THERMOCAPILLARY MIGRATION OF A FLUID DROPLET INSIDE A DROP IN A SPACE LABORATORY

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Abstract--The thermocapillary migration of a fluid droplet located inside a liquid drop in a space laboratory is analyzed. The quasi-static momentum and energy equations are solved at the instant when the droplet passes the center of the drop. Results are presented for prescribed axisymmetric distributions of temperature on the drop surface.

INTRODUCTION

The free fall environment aboard orbiting spacecraft offers a unique opportunity for scientific experimentation as well as the production of new materials. In free fall, materials may be processed without the need for a container. Such containerless processing avoids heterogeneous nucleation and contamination by the container wall and offers the opportunity for making new and useful materials such as optical and laser glasses (Nielson & Weinberg 1977, Weinberg 1978).

The significant reduction of buoyancy in orbit has interesting consequences in multiphase systems such as liquid drops containing a dispersion of bubbles or droplets of a second phase. The migration of the droplets due to the reduced buoyancy is likely to be overshadowed by movement due to interfacial tension gradients, and migration induced by electrical and/or magnetic fields. Since it is desirable in some instances, such as in glass processing in space, to induce the migration of gas bubbles for eliminating them from the melt, we have begun a theoretical study of this phenomenon. We also plan to conduct experiments aboard the Space Shuttle on this subject, at an appropriate future date (Subramanian & Cole 1979; also see Naumann 1978). In the present article, our objective is to construct an approximate theoretical description of the slow migration of a single fluid droplet/bubbler located inside a liquid dropt in free fall under the action of a specified axisymmetric temperature field on the drop surface at the instant when the fluid droplet/bubble is concentric with the drop. This problem is not necessarily limited in scope. First, it will yield useful first order information on the effect of a free outer interface on the thermal migration process of the droplet. In addition, when the center of the droplet is close to the center of the drop, the present analysis will provide a reasonable approximation to the migration velocity of the droplet. This latter case would normally be handled by employing a system of bispherical coordinates. However, this coordinate system has a singularity when the drop and droplet are concentric. This necessitates the present analysis using spherical polar coordinates when the droplet is at the center of the drop. We have, in fact, addressed the more general problem of an eccentrically located bubble using bispherical coordinates. The results will be reported in a subsequent article.

When a fluid droplet (gas or liquid) is located in a liquid medium, the consequence of a temperature gradient at the fluid--liquid interface is to cause a surface tension gradient which results in a tangential stress on the interface causing interfacial movement. By viscous traction, the surrounding fluid is set into motion, and as a result the droplet will move in a liquid medium in the presence of a temperature gradient. The direction of the movement is toward the pole with lower surface tension which is usually the warmer pole. This phenomenon is well

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tThroughout this work, the terms droplet and drop will be used with the connotations designated in this sentence.

illustrated in a film by Trefethan. (In the present case, in addition to this thermocapillary drift, the droplet would also be dragged by the bulk liquid motion induced by the presence of temperature gradients on the drop surface.) Since the appearance of the work of Young *et al.* (1959) wherein both experimental results and a first order theory (valid in the limit of zero Marangoni number) were reported, several articles have appeared on this subject. The literature has been discussed in detail by Subramanian (1981) who has extended the solution of Young *et al.* for small values of the Marangoni number using the method of matched asymptotic expansions.

The problem of steady thermocapillary flow in a drop in "Zero Gravity" was considered by Dragoo (1974), who analyzed it in the creeping flow limit using the method of normal modes.

Happel & Brenner (1965) indicate that many authors have treated the creeping flow problem for the motion of a sphere at the instant it is concentric with an outer spherical container. Calculations for the drag with a solid outer shell have been reported by Cunningham (1910) and Lee (1947) for a solid inner sphere, and by Haberman & Sayre (1958) for a fluid inner sphere. Happel (1958) has treated the case of a solid sphere concentric with a spherical fluid outer boundary.

In the present problem, it is convenient to solve the theoretical model equations for the general case of a fluid droplet of arbitrary viscosity and thermal conductivity located concentrically inside a liquid drop. Thus, at first, the general solutions will be developed. The limiting case of a *gas bubble* present inside a liquid drop is of some interest in applications to glass processing in space. The results therefore will be specialized for this case in the section on discussion.

ANALYSIS

We shall consider the migration of a fluid droplet at the instant it passes the center of a liquid drop with a specified steady axisymmetric temperature distribution on the drop surface. The drop is held fixed in a laboratory in free fall. We shall assume both the fluid droplet and the liquid drop to remain spherical and assume constant physical properties with the exception of the interracial tensions on the two fluid-fluid interfaces which will be assumed to vary linearly with temperature. The flow is assumed to be incompressible and Newtonian. Our objective is to determine the migration velocity of the fluid droplet induced by thermocapillarity at the instant it passes the drop center.

A gas bubble may be modelled by using the appropriate limiting values for the physical properties of the droplet phase. Properties of the droplet phase are denoted by a carat. A sketch of the coordinate system is provided in figure 1. The appropriate scaled equations of motion and

Figure 1. Schematic of the liquid drop-fluid droplet system.

energy may be written as follows. For the liquid in the drop,

$$
N_{\rm Re} \left[\frac{\partial v}{\partial t} + v \cdot \nabla v \right] = - \nabla P + \nabla^2 v \tag{1}
$$

$$
N_{\rm Ma} \left[\frac{\partial T}{\partial t} + v \cdot \nabla T \right] = \nabla^2 T.
$$
 [2]

For the droplet phase,

$$
N_{\rm Re} \left[\frac{\partial \hat{v}}{\partial t} + \hat{v} \cdot \nabla \hat{v} \right] = -\frac{\rho}{\hat{\rho}} \nabla \hat{P} + \frac{\hat{\nu}}{\nu} \nabla^2 \hat{v}
$$
 [3]

$$
N_{\text{Ma}} \left[\frac{\partial \hat{T}}{\partial t} + \hat{v} \cdot \nabla \hat{T} \right] = \frac{\hat{\alpha}}{\alpha} \nabla^2 \hat{T} . \tag{4}
$$

In the above equations, v , T and P are the scaled velocity, temperature, and pressure fields respectively in the liquid drop. The same fields inside the droplet are designated by a carat. ρ , ν and α represent the density, kinematic viscosity, and the thermal diffusivity respectively of the liquid in the drop with similar symbols with a carat designating the droplet phase. The length scale is the drop radius a. A velocity scale obtained from the tangential stress balance at the drop surface which induces the liquid motion, is given by

$$
v_0 = \frac{\left| \frac{d\sigma_d}{dT} \right| \Delta T_{\text{ref}}}{\mu} \,. \tag{5}
$$

The pressure scale P_0 and time scale t_0 are given by

$$
P_0 = \frac{\left| \frac{d\sigma_d}{dT} \right| \Delta T_{\text{ref}}}{a};
$$

$$
t_0 = \frac{a}{v_0} \, . \tag{7}
$$

Here, $d\sigma_d/dT$ is the rate of change of interfacial tension with temperature on the outer drop surface and is assumed to be a constant negative quantity over the temperature range of interest. μ is the dynamic viscosity and ΔT_{ref} is a characteristic temperature difference used in scaling the temperature field. Its definition is deferred till later. The temperature is nondimensionalized by subtracting a reference value T_0 , and dividing by ΔT_{ref} . The Reynolds and Marangoni numbers are defined as follows:

$$
N_{\text{Re}} = \frac{dv_0}{\nu};\tag{8}
$$

$$
N_{\text{Ma}} = \frac{a v_0}{\alpha} = \left| \frac{d \sigma_d}{d T} \right| \frac{\Delta T_{\text{ref}} a}{\mu \alpha} . \tag{9}
$$

It can be seen from [9] that the Marangoni number plays the role of a Peclet number representing the ratio of convective energy transport rate to conduction.

In spite of the conditions initially imposed, the problem still remains fairly complex and is nonlinear. However, a considerable simplification can be effected for molten glasses and other highly viscous fluids because the Reynolds number is expected to be typically several orders of magnitude smaller than unity. Thus, even though the Prandtl number (ν/α) can be on the order of $10³$ or larger, the Marangoni number, being the product of the Reynolds and Prandtl numbers, also will be considerably smaller than unity. Thus, we shall make the quasi-static approximation (Happel & Brenner 1965), and ignore the convective transport of momentum and energy as well as the unsteady accumulation terms, making the transport equations linear.

In view of the axial symmetry of the problem, a stream function ψ may be defined by

$$
v_r = -\frac{1}{r^2 \sin \theta} \frac{\partial \psi}{\partial \theta} \tag{10a}
$$

$$
v_{\theta} = \frac{1}{r \sin \theta} \frac{\partial \psi}{\partial r}.
$$
 [10b]

Similar definitions apply to the droplet phase when symbols with a carat are used. The quasi-static momentum and energy equations for the liquid phase may be written as follows:

$$
E^4\psi = 0 \tag{11}
$$

$$
\nabla^2 T = 0 \tag{12}
$$

Similar equations may be written for the droplet phase. The operator E^2 defined in Happel & Brenner (1965) is given by

$$
E^2 = \frac{\partial^2}{\partial r^2} + \frac{1}{r^2} (1 - s^2) \frac{\partial^2}{\partial s^2}
$$
 [13]

where

$$
s = \cos \theta. \tag{14}
$$

In view of the quasi-static approximations made here, the results will be valid only in the limit $N_{\text{Re}} \rightarrow 0$. However, they may be regarded as useful first approximations for small values of the Reynolds number.

The boundary conditions on the temperature and velocity fields are given below.

The temperature is prescribed on the outer drop surface.

$$
T(1, s) = f(s).
$$
 [15]

Here $f(s)$ is an arbitrary function of s. It may be noted that any arbitrary axisymmetric distribution *f(s)* may be expanded in a series of spherical harmonics. Due to the linearity of the problem, the effect of each of these pure modes of the surface temperature field on the flow inside the drop may be considered independently.

The velocity, temperature, and the heat flux are continuous across the interface between the liquid drop and the fluid droplet.

$$
\mathbf{v}(\kappa, s) = \hat{\mathbf{v}}(\kappa, s) \tag{16}
$$

$$
T(\kappa, s) = \hat{T}(\kappa, s) \tag{17}
$$

$$
k \frac{\partial T}{\partial r}(\kappa, s) = \hat{k} \frac{\partial T}{\partial r}(\kappa, s).
$$
 [18]

Here κ is the scaled radius of the droplet and has the range 0 to 1. k is the thermal conductivity of the liquid in the drop. The normal component of the velocity must vanish on the outer surface of the drop.

$$
v_r(1,s)=0 \qquad \qquad [19]
$$

The droplet is assumed to have a scaled velocity U in the positive z-direction (due to the axial symmetry of the problem). Thus on the fluid droplet-liquid drop interface

$$
\mathbf{v}(\kappa, s) \cdot \mathbf{i}_z = \hat{\mathbf{v}}(\kappa, s) \cdot \mathbf{i}_z = U \tag{20}
$$

The tangential stress balances at the two interfaces may be written as follows:

$$
\tau_{r\theta}(\kappa, s) - \hat{\tau}_{r\theta}(\kappa, s) = -\frac{1}{\kappa} \beta \frac{\partial T}{\partial \theta}\Big|_{r=\kappa};
$$
 [21]

$$
\tau_{r\theta}(1,s) = \frac{\partial T}{\partial \theta}\bigg|_{r=1}.
$$

Here,

$$
\beta = \frac{\left| \frac{d\sigma_f}{dT} \right|}{\left| \frac{d\sigma_d}{dT} \right|},
$$
\n(23)

and $d\sigma_f$ dT, assumed negative, is the rate of change of the interfacial tension at the liquid drop-fluid droplet interface with temperature. In writing [22] it has been assumed that the viscosity of the gas phase surrounding the liquid drop is negligible compared to that of the liquid. With suitable boundedness conditions on the temperature and velocity fields at $r = 0$ and $\theta = 0$, π , the problem statement is complete.

Solution

The general solution of [11] has been summarized by Happel & Brenner (1965). The solution of the conduction equation $[12]$ is well-known and is reported for instance by Carslaw & Jaegar (1959). For the problem under consideration,

$$
\psi(r,s) = \sum_{n=2}^{\infty} \left[A_n r^n + B_n r^{-n+1} + C_n r^{n+2} + D_n r^{-n+3} \right] C_n^{-1/2}(s) ; \qquad [24]
$$

$$
T(r,s) = \sum_{n=0}^{\infty} \left[E_n r^n + F_n r^{-(n+1)} \right] P_n(s) \,. \tag{25}
$$

Similar solutions with carats may be written for the droplet phase. Here, $P_n(s)$ is the Legendre polynomial of order n, and $C_n^{-1/2}$ is the Gegenbauer polynomial of order n and degree $-1/2$. The latter is related to the Legendre polynomial via

$$
C_n^{-1/2}(s) = \frac{P_{n-2}(s) - P_n(s)}{2n-1}.
$$
 [26]

Upon use of the boundary conditions in a tedious but straightforward manner, the twelve constants appearing in the solutions for the stream function and temperature fields for the liquid drop and the fluid droplet phases may be obtained. The details are reported in the appendix.

In the quasi-static limit considered here, inertial terms are assumed to be negligible. Thus, the quasi-static velocity of the fluid droplet may be obtained by setting the net force on the droplet to zero. An expression for this force (in terms of the coefficient D_2) is given by Happel and Brenner for problems with the stream function given by [24] as

$$
F_z = -4\pi D_2. \tag{27}
$$

Thus, setting the force equal to zero (or equivalently, $D_2 = 0$) leads to the following final result for the quasi-static migration velocity of the droplet.

$$
U = G_1 \left[\frac{1}{3} + \frac{2}{\{2(1-\kappa^5) + \eta(3+2\kappa^5)\}} \left(\frac{\beta \kappa (1-\kappa^5)}{\{(2+\kappa^3) + \xi(1-\kappa^3)\}} - \frac{5}{6} \eta \kappa^2 \right) \right].
$$
 [28]

It is interesting to note that only the constant $G₁$ in the expansion of the arbitrary surface temperature field in Legendre polynomials, see [A2], enters the final expression for the net force on the droplet and hence its migration velocity. In other words, only the P_1 -component of the surface temperature field is responsible for droplet migration at the instant the droplet is located concentrically within the drop. Due to their symmetry, the higher modes in the surface temperature field, while contributing to the flow field, do not influence the migration process in this case. However, if the droplet is slightly displaced from the center of the drop along the symmetry axis, it will migrate away from the center if the surface temperature field is a pure $P₂$ -mode. This may be inferred from the results for the flow field illustrated later in this work. The behavior for higher modes is more complex, and can be determined conveniently from an analysis of the eccentric problem.

Limiting case of a gas bubble

In the case of a *gas bubble* located inside a liquid drop, the thermal conductivity and the viscosity of the gas may be considered negligible compared to the same properties of the surrounding liquid. In addition, the gradients of interfacial tension with temperature on the bubble-drop interface and on the outer drop surface (which is assumed to be surrounded by the same gas) are identical. In these limits (ξ , $\eta \rightarrow 0$ and $\beta \rightarrow 1$), [28], for the migration velocity reduces to

$$
U = G_1 \left[\frac{1}{3} + \frac{\kappa}{2 + \kappa^3} \right].
$$
 [29]

DISCUSSION OF RESULTS

Even though general results were derived in the previous section for fluid droplets inside liquid drops, we shall focus our attention here on gas bubbles located inside liquid drops. These systems are of considerable importance in space processing, and furthermore, not much information is available on interfacial tension gradients in liquid-liquid systems of potential importance in space applications.

It was noted earlier that only the P_1 -component of the surface temperature field contributes to the bubble migration process when the bubble is concentric with the drop. Due to the linearity of the problem, it is convenient to illustrate the influence of pure modes of the surface temperature field on the flow inside the drop independently so that the symmetry due to the P_2 and higher modes may be observed. For this, one may let

$$
G_j = \delta_{nj} \tag{30}
$$

where the *n*th mode of the surface temperature field $P_n(s)$ is excited. It should be noted that [30] implies the choice of ΔT_{ref} . For instance, for $n = 1$, ΔT_{ref} is simply the temperature difference between one of the poles and the equator of the drop. Similar interpretations may be made for the higher modes.

Figures 2-7 illustrate isotherms and streamlines for the first three Legendre modes of the temperature field on the drop surface. In all cases, a representative value of the scaled bubble radius $\kappa = 0.5$ has been used. Due to the axial symmetry of the system, sections through meridian planes are shown. It may be observed that in all cases, the isotherms curve to meet the

Figure 2. Isotherms for the drop-bubble system for a P_1 -mode temperature field on the drop surface; scaled bubble radius $\kappa = 0.5$.

STREAMLINES, P₁-MODE $K=0.5$, $\Delta\psi = 0.01$

Figure 3. Streamlines for the drop-bubble system for a P_1 -mode temperature field on the drop surface; scaled bubble radius $\kappa = 0.5$.

Figure 4. Isotherms for the drop-bubble system for a P₂-mode temperature field on the drop surface; scaled bubble radius $\kappa = 0.5$.

Figure 5. Streamlines for the drop-bubble system for a P_2 -mode temperature field on the drop surface; scaled bubble radius $\kappa = 0.5$.

Figure 6. Isotherms for the drop-bubble system for a P_3 -mode temperature field on the drop surface; scaled bubble radius $\kappa = 0.5$.

 $K=0.5, \Delta\psi$ =0.003

Figure 7. Streamlines for the drop-bubble system for a P_3 -mode temperature field on the drop surface; scaled bubble radius $\kappa = 0.5$.

inner (bubble) surface normally to satisfy the requirement of negligible heat flux at the bubble surface. The straight lines in the streamline plots represent the boundaries of the convection cells, and on these lines $\psi = 0$. The stagnation rings lie in planes normal to the axis. The symmetry of the flows generated by the P_2 and P_3 modes of the surface temperature fields is evident from figures 5 and 7. In contrast, from figure 3 it may be seen why the P_1 -mode results in net bubble movement.

For an air bubble in a typical glassmelt, the ratios of viscosities ($\eta = \mu_{air}/\mu_{glass}$) and thermal conductivities ($\xi = k_{air}/k_{glass}$) are typically on the order of 10⁻⁶ and 10⁻¹ respectively. Thus, it is most reasonable to assume $\eta = 0$ while the assumption of $\xi = 0$ in writing [29] may be more suspect. We found that for a choice of $\xi = 0.2$ and $\kappa = 0.5$, the approximate result for a gas bubble given in [29] deviates from the exact result in [28] by less than 4 per cent.

In the general case of a fluid droplet inside a liquid drop, when the surface temperature is specified as being a pure P₁-distribution, that is when $G_i = \delta_{ij}$, an interesting asymptote may be recovered for small values of κ from [28] for the scaled droplet migration velocity. For small values of the scaled droplet radius κ ($\kappa \ll 1$), and $G_1 = 1$, [28] reduces to

$$
U \simeq \frac{1}{3} + \frac{2\beta\kappa}{(2+3\eta)(2+\xi)}.
$$
 [31]

If one were to calculate the streaming velocity at the center of a drop due to the above surface temperature distribution from Dragoo (1974), one obtains

$$
U_1 = \frac{1}{3} \,. \tag{32a}
$$

The quasi-static velocity of a fluid droplet migrating in an infinite body of liquid with a linear temperature field (which corresponds exactly to the P_1 -distribution on the drop surface here) may be obtained from a result given by Young *et al.* (1959) after correcting minor typographical errors appearing in it. In the present notation, this result is

$$
U_2 = \frac{2\beta\kappa}{(2+3\eta)(2+\xi)}.
$$
 [32b]

Thus, in the limit when the size of the droplet is small compared to that of the drop, we recover an asymptote wherein the velocity of the droplet at the center of a liquid drop is simply the sum of its velocity in an equivalent thermal environment in a quiescent fluid and the streaming velocity at the center of an identical drop containing no droplet. It may be observed from [31] that in the limit of small κ , the streaming velocity makes the major contribution to the droplet migration velocity. As the relative size of the fluid droplet is increased, [31] is no longer applicable. It may be expected that the consequence of the tangential stress on the droplet-drop interface due to the interfacial tension gradient will be to oppose the streaming induced by the surface tension gradient on the drop surface. Thus, in general, the droplet migration velocity will be less than the sum of the contributions in [32a] and [32b]. It may be noted that for a gas bubble, even for κ as large as $(0.6, 131)$ predicts a result within 5 per cent of the exact result given in [28].

For simplicity in this treatment, we assumed the interfaces to be spherical in shape, and ignored the balance of normal stresses which must be used to determine the interface shape. When there is no variation of interfacial tension around the surface of a droplet migrating in a large liquid body, Taylor & Acrivos (1964) showed that the droplet remains spherical when inertial effects are negligible. In the present case of a droplet migrating inside a drop due to interfacial tension gradients, it may be verified that only for a drop surface temperature

distribution given by the P_1 -mode, the resulting solutions satisfy the normal stress balances and hence are exact. Similar observations were made by Schechter & Farley (1963) in a different context. These workers analyzed the influence of surfactants on the rate of settling of a fluid droplet in a large liquid body and showed that when there is an interfacial tension gradient, a spherical shape is compatible with the normal stress balance only when that surface tension distribution on the drop surface is assumed to be a pure P_1 -mode. In the presence of a P_2 or higher mode of the surface temperature field, there will be deformation of both the drop and the droplet from a spherical shape in order to satisfy the balance of normal stresses. The principal contributor to this deformation will be the actual variation of surface tension along the interface. The gradient of surface tension with temperature for most common liquids is relatively insensitive to the actual magnitude of the surface tension. A typical value might be $\left|\frac{d\sigma}{dT}\right|\approx 60 \mu\text{N/m} \cdot \text{K}$. Thus, a variation of 50°C on a drop surface will induce a change of approximately $3mN/m$ in surface tension. For molten glasses which possess surface tension values on the order of *300mN[m,* this is a small variation, and the resulting deformation will be small. In the case of fluids of extremely low surface tension, the deformation can be large and can have a strong influence on the results.

Finally, it is useful to present an illustrative set of values to justify the quasi-static assumptions made in this work. If the quasi-static velocity from [29] (which is valid only when a bubble is concentric with the drop) is used as an order of magnitude estimate of the bubble velocity, an air bubble 2 mm diameter, present in a molten glass drop of diameter 1 cm with a viscosity 10 Pa. s will take on the order of 60 s to traverse its own diameter. Using properties characteristic of molten glasses (Morey 1954, Stanek 1981) the Reynolds and Marangoni numbers may be calculated to be 3.3×10^{-5} and 1.75×10^{-2} respectively. The time scale for the velocity field to relax is approx. 0.02 s and the time scale for the relaxation of the temperature distribution is on the order of 11 s. Thus, the quasistatic assumptions may be considered reasonable in this situation.

CONCLUSIONS

The present quasi-static analysis reveals that only the P_1 -component of a general axisymmetric temperature distribution on a drop surface contributes to the thermocapillary motion of a fluid droplet placed concentrically inside the drop. Higher modes induce closed convection cells but exert no net influence on the droplet. It is shown that in the limit when the size of the droplet becomes small compared to that of the drop, the migration velocity of the droplet is given by the sum of the thermocapillary streaming velocity at the drop center and the quasi-static thermocapillary migration velocity of the droplet in a quiescent fluid with the same thermal gradient.

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NOMENCLATURE

- a liquid drop radius
- b fluid droplet radius
- $C_n^{-1/2}(s)$ Gegenbauer polynomial of order *n*, degree $-1/2$, and argument *s*
	- E^2 operator defined in [13]
		- *f(s)* prescribed temperature on the outer drop surface
		- *F.* net force on the fluid droplet
		- G_n expansion coefficient as defined in [A3]
		- *iz* unit vector in the z direction
		- *k* thermal conductivity
- N_{Ma} Marangoni number defined in [9]
- N_{Re} Reynolds number defined in [8]
- P_0 reference pressure defined in [6]
- P scaled pressure field
- *P.(s)* Legendre polynomial of order n and argument s
	- *t* radial coordinate scaled by the drop radius
	- *s* $\cos \theta$
	- *to* reference time defined by [7]
	- *t* modified dimensionless time
	- *T* temperature field scaled by ΔT_{ref} after subtracting a reference temperature T_0
	- *U* fluid droplet/bubble migration velocity scaled by the reference velocity v_0
	- U_1 scaled quasi-static liquid velocity at the drop center for a P_1 surface temperature distribution as obtained by Dragoo (1974)
	- U_2 scaled quasi-static velocity of a fluid droplet migrating in an infinite body of liquid with linear temperature field, as obtained by Young *et al.* (1959)
	- **v** velocity (vector) field scaled by the reference velocity v_0
- v_0 reference velocity defined in [5]
- v_r , v_θ components of the dimensionless velocity in the r and θ directions

Greek symbols

- α thermal diffusivity
- β as defined in [23]
- δ_{ii} Kronecker delta
- η ratio of the dynamic viscosity of the droplet phase to that of the drop phase
- θ polar angle measured from the forward stagnation point
- κ ratio of the droplet to drop radius
- μ dynamic viscosity
- ν kinematic viscosity
- ξ ratio of the thermal conductivity of the droplet phase to that of the drop phase
- ρ density
- σ_d interfacial tension on the outer liquid drop surface
- σ_f interfacial tension at the liquid drop-fluid droplet interface
- $\tau_{r\theta}$ tangential component of the stress scaled by $\mu v_0/a$
- ψ stream function defined in [10a] and [10b]

Special symbols

- ΔT_{ref} reference temperature
	- ∇^2 Laplacian operator
		- designates the droplet phase

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APPENDIX

The temperature field

The boundary conditions on the temperature field may be applied to the general solution, [25], to obtain the four sets of constants, E_n , F_n , \hat{E}_n and \hat{F}_n , as follows.

Boundedness of the temperature field at the origin gives

$$
\hat{F}_n = 0 \tag{A1}
$$

The prescribed temperature field on the drop surface may be expanded in a series of spherical harmonics. Equation [15a] then may be written as

$$
T(1, s) = f(s) = \sum_{n=0}^{\infty} G_n P_n(s).
$$
 [A2]

By using the standard orthogonal relationship for Legendre polynomials, the coefficient G_n may be obtained as

$$
G_n = \frac{2n+1}{2} \int_{-1}^{+1} f(s) P_n(s) \, \mathrm{d} s \,. \tag{A3}
$$

Applying [A2] to [25] we obtain,

$$
E_n + F_n = G_n. \tag{A4}
$$

Continuity of temperature across the droplet-drop interface gives the relation

$$
E_n \kappa^n + F_n \kappa^{-n-1} = \hat{E}_n \kappa^n. \tag{A5}
$$

Continuity of heat flux across the droplet-drop interface results in:

$$
nE_n\kappa^{n-1} - (n+1)F_n\kappa^{-n-2} = \xi n\hat{E}_n\kappa^{n-1} \,. \tag{A6}
$$

Here, ξ is the ratio of the thermal conductivity of the droplet phase to the drop phase. Solving [A4], [A5] and [A6], we obtain the set of constants which determine the temperature field completely,

$$
E_n = \frac{G_n(1 + n(1 + \xi))}{[n(1 + \xi) + 1 + (1 - \xi)n\kappa^{2n+1}]}
$$
 [A7]

$$
F_n = \frac{nG_n\kappa^{2n+1}(1-\xi)}{[n(1+\xi)+1+(1-\xi)n\kappa^{2n+1}]} \tag{A8}
$$

$$
\hat{E}_n = \frac{(2n+1)G_n}{[n(1+\xi)+1+(1-\xi)n\kappa^{2n+1}]}\,. \tag{A9}
$$

The velocity field

The boundary conditions on the velocity field are rewritten in terms of the stream function and then applied to the general solution, [24].

Boundedness of the velocity field at the origin gives

$$
\hat{\mathbf{B}}_n = \hat{\mathbf{D}}_n = 0 \,. \tag{A10}
$$

Continuity of the velocity field across the fluid droplet-liquid drop interface gives the relation,

$$
nA_n\kappa^{n-1} + (1-n)B_n\kappa^{-n} + (n+2)C_n\kappa^{n+1} + (3-n)D_n\kappa^{2-n}
$$

= $n\hat{A}_n\kappa^{n-1} + \hat{C}_n(n+2)\kappa^{n+1}, \quad n \ge 2.$ [A11]

Vanishing of the normal velocity on the drop surface gives,

$$
A_n + B_n + C_n + D_n = 0, \quad n \ge 2.
$$
 [A12]

Using [20], which specifies the velocity of the droplet, we obtain

$$
A_n\kappa^{n-2} + B_n\kappa^{-1-n} + C_n\kappa^n + D_n\kappa^{1-n} = -U\delta_{n2}\bigg\}_{n \geq 2} \qquad [A13]
$$

$$
\hat{A}_n \kappa^{n-2} + \hat{C}_n \kappa^n = -U \delta_{n2} \qquad \Big| \qquad [A14]
$$

Here, δ_{n2} is the Kronecker delta function.

$$
\delta_{ij} = \begin{cases} 1, & i = j \\ 0, & i \neq j \end{cases} .
$$
 [A15]

The tangential stress in the liquid drop written in terms of the stream function is given by

$$
\tau_{r\theta} = \frac{1}{r(1-s^2)^{1/2}} \Biggl\{ \sum_{n=2}^{\infty} \left[n(3-n) A_n r^{n-2} + (1-n)(2+n) B_n r^{-n-1} \right. \\ \left. + (2+n)(1-n) C_n r^n + n(3-n) D_n r^{1-n} \right] C_n^{-1/2}(s) \\ \left. - (1-s^2) \left[\sum_{n=2}^{\infty} A_n r^{n-2} + B_n r^{-n-1} + C_n r^n + D_n r^{1-n} \right] P'_{n-1}(s) \Biggr\} . \tag{A16}
$$

For the droplet phase,

$$
\hat{\tau}_{r\theta} = \frac{\eta}{r(1-s^2)^{1/2}} \left\{ \sum_{n=2}^{\infty} \left[n(3-n) \hat{A}_n r^{n-2} + (n+2)(1-n) \hat{C}_n r^n \right] C_n^{-1/2}(s) - (1-s^2) \left[\sum_{n=2}^{\infty} \hat{A}_n r^{n-2} + \hat{C}_n r^n \right] P'_{n-1}(s) \right\}.
$$
\n(A17)

Here, η is the ratio of the viscosity of the droplet to that of the drop phase. The balance of tangential stress on the drop surface gives

$$
n(3-n)A_n+(1-n)(2+n)B_n+(2+n)(1-n)C_n+n(3-n)D_n=n(1-n)G_{n-1}, n\geq 2.
$$
 [A18]

Similarly, the balance of tangential stress on the drop-droplet interface gives

$$
n(2-n)A_n\kappa^{n-2} + (1-n)(1+n)B_n\kappa^{-n-1} + (1-n)(1+n)C_n\kappa^n + n(2-n)D_n\kappa^{1-n}
$$

= $\eta[n(2-n)\hat{A}_n\kappa^{n-2} + (1-n)(1+n)\hat{C}_n\kappa^n] + \frac{\beta}{2}n(n-1)[E_{n-1}\kappa^{n-1} + F_{n-1}\kappa^{-n}], \quad n \ge 2.$ [A19]

Equations [A11]-[A14], [A18] and [A19] may be solved simultaneously to obtain the six sets of constants, A_n , B_n , C_n , D_n , \hat{A}_n and \hat{C}_n . For $n \ge 2$,

$$
B_{n} = \frac{1}{(2n-1)X_{n}} \left\{ \frac{n(n-1)}{2} \left[\kappa^{2n-2}(\eta - 1) + \kappa \left(1 - \frac{2n-1}{2} \eta \right) \right. \right.\left. + \frac{\eta}{2\kappa} (2n-3) \right\} G_{n-1} - U \delta_{n2} \eta \kappa^{1-n} \left[\frac{2n^{2} - 4n + 3}{2} \right]- \beta (\kappa^{n-2} - \kappa^{1-n}) \frac{n(n-1)}{2} \left[E_{n-1} \kappa^{n-1} + F_{n-1} \kappa^{-n} \right] \right\}. \tag{A20}
$$

Here.

$$
X_n = \kappa^{2n-2}(\eta - 1) + \kappa \left(1 - \frac{2n-1}{2} \eta \right) - \kappa^{-2n} (1 + \eta) + \kappa^{-3} \left(1 + \frac{2n-1}{2} \eta \right)
$$
 [A21]

$$
C_{n} = \frac{1}{(2n-1)X_{n}} \left\{ \frac{-n(n-1)}{2} \left[\frac{1+\eta}{\kappa^{2n}} + \frac{\eta}{2\kappa} (2n-3) - \frac{1}{\kappa^{3}} \left(1 + \frac{2n-1}{2} \eta \right) \right] G_{n-1} + \frac{\beta}{2} n(n-1) [\kappa^{n-2} - \kappa^{1-n}] [E_{n-1} \kappa^{n-1} + F_{n-1} \kappa^{-n}] + \eta \frac{U}{2} \delta_{n2} \kappa^{1-n} (2n^{2} - 4n + 3) \right\}
$$
 [A22]

$$
D_n = \frac{1}{\kappa^{n-2} - \kappa^{1-n}} \left\{ U \delta_{n2} - \frac{\kappa^{n-2} n(n-1)}{2(2n-1)} G_{n-1} + B_n \kappa^{-1-n} + C_n \kappa^n \right\}
$$
 [A23]

$$
A_n = -[B_n + C_n + D_n]
$$
 [A24]

$$
\hat{A}_n = -\frac{1}{2\kappa^n} \left[n A_n \kappa^n + (1-n) B_n \kappa^{1-n} + (n+2) C_n \kappa^{n+2} + (3-n) D_n \kappa^{3-n} + (2+n) U \delta_{n2} \kappa^2 \right] \tag{A25}
$$

$$
\hat{C}_n = \frac{1}{2\kappa^{n+2}} [nA_n\kappa^n + (1-n)B_n\kappa^{1-n} + (n+2)C_n\kappa^{n+2} + (3-n)D_n\kappa^{3-n} + nU\delta_{n2}\kappa^2].
$$
\n[A26]

These constants define the stream function for the liquid drop and the fluid droplet phases. However, they contain the undetermined velocity of the droplet, which may be obtained by setting the net force on the droplet to zero. This is done in [27] and [28] of the text.