

GASES
AND LIQUIDS

On the Thermophoretic Motion of a Heated Spherical Drop in a Viscous Liquid

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Abstract—Expressions for the force and velocity of the thermophoretic motion of a spherical drop in a viscous liquid are derived for arbitrary temperature differences between the surface of the drop and regions away from it. The temperature dependence of the viscosity is taken into account in the form of an exponential–power series. © 2002 MAIK “Nauka/Interperiodica”.

STATEMENT OF THE PROBLEM

The thermophoretic motion of a drop arises when an immobile liquid is subjected to an external temperature gradient. Under the action of the thermocapillary and viscous forces, the drop acquires a constant velocity, the so-called rate of thermophoresis. In general, this motion is related to shear forces appearing on the surface of the drop because of the temperature variation of the surface tension coefficient σ (the Marangoni effect) and the thermal creep of the environment over the surface. In [3], the thermal creep coefficient K_{tc}^e was estimated for a number of liquids. For example, for mercury drops moving in water and glycerol, it was found to be $K_{tc}^e = 0.13$ and 2.5×10^{-5} , respectively. Hereafter, the indices e and i refer to a viscous liquid and drop, respectively; the subscript tc , to liquid parameters away from the drop; the subscript s , to physical quantities taken at the mean temperature of the surface of the drop. Note that the thermal creep coefficient has been so far evaluated with a reasonable accuracy only for gases [4]. This is because a rigorous mathematical theory of inhomogeneous liquids is absent.

Consider a heated drop of a viscous incompressible liquid steadily moving in another viscous incompressible liquid occupying the entire space. The liquids are immiscible with each other. By a heated drop, we mean a drop whose mean surface temperature far exceeds the ambient temperature. The drop may be heated, for example, by a chemical reaction proceeding in its bulk, radioactive decay of the drop material, absorption of electromagnetic radiation, etc. The heated surface of the drop has a significant effect on the thermal physical properties of the environment and thereby affects greatly the velocity and pressure fields in its vicinity. At infinity, the liquid is at rest and is subjected to a given temperature gradient. It is assumed that the densities, thermal conductivities, and specific heats inside and outside the liquids are constant; the viscosity coefficient

of the drop exceeds that of the environmental liquid; and the surface tension coefficient is an arbitrary function of temperature. Also, the drop is assumed to move slowly (small Peclet and Reynolds numbers) and retain the spherical shape (the distortion of sphericity will be discussed later).

Unlike the case studied previously [1–8], we here consider the thermophoretic motion of a spherical drop for an arbitrary temperature difference between the surface of the drop and areas away from it. In the equation of hydrodynamics, the temperature dependence of the viscosity has the form of an exponential–power series; in the equation of heat conduction, only convective terms are taken into account.

For the hydrodynamic problem [9, 10] and for the thermal problem [11], it was shown that the inertial and convective terms away from the sphere become comparable to those responsible for molecular transfer by order of magnitude. Therefore, the standard method of expansion in a small parameter introduces a noticeable error in this case, because it cannot satisfy rigorously boundary conditions at infinity and find an exact unified solution equally valid in the entire flow domain.

It was demonstrated [12] that the heating of the surface of the drop and taking into account the motion of the liquid affect significantly the drag force of the medium. In this work, we study the effect of the motion of the medium on the force and rate of thermophoresis for a heated drop with an arbitrary temperature difference in its vicinity in the presence of a given external temperature gradient.

Of all transport parameters of a liquid, only the viscosity coefficient strongly depends on temperature [13]. To include this dependence, we take advantage of the formula

$$\mu_e = \mu_\infty \left[1 + \sum_{n=1}^{\infty} F_n \left(\frac{T_e}{T_\infty} - 1 \right)^n \right] \exp \left\{ -A \left(\frac{T_e}{T_\infty} - 1 \right) \right\}. \quad (1)$$

which describes the variation of the viscosity over a wide temperature range with any desired accuracy. (At $F_n = 0$, this formula is reduced to the well-known Reynolds relationship [13]).

It is known that the viscosity of a liquid decreases with increasing temperature by the exponential law [13]. The analysis of the semi-empiric formulas of frequent use showed that expression (1) describes the viscosity variation over a wide temperature range most adequately with any desired accuracy. With the coefficients F_n not taken into consideration, the error can be as high as 40%. For illustration, Tables 1 and 2 list the values of F_n for glycerol and water. The relative error does not exceed 3%. The coefficients F_n were calculated with the Maple V software suite.

Let us place the origin of a fixed coordinate system at the instantaneous center of a spherical drop of radius R . We assume that the drop moves with a constant velocity U in the negative OZ direction. The velocity and pressure distributions must be symmetric about the line passing through the center of the drop parallel to the velocity vector \mathbf{U} . In terms of our assumptions, the equations and boundary conditions for the velocity and temperature in the spherical coordinate system are written as [14, 15]

$$\nabla P_e = \mu_e \Delta \mathbf{U}_e + 2(\nabla \mu_e \nabla) \mathbf{U}_e + [\nabla \mu_e \text{rot} \mathbf{U}_e], \quad \text{div} \mathbf{U}_e = 0, \quad (2)$$

$$\mu_i \Delta \mathbf{U}_i = \nabla P_i, \quad \text{div} \mathbf{U}_i = 0, \quad (3)$$

$$\rho_e c_e (\mathbf{U}_e \nabla) T_e = \lambda_e \Delta T_e, \quad (4)$$

$$\rho_i c_i (\mathbf{U}_i \nabla) T_i = \lambda_i \Delta T_i + q_i, \quad (5)$$

$$r = R, \quad T_e = T_i, \quad \lambda_e \frac{\partial T_e}{\partial r} = \lambda_i \frac{\partial T_i}{\partial r},$$

$$U_r^e = U_r^i = -U \cos \Theta, \quad (6)$$

$$U_\Theta^e - U_\Theta^i = K_{tc}^e \frac{v_e}{RT_e} \frac{\partial T_e}{\partial \Theta} - K_{tc}^i \frac{v_i}{RT_i} \frac{\partial T_i}{\partial \Theta} \quad (v = \mu/\rho),$$

$$\begin{aligned} & \mu_e \left[r \frac{\partial}{\partial r} \left(\frac{U_\Theta^e}{r} \right) + \frac{1}{r} \frac{\partial U_r^e}{\partial \Theta} \right] - \frac{1}{r} \frac{\partial \sigma}{\partial T_i} \frac{\partial T_i}{\partial \Theta} \\ & = \mu_i \left[r \frac{\partial}{\partial r} \left(\frac{U_\Theta^i}{r} \right) + \frac{1}{r} \frac{\partial U_r^i}{\partial \Theta} \right], \end{aligned} \quad (7)$$

$$r \rightarrow \infty, \quad \mathbf{U}_e \rightarrow 0, \quad P_e = P_\infty,$$

$$T_e \rightarrow T_\infty + |\nabla T| r \cos \Theta,$$

$$r \rightarrow 0, \quad |\mathbf{U}_i| \neq \infty, \quad P_i \neq \infty, \quad T_i \neq \infty. \quad (8)$$

Here, $q_i(r, \Theta)$ is the density of heat sources nonuniformly distributed in the drop. Specifically, if the drop is heated by absorbing electromagnetic radiation, the nonuniformity depends on the optical constants of the

Table 1.

Glycerol: $A = 17.29, F_1 = -1.228, F_2 = 7.022, T_\infty = 303 \text{ K}$			
$T, \text{ }^\circ\text{C}$	$\mu_{\text{cal}}, \text{ Pa s}$	$\mu_{\text{exp}}, \text{ Pa s}$	$\frac{ \mu_{\text{exp}} - \mu_{\text{cal}} }{\mu_{\text{exp}}} 100\%$
30	0.600000	0.600	0.00
40	0.327979	0.330	0.61
50	0.182001	0.180	1.11
60	0.102619	0.102	0.60
70	0.058797	0.059	0.34
80	0.034212	0.035	2.25
90	0.020189	0.021	3.86

Note: μ_{cal} : dynamic viscosity calculated by formula (1); μ_{exp} : experimental value.

Table 2.

Water: $A = 5.779, F_1 = -2.318, F_2 = 9.118, T_\infty = 273 \text{ K}$			
$T, \text{ }^\circ\text{C}$	$\mu_{\text{cal}}, \text{ Pa s}$	$\mu_{\text{exp}}, \text{ Pa s}$	$\frac{ \mu_{\text{exp}} - \mu_{\text{cal}} }{\mu_{\text{exp}}} 100\%$
0	0.0017525	0.0017525	0.00
10	0.0013151	0.0012992	1.22
20	0.0010089	0.0010015	0.74
30	0.0007943	0.0007971	0.35
40	0.0006433	0.0006513	1.22
50	0.0005359	0.0005441	1.51
60	0.0004581	0.0004630	1.06
70	0.0004002	0.0004005	0.07
80	0.0003556	0.0003509	1.35
90	0.0003199	0.0003113	2.76

drop ($m_i = n_i + ia_i$, where m_i, n_i , and a_i are the complex refractive index, refractive index, and the absorption coefficient of the drop, respectively) and its diffraction parameter $x_d = 2\pi R/\lambda$ (λ is the wavelength). The expression for the radiation energy density converted to heat in the drop can be represented in the form [16]

$$q_i = \frac{4\pi R n_i a_i}{n_e} I B_i,$$

where I is the incident radiation intensity, B_k ($k = e, i$) is a coordinate-dependent function calculated by the Mie theory.

The results of numerical calculation for the B_k distribution in the case of water drops are given in [16]. They show that the nonuniformity of the absorbed energy distribution in a water drop increases with its radius. The nonuniformity is the greatest in the direction of radiation propagation.

Boundary conditions (6) on the surface of the drop ($r = R$) include the impermeability condition for the velocity normal components, equality of the temperatures, heat flux continuity, equality of the shear velocities at the inner and outer surfaces of the drop, and continuity of the shear components of the stress tensor.

Away from the drop ($r \rightarrow \infty$), boundary conditions (7) are valid, while the finiteness of the physical quantities characterizing the drop at $r \rightarrow 0$ is included in (8).

The decisive parameters of the problem are the material constants $\rho_e, \mu_\infty, \lambda_e,$ and $c_e,$ as well as $R, |\nabla T|, T_\infty,$ and $U,$ which remain constant during the motion of the spherical drop. With these parameters, one can compose a three-dimensional combination including $\varepsilon = R|\nabla T|/T_\infty \ll 1$ (this parameter characterizes a temperature difference within the drop), as well as the Peclet and Reynolds numbers.

We will make Eqs. (2)–(5) and boundary conditions (6)–(8) dimensionless by introducing the dimensionless velocity, temperature, and pressure: $\mathbf{V}_k = \mathbf{U}_k/U, t_k = T_k/T_\infty,$ and $p_k = P_k/P_\infty.$ Here, the radius of the drop $R,$ temperature $T_\infty,$ pressure $P_\infty,$ and velocity U are used as units of measure of distance, temperature, pressure, and velocity, respectively ($U \sim |\mu_\infty|\nabla T|/(\rho_e T_\infty)$).

At $\varepsilon \ll 1,$ a solution to the equations of hydrodynamics should be sought in the form

$$\begin{aligned} \mathbf{V} &= \mathbf{V}^{(0)} + \varepsilon \mathbf{V}^{(1)} + \dots, \\ p &= p^{(0)} + \varepsilon p^{(1)} + \dots, \quad t = t^{(0)} + \varepsilon t^{(1)} + \dots \end{aligned} \tag{9}$$

The form of boundary conditions (6)–(8) indicates that an expression for the velocity components V_r and V_Θ is sought as the expansion in Legendre and Gegenbauer polynomials. The force acting on the drop is found by integrating the stress tensor over its surface. Having regard for the properties of Legendre and Gegenbauer polynomials, we can assume that this force depends largely on the first terms of the expansions; therefore,

$$V_r = G(y)\cos\Theta, \quad V_\Theta = -g(y)\sin\Theta, \tag{10}$$

where $G(y)$ and $g(y)$ are arbitrary functions depending on the dimensionless radial coordinate $y = r/R.$

When studying the motion of nonuniformly heated drops under an external temperature gradient in a viscous medium, one should take into account the temperature dependences of both the dynamic viscosity coefficient and the surface tension coefficient. This is because the density q_i of heat sources in the drop is non-uniform. In this work, the surface tension coefficient is taken to be an arbitrary function of temperature. Moreover, for the first time, an attempt is made to take into consideration the effect of motion of the medium on the force and velocity of the thermocapillary drift of a heated drop in a viscous liquid. Therefore, final formulas are of general character and are valid for any tem-

perature difference between the surface of the drop and regions away from it.

TEMPERATURE FIELDS INSIDE AND OUTSIDE A HEATED DROP

When finding the force acting on a nonuniformly heated drop and its velocity, we will consider only first-order corrections. To find them, it is necessary to know temperature fields inside and outside the drop, i.e., to solve Eqs. (4) and (5) with appropriate boundary conditions. The dimensionless equations of heat conduction have the form

$$\varepsilon \text{Pr}_\infty (\mathbf{V}_e \nabla) t_e = \Delta t_e, \tag{11}$$

$$\varepsilon \text{Pr}_\infty \beta (\mathbf{V}_i \nabla) t_i + Q = \Delta t_i, \tag{12}$$

$$\begin{aligned} y = 1, \quad t_i &= t_e, \quad \lambda_e \frac{\partial t_e}{\partial y} = \lambda_i \frac{\partial t_i}{\partial y}, \\ y \rightarrow \infty, \quad t_e &\rightarrow 1 + \varepsilon y \cos \Theta, \\ y \rightarrow 0, \quad t_i &< \infty. \end{aligned} \tag{13}$$

Here, $\text{Pr}_\infty = \mu_\infty c_e / \lambda_e$ is the Prandtl number, $\beta = \chi_e / \chi_i,$ χ is the thermal diffusivity, and $Q = -q_i R^2 / (\lambda_i T_\infty).$ Substituting (9) into Eqs. (11) and (12), we arrive at the following set of equations:

zeroth-order approximation ($\varepsilon = 0$),

$$\Delta t_e^{(0)} = 0, \tag{14}$$

$$\Delta t_i^{(0)} = Q_0, \tag{15}$$

$$\begin{aligned} y = 1, \quad t_e^{(0)} &= t_i^{(0)}, \quad \lambda_e \frac{\partial t_e^{(0)}}{\partial y} = \lambda_i \frac{\partial t_i^{(0)}}{\partial y}, \\ y \rightarrow \infty, \quad t_e^{(0)} &\rightarrow 1, \\ y \rightarrow 0, \quad t_i^{(0)} &< \infty. \end{aligned}$$

first-order approximation ($\sim \varepsilon$),

$$\text{Pr}_\infty \left(B_r \frac{\partial t_e^{(0)}}{\partial y} + \frac{V_\Theta^e \partial t_e^{(0)}}{y \partial \Theta} \right) = \Delta t_e^{(1)}, \tag{16}$$

$$\text{Pr}_\infty \beta \left(V_r^e \frac{\partial t_e^{(0)}}{\partial y} + \frac{V_\Theta^e \partial t_e^{(0)}}{y \partial \Theta} \right) + Q_1 = \Delta t_i^{(1)}, \tag{17}$$

$$\begin{aligned} y = 1, \quad t_i^{(1)} &= t_e^{(1)}, \quad \lambda_i \frac{\partial t_i^{(1)}}{\partial y} = \lambda_e \frac{\partial t_e^{(1)}}{\partial y}, \\ y \rightarrow \infty, \quad t_e^{(1)} &\rightarrow y \cos \Theta, \\ y \rightarrow 0, \quad t_i^{(1)} &< \infty. \end{aligned}$$

When deriving the equation for the temperature distribution inside the drop, we assumed that

$$-\sum_{n=0}^{\infty} q_i(r, \Theta) \frac{R^2}{\lambda_i T_{\infty}} = \sum_{n=0}^{\infty} \varepsilon^n Q_n P_n(x),$$

where

$$Q_n = -\frac{R^2 2n+1}{T_{\infty} 2\lambda_i} \int_{-1}^{+1} q_i(r, \Theta) P_n(x) dx, \quad x = \cos \Theta.$$

Let us find the zeroth-order approximations. The general solutions to (14) and (15) have the form

$$t_e^{(0)}(y) = 1 + \frac{\gamma}{y} + \sum_{n=1}^{\infty} \frac{\Gamma_n}{y^{n+1}} P_n(x), \quad (18)$$

$$t_i^{(0)}(y) = B_0 + \frac{1}{4\pi R T_{\infty} \lambda_i y} \int_V q_i dV + \int_1^y \frac{\Psi_0}{y} dy - \frac{1}{y} \int_1^y \Psi_0 dy, \quad (19)$$

where

$$\Psi_0 = -\frac{R^2}{2\lambda_i T_{\infty}} y^2 \int_{-1}^{+1} q_i(r, \Theta) dx.$$

In (19), integration is over the entire volume of the drop. The constants of integration γ , Γ_n , and B_0 are determined by substituting (18) and (19) into the related boundary condition. After the substitution, we find that

$$\gamma = t_s - 1, \quad B_0 = 1 + \left(1 - \frac{\lambda_e}{\lambda_i}\right) \gamma,$$

$\Gamma_n = 0$ at $n \geq 1$, $t_s = T_s/T_{\infty}$. T_s is the mean temperature on the surface of the drop given by

$$\frac{T_s}{T_{\infty}} = 1 + \frac{1}{4\pi R \lambda_e T_{\infty}} \int_V q_i dV. \quad (20)$$

In (20), integration is also over the entire volume of the drop.

For $\lambda_e < \lambda_i$, we can neglect the Θ dependence of the dynamic viscosity coefficient in the drop-liquid medium system and assume that the viscosity depends only on the temperature $t_e^{(0)}(y)$; i.e., $\mu_e(t_e) = \mu_e(t_e^{(0)})$. With this in mind, expression (1) takes the form

$$\mu_e = \mu_{\infty} \exp \left\{ -A \frac{\gamma}{y} \right\} \left[1 + \sum_{n=1}^{\infty} F_n \frac{\gamma^n}{y^n} \right]. \quad (21)$$

Formula (21) will subsequently be used for finding the velocity and pressure fields in the vicinity of the heated drop.

Now let us find the first-order approximations. Substituting (18) and (19) into (16) and (17), we come to

$$-\omega \frac{V_r^e}{y^2} = \Delta t_e^{(1)}, \quad (22)$$

$$\text{Pr}_{\infty} \beta \frac{V_r^i}{y^2} \left(\int_1^y \Psi_0 dy - \gamma \frac{\lambda_e}{\lambda_i} \right) + Q_1 \cos \Theta = \Delta t_i^{(1)}, \quad (23)$$

where $\omega = \text{Pr}_{\infty} \gamma$.

From (22) and (23), it follows that one must first solve the hydrodynamic problem, i.e., determine the velocity fields inside and outside the drop, in order to find $t_e^{(1)}$ and $t_i^{(1)}$.

DETERMINATION OF THE DRAG FORCE

Substituting (22) into the equations of hydrodynamics, taking into account (10), and separating the variables, we come to an equation similar to that obtained in [17]. Eventually we have the following expressions for the components of the mass velocity and pressure that satisfy boundary conditions (7) and (8):

$$V_r^e(y, \Theta) = \cos \Theta (A_1 G_1 + A_2 G_2),$$

$$V_{\Theta}^e(y, \Theta) = -\sin \Theta (A_1 G_3 + A_2 G_4), \quad (24)$$

$$p_e(y, \Theta) = 1 + \eta_e \cos \Theta (A_1 G_5 + A_2 G_6),$$

$$V_r^i(y, \Theta) = \cos \Theta (A_3 + A_4 y^2),$$

$$V_{\Theta}^i(y, \Theta) = -\sin \Theta (A_3 + 2A_4 y^2), \quad (25)$$

$$p_i(y, \Theta) = p_{0i} + 10\eta_i \cos \Theta y^2 A_4, \quad \eta = \mu/\mu_{\infty},$$

where

$$G_1 = -\frac{1}{y^3} \sum_{n=0}^{\infty} \frac{\Delta_n^{(1)}}{(n+3)y^n}, \quad G_2 = -\frac{1}{y} \sum_{n=0}^{\infty} \frac{\Delta_n^{(2)}}{(n+1)y^n}$$

$$- \frac{\alpha}{y^3} \sum_{n=0}^{\infty} \left[(n+3) \ln \frac{1}{y} - 1 \right] \frac{\Delta_n^{(1)}}{(n+3)^2 y^n},$$

$$G_3 = G_1 + \frac{y}{2} G_1^I,$$

$$G_5 = \frac{y^2}{2} G_1^{III} + y \left(3 + \frac{1}{2} \sum_{n=0}^{\infty} s_n \frac{\gamma^n}{y^n} \right) G_1^{II} \quad (26)$$

$$+ \left(2 + \sum_{n=0}^{\infty} s_n \frac{\gamma^n}{y^n} \right) G_1^I, \quad G_4 = G_2 + \frac{y}{2} G_2^I,$$

$$G_6 = \frac{y^2}{2} G_2^{III} + y \left(3 + \frac{1}{2} \sum_{n=0}^{\infty} s_n \frac{\gamma^n}{y^n} \right) G_2^{II} + \left(2 + \sum_{n=0}^{\infty} s_n \frac{\gamma^n}{y^n} \right) G_2^I,$$

$s_n = AF_{n-1} - nF_n - \sum_{k=1}^n s_{n-k} F_k$, $F_0 = 1$, and $F_n = 0$ at $n < 0$. In (26), G_k^I , G_k^{II} , and G_k^{III} are the first-, second-, and third-order derivatives of the associated functions with respect to y ($k = 1, 2$).

The coefficients $\Delta_n^{(1)}$ and $\Delta_n^{(2)}$ are found from the recurrent relations

$$\Delta_n^{(1)} = -\frac{1}{n(n+5)} \sum_{k=1}^n [(n+4-k) \quad (27)$$

$$\times \{ \alpha_k^{(1)}(n+5-k) - \alpha_k^{(2)} \} + \alpha_k^{(3)}] \gamma^k \Delta_{n-k}^{(1)} \quad (n \geq 1),$$

$$\Delta_n^{(2)} = -\frac{1}{(n+3)(n-2)} \left[-6\alpha_n^{(4)} \gamma^n + \sum_{k=1}^n \{ (n+2-k) \right. \\ \times [(n+3-k)\alpha_k^{(1)} - \alpha_k^{(2)}] + \alpha_k^{(3)} \} \gamma^k \Delta_{n-k}^{(2)} \quad (28)$$

$$\left. + \alpha \sum_{k=0}^n \{ (2n+5-2k)\alpha_k^{(1)} - \alpha_k^{(2)} \} \gamma^k \Delta_{n-k-2}^{(1)} \right] \quad (n \geq 3).$$

Upon calculating the coefficients $\Delta_n^{(1)}$ and $\Delta_n^{(2)}$ by formulas (27) and (28), it should be taken into account that

$$\Delta_0^{(1)} = -3, \quad \Delta_0^{(2)} = -1, \quad \Delta_2^{(2)} = 1, \quad \alpha_0^{(1)} = \alpha_0^{(4)} = 1,$$

$$\alpha_0^{(2)} = 4, \quad \alpha_0^{(3)} = -4, \quad \alpha_n^{(1)} = F_n,$$

$$\alpha_n^{(2)} = (4-n)F_n + AF_{n-1}, \quad \alpha_n^{(4)} = A^n/n!,$$

$$\alpha_n^{(3)} = 2AF_{n-1} - 2(2+n)F_n,$$

$$\Delta_1^{(2)} = -\gamma[6\alpha_1^{(4)} + 2(3\alpha_1^{(1)} - \alpha_1^{(2)}) + \alpha_1^{(3)}],$$

$$\alpha = \frac{\gamma}{15} \{ -6\gamma\alpha_2^{(4)} + [3(4\alpha_1^{(1)} - \alpha_1^{(2)}) + \alpha_1^{(3)}] \Delta_1^{(2)} \\ - [2(3\alpha_2^{(1)} - \alpha_2^{(2)}) + \alpha_2^{(3)}] \gamma \}.$$

Substituting (24) into (22), we have for $t_e^{(1)}$

$$\Delta t_e^{(1)} = -\frac{\omega}{y^2} G(y) \cos \Theta. \quad (29)$$

Here, $G(y) = A_1 G_1 + A_2 G_2$. A solution for $t_e^{(1)}$ is sought

in the form

$$t_e^{(1)} = \tau_e(y) \cos \Theta. \quad (30)$$

Substituting (30) into (29), we check that the variables are separable; then, for $t_e^{(1)}$, we have

$$\frac{d^2 \tau_e}{dy^2} + \frac{2d\tau_e}{y dy} - \frac{2}{y^2} \tau_e = -\frac{\omega}{y^2} G. \quad (31)$$

The general solution to Eq. (31) that satisfies the boundary conditions for $y \rightarrow \infty$ has the form

$$t_e^{(1)}(y, \Theta) = \left\{ \frac{\Gamma}{y^2} + y + \omega \sum_{k=1}^2 A_k \tau_k \right\} \cos \Theta, \quad (32)$$

where

$$\tau_1(y) = \frac{1}{y^3} \sum_{n=0}^{\infty} \frac{\Delta_n^{(1)}}{(n+1)(n+3)(n+4)y^n},$$

$$\tau_2(y) = -\frac{1}{y} \left\{ -\frac{1}{2} + \frac{\Delta_1^{(2)}}{6y} \ln y - \sum_{n=2}^{\infty} \frac{\Delta_n^{(2)}}{(n^2-1)(n+2)y^n} \right.$$

$$\left. - \frac{\alpha}{y^2} \sum_{n=0}^{\infty} \left[(n+1)(n+3)(n+4) \ln \frac{1}{y} - (3n^2 + 16n + 19) \right] \right.$$

$$\left. \times \frac{\Delta_n^{(1)}}{(n+1)^2(n+3)^2(n+4)^2 y^n} \right\}.$$

A solution for $t_i^{(1)}$ is sought in the form

$$t_i^{(1)} = \tau_i(y) \cos \Theta. \quad (33)$$

Substituting (33) into (23), we come to the equation for $t_i^{(1)}$:

$$\text{Pr}_{\infty} \beta \frac{V_r^i}{y^2} \left(\int_1^y \Psi_0 dy - \gamma \frac{\lambda_e}{\lambda_i} \right) + Q_1 \cos \Theta \quad (34)$$

$$= \frac{d^2 \tau_i}{dy^2} + \frac{2d\tau_i}{y dy} - \frac{2}{y^2} \tau_i.$$

The general solution to Eq. (34) that satisfies the boundary condition for $y \rightarrow 0$ is

$$t_i^{(1)} = \cos \Theta \left[By + \frac{C}{y^2} + \omega \frac{\beta \lambda_e}{2 \lambda_i} \left(A_3 - \frac{A_4}{2} y^2 \right) \right. \\ \left. + \frac{1}{3} \left\{ y \int_1^y \frac{\Psi_1}{y^2} dy - \frac{1}{y^2} \int_1^y \Psi_1 y dy \right\} \right] \quad (35)$$

$$+ \frac{1}{3} \left\{ y \int_1^y \Omega \left(A_4 + \frac{A_3}{y^2} \right) dy - \frac{1}{y^2} \int_1^y \Omega (A_3 y + A_4 y^3) dy \right\},$$

where

$$\Psi_1 = -\frac{3R^2}{2\lambda_i T_\infty} y^2 \int_{-1}^{+1} q_i x dx,$$

$$C = \frac{RJ}{3T_\infty \lambda_i} - \frac{\omega \beta \lambda_e}{6 \lambda_i} \left(A_3 + \frac{A_4}{2} \right), \quad V = \frac{4}{3} \pi R^3,$$

$$J = \frac{1}{V} \int_V q_i z dV, \quad \Omega = \text{Pr}_\infty \beta \int_1^y \Psi_0 dy,$$

$$z = r \cos \Theta, \quad \int_V q_i z dV$$

is the dipole moment of the heat source density.

The constants of integration Γ and B are found from the boundary conditions on the surface (the equality of the temperatures and fluxes). Specifically,

$$\Gamma = -\frac{1}{\delta} \left(1 - \frac{\lambda_e}{\lambda_i} \right) + \frac{RJ}{\lambda_i \delta T_\infty} - \frac{\omega}{\delta} \left[A_1 \left(\tau_1 - \frac{\lambda_e}{\lambda_i} \tau_1^1 \right) + A_2 \left(\tau_2 - \frac{\lambda_e}{\lambda_i} \tau_2^1 \right) \right],$$

where $\delta = 1 + 2(\lambda_e/\lambda_i)$ and τ_1^1 and τ_2^1 are the first derivatives of the functions τ_1 and τ_2 with respect to y .

Thus, we determined the temperature fields inside and outside the drop in the first approximation in ε . Now, the constants of integration $A_1, A_2, A_3,$ and A_4 entering into expressions (24) and (25) can be found from the boundary conditions for the velocity components on the surface of the drop.

Below, we give the coefficient A_2 in explicit form, since the total force acting on the drop is expressed through it:

$$A_2 = -\frac{1}{\Delta} \left(N_3 + \frac{\mu_e}{3\mu_i} N_4 \right) - \varepsilon \frac{2G_1}{3U\Delta} \left[3 \frac{\lambda_e}{\delta \lambda_i} + \frac{RJ}{\delta T_\infty \lambda_i} - \frac{\omega \lambda_e}{\delta \lambda_i} \frac{2\tau_1 + \tau_1^1}{G_1} \right] \times \left(K_{tc}^e \frac{V_e^s}{t_s} - K_{tc}^i \frac{V_i^s}{t_s} + \frac{R}{3\mu_i^s} \frac{\partial \sigma}{\partial t_i} \right),$$

where

$$\Delta = N_1 + \frac{\mu_e}{3\mu_i} N_2 + \frac{2}{3\mu_\infty} \rho_e R \frac{\omega \lambda_e}{\delta \lambda_i} [G_1 \Phi_2 - G_2 \Phi_1]$$

$$\times \left(K_{tc}^e \frac{V_e^s}{t_s R} - K_{tc}^i \frac{V_i^s}{t_s R} + \frac{1}{3\mu_i^s} \frac{\partial \sigma}{\partial t_i} \right),$$

$$\Phi_k = 2\tau_k + \tau_k^1 \quad (k = 1, 2).$$

The force acting on the drop is found by integrating the stress tensor over its surface [18]:

$$F = \oint_S \left\{ -P_e \cos \Theta + \sigma_{rr}^e \cos \Theta - \sigma_{r\Theta}^e \sin \Theta \right\} r^2 \sin \Theta d\Theta d\varphi |_{y=1}. \quad (36)$$

where

$$\sigma_{rr}^e = 2\mu_e \frac{\partial U_r^e}{\partial r}, \quad \sigma_{r\Theta}^e = \mu_e \left(\frac{\partial U_\Theta^e}{\partial r} + \frac{1}{r} \frac{\partial U_r^e}{\partial \Theta} - \frac{U_\Theta^e}{r} \right).$$

Substituting expressions (24) into (36) yields after integration

$$F = 4\pi R \mu_\infty U A_2 \exp\{-A\gamma\}. \quad (37)$$

In view of the explicit form of A_2 , the total force acting on a heated drop subjected to an external temperature gradient is the additive sum of the viscous force \mathbf{F}_μ of the medium, thermophoretic force \mathbf{F}_{th} , force \mathbf{F}_q proportional to the dipole moment of the density of heat sources nonuniformly distributed in the bulk of the drop, and force F_m due to the motion of the medium (i.e., the force including convective terms in the heat conduction equation):

$$\mathbf{F} = \mathbf{F}_\mu + \varepsilon \mathbf{F}^{(1)}, \quad \mathbf{F}^{(1)} = \mathbf{F}_{th} + \mathbf{F}_q + \mathbf{F}_d, \quad (38)$$

where $\mathbf{F}_\mu = -6\pi R \mu_\infty U f_\mu \mathbf{n}_z$, $\mathbf{F}_{th} = -6\pi R \mu_\infty f_{th} \mathbf{n}_z$, $\mathbf{F}_q = -6\pi R \mu_\infty f_q \mathbf{J} \mathbf{n}_z$, and $\mathbf{F}_d = -6\pi R \mu_\infty f_m \mathbf{n}_z$.

The coefficients $f_\mu, f_{th}, f_q,$ and f_m can be estimated from the expressions

$$f_\mu = \frac{2}{3\Delta} \left[N_3 + \frac{\mu_e^s}{3\mu_i^s} N_4 \right] \exp\{-A\gamma\},$$

$$f_{th} = \frac{4G_1}{3\Delta R \lambda_i^s \delta} \left(K_{ts}^e \frac{V_e^s}{t_s} - K_{ts}^i \frac{V_i^s}{t_s} + \frac{R}{3\mu_i^s} \frac{\partial \sigma}{\partial t_i} \right) \exp\{-A\gamma\},$$

$$f_q = \frac{4}{9\Delta \lambda_i^s T_\infty \delta} \left(K_{tc}^e \frac{V_e^s}{t_s} - K_{tc}^i \frac{V_i^s}{t_s} + \frac{R}{3\mu_i^s} \frac{\partial \sigma}{\partial t_i} \right) \exp\{-A\gamma\},$$

$$f_m = -\frac{4}{9\Delta R \delta} \frac{\omega \lambda_e^s}{\lambda_i^s} [2\tau_1 + \tau_1^1]$$

$$\times \left(K_{tc}^e \frac{V_e^s}{t_s} - K_{tc}^i \frac{V_i^s}{t_s} + \frac{R}{3\mu_i^s} \frac{\partial \sigma}{\partial t_i} \right) \exp\{-A\gamma\}.$$

In the expressions for the coefficients f_μ , t_{th} , f_q , and f_m , the index s refers to the quantities taken at the mean surface temperature T_s of the drop, which is determined from (20), and the parameters N_1 – N_4 , τ_1 , τ_2 , τ_1^I , τ_2^I , G_1 , and G_2 are taken at $y = 1$ ($N_4 = 2G_1^I + G_1^{II}$, $N_3 = -G_1^I$, $N_2 = [G_2(2G_1^I + G_1^{II}) - G_1(2G_2^I + G_2^{II})]$, and $N_1 = G_1G_1^I - G_2G_1^I$).

Equating the total force to zero, we obtain an expression for the directed velocity of the drop in a given external temperature gradient:

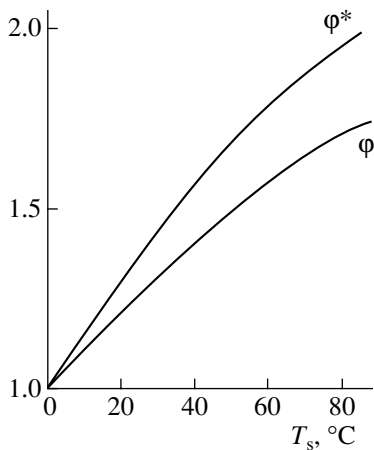
$$\mathbf{U} = -\varepsilon U^{(1)} \mathbf{n}_z, \quad U^{(1)} = U_{th} + U_q + U_d, \quad (39)$$

where $U_{th} = h_{th}$, $U_q = h_q J$, and $U_m = h_m$ ($h_{th} = f_{th}/f_\mu$, $h_q = f_q/f_\mu$, and $h_m = f_m/f_\mu$).

If the drop is heated only slightly, that is, its mean surface temperature is close to the ambient temperature at infinity ($\gamma \rightarrow 0$), the temperature dependence of the viscosity coefficient can be neglected; then, $G_1 = 1$, $G_1^I = -3$, $G_1^{II} = 12$, $G_2 = 1$, $G_2^I = -1$, $G_2^{II} = 2$, $N_1 = 2$, $N_2 = 6$, $N_3 = 3$, $N_4 = 6$, $\tau_1 = -1/4$, $\tau_1^I = 3/4$, $\tau_2 = 1/2$, and $\tau_2^I = -1/2$, and formulas (38) and (39) pass to expressions well-known from the literature [1–6].

If the distribution of heat sources over the volume of the heated drop is known, formulas (38) and (39) allow one to take into account (1) the effect of the motion of the medium on the drag force acting on the drop and (2) the effect of drop surface heating on the thermocapillary force and velocity for arbitrary temperature differences between the surface and regions away from the drop. These formulas also account for the exponential temperature dependence of the viscosity under an external temperature gradient. Emphasize once again that they are of general character.

These formulas imply that the magnitude and the direction of the force $\mathbf{F}^{(1)}$ and velocity $\mathbf{U}^{(1)}$ are also



Functions φ and φ^* vs. mean surface temperature T_s .

affected by the direction of the dipole moment of the heat source density $\int_V q_i z dV$. If, for example, the drop heats up, absorbing electromagnetic energy, the dipole moment can be both negative (a major part of the thermal energy is released in that side of the drop facing the radiation) and positive (a major part of the thermal energy is released in the opposite side), depending on the optical properties of the drop. Note that the surface tension decreases with temperature for most liquids, i.e., $\partial\sigma/\partial t_i < 0$, and that the value of $\int_V q_i z dV$ may be both positive and negative. Accordingly, the magnitude and the direction of the force $\mathbf{F}^{(1)}$ and velocity $\mathbf{U}^{(1)}$ vary.

In addition, these formulas suggest that the forced $\mathbf{F}^{(1)}$ and the velocity $\mathbf{U}^{(1)}$ also depend significantly on the thermal conductivity of the drop. At λ_i approaching infinity, $\mathbf{F}^{(1)}$ and $\mathbf{U}^{(1)}$ tend to zero with the dipole moment of the heat source density fixed.

For $\mu_i \rightarrow \infty$, the above formulas can be used for estimating the force and the velocity of a hard nonuniformly heated particle in a viscous liquid where a constant temperature gradient is maintained.

The figure illustrates the effect of heating the surface of the drop on its drift velocity. The curves relate the function $\varphi = h_{th}/h_{th}|_{T_s=273\text{ K}}$ to the mean surface temperature T_s of the drop. Estimates were obtained for mercury drops suspended in water at $T_\infty = 273\text{ K}$ and $R = 15\ \mu\text{m}$ ($\partial\sigma/\partial T = -2 \times 10^{-4}\text{ N/mK}$, $\text{Pr}_\infty = 12.99$). The value of the function φ^* was estimated by formula (39) for a small temperature difference ($\gamma \rightarrow 0$), but the molecular transfer coefficients were taken at the mean surface temperature T_s .

To estimate the contribution of the motion of the medium to the thermocapillary drift of the drop, it is necessary to clarify the nature of the heat sources. Knowing their nature, one can find an expression for the dipole moment of the heat source density. To analyze the situation qualitatively, let us consider the simplest case. We assume that the drop heats up, absorbing electromagnetic radiation as a black body. Under these conditions, absorption takes place in a thin layer of thickness $\delta R \ll R$ adjacent to the heated part of the drop. In this case, the heat source density within a layer of thickness δR is given by

$$q_i(r, \Theta) = \begin{cases} -\frac{I}{\delta R} \cos \Theta, & \frac{\pi}{2} \leq \Theta \leq \pi, \quad R - \delta R \leq r \leq R, \\ 0, & 0 \leq \Theta \leq \frac{\pi}{2}, \end{cases} \quad (40)$$

where I is the incident radiation intensity. It is related to the mean relative surface temperature of the drop as

$$t_s = 1 + \frac{R}{4\lambda_i T_\infty} I.$$

In view of (40), the integrals entering into the expression for the force acting on the heated drop and the drift velocity are calculated directly:

$$\int_V q_i(r, \Theta) z dV = -\frac{2}{3} \pi R^3 I, \quad \int_V q_i(r, \Theta) dV = \pi R^2 I.$$

Accordingly, formulas (38) and (39) can be represented in a more compact form:

$$\mathbf{F} = -6\pi R \mu_\infty U \varphi_\mu \mathbf{n}_z - \varepsilon 6\pi R \mu_\infty f_{ph} \mathbf{n}_z, \quad \text{where}$$

$$\mathbf{U}_{ph} = -\varepsilon h_{ph} \mathbf{n}_z,$$

$$\varphi_\mu = \frac{2}{3\Delta_h} \left[N_3 + \frac{\mu_e^s}{3\mu_i^s} N_4 \right] \exp\{-A\gamma\}, \quad h_{ph} = f_{ph}/\varphi_\mu,$$

$$f_{ph} = \frac{4}{3\mu_i^s \Delta_h} \frac{G_1 \lambda_e^s}{\delta \lambda_i^s}$$

$$\times \left[1 - \frac{R}{6\lambda_e^s T_\infty} I \left(\frac{\text{Pr}_\infty 2\tau_1 + \tau_1^I}{2G_1} + 1 \right) \right] \frac{\partial \sigma}{\partial t_i} \exp\{-A\gamma\},$$

$$\Delta_h = N_1 + \frac{\mu_e^s}{3\mu_i^s} N_2 + \frac{R^2 \rho_e \text{Pr}_\infty}{6\mu_\infty \mu_i^s} I \frac{\partial \sigma}{\partial t_i} [G_1 \Phi_2 - G_2 \Phi_1].$$

When deriving formulas (41), we took into consideration that the thermal creep coefficient is very small [3]; therefore, it was omitted in numerical calculations. If the motion of the environment is not taken into account ($\omega = 0$),

$$\mathbf{F} = -6\pi R \mu_\infty U \varphi_\mu^* \mathbf{n}_z - \varepsilon 6\pi R \mu_\infty f_{ph}^* \mathbf{n}_z, \quad (41)$$

$$\mathbf{U}_{ph}^* = -\varepsilon h_{ph}^* \mathbf{n}_z,$$

where

$$\varphi_\mu^* = \frac{2}{3\Delta_h^*} \left[N_3 + \frac{\mu_e^s}{3\mu_i^s} N_4 \right] \exp\{-A\gamma\}, \quad h_{ph}^* = f_{ph}^*/\varphi_\mu^*,$$

$$f_{ph}^* = \frac{4}{3\mu_i^s \Delta_h^*} \frac{G_1 \lambda_e^s}{\delta \lambda_i^s} \left[1 - \frac{R}{6\lambda_e^s T_\infty} I \right] \frac{\partial \sigma}{\partial t_i} \exp\{-A\gamma\},$$

$$\Delta_h^* = N_1 + \frac{\mu_e^s}{3\mu_i^s} N_2.$$

Formula (42) lacks the terms responsible for the motion of the medium (convective terms in the heat conduction equation) unlike (41). The convective term depends on the mean surface temperature and the Prandtl number (it is proportional to the Prandtl number times the relative temperature difference between the surface of the drop and regions away from it). Therefore, if the temperature difference is large and the Prandtl number is high (which is a possibility in liquids), convective terms in the heat conduction equation

may change essentially the qualitative picture of the thermocapillary drift of the drop. In gases, the consideration of the environment motion cannot affect the drift of the drop significantly, because the Prandtl number in gases is about unity. If the temperature difference is small ($\gamma \rightarrow 0$), the motion of the surrounding medium should be taken into account when the drop drifts in viscous liquids with a high Prandtl number.

Formulas for the motion of a hard spherical particle can be obtained in a similar way.

To illustrate the contribution of the motion of the medium to the force and velocity of the thermocapillary drift of the heated drop, Tables 3 and 4 summarize the values of the functions $f_{ph} = f_{ph}/f_{ph}|_{T_s = 273 \text{ K}}$ and $h_{ph} = h_{ph}/h_{ph}|_{T_s = 273 \text{ K}}$, respectively. The estimates were made for mercury drops suspended in water at $T_\infty = 273 \text{ K}$. The values of f_{ph}^{av} and h_{ph}^{av} were estimated from formulas (41) for a small temperature difference ($\gamma \rightarrow 0$), but the molecular transport coefficients were taken at the mean surface temperature T_s of the drop.

Table 3

$T_s, \text{ }^\circ\text{C}$	f_{ph}	f_{ph}^{av}
0	1	1
10	0.75679462	1.13172965
20	0.55865389	1.25217591
30	0.40612812	1.35890535
40	0.29192483	1.44815402
50	0.20865530	1.52363269
60	0.14791875	1.58086862
70	0.10410262	1.62387314
80	0.07247662	1.65233041
90	0.04977532	1.66842426

Table 4

$T_s, \text{ }^\circ\text{C}$	h_{ph}	h_{ph}^{av}
0	1	1
10	1.04610555	1.1018381
20	1.06591031	1.1918477
30	1.06609551	1.2690664
40	1.04899473	1.3307655
50	1.01989975	1.3807801
60	0.97702898	1.4155379
70	0.92304912	1.4387383
80	0.85740504	1.4502438
90	0.78130662	1.4518747

DISTORTION OF THE SURFACE SHAPE

The shape of the drop is unknown and should be found from the solution; therefore, boundary conditions (5)–(7) are set for a boundary of unknown shape. Since we restrict our analysis by first-order corrections,

$$\sigma = \sigma_0 + \varepsilon\sigma^{(1)}, \tag{42}$$

where σ_0 is the zeroth-order term in the expansion of the function $\sigma(x)$ in Legendre polynomials $P_n(x)$ ($x = \cos\Theta$).

The shape of the drop is sought in the form [14]

$$r = R[1 + \varepsilon\xi]. \tag{43}$$

Let us expand the quantities $\sigma(\Theta)$ and $\xi(\Theta)$ in Legendre polynomials:

$$\sigma = \sum_{n=0}^{\infty} \sigma_n P_n(\cos\Theta), \quad \xi = \sum_{n=0}^{\infty} \xi_n P_n(\cos\Theta). \tag{44}$$

From the constancy condition for the volume of the drop, it follows that $\xi_0 = 0$. Bearing in mind that the origin of the coordinate system is placed at the center of mass of the heated particle, we have

$$\int_0^\pi \xi \sin^2\Theta d\Theta = 0, \tag{45}$$

hence,

$$\xi_1 \equiv 0. \tag{46}$$

When solving the problem, we did not consider the boundary condition for the normal components of the stress tensor. Up to terms proportional to ε , the boundary condition for the normal stresses on the surface takes the form [17]

$$\sigma_n^{e(1)} - \sigma_n^{i(1)} = \sigma_0 H^{(1)} + 2\frac{\sigma^{(1)}}{R}. \tag{47}$$

Here,

$$2H = \frac{1}{R_1} + \frac{1}{R_2} = \frac{2}{R} + \varepsilon H^{(1)},$$

R_1 and R_2 are the principal radii of curvature of the drop; and H is the mean curvature of the surface, which in the axisymmetric case is given by [17]

$$H^{(1)} = -\frac{2}{R}\xi - \frac{1}{R\sin\Theta} \frac{\partial}{\partial\Theta} \left(\sin\Theta \frac{\partial\xi}{\partial\Theta} \right). \tag{48}$$

In view of (45) and (47), we obtain expression (49) in the form

$$H^{(1)} = \sum_{n=2}^{\infty} \frac{(n+2)(n-1)}{R} \xi_n P_n(\cos\Theta). \tag{49}$$

Thus, as follows from (48) with regard for (50), a nonuniformly heated drop, when moving, retains the spherical shape within the approximation adopted in this work.

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