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THE NATURE OF THERMOPHORESIS OF HIGHLY HEAT-CONDUCTING BODIES IN GASES†

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A method of calculating the rate of thermophoresis of large bodies in gases is briefly described. Particular attention is devoted to the features of the mechanism of the thermophoresis of highly heat-conducting bodies, which were subject to the attempts made over many years to explain the anomalies. A qualitative explanation of the mechanism is given. It is shown that it owes its origin to the presence in the gas region, close to the body surface, of a second (mixed) derivative with respect to the temperature (whereas Epshtein–Maxwell thermophoresis is due to its first derivative). It is emphasized that, whereas classical thermophoresis is independent of the Knudsen number Kn = λ/R (λ is the mean free path of the gas molecules and R is a characteristic dimension of the body), the rate of thermophoresis of highly heat-conducting bodies is directly proportional to it. On the other hand, classical thermophoresis depends very much on the ratio of the thermal conductivities of the body and the gas, while the rate of thermophoresis of highly heat-conducting bodies is independent of this ratio. It is shown that reliable quantitative results can be obtained only if reliable data is available on the accommodation coefficients for collisions between the gas molecules and the body surface. © 2004 Elsevier Ltd. All rights reserved.

The classical representation of the nature of thermophoresis of bodies in gases is based on the discovery by Maxwell [1] of the thermal slip of a gas along the interface. Epshtein [2] made it the basis of the solution of the problem of the thermophoresis of large bodies as a boundary condition which related the problem of the gas flow past a body and the problem of the temperature distribution, when a small constant temperature gradient is maintained far from the body. Epshtein gave an impetus to the study of the nature of the thermophoresis of large bodies in gases. (When we speak of large bodies we mean that the Knudsen number is much less than unity.) Following this, experiments showed that the Maxwell slip mechanism did not exhaust all possible reasons for the phenomenon. In particular, this approach did not enable the experimentally observed unexpectedly high rate of thermophoresis of highly heat-conducting bodies to be explained. A considerable number of publications has already been devoted to a theoretical investigation of this problem (see the reviews in [3, 4]). A variety of approaches and methods of calculation are used in these papers. This situation, and also the existing differences between the final results obtained not only do not help the nature of the phenomenon to be understood, but also make it extremely difficult to use them in practice. It therefore seems useful and opportune to describe a method of calculating the rate of thermophoresis of large bodies in gases which is most accessible for a wide range of specialist, emphasizing the key factors involved.

1. FORMATION OF THE PROBLEM

When solving the problem of the thermophoresis of large bodies Stokes' problem for the gas flow past a sphere is considered, when a small constant temperature gradient is maintained at an infinite distance from the sphere. We will confine ourselves to the case of non-volatile bodies. This means, formally, that the sphere surface is impermeable to gas molecules, i.e. the normal component of the gas velocity on the body surface must vanish. Note that, by considering the case of fairly large bodies, for which the Knudsen number Kn = λR is much less than unity (λ is the mean free path of the gas molecules and R is a characteristic dimension of the body), we can drop certain terms proportional to Kn not only during the calculations but also at the stage of formulating the problem, while taking well-known precautions.

Hence, for the normal component we have

$$v_r = 0 \tag{1.1}$$

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The boundary condition for the tangential component of the gas velocity is less obvious. We will write it in the form [5]

$$v_\theta = \mathrm{Kn}\frac{R}{\eta}C_m\sigma_{r\theta} + k_{\mathrm{TS}}\frac{\eta}{\rho T_0}\frac{1}{R}\frac{\partial}{\partial\theta}\frac{T+T_i}{2} + \frac{3}{2}\frac{\eta}{\rho T_0}\frac{1}{R}\frac{\partial}{\partial\theta}\frac{T-T_i}{2} \tag{1.2}$$

According to Epshtein, the first two terms in expression (1.2) describe viscous and thermal slip. Note, however, that here we have made certain refinements: in the expression for the viscous stress tensor $\sigma_{r\theta}$, in addition to the usual stresses we have taken the thermal stresses $\sigma^{(T)}_{r\theta}$ into account; in the second term, instead of a numerical coefficient, we have used the thermal slip coefficient k_{TS}, which is equal to the Maxwell value of 0.75 only in the special (ideal) case of specular interaction of the gas molecules with the surface of the condensed phase. In addition, the expression for the tangential derivative of the temperature is written in a more general form. However, the most important difference is the presence of the third term, which is not present in Epshtein's scheme.

Here we have also used the following notation: $\mathbf{v}$ is the gas velocity with respect to the body (the subscripts r and θ indicate the radial and tangential components respectively), η is the dynamic viscosity, ρ is the gas density, C_m is the viscous slip coefficient, and T_i and T_0 are temperature inside and at the centre of the sphere respectively.

We will formulate the boundary conditions for determining the temperature in the Laplace problem. The normal component of the heat flux $\mathbf{q}$ on the boundary is continuous:

$$q_r = q_{ir} \quad \text{or} \quad \partial T/\partial r = \xi\partial T_i/\partial r, \quad \xi = \kappa_i/\kappa \tag{1.3}$$

(κ is the thermal conductively of the gas and κ_i is the thermal conductivity of the condensed phase). For the temperature we have (this is essentially the Smoluchowski relation [6] in a more refined form)

$$T - T_i = \mathrm{Kn}\frac{R}{\kappa}C_t\frac{(\mathbf{q}+\mathbf{q}_i)_r}{2} \tag{1.4}$$

where C_t is the temperature jump coefficient.

We will also present an expression for the thermal stresses in a gas which we will need for further calculations. It is most conveniently written in the form

$$\sigma^{(T)}_{r\theta} = \frac{3}{r}\frac{\eta^2}{\rho T_0}\left[\frac{1}{r} - \frac{\partial}{\partial r}\right]\frac{\partial T}{\partial\theta} \tag{1.5}$$

2. THE VELOCITY AND TEMPERATURE DISTRIBUTION

We will write expressions for the components of the gas velocity in the neighbourhood of the body and for the force with which the gas acts on the body in the Stokes approximation in the standard form

$$v_r = v_0\left[1 - 2a\frac{R}{r} + 2b\left(\frac{R}{r}\right)^3\right]\cos\theta, \quad v_\theta = -v_0\left[1 - a\frac{R}{r} - b\left(\frac{R}{r}\right)^3\right]\sin\theta$$

$$\mathbf{F} = 8\pi\eta Ra\mathbf{v}_0$$

where $\mathbf{v}_0$ is the free-stream velocity. In equilibrium, taking into account boundary condition (1.1), we have for the constants of integration: $a = 0$ and $b = -1/2$. We will write the temperature distributions inside and outside the body in the form

$$T_i = T_0 + (\mathbf{A}\mathbf{r}), \quad T = T_0 + (\mathrm{grad}T\mathbf{r}) + (\mathbf{B}\mathbf{r})\left(\frac{R}{r}\right)^3 \tag{2.1}$$

(grad T is the specified constant temperature gradient far from the body). From conditions (1.3) and (1.4) we have a system of two linear equations in the constants of integration $\mathbf{A}$ and $\mathbf{B}$, solving which, we obtain

$$\mathbf{A} = \frac{3}{2}\frac{1}{1+\xi/2}\mathrm{grad}T, \quad \mathbf{B} = \frac{1}{2}\frac{1-\xi}{1+\xi/2}\mathrm{grad}T \tag{2.2}$$

Finally, it follows from expression (1.5) that

$$\sigma_{r\theta}^{(T)} = -\frac{9}{2R}\frac{\eta^2}{\rho T_0}\frac{1-\xi}{1+\xi/2}\sin\theta \operatorname{grad} T$$

3. THE RATE OF THERMOPHORESIS

It is now easy to calculate the quantities occurring in Eq. (1.2). As a result, bearing in mid the fact that the equilibrium velocity of the body (the rate of thermophoresis) is equal to (with the opposite sign) the equilibrium value $\mathbf{v}_0$, we have

$$\mathbf{v}_{TR} = -\frac{\eta}{\rho T_0}k_{\mathrm{TS}}\frac{\operatorname{grad} T}{1+\xi/2}\left\{1+\xi \mathrm{Kn}\left[\frac{1}{2}C_t+\frac{3}{k_{\mathrm{TS}}}\left(\frac{1}{2}C_t-C_m\right)\right]\right\} \tag{3.1}$$

We recall that, since the Knudsen number is small, we dropped a number of terms, including in the final expression (3.1) also. It must, however, be emphasized, that a situation can occur when the thermal conductivity of the body is so high that even for small Knudsen numbers the combination ξKn considerably exceeds unity (such a situation arises, for example, for metal bodies). Hence, terms containing such a product are retained. If the thermal conductivity of the body is not too high compared with the thermal conductivity of the gas, the second term in the braces in expression (3.1) is also small compared with unity and can be neglected. As a result, we arrive at the Epshtein formula (refined in the thermal slip coefficient part)

$$\mathbf{v}_{TR}^{E} = -\frac{\eta}{\rho T_0}k_{\mathrm{TS}}\frac{\operatorname{grad} T}{1+\xi/2} \tag{3.2}$$

On the other hand, in the case of a high thermal conductivity of the body, expression (3.1) becomes

$$\mathbf{v}_{TR} = -\frac{\eta}{\rho T_0}k_{\mathrm{TS}}\operatorname{grad} T\,\mathrm{Kn}\left[C_t+\frac{3}{k_{\mathrm{TS}}}(C_t-2C_m)\right] \tag{3.3}$$

We will make a numerical estimate of this effect. To do this it is necessary to calculate the values of the kinetic coefficients occurring in expression (3.3). We will use the following relations [3, 4]

$$C_m(\varepsilon) = a/\varepsilon+b, \quad C_t(\alpha) = c/\alpha+d, \quad k_{\mathrm{TS}} = 0.75+(g+h\varepsilon)\varepsilon$$

Here α and ε are coefficients characterizing the energy and momentum accommodation respectively, when gas molecules interact with the surface of the condensed phase. The constants, *a*, *b*, *c*, *d*, *g* and *h* are numbers calculated for different models of intermolecular interaction (see the reviews [3, 4]).

To estimate the value of the rate of thermophoresis of highly heat-conducting bodies it is convenient to use the relation

$$\Delta = \frac{\mathbf{v}_{\mathrm{TR}}}{\mathbf{v}_{TR}^{E}} = \frac{1}{2}\xi\,\mathrm{Kn}\left[C_t+\frac{3}{k_{TS}}(C_t-2C_m)\right] \tag{3.4}$$

We will consider two limiting cases, for diffuse interaction (the coefficients α and ε are of the order of unity)

$$\Delta \cong M\xi\mathrm{Kn} \tag{3.5}$$

i.e. the rate of thermophoresis of bodies with a high thermal conductivity may considerably exceed its Epshtein value. For specular interaction (the coefficients α and ε are fairly small)

$$\Delta \cong \xi\mathrm{Kn}\left(\frac{m}{\alpha}-\frac{n}{\varepsilon}\right) \tag{3.6}$$

i.e. the difference of the rate of thermophoresis from the Epshtein value can be as large as desired, on both sides; it can even change sign. Hence M, m and n are certain numbers of the order of unity.

4. DISCUSSION OF THE RESULTS

The essential and basic difference between the boundary conditions derived above the Epshtein conditions is that, in addition to the Maxwell thermal slip, terms due to the presence of a radial heat flux through the interface are also taken into account. The latter is represented in explicit form in the boundary condition for the temperature (1.4). The heat flux is contained in implicit form in all three terms of the boundary condition for the tangential component of the gas velocity on the body surface: in the expression for the thermal stresses, also introduced by Maxwell, in the refined expression for the thermal slip and in the new, third, term. Here it is important to note not simply the presence of a radial heat flux, but its non-uniformity along the body surface (i.e. the non-zero tangential derivative of the thermal flux).

Hence, whereas the classical (Epshtein) thermophoresis is due to Maxwell thermal slip, proportional to the first derivative of the temperature, thermophoresis of highly heat-conducting bodies is determined by a mechanism which has its origin in its second derivative. It is represented by a useful term, introduced by Sone [7], who called this effect second-order slip.

As follows from relations (3.2) and (3.3), the rate of thermophoresis for low values of the thermal conductivity of the body falls sharply as the latter increases, and for high values of the thermal conductivity it ceases to depend on it.

It is worth emphasizing that, despite what is widely asserted in the literature, classical (Epshtein) thermophoresis is independent of the Knudsen number $\mathrm{Kn} = \lambda/R$ (more accurately, it depends to a slight extent as a small correction to the main expression, see formula (3.1)), whereas the rate of thermophoresis of highly heat-conducting bodies is proportional to Kn. At first glance this should indicate that the effect considered is small. However, the factor $\xi = \kappa_i/\kappa$ in expression (3.5) compensates this reduction with interest. In fact, for aerosols of sodium chloride and mercury in air with $\mathrm{Kn} = 0.1$, the measured values exceed the values calculated from Epshtein's formula (3.2) several tens of times, but agree completely with relation (3.5). The same can be said of the remaining measurements, carried out with different kinds of aerosols, the results of which strikingly contradict the predictions of classical theory. (We leave to one side interpolational relations, designed to satisfy purely applied requirements but which have a far from convincing scientific basis).

Formula (3.6) is of particular interest. It follows from this formula that the rate of thermophoresis of highly heat-conducting bodies can take any value, as large or as small as desired, and can even be negative. It always reduces in this case to using some values of the accommodation coefficients of gas molecules in the calculations when they collide with the body surface. For example, if the accommodation of the momentum of the molecules is sufficiently weak, so that the inequality $\varepsilon < \alpha n/m$ is satisfied, the rate of thermophoresis becomes negative. The possibility of the existence of negative thermophoresis has already been suggested in a number of publications (see, for example, [8–11]). Experimental results are interpreted in [12] as an actual demonstration of negative thermophoresis. Note that, to obtain real reliable results when calculating the rate of thermophoresis of highly heat-conducting bodies it is necessary to have reliable data on the nature of the accommodation of gas molecules on the surface of the condensed phase.

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