Thermocapillary Migration of a Drop

An Exact Solution with Newtonian Interfacial Rheology and Stretching/Shrinkage of Interfacial Area Elements for Small Marangoni Numbers

R. BALASUBRAMANIAM\textsuperscript{a} AND R. SHANKAR SUBRAMANIAN\textsuperscript{b}

\textsuperscript{a}National Center for Microgravity Research, NASA Glenn Research Center, Cleveland, Ohio, USA

\textsuperscript{b}Department of Chemical Engineering, Clarkson University, Potsdam, New York, USA

\textbf{Abstract:} In this paper we analyze the effects of the following phenomena associated with the thermocapillary migration of a drop. The first is the influence of Newtonian surface rheology of the interface and the second is that of the energy changes associated with stretching and shrinkage of the interfacial area elements, when the drop is in motion. The former occurs because of dissipative processes in the interfacial region, such as when surfactant molecules are adsorbed at the interface in sufficient concentration. The interface is typically modeled in this instance by ascribing to it a surface viscosity. This is a different effect from that of interfacial tension gradients arising from surfactant concentration gradients. The stretching and shrinkage of interfacial area elements leads to changes in the internal energy of these elements that affects the transport of energy in the fluids adjoining the interface. When an element on the interface is stretched, its internal energy increases because of the increase in its area. This energy is supplied by the neighboring fluids that are cooled as a consequence. Conversely, when an element on the interface shrinks, the adjoining fluids are warmed. In the case of a moving drop, elements of interfacial area are stretched in the forward half of the drop, and are shrunken in the rear half. Consequently, the temperature variation on the surface of the drop and its migration speed are modified. The analysis of the motion of a drop including these effects was first performed by LeVan in 1981, in the limit when convective transport of momentum and energy are negligible. We extend the analysis of LeVan to include the convective transport of momentum by demonstrating that an exact solution of the momentum equation is obtained for an arbitrary value of the Reynolds number. This solution is then used to calculate the slightly deformed shape of the drop from a sphere.

\textbf{Keywords:} thermocapillary migration; Newtonian interfacial rheology; small Marangoni numbers; drop; stretching and shrinking of interface


bala@grc.nasa.gov
INTRODUCTION

The thermocapillary migration of drops was first investigated theoretically by Young, Goldstein, and Block. When a drop is present in an unbounded liquid that has a uniform temperature gradient, a thermocapillary stress is generated at the interface arising from the variation of interfacial tension with temperature. Typically, the drop is propelled toward the warm portion of the continuous phase. Young et al. obtained a result for the steady velocity of a drop, including a contribution from the effect of gravity, in the limit when the Reynolds and Marangoni numbers approach zero, so that the effect of convective transport of momentum and energy can be neglected. To verify their theory, Young et al. performed experiments on air bubbles in silicone oils, in which the effects of thermocapillarity and gravity were counterbalanced, so that the bubbles were nearly stationary.

Balasubramaniam and Chai showed that the solution obtained by Young et al. is an exact solution of the Navier–Stokes equations for any value of the Reynolds number, when the Marangoni number is equal to zero and the effect of gravity is not present. A discussion of other pertinent literature on the motion of drops due to thermocapillarity can be found in Subramanian and Balasubramaniam.

Our objective in this paper is to consider the effect of Newtonian interfacial rheology and stretching/shrinkage of interfacial area elements on the thermocapillary motion of a drop. In the limit when the Reynolds and Marangoni numbers approach zero, this problem has been addressed by LeVan and discussed by Subramanian and Balasubramaniam. The rheology of the surface needs to be considered in special situations, for example, when surfactant molecules are adsorbed on the interface in sufficient quantity so that an interfacial viscosity can be ascribed to the interface. As a consequence the drop encounters additional resistance to its motion. Scriven has provided the framework for a Newtonian model of the interface, in which the contribution to the surface stress depends linearly on the rate of deformation of area elements on the interface. A detailed discussion of surface rheology models is available in Edwards, Brenner, and Wasan.

The second effect that we consider is the consequence of the deformation of interfacial area elements on the energy balance at the interface. Even when a drop moves in a continuous phase fluid that is isothermal, temperature variations can be generated on the interface. This is because elements of surface area grow as they move from the stagnation point to the equatorial region in the forward half of the drop. The increase in area increases the interfacial internal energy, which must be provided by the fluids adjoining the interface. Thus, the neighboring fluids are cooled. In the rear portion of the drop, the situation is opposite, and the adjoining fluids are warmed. Therefore, an interfacial temperature variation can occur, and the consequent thermocapillary stress influences the motion of the drop. This effect has been considered by Harper et al., Kenning, LeVan, and Torres and Herbolzheimer.

One infers from the analysis presented by Subramanian and Balasubramaniam that the thermocapillary migration velocity of the drop is unaffected by the surface shear viscosity, but is influenced by the surface dilatational viscosity and the effect of stretching and shrinkage of interfacial area elements. The velocity of the drop is reduced as a consequence. We shall revisit the analysis performed by LeVan and Subramanian and Balasubramaniam below and show that their results are valid even
when the convective transport of momentum is included in the analysis. The effect of inertia is shown to alter the pressure field, and lead to deformation of the drop. The small change in the shape of the drop from a sphere is obtained using a technique similar to that used by Balasubramaniam and Chai.\textsuperscript{2}

**FORMULATION**

We consider the steady migration of a spherical drop of radius $R_0$ immersed in a liquid that is of infinite extent. The liquid has a density $\rho$, viscosity $\mu$, thermal conductivity $k$, and thermal diffusivity $\kappa$. The corresponding properties in the drop are denoted by $\rho_0$, $\mu_0$, $k_0$, and $\kappa_0$. The rate of change of interfacial tension between the drop and the continuous phase liquid is denoted by $\sigma_T$, and is assumed to be a negative constant. All the physical properties of the fluids, with the exception of the interfacial tension, are assumed to be constant. A temperature gradient of magnitude $G$ is imposed in the continuous phase liquid. We assume that gravitational effects are not present.

The surface shear and dilatational viscosities in the Newtonian model are denoted by $\mu_s^*$ and $\lambda_s^*$, respectively. The corresponding dimensionless surface viscosities, scaled by $\mu R_0$, are denoted by $\mu_s$ and $\lambda_s$. The quantity $e_s - \sigma^*$, where $e_s$ is the surface internal energy per unit area and $\sigma^*$ is the interfacial tension, appears in the energy balance condition at the interface to accommodate the stretching and shrinkage of interfacial area elements. We assume that $e_s - \sigma^*$ is a constant over the drop surface, and define a dimensionless parameter $E_s = (e_s - \sigma^*)/\mu k$.

A reference velocity for motion in the fluids is obtained by balancing the tangential stress at the interface in the continuous phase liquid with the thermocapillary stress. This velocity scale is

$$v_0 = \frac{(-\sigma_T)GR_0}{\mu}. \quad (1)$$

In addition to the property ratios $\alpha$, $\beta$, $\gamma$, and $\lambda$, the dimensionless interfacial viscosities $\mu_s$ and $\lambda_s$, and the interfacial internal energy parameter $E_s$, the dimensionless parameters that are important in determining the motion of the drop are the Reynolds and Marangoni numbers. These parameters are defined using the above velocity scale and the properties of the continuous phase liquid.

$$Re = \frac{v_0 R_0}{\mu} \quad (2)$$

$$Ma = \frac{v_0 R_0}{\kappa}. \quad (3)$$

The problem is analyzed in a reference frame attached to the moving drop. We use a spherical polar coordinate system, with the origin located at the center of the drop. The radial coordinate, scaled by $R_0$, is $r$. The polar coordinate measured from the direction of the temperature gradient is $\theta$. The azimuthal coordinate is $\phi$; however, the problem posed below for the velocity and temperature fields is independent of $\phi$ due to axial symmetry about the direction of the imposed temperature gradient. The scaled radial and tangential velocity fields in the continuous phase liquid are denoted by $u$ and $v$, respectively, and those in the drop by $u'$ and $v'$. The scaled
migration velocity of the drop is denoted by \( v_\infty \). These are obtained by dividing the physical velocities by \( v_0 \). The scaled pressure fields in the two fluids are \( p \) and \( p' \) and are obtained by dividing the physical pressure by \( \rho v_0^2 \). The temperature field in the continuous phase is scaled by subtracting the temperature in the undisturbed continuous phase at the location of the drop and dividing by \( GR_0 \).

\[
T = \frac{\bar{T} - GV_0 v_\infty t}{GR_0},
\]

where \( \bar{T} \) is the fluid temperature and \( t \) denotes time. The scaled temperature \( T' \) in the drop is defined similarly. The governing equations for the velocity and temperature fields in the continuous phase liquid and the drop are given below:

\[
\frac{1}{r^2} \frac{\partial}{\partial r} (r^2 u) + \frac{1}{r \sin \theta} \frac{\partial}{\partial \theta} (v \sin \theta) = 0
\]

\[
\frac{u}{\partial r} + \frac{v}{r \partial \theta} = -\frac{\partial p}{\partial r} + \frac{1}{Re} \left[ \nabla^2 u - \frac{2u}{r^2} - \frac{2v}{r^2} \cot \theta \right]
\]

\[
\frac{u}{\partial r} + \frac{v}{r \partial \theta} = -\frac{1}{Ma} \frac{\partial T}{\partial \theta} + \frac{1}{r^2} \nabla^2 T
\]

\[
\frac{1}{r^2} \frac{\partial}{\partial r} (r^2 u') + \frac{1}{r \sin \theta} \frac{\partial}{\partial \theta} (v' \sin \theta) = 0
\]

\[
\frac{u}{\partial r} + \frac{v'}{r \partial \theta} = -\frac{1}{\gamma r} \frac{\partial p'}{\partial \theta} + \frac{1}{\gamma Re} \left[ \nabla^2 u' - \frac{2u'}{r^2} - \frac{2v'}{r^2} \cot \theta \right]
\]

\[
\frac{u}{\partial r} + \frac{v'}{r \partial \theta} = -\frac{1}{\gamma r} \frac{\partial p'}{\partial \theta} + \frac{1}{\gamma Re} \left[ \nabla^2 v' + \frac{2u'}{r^2} \cot \theta + \frac{v'}{r^2} \right]
\]

The boundary conditions are as follows. Far from the drop, the velocity field is uniform and equal to the migration velocity of the drop (i.e., in the laboratory reference frame, the fluid far away is quiescent). The temperature field far away is the imposed linear field. Therefore, as \( r \to \infty \),

\[
u_\infty \cos \theta, \quad v_\infty \sin \theta
\]

\[
T \to r \cos \theta.
\]

From the analysis by LeVan,\(^4\) it can be shown that when the Reynolds and Marangoni numbers are set equal to zero, the normal stress balance is satisfied by a spherical shape of the drop. This is no longer the case when the Reynolds number is nonzero, the case considered here. Assuming the deformation from the spherical shape is small, it can be shown by using a domain perturbation scheme that it is permissible to obtain the leading order velocity and pressure fields by applying the relevant boundary conditions at the fluid–fluid interface at a spherical boundary, designated here by \( \gamma = 1 \). In the next section, we show how the small perturbation to this shape arising from inertia can be calculated using these leading order fields. At the fluid–
fluid interface, the kinematic boundary condition holds, and the velocity and temperature fields are continuous.

\[ u(1, \theta) = u'(1, \theta) = 0 \]  

(15)  

\[ v(1, \theta) = v'(1, \theta) \]  

(16)  

\[ T(1, \theta) = T'(1, \theta). \]  

(17)

The tangential stress balance and the heat flux balance at the fluid–fluid interface appear below. It is in these boundary conditions that the effects of surface viscosity and surface internal energy appear.

\[
\frac{\partial}{\partial r} \left( \frac{v}{r} \right)(1, \theta) - \alpha \frac{\partial}{\partial r} \left( \frac{v'}{r} \right)(1, \theta) \\
= \frac{\partial T}{\partial \theta}(1, \theta) - 2\mu_s v(1, \theta) - (\lambda_s + \mu_s) \frac{\partial}{\partial \theta} \left[ \frac{1}{r} \frac{\partial}{\partial \theta} (v(1, \theta) \sin \theta) \right] \\
= \frac{E_s}{\sin \theta \partial \theta} [v(1, \theta) \sin \theta]. \tag{18}
\]

(18)  

The migration velocity of the drop is determined from a force balance—at steady state the drop is not accelerating, and hence, the net force acting on it must be zero,

\[
\int_0^{\pi} \left[ \frac{\partial}{\partial r} \left( \frac{v}{r} \right)(1, \theta) \sin^2 \theta + \left( Re \rho(1, \theta) - 2\frac{\partial u}{\partial r}(1, \theta) \right) \cos \theta \sin \theta \right] \sin \theta \, d\theta = 0 \tag{19}
\]

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\]

RESULTS

Exact Solution for Small Marangoni Number

When \( Ma \ll 1 \), convective transport of energy can be neglected compared with conduction, as a first approximation. In this case, the terms on the left side of Equations (8) and (12) may be ignored. Then, it can be shown that the solution first obtained by LeVan4 and given in Subramanian and Balasubramaniam5 for the temperature and velocity fields in the case \( Re = 0 \) continues to apply for arbitrary values of the Reynolds number and other parameters in the problem. Only the pressure field is modified by the inclusion of inertia. This exact solution is given below:

\[
T = \left( r + \frac{B}{r^2} \right) \cos \theta \tag{21}
\]

\[
T' = (1 + B)r \cos \theta \tag{22}
\]

\[
u = -v_{\infty} \left( 1 - \frac{1}{r^2} \right) \cos \theta \tag{23}
\]

\[ v = v_{\infty} \left( 1 + \frac{1}{2r^2} \right) \sin \theta \tag{24}
\]

\[ u' = \frac{3}{2} v_{\infty} (1 - r^2) \cos \theta \tag{25} \]
The constant $A_0$ in the pressure field inside the drop can be determined only when the interfacial normal stress balance is considered. The velocity field outside the drop is irrotational and the pressure field outside the drop satisfies Bernoulli’s equation. The flow field within the drop is the well known Hill’s spherical vortex. The temperature and the velocity fields are coupled by the effects of surface viscosity and interfacial internal energy. The surface shear viscosity $\mu_s$ has no influence on the dynamics. The surface dilatational viscosity $\lambda_s$ and the effect of the interfacial internal energy reduce the migration velocity of the drop. When $\lambda_s$ and $E_s$ are zero, the results given above reduce to those given by Balasubramaniam and Chai.²

Small Deformation of the Drop

We now consider the normal stress balance at the fluid–fluid interface and calculate the slight deformation of the drop from a sphere. Let $R = R_0[1 + f(\theta)]$ denote the shape of the drop. We assume that $f \ll 1$. It will be seen shortly that this requires that the Weber number $We = Re Ca \ll 1$. Here $Ca$ is the Capillary number, defined by $Ca = \mu_0 v_0/\sigma_0$, where $\sigma_0$ is a reference value for the interfacial tension. For a slightly deformed drop, the normal stress balance at the interface can be written as

$$\tau_{rr} - \tau_{rr'} = \frac{2H\sigma}{Ca} + \frac{2\lambda_s}{\sin\theta}\frac{\partial}{\partial\theta} [v(1, \theta) \sin\theta],$$

where $\tau_{rr}$ denotes the dimensionless normal stress at $r = 1$ evaluated from the leading order fields given above, $H$ is the mean curvature of the interface, and $\sigma$ is the interfacial tension scaled by $\sigma_0$. Note that the surface viscosity makes a direct contribution to the normal stress difference (see Scriven⁵). When the deformation from a spherical shape is small,

$$2H \sim 2 - 2f - \frac{1}{\sin\theta}\frac{d}{d\theta}\left(\sin\theta \frac{df}{d\theta}\right).$$

The normal stress balance can then be written as follows:

$$v' = 3v_\infty \left(r^2 - \frac{1}{2}\right) \sin\theta$$  \hspace{1cm} (26)

$$p = p_\infty + \frac{v_\infty^2}{2} \left[1 - \cos^2\theta \left(1 - \frac{1}{r^3}\right) - \sin^2\theta \left(1 + \frac{1}{2r^3}\right)^2\right]$$  \hspace{1cm} (27)

$$p' = A_0 + \frac{9}{8} v_\infty^2 \left[\sin^2\theta(r^4 - r^2) + 2\cos^2\theta \left(r^2 - \frac{r^4}{2}\right)\right] - 15 \frac{v_\infty}{Re} r \cos\theta$$  \hspace{1cm} (28)

$$B = \frac{1 - \beta - \Omega}{2 + \beta + \Omega}$$  \hspace{1cm} (29)

$$\Omega = \frac{2E_s}{2 + 3\alpha + 2\lambda_s}$$  \hspace{1cm} (30)

$$v_\infty = \frac{2}{(2 + 3\alpha + 2\lambda_s)(2 + \beta + \Omega)} = \frac{\Omega}{E_s(2 + \beta + \Omega)}.$$  \hspace{1cm} (31)
From Equation (34), one can conclude that, for a fixed Reynolds number, \( f(\theta) \sim O(Ca) \) as \( Ca \to 0 \). In this asymptotic limit, the leading order differential equation for \( f(\theta) \) is a nonhomogeneous version of the differential equation for the Legendre Polynomial \( P_1(\cos \theta) \). Note the absence of any terms proportional to \( \cos \theta \) in the inhomogeneity. If such a term is present, it can be shown that it will lead to unboundedness of the solution for \( f \) along \( \theta = 0 \) and \( \pi \). In fact, if a force balance on the drop had not been used to determine the speed of the drop, the speed can be obtained by requiring the term proportional to \( \cos \theta \) in the inhomogeneity to be zero to eliminate such singular behavior. We impose the constraints that the volume of the drop is a constant (equal to \( 4\pi R_0^3 \)) and that the origin of the coordinates is the center of mass of the drop. Linearization of these constraints for small \( f \) leads to the following equations:

\[
\int_0^\pi f(\theta) \sin \theta d\theta = 0 \tag{35}
\]
\[
\int_0^\pi f(\theta) \cos \theta \sin \theta d\theta = 0. \tag{36}
\]

The solution can be obtained in a straightforward manner, as shown by Brignell.\(^{10}\)

The unknown constant \( A_0 \) is determined as part of the solution,

\[
A_0 = \frac{2}{Re Ca} + \frac{v_\infty^2}{4} \left( 1 + \frac{3}{2} \gamma \right) \tag{37}
\]

\[
f(\theta) = \frac{3}{32} Re Ca \frac{v_\infty^2}{\gamma} (\gamma - 1)(3 \cos^2 \theta - 1). \tag{38}
\]

The dimensionless surface dilatational viscosity \( \lambda_s \) and the surface internal energy parameter \( E_s \) affect the deformation of the drop via their influence on \( v_\infty \) (given in (31)). Because we have restricted the deformation of the drop to be small, we see that \( We = Re Ca \) must be a small parameter. Drops less dense than the continuous phase liquid are predicted to contract in the direction of the temperature gradient; that is, they attain an oblate shape. Drops more dense than the continuous phase attain a prolate shape.

**CONCLUDING REMARKS**

We have shown that a solution in the limit of zero Reynolds and Marangoni numbers, for the velocity and temperature fields, obtained elsewhere for the thermocapillary migration of a drop accommodating Newtonian surface rheology and the influence of internal energy changes associated with the stretching and shrinkage of surface elements, holds for arbitrary values of the Reynolds number so long as the Marangoni number is set equal to zero. The pressure fields are modified by inertia. The leading order result for the migration speed of the drop is the same as that obtained when the Reynolds number is zero. Furthermore, whereas the drop will be
spherical when the Reynolds number is zero, this is no longer the case for non-zero values of the Reynolds number. A perturbation result for the small deformation of the drop from a spherical shape is given in the case where the Capillary number is asymptotically small, for fixed Reynolds number.

REFERENCES