

**THEORY OF THERMOPHORESIS OF MODERATELY
LARGE DROPS OF CONCENTRATED SOLUTIONS
IN BINARY GASEOUS MIXTURES**

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We have derived a formula for the rate of thermophoresis of a volatile drop of a concentrated solution in a binary gaseous mixture with allowance for known effects that are associated with all types of slipping of the gaseous mixture along the drop surface and jumps of temperature and concentrations. An analysis of this formula has been performed for a number of limiting cases.

A drop of a binary solution that consists of molecules of a solute and a volatile solvent substance is considered. Molecules of the volatile substance can evaporate or condense on the drop surface.

The drop is suspended in a two-component gaseous mixture with a temperature gradient $(\nabla T_e)_\infty$ prescribed at infinity. The first component of the gaseous mixture is composed of molecules of vapor of the solvent of the drop, while the molecules of the second component of the mixture do not undergo phase transitions on the drop surface.

By virtue of the smallness of the relative temperature differences in the vicinity of the drop, we will consider the transfer coefficients (the coefficients of viscosity, thermal conductivity, and diffusion) to be constant quantities.

The motion will be described within the framework of approximations of [1, 2]. The basic approximation is a condition for linearization of the equations of hydrodynamics, heat conduction, and diffusion that describe the distribution of velocities and concentrations outside and inside the particle. The nonlinear terms in these equations are much smaller the linear ones on condition that $R|(\nabla T_e)_\infty| \ll 1$. For actual aerosol systems this condition is realized with a large margin. For example, with radii of the aerosol particles in the range of $10^{-6} \text{ m} \ll R \ll 100 \cdot 10^{-6} \text{ m}$ and with temperatures of $T_e \approx 300 \text{ K}$, in order to violate the indicated condition, ∇T_e must vary in the range of $3 \cdot 10^6 \text{ K/m} \ll |(\Delta T_e)_\infty| \ll 3 \cdot 10^8 \text{ K/m}$. Under actual conditions such temperatures are impracticable. Therefore, linearization of the equations is legitimate. To solve the problem, we use a spherical coordinate system (r, Θ, φ) whose origin is rigidly tied to the center of the particle (see Fig. 1). The rate of thermophoresis is $\mathbf{U}_{th} = -\mathbf{U}$.

The distributions of the mass velocity \mathbf{v} , pressure p , and temperature T outside and inside the drop are described by the system of linearized equations [1, 2]

$$\begin{aligned} \eta_e \nabla^2 \mathbf{v}^{(e)} &= \nabla p^{(e)}, \quad \eta_i \nabla^2 \mathbf{v}^{(i)} = \nabla p^{(i)}; \\ \operatorname{div} \mathbf{v}^{(e)} &= 0, \quad \operatorname{div} \mathbf{v}^{(i)} = 0; \\ \nabla^2 T_e &= 0, \quad \nabla^2 T_i = 0; \\ \nabla^2 C_{1e} &= 0, \quad \nabla^2 C_{1i} = 0. \end{aligned} \tag{1}$$

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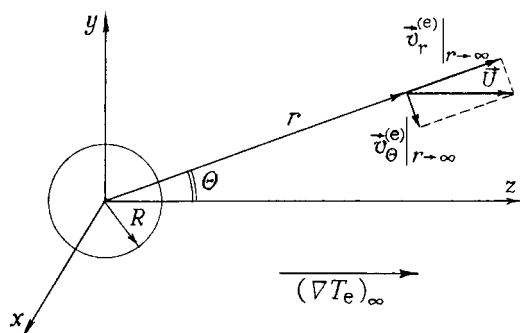


Fig. 1. Aerosol particle in a spherical coordinate system whose origin coincides with the center of the particle.

The index e refers to quantities that characterize the medium outside the drop, and the index i , inside the drop. In Eqs. (1) η_e and η_i are the coefficients of dynamic viscosity; $C_{1e} = n_{1e}/n_e$; $C_{2e} = n_{2e}/n_e$; $n_e = n_{1e} + n_{2e}$; $C_{1i} = m_1 n_{1i}/\rho_{0i}$; $C_{3i} = m_3 n_{3i}/\rho_{0i}$; $\rho_{0i} = m_1 n_{1i} + m_3 n_{3i}$; $C_{1e} + C_{2e} = 1$; $C_{1i} + C_{3i} = 1$.

Solutions of Eqs. (1) can be found with the following conditions at infinity:

a) the radial $v_r^{(e)}$ and tangential $v_\Theta^{(e)}$ components of the mass velocity at $r \rightarrow \infty$ are, respectively,

$$v_r^{(e)} = |\mathbf{U}| \cos \Theta, \quad v_\Theta^{(e)} = |\mathbf{U}| \sin \Theta; \quad (2)$$

b) the pressure at $r \rightarrow \infty$ is

$$p^{(e)} = p_0^{(e)}; \quad (3)$$

c) the temperature at $r \rightarrow \infty$ is

$$T_e = T_{0e} + |(\nabla T_e)_\infty| r \cos \Theta; \quad (4)$$

d) the relative concentration at $r \rightarrow \infty$ is

$$C_{1e} = C_{01e}. \quad (5)$$

On the surface of the spherical drop of radius R the following boundary conditions are fulfilled:

a) slipping of the gaseous mixture on the interface of drop-external medium [1]

$$\begin{aligned} (v_\Theta^{(e)} - v_\Theta^{(i)})|_{r=R} = c_m^* \left[r \frac{\partial}{\partial r} \left(\frac{v_\Theta^{(e)}}{r} \right) + \frac{1}{r} \frac{\partial v_r^{(e)}}{\partial \Theta} \right] \Big|_{r=R} + \\ + \frac{k_{Tsl}^*}{T_{0e} R} \left(1 + \frac{\sigma_T \beta_R^{*(T)}}{R} + \frac{\beta_R'^{*(T)}}{R} \right) \frac{\partial T_e}{\partial \Theta} \Big|_{r=R} + \frac{k_{Tsl}^* \beta_B^{*(T)}}{T_{0e} R} \left(\frac{\partial^2 T_e}{\partial r \partial \Theta} - \frac{1 \partial T_e}{r \partial \Theta} \right) \Big|_{r=R} + \\ + \frac{k_{Dsl} D_{12}^{(e)}}{R} \left(1 + \frac{\sigma_C \beta_R^{*(D)}}{R} + \frac{\beta_R'^{*(D)}}{R} \right) \frac{\partial C_{1e}}{\partial \Theta} \Big|_{r=R} + \\ + \frac{k_{Dsl} D_{12}^{(e)} \beta_B^{*(D)}}{R} \left(\frac{\partial^2 C_{1e}}{\partial r \partial \Theta} - \frac{1 \partial C_{1e}}{r \partial \Theta} \right) \Big|_{r=R}, \quad (6) \end{aligned}$$

in condition (6) the terms containing c_m^* , k_{Tsl}^* , and k_{Dsl} allow for isothermal, thermal, and diffusion slipping; the terms containing $\beta_R^{*(T)}$ and $\beta_R^{*(D)}$ allow for the influence of the curvature and deformation of the temperature

field; the terms containing $\beta_R^{*(T)}$ and $\beta_R^{*(D)}$ allow for the influence of just the curvature of the temperature field; the terms containing $\beta_B^{*(T)}$ and $\beta_B^{*(D)}$ allow for the influence of the Barnett effects on the mixture slipping; the coefficients σ_T and σ_C are determined from the formulas [3]

$$\sigma_T = \left(\frac{\partial^2 \ln T_e}{\partial r \partial \Theta} \right) \left(\frac{\partial \ln T_e}{r \partial \Theta} \right)^{-1}, \quad \sigma_C = \left(\frac{\partial^2 \ln C_{1e}}{\partial r \partial \Theta} \right) \left(\frac{\partial \ln C_{1e}}{r \partial \Theta} \right)^{-1};$$

b) continuity of the radial and tangential components of the total-stress tensor:

$$\left(-p^{(e)} + 2\eta_{0e} \frac{\partial v_r^{(e)}}{\partial r} \right) \Big|_{r=R} - \frac{2}{R} \frac{\partial \sigma}{\partial T_i} \Big|_{\substack{T_i=T_{0i} \\ C_{1i}=C_{01i}}} (T_i - T_{0i}) \Big|_{r=R} -$$

$$- \frac{2}{R} \frac{\partial \sigma}{\partial C_{1i}} \Big|_{\substack{T_i=T_{0i} \\ C_{1i}=C_{01i}}} (C_{1i} - C_{01i}) \Big|_{r=R} - \frac{2\sigma}{R} \Big|_{\substack{T_i=T_{0i} \\ C_{1i}=C_{01i}}} = \left(-p^{(i)} + 2\eta_{0i} \frac{\partial v_r^{(i)}}{\partial r} \right) \Big|_{r=R}, \quad (7)$$

$$\eta_{0e} \left(\frac{1}{r} \frac{\partial v_r^{(e)}}{\partial \Theta} + \frac{\partial v_\Theta^{(e)}}{\partial r} - \frac{v_\Theta^{(e)}}{r} \right) \Big|_{r=R} + \frac{1}{R} \frac{\partial \sigma}{\partial T_i} \Big|_{\substack{T_i=T_{0i} \\ C_{1i}=C_{01i}}} \frac{\partial T_i}{\partial \Theta} \Big|_{r=R} +$$

$$+ \frac{1}{R} \frac{\partial \sigma}{\partial C_{1i}} \Big|_{\substack{T_i=T_{0i} \\ C_{1i}=C_{01i}}} \frac{\partial C_{1i}}{\partial \Theta} \Big|_{r=R} = \eta_{0i} \left(\frac{1}{r} \frac{\partial v_r^{(i)}}{\partial \Theta} + \frac{\partial v_\Theta^{(i)}}{\partial r} - \frac{v_\Theta^{(i)}}{r} \right) \Big|_{r=R}; \quad (8)$$

in conditions (7) and (8) the terms containing $\partial\sigma/\partial C_{1i}$, $\partial\sigma/\partial T$, and σ/R take into account the influence of the surface tension on the particle motion;

c) continuity of the radial flux of the first (volatile) component at each point of the drop surface

$$n_{1e} v_r^{(e)} \Big|_{r=R} - \frac{D_{12}^{(e)} n_e^2 m_2}{\rho_{0e}} \frac{\partial C_{1e}}{\partial r} \Big|_{r=R} = n_{1i} v_r^{(i)} \Big|_{r=R} - \frac{D_{13}^{(i)} \rho_{0i}}{m_1} \frac{\partial C_{1i}}{\partial r} \Big|_{r=R}; \quad (9)$$

condition (9) is written without allowance for the volume thermal diffusion or spreading of the molecules of the first component of the gaseous mixture in the Knudsen layer;

d) impermeability of the drop surface to the second component of the external binary mixture

$$n_{2e} v_r^{(e)} \Big|_{r=R} + \frac{D_{12}^{(e)} n_e^2 m_1}{\rho_{0e}} \frac{\partial C_{1e}}{\partial r} \Big|_{r=R} = 0; \quad (10)$$

condition (10) is written without allowance for the volume diffusion or spreading of the molecules of the second component in the Knudsen layer;

e) impermeability of the drop surface to the substance dissolved in the drop

$$n_{3i} v_r^{(i)} \Big|_{r=R} + \frac{D_{13}^{(i)} \rho_{0i}}{m_3} \left(\frac{\partial C_{1i}}{\partial r} \right) \Big|_{r=R} = 0; \quad (11)$$

f) continuity of the radial heat flux through the drop surface

$$n_{3i} v_r^{(i)} \Big|_{r=R} + \frac{D_{13}^{(i)} \rho_{0i}}{m_3} \left(\frac{\partial C_{1i}}{\partial r} \right) \Big|_{r=R} = 0; \quad (11)$$

$$\left(-\chi_e \frac{\partial T_e}{\partial r} + \chi_i \frac{\partial T_i}{\partial r} \right) \Big|_{r=R} = \frac{Ln_e^2 m_1 m_2 D_{12}^{(e)}}{\rho_{0e}} \frac{\partial C_{1e}}{\partial r} \Big|_{r=R}; \quad (12)$$

condition (12) is written without allowance for spreading of the heat flux in the Knudsen layer;

g) the temperature jump of the first (volatile) component of the gaseous mixture in the Knudsen layer

$$(T_e - T_i) \Big|_{r=R} = k_T^{(T)} \frac{\partial T_e}{\partial r} \Big|_{r=R} + k_T^{(n)} T_{0e} \frac{\partial C_{1e}}{\partial r} \Big|_{r=R}, \quad (13)$$

where $k_T^{(T)}$ and $k_T^{(n)}$ are the coefficients of the temperature jump and the relative concentration;

h) the jump of the relative concentration of the first component of the gaseous mixture in the Knudsen layer

$$C_{1e} \Big|_{r=R} = \Phi(C_{1i}, T_i) \Big|_{\substack{T_i=T_{0i} \\ C_{1i}=C_{01i}}} + \frac{\partial \Phi}{\partial T_i} \Big|_{\substack{T_i=T_{0i} \\ C_{1i}=C_{01i}}} (T_i - T_{0i}) \Big|_{r=R} + \\ + \frac{\partial \Phi}{\partial C_{1i}} \Big|_{\substack{T_i=T_{0i} \\ C_{1i}=C_{01i}}} (C_{1i} - C_{01i}) \Big|_{r=R} + k_n^{(n)} \frac{\partial C_{1e}}{\partial r} \Big|_{r=R} + k_n^{(T)} \frac{1}{T_{0e}} \frac{\partial T_e}{\partial r} \Big|_{r=R}, \quad (14)$$

where $k_n^{(n)}$ and $k_n^{(T)}$ are the coefficients of the jump of the relative concentration and the temperature.

In the boundary conditions (6)-(14), $\sigma(C_{1i}, T_i)$ and $\Phi(C_{1i}, T_i)$ are the coefficient of surface tension and the relative concentration of saturated vapor of the volatile component, which depend on the values of C_{1i} and T_i at each point of the drop surface; $D_{12}^{(e)}$ and $D_{13}^{(i)}$ are the coefficients of interdiffusion of the gaseous mixture and the solution; c_m^* , $\beta_R^{*(T)}$, $\beta_R^{*(T)}$, $\beta_B^{*(T)}$, $\beta_B^{*(T)}$, $k_n^{(n)}$, $k_n^{(T)}$, $k_T^{(n)}$, and $k_T^{(T)}$ are coefficients for the two-component gaseous mixture with arbitrary ratios of the masses and concentrations of the components whose analytical form and computational procedure are presented in [1, 3-16].

All the boundary conditions at large distances from the surface and on the particle surface itself, with allowance for all effects that are associated with the presence of the Knudsen layer, in the case of thermophoresis of moderately large drops of concentrated solutions are given in the monograph [1].

Similar boundary conditions were used in [2] to construct a theory of thermophoresis of the same drops. However, the boundary conditions and the result for the rate are represented there in a form that is valid for the case where $C_{1e} \ll C_{2e}$. Therefore, in this work we use boundary conditions that correspond to [1], which gives a more general form for the rate of thermophoresis than in [2]. It should also be noted that in this work, as compared to [1], the dependence of the surface-tension coefficient on the concentration is taken into account in formulas (7) and (8).

Solutions of Eqs. (1) are sought in the form [1]

$$T_e = T_{0e} + \frac{\Phi_1}{r} + \frac{\mu_1}{r^2} \cos \Theta + |(\nabla T_e)_\infty| r \cos \Theta, \quad (15)$$

$$T_i = T_{0i} + \mu_2 r \cos \Theta, \quad (16)$$

$$C_{1e} = C_{01e} + \frac{\Phi_2}{r} + \frac{\mu_3}{r^2} \cos \Theta, \quad (17)$$

$$C_{1i} = C_{01i} + \mu_4 r \cos \Theta, \quad (18)$$

$$v_r^{(e)} = \left(\frac{A_e}{r^3} + \frac{B_e}{r} + |\mathbf{U}| \right) \cos \Theta + \frac{\gamma_e}{r}, \quad (19)$$

$$v_r^{(i)} = (Q_i + D_i r^2) \cos \Theta, \quad (20)$$

$$v_\Theta^{(e)} = \left(\frac{A_e}{2r^3} - \frac{B_e}{2r} - |\mathbf{U}| \right) \sin \Theta, \quad (21)$$

$$v_\Theta^{(i)} = -(Q_i + 2D_i r^2) \sin \Theta, \quad (22)$$

$$p^{(e)} = p_0^{(e)} + \eta_{0e} \frac{B_e}{r^2} \cos \Theta, \quad (23)$$

$$p^{(i)} = p_0^{(i)} + 10\eta_{0i} D_i r \cos \Theta. \quad (24)$$

By substituting functions (15)-(24) into the boundary conditions (6)-(14), it is possible to find the quantities Φ_1 , Φ_2 , μ_1 , μ_2 , μ_3 , μ_4 , A_e , B_e , γ_e , Q_i , D_i , and $|\mathbf{U}|$.

The objective of the investigations carried out is to determine the rate $\mathbf{U}_{th} = -\mathbf{U}$; therefore, without giving unwieldy expressions for the remaining enumerated quantities, for \mathbf{U}_{th} we write

$$\mathbf{U}_{th} = - \frac{2k_{Tsl}^* (\nabla T_e)_\infty}{\left(\frac{2}{3} \frac{\eta_{0e}}{\eta_{0i}} + 1 + 2 \frac{c_m}{R} \right) T_{0e} g} F_{res}^{(th)}, \quad (25)$$

where

$$\begin{aligned} F_{res}^{(th)} &= F_{Tsl} + F_{\sigma T} + F_{\sigma C} + F_{Dsl} + F_{react}; \\ F_{Tsl} &= \left(1 + \frac{\beta_R^{*(T)}}{R} \right) f_1 + \frac{\beta_R^{*(T)}}{R} f_2 + \frac{\beta_B^{*(T)}}{R} f_3; \\ F_{\sigma T} &= \frac{T_{0e} R}{3k_{Tsl}^* \eta_{0i}} \frac{\partial \sigma}{\partial T_i} f_4; \quad F_{\sigma C} = \frac{T_{0e} R}{3k_{Tsl}^* \eta_{0i}} \frac{\partial \sigma}{\partial C_{1i}} f_5; \\ F_{Dsl} &= \frac{T_{0e} k_{Dsl} D_{12}^{(e)}}{k_{Tsl}^*} \left(1 + \frac{\beta_R^{*(D)}}{R} - \frac{2\beta_R^{*(D)}}{R} - \frac{3\beta_B^{*(D)}}{R} \right) f_6; \end{aligned}$$

$$\begin{aligned}
F_{\text{react}} &= \frac{T_{0e} D_{12}^{(e)} n_e^2}{k_{\text{Tsl}}^* \rho_{0e}} \left[\left(1 + \frac{2\eta_{0e}}{\eta_{0i}} + \frac{2\rho_{0e}}{\rho_{0i}} + \frac{6c_m^*}{R} \right) \frac{m_1}{n_{2e}} - \left(1 + \frac{2\eta_{0e}}{\eta_{0i}} + \frac{6c_m^*}{R} \right) \frac{m_2}{n_{1e}} \right] f_6; \\
f_1 &= \left(1 + \frac{2k_n^{(n)}}{R} \right) \left(\frac{k_T^{(T)}}{R} + \frac{\chi_e}{\chi_i} \right) - \frac{2k_n^{(T)}}{T_{0e} R} \left(\frac{Lm_1 m_2 n_e^2 D_{12}^{(e)}}{\rho_{0e} \chi_i} + \frac{T_{0e} k_T^{(n)}}{R} \right) + \\
&\quad + \frac{2\partial\Phi}{\partial T_i} \left(\frac{Lm_1 m_2 n_e^2 D_{12}^{(e)} k_T^{(T)}}{\rho_{0e} \chi_i R} - \frac{T_{0e} k_T^{(n)} \chi_e}{R \chi_i} \right) + \\
&\quad + \frac{2\partial\Phi}{\partial C_{li}} \frac{n_e^2 \rho_{1e} \rho_{3i} D_{12}^{(e)}}{n_{1e} n_{2e} \rho_{0i}^2 D_{13}^{(i)}} \left(\frac{k_T^{(T)}}{R} + \frac{\chi_e}{\chi_i} \right); \\
f_2 &= 1 + \frac{2k_n^{(n)}}{R} + \frac{2\partial\Phi}{\partial C_{li}} \frac{D_{12}^{(e)} n_e^2 \rho_{1e} \rho_{3i}}{n_{2e} n_{1e} \rho_{0i}^2 D_{13}^{(i)}} + \frac{2\partial\Phi}{\partial T_i} \frac{Lm_1 m_2 n_e^2 D_{12}^{(e)}}{\rho_{0e} \chi_i}; \\
f_3 &= \left(1 + \frac{2k_n^{(n)}}{R} \right) \left(1 - \frac{k_T^{(T)}}{R} - \frac{\chi_e}{\chi_i} \right) + \frac{2k_n^{(T)}}{T_{0e} R} \left(\frac{Lm_1 m_2 n_e^2 D_{12}^{(e)}}{\rho_{0e} \chi_i} + \frac{T_{0e} k_T^{(n)}}{R} \right) - \\
&\quad - \frac{2\partial\Phi}{\partial T_i} \left[\left(-1 + \frac{k_T^{(T)}}{R} \right) \frac{Lm_1 m_2 n_e^2 D_{12}^{(e)}}{\rho_{0e} \chi_i} - \frac{T_{0e} k_T^{(n)} \chi_e}{R \chi_i} \right] + \\
&\quad + \frac{2\partial\Phi}{\partial C_{li}} \frac{n_e^2 \rho_{1e} \rho_{3i} D_{12}^{(e)}}{n_{1e} n_{2e} \rho_{0i}^2 D_{13}^{(i)}} \left(1 - \frac{k_T^{(T)}}{R} - \frac{\chi_e}{\chi_i} \right); \\
f_4 &= \left(1 + \frac{2k_n^{(n)}}{R} \right) \frac{\chi_e}{\chi_i} - \frac{2D_{12}^{(e)} Lm_1 m_2 n_e^2 k_n^{(T)}}{RT_{0e} \rho_{0e} \chi_i} + \frac{2\partial\Phi}{\partial C_{li}} \frac{n_e^2 \rho_{1e} \rho_{3i} D_{12}^{(e)} \chi_e}{n_{1e} n_{2e} \rho_{0i}^2 D_{13}^{(i)} \chi_i}; \\
f_5 &= - \frac{2n_e^2 \rho_{1e} \rho_{3i} D_{12}^{(e)}}{n_{1e} n_{2e} \rho_{0i}^2 D_{13}^{(i)}} \left(\frac{k_n^{(T)}}{T_{0e} R} + \frac{\partial\Phi}{\partial T_i} \frac{\chi_e}{\chi_i} \right); \quad f_6 = \frac{k_n^{(T)}}{T_{0e} R} + \frac{\partial\Phi}{\partial T_i} \frac{\chi_e}{\chi_i}.
\end{aligned}$$

In formula (25) we introduce the notation

$$\begin{aligned}
g &= 1 + \frac{2k_n^{(n)}}{R} + \frac{2k_T^{(T)}}{R} + \frac{4k_n^{(n)} k_T^{(T)}}{R^2} + \frac{2\chi_e}{\chi_i} + \frac{4k_n^{(n)} \chi_e}{R \chi_i} - \\
&\quad - \frac{4k_n^{(T)}}{R} \frac{Lm_1 m_2 n_e^2 D_{12}^{(e)}}{T_{0e} \rho_{0e} \chi_i} - \frac{4k_T^{(n)} k_n^{(T)}}{R^2} + \frac{2\partial\Phi}{\partial T_i} \frac{Lm_1 m_2 n_e^2 D_{12}^{(e)}}{\rho_{0e} \chi_i} + \\
&\quad + \frac{4\partial\Phi}{\partial T_i} \frac{Lm_1 m_2 n_e^2 D_{12}^{(e)} k_T^{(T)}}{\rho_{0e} \chi_i R} - \frac{4\partial\Phi}{\partial T_i} \frac{\chi_e T_{0e} k_T^{(n)}}{\chi_i R} + \\
&\quad + \frac{2\partial\Phi}{\partial C_{li}} \frac{D_{12}^{(e)} n_e^2 m_1 m_3 n_{3i}}{n_{2e} D_{13}^{(i)} \rho_{0i}^2} + \frac{4\partial\Phi}{\partial C_{li}} \frac{D_{12}^{(e)} k_T^{(T)} n_e^2 m_1 m_3 n_{3i}}{D_{13}^{(i)} R n_{2e} \rho_{0i}^2}
\end{aligned}$$

$$+ \frac{4\partial\Phi}{\partial C_{li}} \frac{D_{12}^{(e)} n_e^2 \chi_e m_1 m_3 n_{3i}}{D_{13}^{(i)} n_{2e} \chi_i \rho_{0i}^2}.$$

Formula (25) provides the possibility of determining the rate of thermophoresis of the solution drop for an arbitrary concentration C_{3i} of the dissolved substance. When $C_{3i} \rightarrow 0$, this formula becomes an expression for the rate of thermophoresis of moderately large volatile drops of a pure substance:

$$\mathbf{U}_{th}^{(0)} = - \frac{2k_{Tsl}^* (\nabla T_e)_\infty}{\left(\frac{2}{3} \frac{\eta_{0e}}{\eta_{0i}} + 1 + 2 \frac{c_m^*}{R} \right) T_{0e} g^{(0)}} F_{res}^{(0)}, \quad (26)$$

where

$$F_{res}^{(0)} = F_{Tsl}^{(0)} + F_{\sigma T}^{(0)} + F_{Dsl}^{(0)} + F_{react}^{(0)};$$

$$F_{Tsl}^{(0)} = \left(1 + \frac{\beta_R'^{(T)}}{R} \right) f_1^{(0)} + \frac{\beta_R^{*(T)}}{R} f_2^{(0)} + \frac{\beta_B^{*(T)}}{R} f_3^{(0)};$$

$$F_{\sigma T}^{(0)} = \frac{T_{0e} R}{3k_{Tsl}^* \eta_{0i}} \frac{\partial \sigma}{\partial T_i} f_4^{(0)};$$

$$F_{Dsl}^{(0)} = \frac{T_{0e} k_{Dsl} D_{12}^{(e)}}{k_{Tsl}^*} \left(1 + \frac{\beta_R'^{(D)}}{R} - \frac{2\beta_R^{*(D)}}{R} - \frac{3\beta_B^{*(D)}}{R} \right) f_6^{(0)};$$

$$F_{react}^{(0)} = \frac{T_{0e} D_{12}^{(e)} n_e^2}{k_{Tsl}^* \rho_{0e}} \left[\left(1 + \frac{2\eta_{0e}}{\eta_{0i}} + \frac{2\rho_{0e}}{\rho_{0i}} + \frac{6c_m^*}{R} \right) \frac{m_1}{n_{2e}} - \left(1 + \frac{2\eta_{0e}}{\eta_{0i}} + \frac{6c_m^*}{R} \right) \frac{m_2}{n_{1e}} \right] f_6^{(0)};$$

$$f_1^{(0)} = \left(1 + \frac{2k_n^{(n)}}{R} \right) \left(\frac{k_T^{(T)}}{R} + \frac{\chi_e}{\chi_i} \right) - \frac{2k_n^{(T)}}{T_{0e} R} \left(\frac{Lm_1 m_2 n_e^2 D_{12}^{(e)}}{\rho_{0e} \chi_i} + \frac{T_{0e} k_T^{(n)}}{R} \right) + \frac{2\partial\Phi}{\partial T_i} \left(\frac{Lm_1 m_2 n_e^2 D_{12}^{(e)} k_T^{(T)}}{\rho_{0e} \chi_i R} - \frac{T_{0e} k_T^{(n)} \chi_e}{R \chi_i} \right);$$

$$f_2^{(0)} = 1 + \frac{2k_n^{(n)}}{R} + \frac{2\partial\Phi}{\partial T_i} \frac{Lm_1 m_2 n_e^2 D_{12}^{(e)}}{\rho_{0e} \chi_i};$$

$$f_3^{(0)} = \left(1 + \frac{2k_n^{(n)}}{R} \right) \left(1 - \frac{k_T^{(T)}}{R} - \frac{\chi_e}{\chi_i} \right) + \frac{2k_n^{(T)}}{T_{0e} R} \left(\frac{Lm_1 m_2 n_e^2 D_{12}^{(e)}}{\rho_{0e} \chi_i} + \frac{T_{0e} k_T^{(n)}}{R} \right) - \frac{2\partial\Phi}{\partial T_i} \left[\left(-1 + \frac{k_T^{(T)}}{R} \right) \frac{Lm_1 m_2 n_e^2 D_{12}^{(e)}}{\rho_{0e} \chi_i} - \frac{T_{0e} k_T^{(n)} \chi_e}{R \chi_i} \right];$$

$$\begin{aligned}
f_4^{(0)} &= \left(1 + \frac{2k_n^{(n)}}{R}\right) \frac{\chi_e}{\chi_i} - \frac{2D_{12}^{(e)} L m_1 m_2 n_e^2 k_n^{(T)}}{T_{0e} R \rho_{0e} \chi_i}; \\
f_6^{(0)} &= \frac{k_n^{(T)}}{T_{0e} R} + \frac{\partial \Phi}{\partial T_i} \frac{\chi_e}{\chi_i}; \\
g^{(0)} &= 1 + \frac{2k_n^{(n)}}{R} + \frac{2k_T^{(T)}}{R} + \frac{4k_n^{(n)} k_T^{(T)}}{R^2} + \frac{2\chi_e}{\chi_i} + \frac{4k_n^{(n)}}{R} \frac{\chi_e}{\chi_i} - \\
&\quad - \frac{4k_n^{(T)} L m_1 m_2 n_e^2 D_{12}^{(e)}}{R T_{0e} \rho_{0e} \chi_i} - \frac{4k_T^{(n)} k_n^{(T)}}{R^2} + \frac{2\partial \Phi}{\partial T_i} \frac{L m_1 m_2 n_e^2 D_{12}^{(e)}}{\rho_{0e} \chi_i} + \\
&\quad + \frac{4\partial \Phi}{\partial T_i} \frac{L m_1 m_2 n_e^2 D_{12}^{(e)} k_T^{(T)}}{\rho_{0e} \chi_i R} - \frac{4\partial \Phi}{\partial T_i} \frac{\chi_e T_{0e} k_T^{(n)}}{\chi_i R}.
\end{aligned}$$

In the case $C_{3i} \rightarrow 1$, from Eq. (25) we can obtain the thermophoresis rate of a nonvolatile liquid particle $\mathbf{U}_{th}^{(liq)}$:

$$\mathbf{U}_{th}^{(liq)} = - \frac{2k_{Tsl}^* (\nabla T_e)_\infty}{\left(\frac{2}{3} \frac{\eta_{0e}}{\eta_{0i}} + 1 + 2 \frac{c_m^*}{R}\right) T_{0e} g^{(liq)}} F_{res}^{(liq)}, \quad (27)$$

where

$$\begin{aligned}
F_{res}^{(liq)} &= F_{Tsl}^{(liq)} + F_{\sigma T}^{(liq)}; \\
F_{Tsl}^{(liq)} &= \left(1 + \frac{\beta_R^{*(T)}}{R}\right) f_1^{(liq)} + \frac{\beta_R^{*(T)}}{R} f_2^{(liq)} + \frac{\beta_B^{*(T)}}{R} f_3^{(liq)}; \\
F_{\sigma T}^{(liq)} &= \frac{T_{0e} R}{3k_{Tsl}^* \eta_{0i}} \frac{\partial \sigma}{\partial T_i} f_4^{(liq)}; \\
f_1^{(liq)} &= \frac{k_T^{(T)}}{R} + \frac{\chi_e}{\chi_i}; \quad f_2^{(liq)} = 1; \\
f_3^{(liq)} &= 1 - \frac{k_T^{(T)}}{R} - \frac{\chi_e}{\chi_i}; \quad f_4^{(liq)} = \frac{\chi_e}{\chi_i}; \\
g^{(liq)} &= 1 + \frac{2\chi_e}{\chi_i} + \frac{2k_T^{(T)}}{R}.
\end{aligned}$$

For $C_{3i} \rightarrow 1$, $\eta_{0e}/\eta_{0i} \rightarrow 0$, and $\rho_{0e}/\rho_{0i} \rightarrow 0$, from Eq. (25) we can obtain a formula for the thermophoresis of a solid nonvolatile particle:

$$\mathbf{U}_{th}^{(sol)} = - \frac{2k_{Tsl}^* (\nabla T_e)_\infty}{\left(1 + 2 \frac{c_m^*}{R}\right) T_{0e} g^{(sol)}} F_{res}^{(sol)}, \quad (28)$$

where

$$F_{\text{res}}^{(\text{sol})} = F_{\text{Tsl}}^{(\text{sol})} ;$$

$$F_{\text{Tsl}}^{(\text{sol})} = \left(1 + \frac{\beta_R^{*(T)}}{R} \right) f_1^{(\text{sol})} + \frac{\beta_R^{*(T)}}{R} f_2^{(\text{sol})} + \frac{\beta_B^{*(T)}}{R} f_3^{(\text{sol})} ;$$

$$f_1^{(\text{sol})} = \frac{k_T^{(T)}}{R} + \frac{\chi_e}{\chi_i} ; f_2^{(\text{sol})} = 1 ; f_3^{(\text{sol})} = 1 - \frac{k_T^{(T)}}{R} - \frac{\chi_e}{\chi_i} ; g^{(\text{sol})} = g^{(\text{liq})} .$$

In the case of a large volatile drop the formula for the rate of thermophoresis takes the form

$$\mathbf{U}_{\text{th}}^{(\text{large})} = - \frac{2k_{\text{Tsl}}^* (\nabla T_e)_\infty}{\left(1 + \frac{2\eta_{0e}}{3\eta_{0i}} \right) T_{0e} g^{(\text{large})}} F_{\text{res}}^{(\text{large})} , \quad (29)$$

where

$$F_{\text{res}}^{(\text{large})} = F_{\text{Tsl}}^{(\text{large})} + F_{\sigma T}^{(\text{large})} + F_{\sigma C}^{(\text{large})} + F_{\text{Dsl}}^{(\text{large})} + F_{\text{react}}^{(\text{large})} ;$$

$$F_{\text{Tsl}}^{(\text{large})} = f_1^{(\text{large})} ; F_{\sigma T}^{(\text{large})} = \frac{T_{0e} R}{3k_{\text{Tsl}}^* \eta_{0i}} \frac{\partial \sigma}{\partial T_i} f_4^{(\text{large})} ;$$

$$F_{\sigma C}^{(\text{large})} = \frac{T_{0e} R}{3k_{\text{Tsl}}^* \eta_{0i}} \frac{\partial \sigma}{\partial C_{1i}} f_5^{(\text{large})} ; F_{\text{Dsl}}^{(\text{large})} = \frac{T_{0e} k_{\text{Dsl}} D_{12}^{(e)}}{k_{\text{Tsl}}^*} f_6^{(\text{large})} ;$$

$$F_{\text{react}}^{(\text{large})} = \frac{T_{0e} D_{12}^{(e)} n_e^2}{k_{\text{Tsl}}^* \rho_{0e}} \left[\left(1 + \frac{2\eta_{0e}}{\eta_{0i}} + \frac{2\rho_{0e}}{\rho_{0i}} \right) \frac{m_1}{n_{2e}} - \left(1 + \frac{2\eta_{0e}}{\eta_{0i}} \right) \frac{m_2}{n_{1e}} \right] f_6^{(\text{large})} ;$$

$$f_1^{(\text{large})} = f_4^{(\text{large})} = \frac{\chi_e}{\chi_i} \left\{ \frac{2\partial \Phi}{\partial C_{1i}} \frac{n_e^2 \rho_{1e} \rho_{3i} D_{12}^{(e)}}{n_{1e} n_{2e} \rho_{0i}^2 D_{13}^{(i)}} + 1 \right\} ;$$

$$f_5^{(\text{large})} = \frac{n_e^2 \rho_{1e} \rho_{3i}}{n_{1e} n_{2e} \rho_{0i}^2 D_{13}^{(i)}} \left[-2D_{12}^{(e)} \frac{\partial \Phi}{\partial T_i} \frac{\chi_e}{\chi_i} \right] ; f_6^{(\text{large})} = \frac{\partial \Phi}{\partial T_i} \frac{\chi_e}{\chi_i} ;$$

$$g^{(\text{large})} = 1 + \frac{2\chi_e}{\chi_i} + \frac{4\partial \Phi}{\partial C_{1i}} \frac{D_{12}^{(e)} m_1 m_3 n_{3i} n_e^2 \chi_e}{n_{2e} D_{13}^{(i)} \rho_{0i}^2 \chi_i} +$$

$$+ \frac{2\partial \Phi}{\partial C_{1i}} \frac{D_{12}^{(e)} n_e^2 m_1 m_3 n_{3i}}{n_{2e} D_{13}^{(i)} \rho_{0i}^2} + \frac{2\partial \Phi}{\partial T_i} \frac{L m_1 m_2 n_e^2 D_{12}^{(e)}}{\rho_{0e} \chi_i} .$$

In the case of a large solid nonvolatile particle, when $\eta_{0e}/\eta_{0i} \rightarrow 0$ and $\rho_{0e}/\rho_{0i} \rightarrow 0$, formula (25) converts to

$$\mathbf{U}_{\text{th}}^{(\text{large sol})} = - \frac{2k_{\text{Tsl}}^* (\nabla T_e)_{\infty}}{T_{0e} g^{(\text{large sol})}} F_{\text{res}}^{(\text{large sol})}; \quad (30)$$

where

$$F_{\text{res}}^{(\text{large sol})} = F_{\text{Tsl}}^{(\text{large sol})}; \quad F_{\text{Tsl}}^{(\text{large sol})} = f_1^{(\text{large sol})};$$

$$f_1^{(\text{large sol})} = \frac{\chi_e}{\chi_i}; \quad g^{(\text{large sol})} = 1 + \frac{2\chi_e}{\chi_i}.$$

An analysis of the results obtained for moderately large particles showed that the total velocity of thermophoretic motion \mathbf{U}_{th} is determined by the combined influence of the separate effects: the thermal ($\sim F_{\text{Tsl}}$) and diffusion ($\sim F_{\text{Dsl}}$) slippings, the variable surface tension ($\sim F_{\sigma T}$, $\sim F_{\sigma C}$), and the reactive effect ($\sim F_{\text{react}}$) from the uncompensated phase transition of the volatile component, and since $F_{\text{Tsl}} > 0$, the effect of the thermal slipping causes the drop to move toward a decrease in the temperature.

In view of the fact that $\partial\sigma/\partial T_i < 0$, we have $F_{\sigma T} < 0$ and the effect that is associated with the dependence of the interphase surface tension on the temperature causes the particle to move toward an increase in the temperature.

The sign of the quantity $F_{\sigma C}$ depends on the sign of $\partial\sigma/\partial C_{li}$. If $\partial\sigma/\partial C_{li} > 0$, then $F_{\sigma C} < 0$, i.e., the particle motion occurs toward an increase in the temperature. But if $\partial\sigma/\partial C_{li} < 0$, then $F_{\sigma C} > 0$, and the particle moves toward a decrease in the temperature.

The sign of F_{Dsl} is determined by the sign of k_{Dsl} . It is known [1] that $k_{\text{Dsl}} \sim (m_2 - m_1)$. If $m_2 > m_1$, then $k_{\text{Dsl}} > 0$ and $F_{\text{Dsl}} > 0$, and, consequently, the effect of the diffusion slipping causes the particle to move in a direction opposite to the direction of the external temperature gradient $(\nabla T_e)_{\infty}$. But if $m_1 > m_2$, then $F_{\text{Dsl}} < 0$ and the particle moves in the direction of the external temperature gradient $(\nabla T_e)_{\infty}$. For example, in the case of evaporation of a water drop in a vapor-air mixture the molar mass of water is $\mu_1 = 18 \cdot 10^{-3}$ kg/mole, the molar mass of air is $\mu_2 = 29 \cdot 10^{-3}$ kg/mole, $m_1 = \mu_1/N_A$, $m_2 = \mu_2/N_A$, $(m_2 - m_1) > 0$, and in this case the particle moves toward a decrease in the temperature.

The sign of F_{react} is determined by the difference

$$\left(1 + \frac{2\eta_{0e}}{\eta_{0i}} + \frac{2\rho_{0e}}{\rho_{0i}} + \frac{6c_m^*}{R} \right) \frac{m_1}{n_{2e}} - \left(1 + \frac{2\eta_{0e}}{\eta_{0i}} + \frac{6c_m^*}{R} \right) \frac{m_2}{n_{1e}},$$

whose sign depends on m_1 , m_2 , n_{1e} , and n_{2e} . For example, in the case of motion of a drop of an aqueous solution in a vapor-air mixture, $m_1/n_{2e} < m_2/n_{1e}$, since under normal conditions $m_1 < m_2$ and $n_{2e} \gg n_{1e}$. Here $F_{\text{react}} < 0$ and the particle moves toward an increase in the temperature.

From the aforesaid it follows that the general direction of particle motion in a two-component gaseous mixture depends on the relation of the numerical values of F_{Tsl} , $F_{\sigma T}$, $F_{\sigma C}$, F_{Dsl} , and F_{react} . Specific results can be obtained by performing calculations for selected mixtures. Evaluations show that the main contribution to particle motion is made by capillary effects that are associated with the dependence of the surface tension on the temperature ($\sim F_{\sigma T}$) and the concentration of the substance ($\sim F_{\sigma C}$) dissolved in the drop.

NOTATION

\mathbf{U}_{th} , rate of thermophoresis, m/sec; \mathbf{U} , velocity of motion of the mixture relative to the drop, m/sec; $(\nabla T_e)_{\infty}$, constant temperature gradient of the external gaseous mixture at a large distance from the particle (the gradient is prescribed at infinity), K/m; $\mathbf{v}_r^{(e)}$ and $\mathbf{v}_{\Theta}^{(e)}$, radial and tangential components of the mass velocity, m/sec; r , distance from the center of the particle to the point of the medium, m; R , radius of the aerosol particle, m; (r, Θ, φ) , spherical coordinate system, m, rad, rad; η_{0e} and η_{0i} , mean value of the viscosities of the

external gaseous mixture and inside the drop, N·sec/m²; ρ_{0e} and ρ_{0i} , density of the external gaseous mixture and the solution drop, kg/m³; $\mathbf{v}^{(e)}$ and $\mathbf{v}^{(i)}$, velocities of the centers of inertia of the mixture outside and inside the drop, m/sec; $p^{(e)}$ and $p^{(i)}$, pressures outside and inside the drop, Pa; $p_0^{(e)}$ and $p_0^{(i)}$, the same, the mean values, Pa; χ_e and χ_i , thermal-conductivity coefficients of the external medium and the drop, W/(K·m); T_e and T_i , temperatures of the external medium and the drop, K; T_{0e} and T_{0i} , the same, the mean values, K; C_{1e} and C_{1i} , dimensionless relative concentrations of the first component of the gaseous mixture (solvent) outside and inside the drop; C_{01e} and C_{01i} , the same, the mean values; C_{2e} , dimensionless relative concentration of the second component of the gaseous mixture; C_{3i} , the same, of the solute in the drop (dimensionless); m_1 , m_2 , and m_3 , masses of the molecules of the first component (solvent), the second component of the gaseous mixture, and the solute in the drop, kg; n_{1e} and n_{2e} , concentrations of the molecules of the first and second components of the external gaseous mixture, 1/m³; n_{1i} and n_{3i} , concentrations of the molecules of the volatile substance and the solute in the drop, 1/m³; L , specific heat of the phase transition, J/kg; $N_A = 6.02 \cdot 10^{23}$ mole⁻¹, Avogadro constant; Φ_1 , Φ_2 , μ_1 , μ_2 , μ_3 , μ_4 , A_e , B_e , γ_e , Q_i , and D_i , dimensional coefficients that depend on the parameters of the mixture and the drop; F_{res} , F_{Tsl} , F_{Dsl} , $F_{\sigma T}$, $F_{\sigma C}$, and F_{react} , coefficients of the temperature gradient that compose the expression for the rate; σ_T and σ_C , dimensionless coefficients that depend on the parameters of the mixture and the drop.

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