mathcal{P}_r + \mu_2\frac{1}{r}\nabla^2(rU), \tag{A5a}$$

$$\rho_2 V_t = -\frac{1}{r}\mathcal{P}_\theta + \mu_2\left(\nabla^2 V + \frac{2}{r^2}U_\theta - \frac{V}{r^2\sin^2\theta}\right). \tag{A5b}$$

Let $x(\theta,t)$ be the displacement of the inner interface from the initial spherical configuration radius $R_i$; similarly, let $y(\theta,t)$ be the displacement from the initial radius $R_o$ of the compound drop. The boundary conditions expressing the balance of forces at the two interfaces involve pressure, viscous stress, and surface tension forces.

Normal Stress on $r = R_i$:

$$-p + 2\mu_1 u_r = -\mathcal{P} + 2\mu_2 U_r + \frac{\sigma_i}{R_i^2}\left[\frac{1}{\sin\theta}(\sin\theta x_\theta)_\theta + 2x\right]. \tag{A6}$$

Tangential Stress on $r = R_i$:

$$\mu_1(u_\theta + R_i v_r - v) = \mu_2(U_\theta + R_i V_r - V). \tag{A7}$$

Normal Stress on $r = R_o$:

$$-\mathcal{P} + 2\mu_2 U_r = \frac{\sigma_o}{R_o^2}\left[\frac{1}{\sin\theta}(\sin\theta y_\theta)_\theta + 2y\right]. \tag{A8}$$

Tangential Stress on $r = R_o$:

$$U_\theta + R_o V_r - V = 0 \tag{A9}$$

Furthermore, since the fluids are viscous, both the radial and tangential velocity components must be continuous at the interface between the core and shell fluids. Also a kinematic condition at both interfaces requires the radial velocity of the interface to equal the radial velocity of the fluid at the interface.

Continuity of velocity components and kinematic condition on $r = R_i$:

$$u = U \tag{A10}$$

$$v = V \tag{A11}$$

$$u = x_t. \tag{A12}$$

Kinematic Condition at $r = R_o$:

$$U = y_t. \tag{A13}$$

It will be assumed that the flow in the two fluids has low Reynolds number, that is, $|\rho_1 u_t/\mu_1 r^{-1}\nabla^2(ru)|$ and $|\rho_2 U_t/\mu_2 r^{-1}\nabla^2(rU)|$ are both much less than unity (similar expressions involving $v_t$ and $V_t$ must also be valid). Then the lefthand sides of Eqs. A3 and A5 can be approximated by zero. These equations are then the linearized, viscous limit of the Navier-Stokes equations for incompressible fluids. These equations can be rewritten to give the pressures as harmonic functions. Solutions to the modified Eqs. A3 and A5 can be expressed in terms of spherical harmonics, the Legendre polynomials $P_n(\cos\theta)$.

For the core fluid in the region $r < R_i$,

$$p = \sum_{n=0}^{\infty} e^{\lambda_n t} F_n \left(\frac{r}{R_i}\right)^n P_n(\cos\theta)$$

$$u = \sum_{n=1}^{\infty} e^{\lambda_n t}\left[A_n\left(\frac{r}{R_i}\right)^{n-1} + \frac{nF_n}{2\mu_1(2n+3)}\left(\frac{r}{R_i}\right)^{n+1}\right]P_n(\cos\theta), \tag{A14}$$

$$v = \sum_{n=1}^{\infty} e^{\lambda_n t}\left[\frac{A_n}{n}\left(\frac{r}{R_i}\right)^{n-1} + \frac{(n+3)F_n}{2\mu_1(n+1)(2n+3)}\left(\frac{r}{R_i}\right)^{n+1}\right]\frac{d}{d\theta}P_n(\cos\theta),$$

and for the shell fluid in the region $R_i < r < R_o$,

$$P = \sum_{n=0}^{\infty} e^{\lambda_n t}\left[G_n\left(\frac{r}{R_i}\right)^n + H_n\left(\frac{r}{R_i}\right)^{-n-1}\right]P_n(\cos\theta),$$

$$U = \sum_{n=1}^{\infty} e^{\lambda_n t}\left[C_n\left(\frac{r}{R_i}\right)^{n-1} + D_n\left(\frac{r}{R_i}\right)^{-n-2} + \frac{nG_n}{2\mu_2(2n+3)}\left(\frac{r}{R_i}\right)^{n+1} + \frac{(n+1)H_n}{2\mu_2(2n-1)}\left(\frac{r}{R_i}\right)^{-n}\right]P_n(\cos\theta)$$

$$V = \sum_{n=1}^{\infty} e^{\lambda_n t}\left[\frac{C_n}{n}\left(\frac{r}{R_i}\right)^{n-1} - \frac{D_n}{n+1}\left(\frac{r}{R_i}\right)^{-n-2} + \frac{(n+3)G_n}{2\mu_2(n+1)(2n+3)}\left(\frac{r}{R_i}\right)^{n+1} - \frac{(n-2)H_n}{2\mu_2 n(2n-1)}\left(\frac{r}{R_i}\right)^{-n}\right]\frac{d}{d\theta}P_n(\cos\theta), \tag{A15}$$

where $A_n$, $C_n$, $D_n$, $F_n$, $G_n$, and $H_n$ are arbitrary real constants.

Solutions to the surface disturbances are expressed as

$$x = \sum_{n=2}^{\infty} e^{\lambda_n t} X_n P_n(\cos\theta),$$

$$y = \sum_{n=2}^{\infty} e^{\lambda_n t} Y_n P_n(\cos\theta). \tag{A16}$$

(There are no modes corresponding to $n = 0, 1$ since the volume and position of center of gravity of the perturbed droplet must remain fixed.)

These expressions are now substituted into all the boundary conditions, except for the kinematic conditions, Eqs. A12 and A13, and then $A_n$ through $H_n$ can be solved in terms of $X_n$ and $Y_n$. Here $M = R_o/R_i$.

Equation A6:

$$-2\mu_1(n-1)A_n - \frac{(n^2-n-3)}{(2n+3)}F_n + 2\mu_2(n-1)C_n - 2\mu_2(n+2)D_n + \frac{(n^2-n-3)}{(2n+3)}G_n - \frac{(n^2+3n-1)}{(2n-1)}H_n = \frac{\sigma_i}{R_i}(n+2)(n-1)X_n,$$

Equation A7:

$$\frac{2\mu_1(n-1)}{n}A_n + \frac{n(n+2)}{(n+1)(2n+3)}F_n - \frac{2\mu_2(n-1)}{n}C_n - \frac{2\mu_2(n+2)}{(n+1)}D_n - \frac{n(n+2)}{(n+1)(2n+3)}G_n - \frac{(n^2-1)}{n(2n-1)}H_n = 0,$$

Equation A8:

$$2\mu_2(n-1)C_nM^{n-1} - 2\mu_2(n+2)D_nM^{-n-2} + \frac{(n^2-n-3)}{(2n+3)}G_nM^{n+1} - \frac{(n^2+3n-1)}{(2n-1)}H_nM^{-n} = -\frac{\sigma_o}{R_o}(n+2)(n-1)Y_n, \tag{A17}$$

Equation A9:

$$\frac{2(n-1)}{n}C_nM^{n-1} + \frac{2(n+2)}{(n+1)}D_nM^{-n-2} + \frac{n(n+2)}{\mu_2(n+1)(2n+3)}G_nM^{n+1} + \frac{(n^2-1)}{\mu_2 n(2n-1)}H_nM^{-n} = 0,$$

Equation A10

$$A_n + \frac{nF_n}{2\mu_1(2n+3)} - C_n - D_n - \frac{nG_n}{2\mu_2(2n+3)} - \frac{(n+1)}{2\mu_2(2n-1)}H_n = 0,$$

Equation A11:

$$\frac{A_n}{n} + \frac{(n+3)}{2\mu_1(n+1)(2n+3)}F_n - \frac{C_n}{n} + \frac{D_n}{(n+1)} - \frac{(n+3)}{2\mu_2(n+1)(2n+3)}G_n + \frac{(n-2)}{2\mu_2 n(2n-1)}H_n = 0.$$

Finally, each of the six coefficients, being expressed in terms of $X_n$ and $Y_n$ can be substituted into the kinematic conditions, Eqs. A12 and A13:

$$A_n + \frac{nF_n}{2\mu_1(2n+3)} = \lambda_n X_n,$$

$$C_n M^{n-1} + D_n M^{-n-2} + \frac{nG_n}{2\mu_2(2n+3)} M^{n+1} + \frac{(n+1)}{2\mu_2(2n-1)} H_n M^{-n} = \lambda_n Y_n; \quad \text{(A18)}$$

this results in the linear system, which must be solved for $n \geq 2$:

$$\begin{bmatrix} a_{11} - \lambda_n & a_{12} \\ a_{21} & a_{22} - \lambda_n \end{bmatrix} \begin{bmatrix} Y_n \\ X_n \end{bmatrix} = \begin{bmatrix} 0 \\ 0 \end{bmatrix}. \quad \text{(A19)}$$

The coefficients $a_{ij}$ are functions of $\mu_1, \mu_2, \sigma_i, \sigma_o, M = R_o/R_i$, and $n$. They are omitted here for brevity but can be found in the discussion section of the paper.

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