

THERMOPHORESIS OF LARGE VOLATILE ONE-COMPONENT DROPS

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The work is devoted to the theory of motion of aerosol particles in temperature-inhomogeneous gases. An investigation is made of the influence of the Dufour effect on the thermophoresis rate of large volatile one-component drops. Estimates of the resultant formula show that this effect exerts an insignificant influence on the velocity of motion of the drops under actual conditions.

The theory of thermophoresis of drops in the presence of a phase transition has been actively developed in a series of investigations [1-10]. Three candidate's dissertations on the indicated subject have been defended [11-13] in which possible variants of the theory were analyzed in detail: both with and without allowance for the influence of internal flows on the thermophoresis rate of drops and also with a nonvolatile substance present in the drop whose relative concentration varies from 0 to 1. A drop in which the concentration of the dissolved substance is equal to zero is considered to be pure. As the concentration of the nonvolatile substance approaches unity, the drop automatically passes to the category of nonvolatile drops.

Ten years after publication of the monograph [10], in which different variants of the theory of motion of volatile particles are investigated, two works [14, 15] appeared on the same problem of thermophoresis of volatile spherical aerosol particles, the solution of which repeats, in fact, the results of the above works without sufficient reference to the original works.

Below we provide a more in-depth analysis of the situation described. In the work, an investigation is made of the thermophoresis of large volatile drops without a dissolved substance in the latter. Precisely this particular case is considered in [14, 15].

Formulation of the Problem. We present rather fully the essence of the problem of the thermophoresis of a large volatile one-component drop, which was suggested and used for the first time in [1-13]. We assume that a spherical drop of radius R is suspended in a viscous binary mixture that is inhomogeneous with respect to the temperature. On the drop surface, one component of the external mixture undergoes a phase change. The drop radius is substantially larger than both mean free paths of the molecules of the mixture components. Therefore corrections to the main effects proportional to the Knudsen number will not be taken into account. We consider the case where the external binary mixture is a gas with comparable concentrations of the components. Here, thermodiffusion effects can play a role. The external medium will be characterized by the mean viscosity η_{0e} , density ρ_{0e} , and temperature T_{0e} at a large distance from the drop. We assume that the mean distance between the drops is substantially larger than their radius R . By virtue of this the interaction between the drops, in the context of their mutual diffusional-hydrodynamic influence, can be neglected. Since the drop represents a sphere, it is convenient to carry out calculations in a spherical coordinate system. We choose the origin of the spherical coordinates r, θ, φ at the center of the particle. We assume that at a relatively large distance from the particle there is a constant temperature gradient $(\nabla T_e)_\infty$ in the volume of the external mixture and that it is directed along the polar axis $z = r \cos \theta$. With this choice of the position of the origin of coordinates it is convenient to consider the particle to be at rest, while the center of gravity of the gas mixture moves relative to the particle with the velocity \vec{U} at $r \rightarrow \infty$.

The distribution of the velocities, pressures, temperatures, and concentrations outside and inside each individual drop satisfies the following system of linearized differential equations:

$$\eta_e \nabla^2 \vec{V}^{(e)} = \nabla p^{(e)}, \quad (1)$$

$$\operatorname{div} \vec{V}^{(e)} = 0, \quad (2)$$

$$\nabla^2 T_e = 0, \quad (3)$$

$$\eta_i \nabla^2 \vec{V}^{(i)} = \nabla p^{(i)}, \quad (4)$$

$$\operatorname{div} \vec{V}^{(i)} = 0, \quad (5)$$

$$\nabla^2 T_i = 0, \quad (6)$$

$$\nabla^2 C_{1e} = 0. \quad (7)$$

Boundary Conditions. Now we will pass to formulation of the boundary conditions at infinity and on the drop surface. For those for the radial $V_r^{(e)}$ and tangential $V_\theta^{(e)}$ velocity components we can write for $r \rightarrow \infty$:

$$V_r^{(e)} = |\vec{U}| \cos \theta, \quad (8)$$

$$V_\theta^{(e)} = -|\vec{U}| \sin \theta. \quad (9)$$

Next, for $p^{(e)}$ and T_e , the relations

$$p^{(e)} = p_0^{(e)}, \quad (10)$$

$$T_e = T_{0e} + |(\nabla T_e)_\infty| r \cos \theta \quad (11)$$

hold for $r \rightarrow \infty$.

The drop surface is impermeable to the radial flow of the second component of the external binary mixture. This condition can be represented in the form

$$\left(n_{02e} V_r^{(e)} + D_{12}^{(e)} \beta_1 \frac{\partial C_{1e}}{\partial r} + \beta_1 D_{12}^{(e)} \frac{K_{\text{th.dif}}^{(e)}}{T_{0e}} \frac{\partial T_e}{\partial r} \right) \Big|_{r=R} = 0. \quad (12)$$

On the drop surface, the radial flow of the first component, undergoing a phase transition, is continuous:

$$\left(n_{01e} V_r^{(e)} - D_{12}^{(e)} \beta_2 \frac{\partial C_{1e}}{\partial r} - \beta_2 D_{12}^{(e)} \frac{K_{\text{th.dif}}^{(e)}}{T_{0e}} \frac{\partial T_e}{\partial r} \right) \Big|_{r=R} = (n_{01i} V_r^{(i)}) \Big|_{r=R}. \quad (13)$$

Here

$$\beta_1 = n_{0e}^2 \frac{m_1}{\rho_{0e}}, \quad \beta_2 = n_{0e}^2 \frac{m_2}{\rho_{0e}}, \quad n_{0e} = n_{01e} + n_{02e}. \quad (14)$$

The difference of the tangential components of the external and internal velocities on the drop surface satisfies the slip condition

$$(V_\theta^{(e)} - V_\theta^{(i)})|_{r=R} = \frac{K_{th,s} \eta_{0e}}{T_{0e} R \rho_{0e}} \frac{\partial T_e}{\partial \theta} \Big|_{r=R} + \frac{K_{dif,s}}{R} D_{12}^{(e)} \frac{\partial C_{1e}}{\partial \theta} \Big|_{r=R}. \quad (15)$$

The following two boundary conditions express, respectively, the continuity conditions of the temperature and the heat flux:

$$T_e|_{r=R} = T_i|_{r=R}, \quad (16)$$

$$\begin{aligned} & \left(-\chi_e \frac{\partial T_e}{\partial r} + \chi_i \frac{\partial T_i}{\partial r} - Lm_1 \beta_2 D_{12}^{(e)} \left(\frac{\partial C_{1e}}{\partial r} + \frac{K_{th,dif}^{(e)}}{T_{0e}} \frac{\partial T_e}{\partial r} \right) - \right. \\ & \left. - \frac{K_{th,dif}^{(e)} D_{12}^{(e)}}{C_{sat} (1 - C_{sat})} p \frac{\partial C_{1e}}{\partial r} \right) \Big|_{r=R} = 0. \end{aligned} \quad (17)$$

Allowance for the Dufour effect results in the appearance of an additional component in the boundary condition for continuity of the heat flux. This effect is contained in the form of the fourth term of formula (17).

The boundary conditions for continuity of the radial and tangential components of the stresses on the drop surface are of the form

$$\begin{aligned} & \left(-p^{(e)} + 2\eta_{0e} \frac{\partial V_r^{(e)}}{\partial r} \right) \Big|_{r=R} - \frac{2\sigma_0}{R} - \frac{2}{R} \frac{\partial \sigma_0}{\partial T_e} \Big|_{T_e=T_{e0}} (T_e - T_{0e})|_{r=R} = \\ & = \left(-p^{(i)} + 2\eta_{0i} \frac{\partial V_r^{(i)}}{\partial r} \right) \Big|_{r=R}, \end{aligned} \quad (18)$$

$$\begin{aligned} & \eta_{0e} \left(\frac{1}{r} \frac{\partial V_r^{(e)}}{\partial \theta} + \frac{\partial V_\theta^{(e)}}{\partial r} - \frac{V_\theta^{(e)}}{r} \right) \Big|_{r=R} + \frac{1}{r} \frac{\partial \sigma_0}{\partial T_e} \Big|_{T_e=T_{0e}} \frac{\partial T_e}{\partial \theta} \Big|_{r=R} = \\ & = \eta_{0i} \left(\frac{1}{r} \frac{\partial V_r^{(i)}}{\partial \theta} + \frac{\partial V_\theta^{(i)}}{\partial r} - \frac{V_\theta^{(i)}}{r} \right) \Big|_{r=R}. \end{aligned} \quad (19)$$

The molecular concentration of the first component, undergoing a phase transition, of the external mixture at the drop surface must satisfy the condition

$$C_{1e}|_{r=R} = C_{0sat} + \frac{\partial C_{sat}}{\partial T_e} \Big|_{\substack{T_e=T_{0e} \\ C_{sat}=C_{0sat}}} (T_e - T_{0e})|_{r=R}. \quad (20)$$

Solution of the Problem. The system of differential equations (1)-(7), being represented in a spherical coordinate system, in combination with the boundary conditions (8)-(20) allows us to seek solutions in the form [10]

$$V_r^{(e)} = \left(\frac{A_e}{r^3} + \frac{B_e}{r} + |\vec{U}| \right) \cos \theta + \frac{\gamma_e}{r}, \quad (21)$$

$$V_\theta^{(e)} = \left(\frac{A_e}{2r^3} - \frac{B_e}{2r} - |\vec{U}| \right) \sin \theta, \quad (22)$$

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

$$V_r^{(i)} = (Q_i + D_i r^2) \cos \theta, \quad (24)$$

$$V_\theta^{(i)} = -(Q_i + 2D_i r^2) \sin \theta, \quad (25)$$

$$p^{(i)} = p_0^{(i)} + 10\eta_i D_i r \cos \theta, \quad (26)$$

$$T_e = T_{0e} + |(\nabla T_e)_\infty| r \cos \theta + \frac{\mu_1}{r^2} \cos \theta + \frac{\Phi_1}{r}, \quad (27)$$

$$T_i = T_{0i} + \mu_2 r \cos \theta, \quad (28)$$

$$C_{1e} = C_{01e} + \frac{\mu_3}{r^2} \cos \theta + \frac{\Phi_2}{r}. \quad (29)$$

Further solution of the problem is reduced to substitution of the solutions (21)-(29) into the boundary conditions (12)-(20) and determination of the constants A_e , B_e , γ_e , Q_i , D_i , μ_1 , μ_2 , μ_3 , Φ_1 , Φ_2 , and $|\vec{U}|$.

In formulating the problem, by \vec{U} was meant the velocity of motion of the center of inertia of the external binary mixture relative to the drop at rest. Based on this assumption, the thermophoresis rate of the drop relative to the center of inertia of the binary mixture is

$$\vec{U}_{tp} = -\vec{U}. \quad (30)$$

As a result, we arrive at

$$\begin{aligned} \vec{U}_{tp} = & -\frac{6\eta_{0i} \eta_{0e} K_{th.s}}{(2\eta_{0e} + 3\eta_{0i}) T_{0e} \rho_{0e}} \left(\chi_e + Lm_1 \beta_2 \frac{D_{12}^{(e)}}{T_{0e}} K_{th.dif}^{(e)} \right) \frac{(\nabla T_e)_\infty}{\Delta} - \\ & - \left(\frac{6\eta_{0i}}{2\eta_{0e} + 3\eta_{0i}} \right) K_{dif.s} D_{12}^{(e)} \left(\chi_e \frac{\partial C_{sat}}{\partial T_e} + Lm_1 \beta_2 \frac{D_{12}^{(e)}}{T_{0e}} K_{th.dif}^{(e)} \frac{\partial C_{sat}}{\partial T_e} \right) \frac{(\nabla T_e)_\infty}{\Delta} - \end{aligned}$$

$$\begin{aligned}
& - \left(\frac{2R}{2\eta_{0e} + 3\eta_{0i}} \right) \frac{\partial \sigma_0}{\partial T_e} \left(\chi_e + Lm_1 \beta_2 \frac{D_{12}^{(e)}}{T_{0e}} K_{th.dif}^{(e)} \right) \frac{(\nabla T_e)_\infty}{\Delta} + \\
& + \left(\frac{2\eta_{0e} + \eta_{0i}}{2\eta_{0e} + 3\eta_{0i}} \right) \frac{3\beta_1 D_{12}^{(e)} K_{th.dif}^{(e)}}{n_{02e} T_{0e}} (\nabla T_e)_\infty - \\
& - \left(\frac{2\eta_{0e} + \eta_{0i}}{2\eta_{0e} + 3\eta_{0i}} \right) \frac{6\beta_1 D_{12}^{(e)}}{n_{02e}} \left(\frac{K_{th.dif}^{(e)}}{T_{0e}} + \frac{\partial C_{sat}}{\partial T_e} \right) \left(\chi_e + Lm_1 \beta_2 \frac{D_{12}^{(e)}}{T_{0e}} K_{th.dif}^{(e)} \right) \frac{(\nabla T_e)_\infty}{\Delta}. \tag{31}
\end{aligned}$$

In expression (31) we have

$$\begin{aligned}
\Delta = & \left(2\chi_e + 2Lm_1 \beta_2 \frac{D_{12}^{(e)}}{T_{0e}} K_{th.dif}^{(e)} + \chi_i + 2Lm_1 \beta_2 D_{12}^{(e)} \frac{\partial C_{sat}}{\partial T_e} + \right. \\
& \left. + \frac{2K_{th.dif}^{(e)} D_{12}^{(e)} p}{C_{sat} (1 - C_{sat})} \frac{\partial C_{sat}}{\partial T_e} \right). \tag{32}
\end{aligned}$$

To carry out numerical evaluations by formula (31), we will consider, as an example, the motion of a water drop in a binary gas mixture of air–water vapor at atmospheric pressure of the mixture. Calculations were carried out for the following values of the quantities entering expression (31): $n_{0e} = 2.7 \cdot 10^{25} \text{ m}^{-3}$; $n_{02e} = 2.67 \cdot 10^{25} \text{ m}^{-3}$; $m_1 = 2.88 \cdot 10^{-26} \text{ kg}$; $m_2 = 4.7 \cdot 10^{-26} \text{ kg}$; $\rho_{0e} = 1.3 \text{ kg/m}^3$; $\eta_{0e} = 1.81 \cdot 10^{-5} \text{ N}\cdot\text{sec/m}^2$; $\eta_{0i} = 10^{-3} \text{ N}\cdot\text{sec/m}^2$; $\chi_0 = 2.6 \cdot 10^{-2} \text{ J/(m}\cdot\text{sec}\cdot\text{K)}$; $\chi_i = 0.6 \text{ J/(m}\cdot\text{sec}\cdot\text{K)}$; $D_{12}^{(e)} = 2.5 \cdot 10^{-5} \text{ m}^2/\text{sec}$; $K_{th.s} = 1.17$; $K_{dif.s} = 0.3$; $L = 2.3 \cdot 10^6 \text{ J/kg}$; $K_{th.dif}^{(e)} = 5.5 \cdot 10^{-3}$; $\partial \sigma_0 / \partial T_e = -1.6 \cdot 10^{-4} \text{ N/(K}\cdot\text{m)}$; $\partial C_{sat} / \partial T_e = 1.67 \cdot 10^{-3} \text{ K}^{-1}$.

We represent formula (31) in the form

$$\vec{U}_{tp} = (U_{tp}^{(1)} + U_{tp}^{(2)} + U_{tp}^{(3)} + U_{tp}^{(4)} + U_{tp}^{(5)}) (\nabla T_e)_\infty, \tag{33}$$

where

$$U_{tp}^{(1)} = - \frac{6\eta_{0i} K_{th.s} \eta_{0e}}{(2\eta_{0e} + 3\eta_{0i}) T_{0e} \rho_{0e} \Delta} \left(\chi_e + Lm_1 \beta_2 \frac{D_{12}^{(e)}}{T_{0e}} K_{th.dif}^{(e)} \right), \tag{34}$$

$$U_{tp}^{(2)} = - \left(\frac{6\eta_{0i}}{2\eta_{0e} + 3\eta_{0i}} \right) \frac{K_{dif.s} D_{12}^{(e)}}{\Delta} \left(\chi_e + Lm_1 \beta_2 \frac{D_{12}^{(e)}}{T_{0e}} K_{th.dif}^{(e)} \right) \frac{\partial C_{sat}}{\partial T_e}, \tag{35}$$

$$U_{tp}^{(3)} = - \frac{2R}{(2\eta_{0e} + 3\eta_{0i}) \Delta} \frac{\partial \sigma_0}{\partial T_e} \left(\chi_e + Lm_1 \beta_2 \frac{D_{12}^{(e)}}{T_{0e}} K_{th.dif}^{(e)} \right), \tag{36}$$

$$\begin{aligned}
U_{tp}^{(4)} = & - \left(\frac{2\eta_{0e} + \eta_{0i}}{2\eta_{0e} + 3\eta_{0i}} \right) \frac{6\beta_1 D_{12}^{(e)}}{n_{02e} \Delta} \left(\frac{K_{th.dif}^{(e)}}{T_{0e}} + \frac{\partial C_{sat}}{\partial T_e} \right) \times \\
& \times \left(\chi_e + Lm_1 \beta_2 \frac{D_{12}^{(e)}}{T_{0e}} K_{th.dif}^{(e)} \right), \tag{37}
\end{aligned}$$

TABLE 1. Temperature Dependence of the Coefficients of $(\nabla T_e)_\infty$ in the Thermophoresis Rate

T_{0e}	$U_{tp}^{(1)}$	$U_{tp}^{(2)}$	$U_{tp}^{(3)}$	$U_{tp}^{(4)}$	$U_{tp}^{(5)}$
303	$-3.555 \cdot 10^{-9}$	$-8.283 \cdot 10^{-10}$	$3.526 \cdot 10^{-3}R$	$-1.749 \cdot 10^{-9}$	$2.81 \cdot 10^{-10}$
323	$-3.333 \cdot 10^{-9}$	$-8.276 \cdot 10^{-10}$	$3.524 \cdot 10^{-3}R$	$-1.747 \cdot 10^{-9}$	$2.64 \cdot 10^{-10}$
353	$-3.044 \cdot 10^{-9}$	$-8.261 \cdot 10^{-10}$	$3.518 \cdot 10^{-3}R$	$-1.742 \cdot 10^{-9}$	$2.41 \cdot 10^{-10}$

TABLE 2. Contribution of the Dufour Effect to the Thermophoresis Rate at Different Temperatures

T_{0e}	Δ	D
303	$8.006 \cdot 10^{-1}$	$1.289 \cdot 10^{-3}$
323	$7.998 \cdot 10^{-1}$	$5.103 \cdot 10^{-4}$
353	$7.993 \cdot 10^{-1}$	$1.993 \cdot 10^{-4}$

$$U_{tp}^{(5)} = \left(\frac{2\eta_{0e} + \eta_{0i}}{2\eta_{0e} + 3\eta_{0i}} \right) \frac{3\beta_1 D_{12}^{(e)} K_{th,dif}^{(e)}}{n_{02e} T_{0e}}. \quad (38)$$

In Table 1, numerical values of the quantities defined by relations (34)-(38) are given for three temperatures of the mixture T_{0e} . The drop radius is R .

It is pertinent to note that expression (31) is obtained in [10] as a particular case of a formula for volatile drops of concentrated solutions with a solute concentration tending to zero and coincides, with an accuracy up to the Dufour effect, with the resultant formula of [14, 15] for $(\eta_{0e}/\eta_{0i}) \rightarrow 0$. In formula (31), just as in the resultant relations of [14, 15], allowance is made for the Dufour effect, which is contained in the form of the fifth term in the denominator Δ of the resultant formula (31) for \vec{U}_{tp} , i.e., in the form

$$D = \frac{2K_{th,dif}^{(e)} D_{12}^{(e)}}{C_{sat} (1 - C_{sat})} \frac{\partial C_{sat}}{\partial T_e}. \quad (39)$$

Table 2 provides numerical values of the quantities determined by relations (32) and (39). As is seen from the table, Δ is three orders of magnitude larger than D , i.e., the Dufour effect makes an insignificant contribution to the total rate in actual situations.

If we go back to the fundamental theory of thermophoresis, formula (31) without allowance for the mentioned Dufour effect was derived in works published more than ten years ago. Ignoring this fact in [14, 15] misleads readers as to the question of priority of the school of Yu. I. Yalamov and his colleagues in the field of the theory of thermophoresis of volatile aerosol particles.

As for the analysis made in [14, 15], it is absolutely trivial, and the physical aspects of the effects discussed in these papers are easily tracked in [5, 6, 10-13].

NOTATION

η_{0e} , mean coefficient of dynamic viscosity of the external medium, N·sec/m²; ρ_{0e} , mean density of the external medium, kg/m³; T_{0e} , mean temperature of the external medium at a large distance from the drop, K; R , drop radius, m; r, θ, φ , spherical coordinates; $(\nabla T_e)_\infty$, temperature gradient at a relatively large distance from the drop, K/m; \vec{U} , velocity of motion of the center of inertia of the external binary mixture relative to the drop at rest, m/sec; η_e , coefficient of dynamic viscosity of the external medium, N·sec/m²; $\vec{V}^{(e)}$, velocity of the center of inertia of the external medium, m/sec; $p^{(e)}$, pressure of the external medium, Pa; T_e , temperature of the external medium, K; η_i , coefficient of dynamic viscosity of the drop substance, N·sec/m²; η_{0i} , mean coefficient of dynamic viscosity of the drop substance, N·sec/m²; $\vec{V}^{(i)}$, velocity of the center of inertia of the drop substance, m/sec; $p^{(i)}$, pressure inside the drop, Pa; T_i , temperature inside the drop, K; C_{1e} , dimensionless relative concentration of the first component of the external mixture; $V_r^{(e)}$ and $V_\theta^{(e)}$, radial and tangential compo-

nents of the velocity of the center of inertia of the external mixture in the spherical coordinate system, m/sec; $p_0^{(e)}$, mean pressure of the external mixture at a large distance from the drop, Pa; n_{01e} , mean molecular concentration of the first component of the external mixture, m^{-3} ; n_{02e} , mean molecular concentration of the second component of the external mixture, m^{-3} ; n_{0e} , mean molecular concentration of the external mixture, m^{-3} ; n_{01i} , mean molecular concentration inside the drop, m^{-3} ; β_1 and β_2 , coefficients that depend on the parameters of the external mixture, m^{-3} ; m_1 and m_2 , molecular masses of the first and second components of the external mixture, kg; $D_{12}^{(e)}$, mutual-diffusion coefficient of the external mixture, m^2/sec ; $K_{\text{th.dif}}^{(e)}$, dimensionless thermomodification ratio; $V_r^{(i)}$ and $V_\theta^{(i)}$, radial and tangential components of the velocity of the center of inertia of the drop substance in the spherical coordinate system, m/sec; $K_{\text{th.s}}$, dimensionless thermal-slip coefficient; $K_{\text{dif.s}}$, dimensionless diffusional-slip coefficient; χ_e , thermal conductivity of the external mixture, $J/(\text{m}\cdot\text{sec}\cdot\text{K})$; χ_i , thermal conductivity of the drop substance, $J/(\text{m}\cdot\text{sec}\cdot\text{K})$; L , specific heat of the phase transition of the drop substance, J/kg ; C_{sat} , dimensionless relative concentration of the saturated vapor of the drop substance; $C_{0\text{sat}}$, relative saturating concentration of the first component of the external mixture at temperature T_{0e} ; p , pressure, Pa; σ_0 , mean coefficient of surface tension at the interface of drop-external mixture, N/m ; A_e , B_e , γ_e , Q_i , D_i , μ_1 , μ_2 , μ_3 , Φ_1 , and Φ_2 , coefficients that depend on the parameters of the external mixture and the drop; \vec{U}_{tp} , thermophoresis rate, m/sec; Δ , common denominator in the formula for the thermophoresis rate, $J/(\text{K}\cdot\text{m}\cdot\text{sec})$; $U_{\text{tp}}^{(1)}$, $U_{\text{tp}}^{(2)}$, $U_{\text{tp}}^{(3)}$, $U_{\text{tp}}^{(4)}$, and $U_{\text{tp}}^{(5)}$, coefficients of $(\nabla T_e)_\infty$ in the thermophoresis rate, $m^2/(\text{sec}\cdot\text{K})$; D , contribution of the Dufour effect to the thermophoresis rate, $J/(\text{K}\cdot\text{m}\cdot\text{sec})$. Subscripts: e, external; 0, mean; i, internal; tp, thermophoresis; th, thermal; dp, diffusiophoresis; s, slip; sat, saturated.

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