Unsteady thermocapillary migration of isolated drops in creeping flow

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The problem of an isolated immiscible drop that slowly migrates due to unsteady thermocapillary stresses is considered. All physical properties except for interfacial tension are assumed constant for the two Newtonian fluids. Explicit expressions are found for the migration rate and stream functions in the Laplace domain. The resulting microgravity theory is useful, e.g., in predicting the distance a drop will migrate due to an impulsive interfacial temperature gradient as well as the time required to attain steady flow conditions from an initially resting state.

Keywords: drops; thermocapillary flow; transient flow; creeping flow; microgravity; Newtonian fluids

Introduction

A gradient in temperature along the interface of an immiscible drop will generally cause migration toward warmer fluid. This effect derives from nonuniform capillary stresses generated by the temperature gradient and the resulting Marangoni flow. The direction of movement is governed by the sign of the variation of interfacial tension with temperature, which is usually negative. Wozniak et al.[1] and Subramanian[2] have recently reviewed this subject.

Because migration rates caused by these thermocapillary stresses are in general much less than those caused by Archimedes force, interest in this phenomenon is usually restricted to reduced-gravity environments. In an orbiting spacecraft, thermocapillary stresses are expected to be a primary mechanism for drop movement in otherwise quiescent fluids. Examples in which such migration may be important include the fining of molten glasses during containerless processing,[3,4] the formation of uniform composites from binary fluids possessing a miscibility gap,[5] and the rejection of gas bubbles from melt–solid interfaces during the growth of crystals.[2] (The term drop here includes gas bubbles as well as liquid drops; the former may be viewed as drops of negligible viscosity, density, and thermal conductivity compared with these properties of the external liquid phase.)

In contrast with most prior investigations,[6–12] the present study considers how an unsteady temperature gradient along the drop interface affects the rate of migration. In applications, unsteady interfacial temperature gradients are expected to occur far more frequently than steady ones. For example, simple heating or cooling of an initially isothermal fluid containing a drop will usually generate an unsteady interfacial temperature gradient and result in an unsteady thermocapillary migration.

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In this class of problems, only the interfacial temperature, or more precisely, its surface gradient, is important in establishing the flows in the internal and external phases. We here assume the scaled interfacial temperature T_s is given by

$$T_s(\theta, t) = T_0(t) + A(t)\cos\theta \tag{1}$$

This is an axisymmetric distribution with the polar angle θ defined by the relation $\mathbf{n}\cdot\hat{\mathbf{x}} = \cos\theta$. Here, $\mathbf{n}$ is a unit normal on the spherical interface, and $\hat{\mathbf{x}}$ is a unit vector pointing along the axis of symmetry. Tangential thermocapillary stresses, which are proportional to $A(t)$, drive the flow and derive from the surface gradient of T_s. Due to passage into warmer fluid, the mean interfacial temperature T_0 varies with time t, but does not affect the tangential thermocapillary stresses.

Equation 1 contains only the first two terms of an infinite Legendre series that can represent any unsteady axisymmetric temperature distribution along a spherical surface. These higher terms affect the fluid flow in the vicinity of the drop, but do not influence the rate of migration. The main analysis is restricted to the above interfacial temperature distribution. An analysis for the general unsteady axisymmetric distribution is provided in the Appendix.

Laplace transforms are used here to solve the unsteady Stokes equations. Analysis yields explicit expressions for the migration velocity and stream functions in the transform space. The theory is illustrated in two specific problems that predict distances of migration under transient conditions.

Analysis

Consider a viscous drop (density = ρ_1, viscosity = μ_1) of constant radius R within a resting viscous fluid (density = $\rho_2 = \rho_1/\rho^*$, viscosity = $\mu_2 = \mu_1/\mu^*$) of effectively infinite extent. Through an appropriate mechanism, thermal energy is transferred to the interface such that its temperature is given by Equation 1, resulting in an unsteady thermocapillary flow.

Our primary goal is to predict the transient migration velocity $\mathbf{U}(t) = \hat{\mathbf{x}}U(t)$ by solution of the governing equations of motion. To keep the analysis tractable, we assume creeping flow, that all physical properties other than interfacial tension are independent of temperature, and that the drop is a perfect sphere. The latter assumption is subject to *a posteriori* justification. Interfacial tension is assumed to decrease linearly with increasing temperature, a property shared by many fluids.

A schematic diagram of the type of physical system under consideration is shown in Figure 1. To be concrete, the figure shows the fluid temperature far from the drop linearly increasing with distance in the $\hat{\mathbf{x}}$ direction. With appropriate auxiliary assumptions, the transient interfacial temperature will possess the $\cos\theta$ dependence required by Equation 1. Specific calculations involving this type of system are provided in the next section.

Reference scales selected to nondimensionalize the governing equations include the drop radius R for length, $\mu_i U_R/R$ $(i = 1, 2)$ for pressure and $U_R = (-\sigma_T)T_R/\mu_2$ for velocity. Here, σ_T is the derivative of interfacial tension with temperature, and the temperature scale T_R represents a characteristic temperature difference along the interface. We denote the reference scale for time by t_R, but only assign its value in specific applications of the theory. In a reference frame attached to the drop, it follows that the scaled velocity $\mathbf{v}_i$ and pressure p_i for phase i (drop = 1, host fluid = 2) satisfy the continuity equation

$$\nabla\cdot\mathbf{v}_i = 0 \quad \text{for } i = 1, 2 \tag{2}$$

and the unsteady Navier-Stokes equation

$$\varepsilon_i \frac{\partial}{\partial t}(\mathbf{v}_i + \mathbf{U}) + \mathrm{Re}_i\, \mathbf{v}_i\cdot\nabla\mathbf{v}_i = -\nabla p_i + \nabla^2\mathbf{v}_i \quad \text{for } i = 1, 2 \tag{3}$$

at every position $\mathbf{r}$ within the respective phases and at every time $t > 0$. The additional inertial term on the left-hand side of the latter equation represents a force due to the drop's noninertial reference frame. That equation also possesses two dimensionless parameters. The first one, ε_i, is the ratio of the characteristic viscous time R^2/ν_i to t_R. The second one is the Reynolds number $\mathrm{Re}_i = U_R R/\nu_i$, which represents the relative importance of inertial to viscous forces in the fluid. Our assumption of creeping flow permits this nonlinear term to be neglected in the following analysis. A condition on the tangential interfacial stresses,

$$\mathbf{n}\cdot(\mathbf{P}_2 - \mu^*\mathbf{P}_1)\times\mathbf{n} = \nabla_s T_s \times \mathbf{n} \quad \text{at } |\mathbf{r}| = 1 \tag{4}$$

provided the above thermocapillary velocity scale. In this equation, $\mathbf{P}_i = -\mathbf{I}p_i + [\nabla\mathbf{v}_i + (\nabla\mathbf{v}_i)^t]$ is the pressure tensor, and $\nabla_s = (\mathbf{I} - \mathbf{nn})\cdot\nabla$ is the surface gradient operator. The velocity fields must also satisfy conditions corresponding to both fluids initially at rest, $\mathbf{v}_1 = \mathbf{v}_2 = \mathbf{U} = \mathbf{0}$ at $t = 0$; a kinematic condition for a spherical interface, $\mathbf{n}\cdot\mathbf{v}_i = 0$ $(i = 1, 2)$ at $|\mathbf{r}| = 1$; continuity of the tangential interfacial velocities $\mathbf{n}\times\mathbf{v}_1 = \mathbf{n}\times\mathbf{v}_2$ at $|\mathbf{r}| = 1$; far-field conditions, $(\mathbf{v}_2, p_2) \to (-\mathbf{U}, p_\infty)$ as $|\mathbf{r}| \to \infty$; and Newton's law governing the motion

Notation

$A(t)$	See Equation 1
A'	A constant
B	See Equation 32
C_{ij}	See Equation 10
D	Defined in Equation 25
D_{ij}	See Equation 33
d	Scaled stopping distance, Equation 44
Δd	Scaled difference in travel distance, Equation 39
E^2	Partial differential operator, Equation 8
F	See Equation 9
$I_{n/2}$	Modified Bessel function of first kind of order $n/2$
$K_{n/2}$	Modified Bessel function of second kind of order $n/2$
k	Thermal conductivity
Ma	Marangoni number, $U_R R/\alpha$
$\mathbf{n}$	Unit vector normal to drop's interface
$\mathbf{P}$	Scaled pressure tensor
Pr	Prandtl number, ν/α
p	Scaled pressure
p_∞	Scaled pressure at infinity
q_{In}, q_{Kn}	Functions defined in Equations 26 and 27
R	Radius of drop
Re	Reynolds number, $U_R R/\nu$
r	Scaled radial coordinate
$\mathbf{r}$	Scaled position vector
s	Laplace variable
T	Scaled temperature
t	Scaled time
$\mathbf{U}$, U	Scaled migration velocity and its magnitude, $U = \lvert\mathbf{U}\rvert$
U_∞	Scaled steady-state migration velocity
$\mathbf{v}$	Scaled fluid velocity
v_r, v_θ	Scaled radial and polar velocity components
$\hat{\mathbf{x}}$	Unit vector along axis of symmetry
z	Complex number defined as $(s\varepsilon)^{1/2}$
z_α	Complex number defined as $(s\varepsilon_\alpha)^{1/2}$

Greek symbols

α	Thermal diffusivity
ε	Ratio of viscous time R^2/ν to reference time t_R
ε_M	Ratio of viscous-momentum time $\rho_1 R^2/\mu_2$ to reference time t_R
ε_α	Ratio of thermal time R^2/α to reference time t_R
θ	Polar angle defined by $\cos\theta = \hat{\mathbf{x}}\cdot\mathbf{n}$
μ	Viscosity
ν	Kinematic viscosity
ρ	Density
σ_T	Rate of change of interfacial tension with temperature
ψ	Scaled stream function

Subscripts

1	Drop phase
2	Host phase
R	Reference scale
s	Interfacial quantity

Superscript

*	Drop property/host property, e.g., $k^* = k_1/k_2$

Miscellaneous

‾	(overbar) Laplace transform
∇	Scaled gradient operator

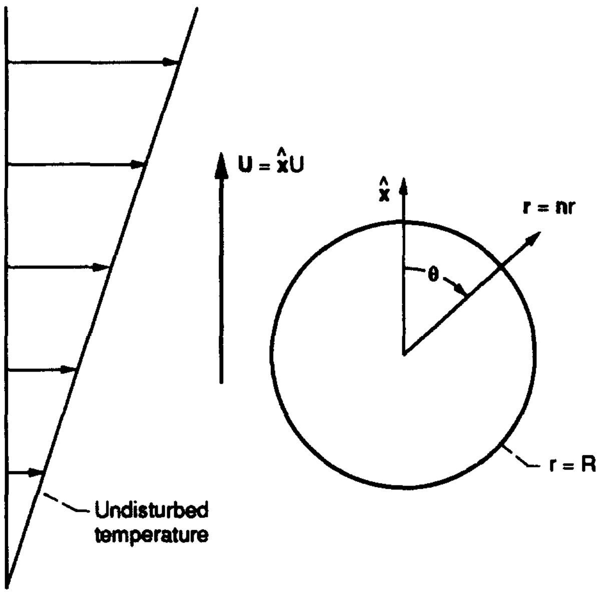

Figure 1 Thermocapillary migration of a drop. Here, a temperature gradient in fluid far from the drop induces a temperature gradient along the interface. The resulting gradient in interfacial tension causes the drop to migrate

of the drop:

$$\int_{|\mathbf{r}|=1} d\mathbf{S}\cdot\mathbf{P}_2 = \tfrac{4}{3}\pi\varepsilon_M \frac{d\mathbf{U}}{dt} \tag{5}$$

This last equation introduces another dimensionless time parameter, $\varepsilon_M = \rho^*\varepsilon_2$, which is associated with the drop's momentum. Here, $\rho^* = \rho_1/\rho_2$.

After setting Re_i $(i = 1, 2)$ to zero, four dimensionless parameters remain: ε_1, ε_2, ε_M, and μ^*. Only three parameters of this set are independent because ε_M may also be expressed as $\mu^*\varepsilon_1$. In the following, we use the alternative dependent set $[\varepsilon_1, \varepsilon_2, \mu^*, \rho^*]$ for notational convenience.

To solve this axisymmetric problem, we introduce a stream function ψ_i for each phase. The two scalar relations

$$\mathrm{v}_{ri} = \frac{-1}{r^2\sin\theta}\frac{\partial\psi_i}{\partial\theta}, \qquad v_{\theta i} = \frac{1}{r\sin\theta}\frac{\partial\psi_i}{\partial r} \quad \text{for } i = 1, 2 \tag{6a,b}$$

define ψ_i. Here, v_{ri} and $\mathrm{v}_{\theta i}$ are, respectively, the radial and polar components of the fluid velocity in phase i, and $r = |\mathbf{r}|$ is the radial coordinate of a spherical coordinate system (r, θ, ϕ), with corresponding orthogonal unit vectors $(\mathbf{n}, \mathbf{i}_\theta, \mathbf{i}_\phi)$. Each ψ_i satisfies the dimensionless partial differential equation

$$E^2\left(E^2 - \varepsilon_i\frac{\partial}{\partial t}\right)\psi_i = 0 \quad \text{for } i = 1, 2 \tag{7}$$

where the second-order partial differential operator E^2 is defined by

$$E^2 = \frac{\partial^2}{\partial r^2} + \frac{\sin\theta}{r^2}\frac{\partial}{\partial\theta}\left(\frac{1}{\sin\theta}\frac{\partial}{\partial\theta}\right) \tag{8}$$

The tangential stress condition suggests that a suitable trial solution for ψ_i is

$$\psi_i(r, \theta, t) = F_i(r, t)\sin^2\theta \quad \text{for } i = 1, 2 \tag{9}$$

wherein the function $F_i(r, t)$ must be determined. Following a Laplace transformation, the resulting ordinary differential equation possesses the general solution

$$\bar{F}_i(r; s) = C_{i1}(s)r^2 + C_{i2}(s)r^{-1} + C_{i3}(s)\frac{r^{1/2}I_{3/2}(z_i r)}{I_{3/2}(z_i)} + C_{i4}(s)\frac{r^{1/2}K_{3/2}(z_i r)}{K_{3/2}(z_i)} \quad \text{for } i = 1, 2 \tag{10}$$

Four unknown functions of s, C_{i1}, C_{i2}, C_{i3}, and C_{i4}, appear in the previous solution for each of the two phases. The transform of the migration velocity, $\bar{U}$, which appears within the boundary conditions, constitutes another unknown. Three of these, C_{12}, C_{14}, and C_{23}, vanish identically because of regularity requirements at the origin and at infinity. The remaining ones are determined from the following six boundary conditions:

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$$\bar{F}_2 \to \tfrac{1}{2}\bar{U}r^2 \quad \text{as } r \to \infty \tag{11}$$

$$\bar{F}_i = 0 \quad \text{at } r = 1 \quad \text{for } i = 1, 2 \tag{12}$$

$$\frac{d\bar{F}_1}{dr} = \frac{d\bar{F}_2}{dr} \quad \text{at } r = 1 \tag{13}$$

$$\frac{d}{dr}\left[r^{-2}\frac{d}{dr}(\bar{F}_2 - \mu^*\bar{F}_1)\right] = -\bar{A} \quad \text{at } r = 1 \tag{14}$$

$$\frac{d^2\bar{F}_2}{dr^2} = -\left(\frac{1 + 2\rho^*}{6}\right)\varepsilon_2 s\bar{U} \quad \text{at } r = 1 \tag{15}$$

The previous equations, respectively, correspond to the far-field condition on the fluid velocity, the vanishing of both radial velocities at the interface, continuity of tangential velocities at the interface, the tangential stress jump condition, and Newton's law as applied to the entire drop. These conditions lead to the following solution:

$$C_{11} = -C_{13} \tag{16}$$

$$C_{12} = 0 \tag{17}$$

$$C_{13} = \bar{U}\left[\frac{q_{K1}(z_2)(1 + 2\rho^*) + 9}{6q_{I2}(z_1)}\right] \tag{18}$$

$$C_{14} = 0 \tag{19}$$

$$C_{21} = \tfrac{1}{2}\bar{U} \tag{20}$$

$$C_{22} = -(C_{21} + C_{24}) \tag{21}$$

$$C_{23} = 0 \tag{22}$$

$$C_{24} = -\bar{U}\left(\frac{1 + 2\rho^*}{6}\right) \tag{23}$$

$$\bar{U}(s) = \frac{6\bar{A}(s)}{D(s)} \tag{24}$$

Here, $D(s)$, $q_{Kn}(z)$, and $q_{In}(z)$ are functions defined as

$$D(s) = s\varepsilon_2(1 + 2\rho^*) + \{2 + \mu^*[3 + q_{I3}(z_1)]\} \times [9 + q_{K1}(z_2)(1 + 2\rho^*)] \tag{25}$$

$$q_{Kn}(z) = \frac{zK_{n-1/2}(z)}{K_{n+1/2}(z)} \quad \text{for } n = 1, 2, 3, \ldots \tag{26}$$

$$q_{In}(z) = \frac{zI_{n+1/2}(z)}{I_{n-1/2}(z)} \quad \text{for } n = 1, 2, 3, \ldots \tag{27}$$

Cancellation of common exponential factors in $K_{n-1/2}(z)$ and $K_{n+1/2}(z)$ permits $q_{Kn}(z)$ to be expressed as ratios of two polynomials in z. For example, $q_{K1}(z)$ is given by

$$q_{K1}(z) = \frac{z^2}{1+z} \tag{28}$$

For small z, it can be shown that $q_{Kn}(z)$ and $q_{In}(z)$ are proportional to z^2. It follows that at $s = 0$ the denominator $D(0)$ equals $9(2 + 3\mu^*)$.

If the interfacial temperature is given by Equation 1, with $A(t)$ equal to some constant, say A′, the present unsteady theory is expected to pass over to the steady theory of Young et al.[6] (with gravity neglected) as $t \to \infty$. The terminal migration velocity U_∞ is related to $\bar{U}$ by the final value theorem of Laplace transforms:

$$U_\infty = \lim_{t\to\infty} U(t) = \lim_{s\to 0} s\bar{U}(s)$$

Introduction of Equation 24 yields

$$U_\infty = \frac{2}{3}\frac{A'}{(2 + 3\mu^*)} \tag{29}$$

for U_∞. The dimensional result of Young et al. follows directly on setting the temperature scale T_R to RG and A′ to $3/(2 + k^*)$. Here, G is the magnitude of the temperature gradient at infinity, and $k^* = k_1/k_2$ is the thermal conductivity ratio. This value of A′ derives from solution of the steady-state energy equations. The flow field can similarly be shown to asymptotically approach the result of Young et al. as $t \to \infty$.

Because of the assumed spherical shape of the drop, we have not required satisfaction of the normal stress condition. According to Taylor and Acrivos,[15] the present solution can be used to determine small deviations from the spherical shape via examination of predicted excess normal stresses. We calculated these excess stresses in the Laplace domain and found them to be identically zero at every interfacial point independently of the value of s. This leads to the conclusion that the drop remains spherical even during transient flow. Proof that the normal stress condition in the Laplace domain,

$$\mathbf{n}\cdot(\bar{\mathbf{P}}_2 - \mu^*\bar{\mathbf{P}}_1)\cdot\mathbf{n} = 2\bar{\sigma} \quad \text{at } r = 1$$

is exactly satisfied is rather lengthy, and so we refer the reader to Chisnell,[16] who performed a similar calculation for unsteady buoyancy-driven migration. (In the above equation, $\bar{\sigma}$ is the transform of a dimensionless interfacial tension. The scaling factor is $-\sigma_T T_R$.)

The normal stress condition is expected to be violated during transient motion if the interfacial temperature distribution is not given by the truncated Legendre series (Equation 1). Levan[17] indicates that if additional terms are required in the temperature series, the normal stress condition is not satisfied under steady conditions. For such temperature distributions, therefore, we should expect some deformation under unsteady conditions as well.

Analytic inversion of the above transforms appears difficult. Crump,[18] however, gives an efficient numerical method for inversion of Laplace transforms that provides an approximation of the error. We used a standard IMSL routine based on this method to investigate possible difficulties associated with numerical inversion. It was found that calculations of ratios of modified Bessel functions of the first kind (e.g., $q_{In}(z)$) could cause an overflow condition for large values of real (z). This problem can be overcome by letting $z = x + iy$ and expressing the complex hyperbolic functions in terms of $\sinh(x)$, $\cosh(x)$, $\sin(y)$, and $\cos(y)$.[14] As $x \to \infty$, exponentially small terms decay and the two hyperbolic functions become asymptotically equal. For $x = 21$, e.g., the relative difference between $\sinh(x)$ and $\cosh(x)$ is $0(10^{-18})$. Setting these quantities as equal for large x permits cancellation of exponentially large factors in both numerator and denominator and thereby avoids the problem of overflow. The remaining numerical code follows directly from the formulas provided.

Discussion

Two applications of the above theory are provided here: (1) a drop migrating from rest within a fluid possessing a uniform temperature gradient at infinity and (2) a drop subjected to a brief interfacial temperature gradient. In both cases, distances traversed in transient motion are calculated.

Migration from rest to steady flow

Consider a drop spontaneously appearing within a resting fluid that possesses a uniform temperature gradient. This may occur, e.g., due to homogeneous nucleation from a supersaturated solution. Alternatively, the drop may appear from a simple injection of one fluid into another. However the drop appears, we assume disturbances associated with the generation process decay extremely fast so that the thermocapillary flow may be assumed to begin from rest. Another assumption is that the initial (scaled) temperature of the drop $T_0(0)$ equals the undisturbed temperature corresponding to the drop's center. This ensures that the drop's interfacial temperature can be represented by Equation 1. Our problem is to determine the interfacial temperature as a function of time (i.e., $A(t)$, or more precisely, $\bar{A}$) so as to calculate the instantaneous migration velocity $U(t)$ (Equation 24).

Let $\mathbf{G} = G\hat{\mathbf{x}}$ be the undisturbed temperature gradient, and RG the reference temperature scale T_R. Suppose the scaled temperature of the drop phase is given by $T_0(t) + T_1(\mathbf{r}, t)$, and that of the host fluid by $T_0(t) + \mathbf{r}\cdot\hat{\mathbf{x}} + T_2(\mathbf{r}, t)$; these expressions are definitions of the two unknown thermal disturbance fields T_i $(i = 1, 2)$. Here, $T_0(t)$ is the temperature at the center of the drop at time t. Introduction of the above expressions for temperature into the energy equation yields

$$\varepsilon_{\alpha 1}\frac{\partial T_1}{\partial t} + \mathrm{Ma}_1(U + \mathbf{v}_1\cdot\nabla T_1) = \nabla^2 T_1 \tag{30}$$

for the drop fluid and

$$\varepsilon_{\alpha 2}\frac{\partial T_2}{\partial t} + \mathrm{Ma}_2[U + \mathbf{v}_2\cdot(\hat{\mathbf{x}} + \nabla T_2)] = \nabla^2 T_2 \tag{31}$$

for the host fluid. For the two phases, these equations possess four dimensionless parameters: two thermal time scales $\varepsilon_{\alpha i} = R^2/\alpha_i t_R$ $(i = 1, 2)$ and two Marangoni numbers (or equivalently, thermal Peclet numbers) $\mathrm{Ma}_i = U_R R/\alpha_i$ $(i = 1, 2)$. Another parameter, the conductivity ratio k^*, enters through the flux boundary condition. The assumption of negligible convective transport is equivalent to both Marangoni numbers being small compared with unity and leads to a linear system of unsteady equations commonly encountered in studies of heat conduction in composite solids.[19] The resulting equations depend on the three remaining parameters, which are independent.

Imposed boundary conditions are as follows. At the interface, both temperature and thermal flux are continuous for $t > 0$. (These interfacial conditions require jumps in the disturbance temperatures and in their fluxes.) The initial conditions $T_i(\mathbf{r}, 0) = 0$ $(i = 1, 2)$ correspond to the drop's initial temperature being $T_0(0)$ and that of the host fluid as $T_0(0) + \mathbf{r}\cdot\hat{\mathbf{x}}$. (Note the momentary discontinuity in temperature along the interface.) Lastly, the far-field condition is $T_2(\mathbf{r}, t) \to 0$ as $|\mathbf{r}| \to \infty$.

Let

$$T_i(r, \theta, t) = B_i(r, t) \cos\theta \quad \text{for } i = 1, 2 \tag{32}$$

be trial solutions for the dimensionless thermal disturbance fields. Following a Laplace transformation, the $\bar{B}_i$ functions are found to obey a second-order ordinary differential equation with the general solution

$$\bar{B}_i(r; s) = D_{i1}(s) \frac{I_{3/2}(z_{\alpha i} r)}{r^{1/2} I_{3/2}(z_{\alpha i})} + D_{i2}(s) \frac{K_{3/2}(z_{\alpha i} r)}{r^{1/2} K_{3/2}(z_{\alpha i})} \quad \text{for } i = 1, 2 \tag{33}$$

Here, $z_{\alpha i} = (s\varepsilon_{\alpha i})^{1/2}$. Regularity at the origin and at infinity requires that functions $D_{12}(s)$ and $D_{21}(s)$ vanish identically. Application of the interfacial boundary conditions yields

$$D_{11} = \frac{1}{s} \frac{3 + q_{K1}(z_{\alpha 2})}{2 + q_{K1}(z_{\alpha 2}) + k^*[1 + q_{I2}(z_{\alpha 1})]} \tag{34}$$

$$D_{22} = D_{11} - \frac{1}{s} \tag{35}$$

for the remaining two functions of s. Inversion of the above solution gives the temperature field at any point for $t > 0$.

This solution yields $\bar{A}$, which is closely related to the interfacial temperature gradient. Set $r = 1$ in Equation 32 and compare with Equation 1 to find

$$\bar{A}(s) = \bar{B}_1(1; s) \equiv D_{11}(s) \tag{36}$$

Introduction into Equation 24 then yields an explicit formula for the transformed thermocapillary migration velocity $\bar{U}$. Observe that, according to the final value theorem, $A(t) \to 3/(2 + k^*)$ as $t \to \infty$; i.e., the known steady-state result is recovered. Both thermal and flow fields are now known in the Laplace domain. These results may now be used to calculate various quantities of physical interest.

We note that the above results are consistent with those previously calculated for a gas bubble wherein the material property ratios μ^*, k^*, and ρ^* were taken as identically zero.[20]

One interesting prediction of this theory is that during the transient regime a drop may migrate faster than its terminal velocity. Expansion of $\bar{A}$ for small s followed by inversion leads to the long-time result[21]

$$A(t) \simeq \frac{3}{2 + k^*}\left[1 - \frac{1}{6\pi^{1/2}}\left(\frac{1 - k^*}{2 + k^*}\right)\left(\frac{\varepsilon_{\alpha 2}}{t}\right)^{3/2} + 0(t^{-5/2})\right] \quad \text{as } t \to \infty \tag{37}$$

According to this expression, if the thermal conductivity ratio k^* exceeds unity, $A(t)$ will exceed its steady-state value of $3/(2 + k^*)$ so that tangential capillary stresses will also exceed their terminal magnitude. A similar inversion for $\bar{U}$ gives

$$U(t) \simeq \frac{2}{(2 + k^*)(2 + 3\mu^*)} \times \left\{1 - \frac{1}{6\pi^{1/2}}\left[\frac{(1 - k^*)}{(2 + k^*)} \mathrm{Pr}_2^{3/2} + \frac{(1 + 2\rho^*)}{3}\right] \times \left(\frac{\varepsilon_2}{t}\right)^{3/2} + 0(t^{-5/2})\right\} \quad \text{as } t \to \infty \tag{38}$$

wherein $\mathrm{Pr}_2 = \nu_2/\alpha_2 = \varepsilon_{\alpha 2}/\varepsilon_2$ is the Prandtl number of phase 2. The expression predicts the terminal velocity, $U_\infty = 2[(2 + k^*)(2 + 3^*)]^{-1}$, will be exceeded if the quantity in large square brackets is negative. The latter is favored by $k^* > 1$ with $\mathrm{Pr}_2 \gg 1$. If Pr_2 is large, the migration rate responds quasistatically to a slowly developing thermal field.

The above conclusion is supported by an exact calculation of the difference in distance traveled by one drop moving at its terminal velocity and an identical one starting from rest. Let Δd be this distance scaled with the length $U_R t_R$. From its definition,

$$\Delta d = \int_0^\infty [U(t) - U_\infty] dt \tag{39}$$

and standard Laplace transform theory, we find the equivalent relation

$$\Delta d = \lim_{s \to 0} (\bar{U} - U_\infty/s) \tag{40}$$

Use of Equations 24, 28, and 36 leads to the result

$$\Delta d = U_\infty\left[\frac{5\varepsilon_{\alpha 2}(k^* - 1) - 3\varepsilon_{\alpha 1}k^*}{15(k^* + 2)} - \frac{(1 + 2\rho^*)(1 + \mu^*)\varepsilon_2 + \frac{3}{7}\mu^*\varepsilon_1}{3(2 + 3\mu^*)}\right] \tag{41}$$

A negative value for the bracketed expression corresponds to the drop in unsteady motion lagging the one in steady motion.

The first term in this expression involves only thermal properties of the two fluids; it is negative unless $k^* > 1$ and the thermal diffusivity ratio $\alpha^* = \varepsilon_{\alpha 2}/\varepsilon_{\alpha 1}$ is sufficiently large. The second term involves only hydrodynamic quantities; it is negative for all values of parameters. According to the complete expression, for most parametric ranges a drop starting from rest will move a smaller distance than one moving at the terminal velocity. However, if the thermal conductivity ratio exceeds unity, the drop not only can temporarily exceed the terminal migration rate (as predicted by Equation 38), but can outdistance a steadily moving drop. This latter behavior is favored if $\varepsilon_{\alpha 2}$ is much slower (i.e., much greater in magnitude) than the other four natural time scales.

Except for the case $\rho^* \gg 1$, Equation 38 indicates that the drop attains a steady rate of migration prior to moving one radius. To see this, let t_R equal the convective time scale R/U_R. Then $t = U_\infty^{-1}$ is the time required for a drop moving at U_∞ to traverse the distance of one radius. The dimensionless time parameters ε_i and $\varepsilon_{\alpha i}$ here equal Re_i and Ma_i, respectively, and are then by assumption much less than unity. According to Equation 38, $U(U_\infty^{-1})$ can differ significantly from U_∞ only if $\rho^* \gg 1$. (The product $\mathrm{Pr}_2\varepsilon_2$ equals Ma_2, which is by assumption small.)

Equation 41 similarly indicates that, unless $\rho^* \gg 1$, the absolute difference in distance traveled by the two drops is less than a single radius. With the above selection for t_R, Δd is scaled by the radius R. The absolute value of the right-hand side of this expression can only exceed unity if $\rho^* \gg 1$. Physically, this exceptional case corresponds to a liquid drop migrating in a gas phase. We note that similar though qualitative conclusions could have been drawn from consideration of the original transport equations.

For completeness, we also provide asymptotic results for very small times. Expansion of the Laplace transforms $\bar{A}$ and $\bar{U}$ for large values of s followed by inversion gives

$$A(t) = \left(1 + \frac{k^*}{\alpha^{*1/2}}\right)^{-1} + 0(t^{1/2}) \quad \text{for } t \ll 1 \tag{42}$$

$$U(t) = 6\left\{\left(1 + \frac{k^*}{\alpha^{*1/2}}\right)[1 + (\mu^*\rho^*)^{1/2}](1 + 2\rho^*)\right\}^{-1} \times \frac{t}{\varepsilon_2} + 0(t^{3/2}) \quad \text{for } t \ll 1 \tag{43}$$

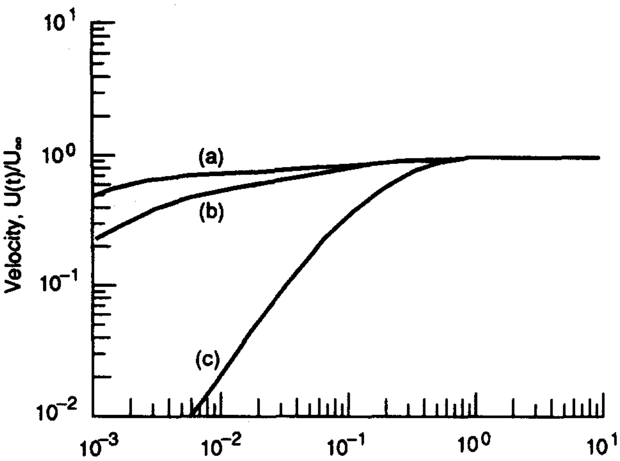

Figure 2 Transient velocities for (a) gas bubble in liquid host ($\varepsilon_1 = \varepsilon_2 = 10^{-2}$, $\mu^* = \rho^* = 10^{-3}$, $\varepsilon_{\alpha 1} = 10^{-2}$, $\varepsilon_{\alpha 2} = 1$, $k^* = 0.1$), (b) liquid drop in liquid host ($\varepsilon_1 = \varepsilon_2 = 10^{-2}$, $\mu^* = \rho^* = 1 = \varepsilon_{\alpha 1} = \varepsilon_{\alpha 2} = k^* = 1.0$), and (c) liquid drop in gaseous host ($\varepsilon_1 = \varepsilon_2 = 10^{-3}$, $\mu^* = \rho^* = 10^3$, $\varepsilon_{\alpha 1} = 0.1$, $\varepsilon_{\alpha 2} = 10^{-3}$, $k^* = 10$)

Equation 42 correctly predicts that for every position along the interface the interfacial temperature at the initial moment lies between the prescribed values for the inner and outer phases. Observe that this initial value may be larger or smaller than the terminal value predicted by Equation 37.

The migration velocity $U(t)$ as a function of time was investigated for a wide range of the five independent parameters. Figure 2 provides representative plots of transient migration velocities, scaled by the associated terminal velocity U_∞, for a gas bubble, a liquid drop immersed in another liquid, and a liquid drop in a gas. In each case, the slowest of the five natural time scales was used for t_R. In general, the slow scale was found to govern the approach to steady state. As expected, if either of the inner time scales, ε_1 or $\varepsilon_{\alpha 1}$, is the slowest, and μ^* or k^*, respectively, is small, the slowest of the remaining time scales determines the time for U to approach steady state. In these cases, the migration attains steady state prior to transport processes within the interior of the drop phase.

Figure 3 also shows the migration velocity as a function of time for a liquid drop within a liquid host phase. Shown too are the relevant short- and long-time asymptotic predictions based on Equations 43 and 38, respectively. The displayed results appear to be typical of the several cases investigated: the two asymptotic expressions are valid in their appropriate limits, but unfortunately their domains of validity do not overlap.

Experiments conducted under low gravity conditions frequently are severely limited in the time available for experimentation. Drop towers, e.g., provide little more than 5 seconds to complete the entire experiment. The above theory can provide quantitative guidance to predict the time required to approach steady state. A rational approach might be to calculate the quantity $(U(t) - U_\infty)/U_\infty$ as a measure of deviation from steady migration. When this ratio decreases below a level governed by experimental uncertainty, the radius of the largest suitable drop can be easily calculated. In most test calculations, this ratio was less than 0.5 percent when $t > 5$.

Migration from rest to rest

Consider the problem of predicting the distance a drop will travel due to the brief imposition of an interfacial thermal gradient. This could occur, e.g., by briefly warming a drop in an otherwise isothermal fluid with a laser.[22] As before, we

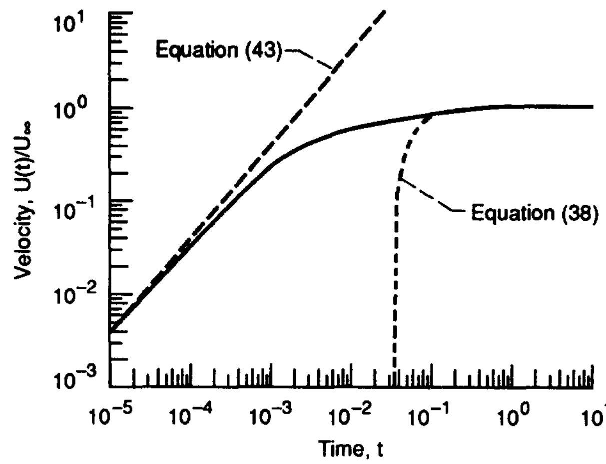

Figure 3 Transient velocity of a liquid drop in a liquid host ($\varepsilon_1 = \varepsilon_2 = 0.01$, $\mu^* = \rho^* = \varepsilon_{\alpha 1} = \varepsilon_{\alpha 2} = 1$, $k^* = 0.8$). Results from numerical inversion (solid line) are compared with those from short- and long-time asymptotic expansions

assume the fluids are at rest prior to $t = 0$. For $t > 0$, we merely require that $A(t)$ (cf. Equation 1) decays sufficiently fast that the improper integral

$$\int_0^\infty A(t)\,dt$$

exists. Let d be the distance between the initial and final resting positions of the drop, made nondimensional with the length scale $U_R t_R$. Here, t_R is a time scale associated with the duration of $A(t)$. By definition, d is given by the integral

$$d = \int_0^\infty U(t)\,dt \tag{44}$$

A procedure similar to that used for Δd leads to the expression

$$d = \frac{2}{3(2 + 3\mu^*)} \int_0^\infty A(t)\,dt \tag{45}$$

This result is interesting in that, though the viscous time scales ε_1, ε_2, and ε_M (with μ^* constant) affect the instantaneous migration velocity, they have no influence on the total distance traveled.

Observe that if A′ is defined as the value

$$A' = \int_0^\infty A(t)\,dt \tag{46}$$

the above expression can be written simply as

$$d = U_\infty \tag{47}$$

Here, U_∞ is the steady-state velocity associated with A′ (Equation 29).

Conclusions

The viscous flow associated with a drop, migrating due to capillary forces that result from an unsteady axisymmetric interfacial temperature distribution, is solved analytically in the Laplace domain. The theory is useful in predicting transient fluid motion for a wide range of thermocapillary migration problems.

References

1 Wozniak, G., Siekmann, J. and Srulijes, J. Thermocapillary bubble and drop dynamics under reduced gravity—Survey and prospects. *Z. Flugwiss. Weltraumforsch.*, 1988, **12**, 137–144

2 Subramanian, R. S. *Transport Processes in Bubbles, Drops, and Particles* (R. P. Chhabra and D. DeKee, Eds.), Hemisphere Publishing, New York, 1992

3 Annamalai, P., Shankar, N., Cole, R. and Subramanian, R. S. Bubble migration inside a liquid drop in a space laboratory. *Appl. Sci. Res.*, 1982, **38**, 179–186

4 Mattox, D. M., Smith, H. D., Wilcox, W. R. and Subramanian, R. S. Thermal-gradient-induced migration of bubbles in molten glass. *J. Am. Ceram. Soc.*, 1982, **65**, 437–442

5 Gelles, S. H. and Markworth, A. J. Microgravity studies in the liquid-phase immiscible system: Aluminum-indium. *AAIA J.*, 1978, **16**, 431–438

6 Young, N. O., Goldstein, J. S. and Block, M. J. The motions of bubbles in a vertical temperature gradient. *J. Fluid Mech.*, 1959, **6**, 350–356

7 Balasubramaniam, R. and Chai, A.-T. Thermocapillary migration of droplets: An exact solution for small Marangoni numbers. *J. Colloid Interface Sci.*, 1987, **119**, 531–538

8 Subramanian, R. S. Slow migration of a gas bubble in a thermal gradient. *AIChE J.*, 1981, **27**, 646–654

9 Subramanian, R. S. Thermocapillary migration of bubbles and droplets. *Adv. Space Res.*, 1983, **3**, 145–153

10 Shankar, N. and Subramanian, R. S. The Stokes motion of a gas bubble due to interfacial tension gradients at low to moderate Marangoni numbers. *J. Colloid Interface Sci.*, 1988, **123**, 512–522

11 Szymczyk, J. and Siekmann, J. Numerical calculation of the thermocapillary motion of a bubble under microgravity. *Chem. Eng. Comm.*, 1988, **69**, 129–147

12 Balasubramaniam, R. and Lavery, J. E. Numerical simulation of thermocapillary bubble migration under microgravity for large Reynolds and Marangoni numbers. *Numer. Heat Trans.*, 1989, **16**, 175–187

13 Happel, J. and Brenner, H. *Low Reynolds Number Hydrodynamics*, Noordhoff International Publishing, Leyden, The Netherlands, 1973

14 Abramovitz, M. and Stegun, I. A., Eds. *Handbook of Mathematical Functions with Formulas, Graphs, and Mathematical Tables*, National Bureau of Standards, Applied Mathematics Series 55, Washington, DC, 1972

15 Taylor, T. D. and Acrivos, A. On the deformation and drag of a falling viscous drop at low Reynolds number. *J. Fluid Mech.*, 1964, **18**, 466–476

16 Chisnell, R. F. The unsteady motion of a drop moving vertically under gravity. *J. Fluid Mech.*, 1987, **176**, 443–464

17 Levan, M. D. Motion of a droplet with a Newtonian interface. *J. Colloid Interface Sci.*, 1981, **83**, 11–17

18 Crump, K. S. Numerical inversion of Laplace transforms using a Fourier series approximation. *J. Assoc. Comput. Machinery*, 1976, **23**, 89–96

19 Carslaw, H. S. and Jaeger, J. C. *Conduction of Heat in Solids*, 2nd ed. Oxford University Press, Oxford, 1986

20 Dill, L. H. and Balasubramaniam, R. Unsteady thermocapillary migration of bubbles. *Drops and Bubbles, Third International Colloquium, AIP Conference Proceeding 197* (T. G. Wang, Ed.), American Institute of Physics, New York, 1989, 481–486 (Also, NASA TM 101338, 1988)

21 Arpaci, V. S. *Conduction Heat Transfer*. Addison-Wesley, Reading, MA, 1966

22 Oliver, D. L. R. and DeWitt, K. J. Surface tension driven flows for a droplet in a micro-gravity environment. *Int. J. Heat Mass Trans.*, 1988, **31**, 1534–1537

Appendix: Flow due to a general unsteady axisymmetric interfacial temperature gradient

An arbitrary unsteady axisymmetric temperature along the surface of a spherical drop can be expressed by the series

$$T_s(x,t) = \sum_{n=0}^{\infty} A_n(t) P_n(x) \tag{A1}$$

Here, x equals $\cos\theta$ and $P_n(x)$ is the Legendre polynomial of order n. Unsteady coefficients in the above series are related to the surface temperature by

$$A_n(t) = \frac{2n+1}{2}\int_{-1}^{1} T_s(x,t) P_n(x)\,dx \quad \text{for } n = 0, 1, 2, \ldots \tag{A2}$$

In the following, each $A_n(t)$ is assumed to possess a Laplace transform.

We here determine the viscous flow in the Laplace domain that results from the above interfacial temperature. Velocity fields $\mathbf{v}_i$ $(i = 1, 2)$ are assumed to satisfy the same conditions as before, i.e., Equations 2–5 and associated initial and boundary conditions. The stream function for phase i is given by the series[13]

$$\psi_i(r,x,t) = \sum_{n=2}^{\infty} F_i^{(n)}(r,t)\mathscr{C}_n^{-1/2}(x) \quad \text{for } i = 1, 2 \tag{A3}$$

and satisfies differential Equation 7. Here, the $F_i^{(n)}(r,t)$ are functions to be determined and $\mathscr{C}_n^{-1/2}(x)$ are Gegenbauer polynomials of order n and degree $-1/2$. Happel and Brenner[13] provide several properties of Gegenbauer polynomials, including their relation to Legendre polynomials. The lower-order Gegenbauer polynomials are given by $\mathscr{C}_0^{-1/2}(x) = 1$, $\mathscr{C}_1^{-1/2}(x) = -x = -\cos\theta$, and $\mathscr{C}_2^{-1/2}(x) = 1/2\,(1-x^2)^{1/2} = 1/2\sin^2\theta$. Following a Laplace transformation, the $\bar{F}_i^{(n)}$ are found to possess the general solution

$$\bar{F}_i^{(n)}(r;s) = C_{i1}^{(n)}(s)r^n + C_{i2}^{(n)}(s)r^{-n+1} + C_{i3}^{(n)}(s)r^{1/2}\frac{I_{n-1/2}(z_i r)}{I_{n-1/2}(z_i)} + C_{i4}^{(n)}(s)r^{1/2}\frac{K_{n-1/2}(z_i r)}{K_{n-1/2}(z_i)}$$
$$\text{for } i = 1, 2, 3, \ldots;\ n = 2, 3, 4, \ldots \tag{A4}$$

This equation obviously reduces to the solution given in Equation 10 for the case $n = 2$. The $C_{ij}^{(n)}(s)$ are unknown functions of s that are determined from the boundary conditions.

In Stokes flow, only the first term of Equation A3 (the $n = 2$ term) affects the hydrodynamic force on the drop. The remaining terms contribute to the flow field, but because of symmetries with respect to the polar angle, their net contribution to the hydrodynamic force is identically zero.[13] Furthermore, standard identities give

$$\nabla_s T_s = -\mathbf{i}_\theta \sum_{n=2}^{\infty} A_{n-1}\frac{n(n-1)\mathscr{C}_n^{-1/2}(x)}{(1-x^2)^{1/2}} \tag{A5}$$

for the surface gradient of temperature. Introduction of this expression into Equation 4 leads to the tangential stress condition given below in Equation A9. It is clear from the latter equation that the $n-1$ harmonic of the temperature distribution is linked solely with the n^{th} harmonic of the stream function. From this we conclude that the migration velocity for a drop possessing the above unsteady interfacial temperature is given by the same expression as before, i.e., Equation 24. Also, the coefficients $C_{ij}^{(2)}$ are seen to equal $2C_{ij}$, which are defined in Equations 16–24. The factor of 2 derives from the relation $\mathscr{C}_2^{-1/2}(\cos\theta) = 1/2\sin^2\theta$.

For $n = 3, 4, 5, \ldots$, the $\bar{F}_i^{(n)}$ must satisfy the following boundary conditions:

$$\bar{F}_2^{(n)} \to 0 \quad \text{as } r \to \infty \tag{A6}$$

$$\bar{F}_i^{(n)} = 0 \quad \text{at } r = 1 \quad \text{for } i = 1, 2 \tag{A7}$$

$$\frac{d\bar{F}_1^{(n)}}{dr} = \frac{d\bar{F}_2^{(n)}}{dr} \quad \text{at } r = 1 \tag{A8}$$

$$\frac{d}{dr}\left[r^{-2}\frac{d}{dr}(F_2^{(n)} - \mu^* F_1^{(n)})\right] = -n(n-1)\bar{A}_{n-1} \quad \text{at } r = 1 \tag{A9}$$

These equations may be compared with the boundary conditions on $\bar{F}_i$, i.e., Equations 11–15. Regularity conditions, including Equation A6, require four of these coefficient functions of s to vanish. The four interfacial conditions, Equations A7–A9, respectively, define the remaining four functions. These conditions yield the solution

$$C_{11}^{(n)} = \frac{n(n-1)\bar{A}_{n-1}}{\dfrac{q_{In}(z_1)}{q_{K(n-1)}(z_2)}[2q_{K(n-1)}(z_2) + z_2^2] - \mu^*[2q_{In}(z_1) - z_1^2]} \tag{A10}$$

$$C_{12}^{(n)} = 0 \tag{A11}$$

$$C_{13}^{(n)} = -C_{11}^{(n)} \tag{A12}$$

$$C_{14}^{(n)} = 0 \tag{A13}$$

$$C_{21}^{(n)} = 0 \tag{A14}$$

$$C_{22}^{(n)} = \frac{-q_{In}(z_1)C_{11}^{(n)}}{q_{Kn}(z_2)} \tag{A15}$$

$$C_{23}^{(n)} = 0 \tag{A16}$$

$$C_{24}^{(n)} = -C_{22}^{(n)} \tag{A17}$$

Together with Equations 16–24 (and noting the factor of 2 between $C_{ij}^{(2)}$ and C_{ij}), the above analysis represents a complete solution of the flow fields in both phases.