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Abstract

Full Text

PHYSICAL CHEMISTRY

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THEORY OF THERMOPHORESIS OF MODERATELY LARGE AEROSOL PARTICLES

The theory of thermophoresis of aerosol particles developed by one of us, corresponding to the limiting case ⁽¹⁾

$$\text{Kn} \equiv \lambda/R \rightarrow 0, \tag{1}$$

where Kn is the Knudsen number, λ is the mean free path of gas molecules, and R is the radius of the aerosol particle, agrees much better with experimental data than Epstein's theory ⁽²⁾.* Brock ^(4,5) attempted to refine Epstein's theory by taking into account terms of order λ/R that depend on the jumps of temperature and velocity at the surface of aerosol particles. In order to judge objectively which of the two different thermophoresis mechanisms considered in ^(1,2), volume or surface, more correctly reflects reality, it is evidently necessary in theory ⁽¹⁾ as well to pass from the zeroth to the first-order approximation in Kn. The present work is devoted to this.

Analogously to work ⁽¹⁾, let us consider a highly porous partition formed from randomly arranged spheres of radius R , rigidly fixed in space at distances much greater than the radius. At the ends of the partition, temperature and pressure differences ΔT and Δp are maintained. Let us consider the stationary fluxes of heat Q and matter I_M passing through a unit cross section of the partition. Under the condition $\Delta p = 0$ we have ^(1,6)

$$I_M|_p = -a_{12} \Delta T/T^2. \tag{2}$$

Under the condition

$$\Delta T = 0 \tag{3}$$

we have ^(1,6)

$$Q^*|_T = -a_{21} \Delta p/\rho T. \tag{4}$$

In (2) and (4), p is the pressure, ρ is the gas density, and T is the absolute temperature. According to Onsager's principle, $a_{12} = a_{21}$, and consequently,

$$I_M|_p = \frac{\rho Q^*|_T}{\Delta p} \frac{\Delta T}{T}. \quad (5)$$

The mean linear velocity of the gas through the partition, equal (with the opposite sign) to the thermophoretic velocity v_T , for $\Delta T \neq 0$ and $p = \text{const}$:

$$v = -v_T = I_M|_p / \rho. \quad (6)$$

The Chapman-Enskog method for solving the kinetic equation makes it possible to calculate (as a third approximation) the heat transfer in an isothermal gas flow when the spatial derivatives of the velocity-gradient are not zero⁽⁷⁾. This heat flux, taking into account the Navier-Stokes equations

$$\eta \Delta \mathbf{V} = \text{grad } p, \quad (7)$$

$$\text{div } \mathbf{V} = 0 \quad (8)$$

is equal to

$$\mathbf{Q}_e = \frac{3}{2} \frac{\eta}{\rho} \text{grad } p. \quad (9)$$

* For the case $\lambda/R \gg 1$, the theory of thermophoresis was published earlier⁽³⁾.

The isothermal transfer of heat in the Knudsen layer of thickness λ around each sphere can, as was shown in⁽⁸⁾, be neglected. Let us consider the mean heat transfer through a unit area of an imaginary section of a porous partition (Fig. 1), parallel to its surfaces. From (9), for a small filling of the space by spheres, it follows that

$$\overline{Q_e} = \frac{3}{2} \frac{\eta}{\rho} \text{grad}_n p = \frac{3}{2} \frac{\eta}{\rho} \frac{\Delta p}{H}, \quad (10)$$

where the subscript n denotes the direction of the normal to the plane of the section, and H is the thickness of the partition. However, $\overline{Q_e}$ cannot be substituted for Q^* in formula (5), despite the fact that in the gas volume, as follows from (7) and (8), $\text{div } Q_e = 0$, and consequently no heat is released. It is, however, released on the surfaces of the spheres swept by the gas flow. To determine the corresponding sources and sinks of heat, we shall now solve equations (7) and (8) for the neighborhood of one sphere, taking into account the gas-kinetic slip of velocity at its surface and, consequently, with the boundary conditions:

$$v_r = 0 \quad \text{at } r = R,$$

$$v_\theta = C_m \lambda \left[r \frac{\partial}{\partial r} \left(\frac{v_\theta}{r} \right) + \frac{1}{r} \frac{\partial v_r}{\partial \theta} \right]_{r=R}, \quad (11)$$

where r and θ are polar coordinates with the axis OZ parallel to the velocity of the gas flow far from the sphere \mathbf{u} ; C_m , according to ⁽¹³⁾, for completely diffuse reflection of molecules is equal to 1.09. A detailed analysis of boundary condition (11) is given in ^(4, 10, 11). The solution gives

$$\text{grad } p = 2A\eta \left[\frac{3(\mathbf{u}\mathbf{r})\mathbf{r}}{r^5} - \frac{\mathbf{u}}{r^3} \right], \quad (12)$$

Fig. 1. Temperature distribution around spheres

where \mathbf{r} is the radius vector of the point under consideration, (\mathbf{u}, \mathbf{r}) is the scalar product of the vectors \mathbf{u} and \mathbf{r} ,

$$A = \frac{3}{4} R (1 + 2C_m \lambda / R) / (1 + 3C_m \lambda / R). \quad (13)$$

Then from (9), for the heat released per unit time per unit area of each element of the surface of the sphere, we obtain

$$Q_R = \frac{6A\eta^2}{\rho R^3} |\mathbf{u}| \cos \theta. \quad (14)$$

Since $H \gg R$, the overwhelming part of the heat from the surface sources and sinks is neutralized by thermal conduction in the local temperature field in the neighborhoods of each sphere (see (1)). These heat flows penetrate the section of the partition under consideration (Fig. 1) in the direction opposite to the heat of isothermal transfer Q_e , and consequently reduce it by the amount Q_i .

The contribution to Q_i from heat exchange inside and in the neighborhoods of some sphere between its surface sources and sinks of heat is equal to the total power of heat release of the sources located on one side of the section of the sphere by the plane under consideration:

$$\Delta Q_i = \frac{6A\eta^2}{\rho R^3} |\mathbf{u}| \int \cos \theta dS = \frac{6A\eta^2}{\rho R^3} |\mathbf{u}| \Delta S, \quad (15)$$

where dS is the area of an elementary spherical zone, and ΔS is the area of the section by the plane of one sphere. Obviously, $S = \sum \Delta S$ —the fraction of the secant plane located inside the spheres—is, owing to statistical disorder, always the same, independently of the position of the plane, and is equal to the volume

filling of the space of the partition by spheres, $\frac{4}{3}\pi R^3 N$, where N is the number of spheres per unit volume. Therefore, summing (15) term by term, we obtain

$$Q_t = 8\pi N A \eta^2 |\mathbf{u}| / \rho. \quad (16)$$

As a result, according to (10) and (16), the total heat transfer through a unit cross section of the partition at $\Delta T_i^* = 0$ is equal to

$$|Q|_T = \frac{3}{2} \frac{\eta}{\rho} \frac{\Delta p}{H} - 8\pi N A \eta^2 |\mathbf{u}| / \rho. \quad (17)$$

Let F be the force of resistance to the flow of one sphere. Determining it by the usual method ⁽⁹⁾ from equations (7) and (8) with the boundary conditions (11), we find:

$$\Delta p / NH \equiv F = 6\pi \eta R |\mathbf{u}| (1 + 2C_m \lambda / R) / (1 + 3C_m \lambda / R), \quad (18)$$

an expression in good agreement with those obtained by other methods ⁽¹²⁾. Substituting (18) into (17), we obtain

$$|Q|_T = \frac{1}{2} \frac{\eta}{\rho} \frac{\Delta p}{H}. \quad (19)$$

Although the temperature field of the heat sources on the surface of each sphere is mainly localized in its vicinity, a small part of the heat fluxes reaching both surfaces of the porous partition, when summed with the analogous actions of other spheres, can substantially disturb condition (3).

To determine ΔT , let us first find the temperature distribution near one sphere caused by its surface sources at small Peclet numbers from the equation:

$$\frac{\partial^2 T}{\partial x^2} + \frac{\partial^2 T}{\partial y^2} + \frac{\partial^2 T}{\partial z^2} = 0 \quad (20)$$

with the boundary conditions ^(1,4)

$$T_e|_{r \rightarrow \infty} = 0; \quad T_i|_{r \rightarrow 0} < \infty; \quad (21)$$

$$(T_e - T_i)|_{r=R} = C_t \lambda (\partial T_e / \partial r)|_{r=R}; \quad (22)$$

$$(-\chi_e \partial T_e / \partial r + Q_r)|_{r=R} = -\chi_i (\partial T_i / \partial r)|_{r=R}, \quad (23)$$

where T_e and T_i are the temperatures outside and inside the sphere; C_t is a constant depending on the coefficient of thermal accommodation when gas molecules strike the surface of the particle, and χ_e and χ_i are the thermal conductivities of the gas and the particle, respectively. Condition (22) takes into account the existence of a temperature jump at the surface of the sphere^(14,15). The solutions of equation (20) with the written boundary conditions have, if (13), (14), and (18) are taken into account, the form:

$$T_e = - \left(\frac{\Delta p}{NH} \right) \frac{3\eta \cos \theta}{4\pi\rho r^2} \left/ \left(2\chi_e + \chi_i + \frac{2C_t\lambda}{R}\chi_i \right) \right., \quad (24)$$

$$T_i = - \left(\frac{\Delta p}{NH} \right) \frac{3\eta r \cos \theta}{4\pi\rho R^3} \left(1 + 2C_t \frac{\lambda}{R} \right) \left/ \left(2\chi_e + \chi_i + \frac{2C_t\lambda}{R}\chi_i \right) \right.. \quad (25)$$

By analogy with electrostatics we may say that the temperature field that arises is produced by thermal dipoles with moment:

$$\vec{\mu} = - \frac{6A\eta^2 \mathbf{u}}{\rho} \left/ \left(2\chi_e + \chi_i + \frac{2C_t\lambda}{R}\chi_i \right) \right.. \quad (26)$$

As a result the entire partition acquires a moment of thermal polarization:

$$\mathbf{M} = N\vec{\mu}. \quad (27)$$

The mean temperature gradient in the direction normal to the partition, in the volume of the gas, will be

$$\overline{\text{grad}_n T_e} = -4\pi|\mathbf{M}|, \quad (28)$$

and the temperature difference at the ends of the partition corresponding to (28) and (27) is

$$\Delta T = -4\pi|M|H, \quad (29)$$

where H is the width of the partition.

To satisfy condition (3), it is necessary to create on the surfaces of the partition heat sources and sinks that compensate ΔT from (29). The heat carried by the additional sources, according to (28), (27), (26), (13), and (18), is equal to

$$|Q_a| = \varkappa_e \overline{\text{grad}_n T_e} = 3\varkappa_e \eta \frac{\Delta p}{H} \left/ \rho \left(2\varkappa_e + \varkappa_i + \frac{2C_t\lambda}{R}\varkappa_i \right) \right.. \quad (30)$$

As a result, the true heat of transfer is equal to:

$$Q_T^* = |Q|_T + |Q_a| = \left(\frac{4\kappa_e + \frac{1}{2}\kappa_i + C_t \frac{\lambda}{R} \kappa_i}{2\kappa_e + \kappa_i + 2C_t \frac{\lambda}{R} \kappa_i} \right) \frac{\eta \Delta p}{\rho H}. \quad (31)$$

Consequently, according to (5), (6), and (31), the thermophoretic velocity is:

$$v_T = - \frac{\left(4\kappa_e + \frac{1}{2}\kappa_i + C_t \frac{\lambda}{R} \kappa_i \right)}{\left(2\kappa_e + \kappa_i + \frac{2C_t \lambda}{R} \kappa_i \right)} \frac{\eta}{\rho T} \text{grad } T. \quad (32)$$

For $\lambda/R \rightarrow 0$, result (32) goes over into the formula of paper ⁽¹⁾. We see that the correction for the Knudsen number depends only on C_t , but not on C_m .

According to Brock ⁽⁴⁾, the thermophoretic force has the form

$$F_T^{(B)} = - \frac{9\pi\eta^2 R \left(\kappa_e + \frac{C_t \lambda}{R} \kappa_i \right) \text{grad } T}{\rho T \left(2\kappa_e + \kappa_i + \frac{2C_t \lambda}{R} \kappa_i \right) \left(1 + \frac{3C_m \lambda}{R} \right)}. \quad (33)$$

To this force, according to (18), there corresponds the thermophoretic velocity

$$v_T^{(B)} = - \frac{3}{2} \frac{\eta}{\rho T} \frac{\left(\kappa_e + C_t \frac{\lambda}{R} \kappa_i \right) \text{grad } T}{\left(2\kappa_e + \kappa_i + \frac{2C_t \lambda}{R} \kappa_i \right) \left(1 + \frac{3C_m \lambda}{R} \right)}. \quad (34)$$

For $\lambda/R \rightarrow 0$, (34) goes over into Epstein's formula ⁽²⁾.

It should be noted that taking into account the terms with C_t has lowered v_T in comparison with the Bakanov-Derjaguin formula and raised it in comparison with Epstein's formula, reducing the discrepancy between the two theories. Nevertheless, it is still sufficiently large that, on the basis of the available experimental data, preference should be given to formula (32). In contrast to (34), formula (32) for $\lambda/R \rightarrow \infty$ is close to the exact formula ⁽³⁾.

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