## Thermophoresis in gases at small Knudsen numbers

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Theoretical and experimental studies characterizing the status of the science of thermophoresis in gases at small Knudsen numbers (relative to the dimensions of the object) are analyzed. The known modifications of the theory of thermophoresis amount to an approximation linear in the Knudsen number, while the zero-order approximation corresponds to the result of Epstein. Analysis of the experimental data on thermophoresis of aerosols of high heat conductivity shows that, among all the experiments described in the literature, not one satisfies the requirements that allow one to perform a correct comparison with the theory. Also the results of experiments with aerosols (objects) of low heat conductivity are unsatisfactory. An original method of studying thermophoresis in gases is proposed.

### **1.INTRODUCTION**

The phenomenon of thermophoresis (TP) was discovered experimentally more than 100 years ago.<sup>1</sup> Upon placing a foreign object in a gas where a temperature gradient was maintained with external heat sources, the observer found that, despite the absence of ordinary external forces, it goes into motion in the direction of the temperature variation. The measurements showed that the velocity of established motion is proportional to the temperature gradient, at least for small gradients, and is directed in the same direction as the heat flux arising from the heat conductivity of the gas.

Classical hydrogasdynamics proved powerless to explain this phenomenon. Actually, the distribution of velocities and temperatures in the gas-object system according to classical hydrodynamics is determined by the solution of the Navier-Stokes-Laplace equations with the boundary conditions

$$v_{\mathbf{r}} = 0, \quad v_{\mathbf{r}} = 0,$$

$$T_{\mathbf{i}} = T_{\mathbf{e}}, \quad \varkappa_{\mathbf{i}} \frac{\delta T_{\mathbf{i}}}{\delta \mathbf{r}} = \varkappa_{\mathbf{e}} \frac{\delta T_{\mathbf{e}}}{\delta \mathbf{r}},$$
(1)

and the condition  $\nabla T = \nabla T_{\infty}$  at infinity ( $\nabla T_{\infty}$  is the specified temperature gradient,  $\varkappa_i$  and  $\varkappa_e$  are the heat conductivities of the object and the gas, **r** and  $\tau$  are the unit vectors of the normal and the tangent to the surface of the object). We can easily see that the fields of velocities and temperatures in this formulation are completely independent. Consequently, relative motion of the gas and the object in the absence of external forces does not occur.

A relationship between the flow field and the temperature was first found theoretically by Maxwell<sup>2</sup> and experimentally by Reynolds.<sup>3</sup> It was established that, if one maintains the temperature gradient  $\nabla T$  in the gas along its boundary with the solid object, then a motion of the gas arises in the direction of this gradient. This phenomenon was called creep or thermal slip. The velocity of slip according to Maxwell is

$$\mathbf{v}_{c} = -\frac{3}{4} \nu \, \nabla \ln T; \tag{2}$$

v is the kinematic viscosity of the gas.

Maxwell calculated the value of the coefficient of thermal slip (3/4) under rather crude assumptions by using the kinetic theory of gases that he had created.

These studies originated the development of a new branch of hydrogasdynamics—hydrogasdynamics "with slip," in which the classical conditions of "adhesion" of the gas to the wall are replaced by conditions of "slip."

This result proved sufficient, when the fields of velocities and temperature were correlated thereby, to obtain a nonzero velocity in the problem of TP. Actually, in the system of boundary conditions (1), instead of  $\mathbf{v}_{\bar{\tau}} = 0$  the following equation arises

$$v_{\tau} = \mathbf{v}_{c} = -\frac{3}{4}\nu \nabla \ln T.$$

Integration of the Navier–Stokes–Laplace equations with these boundary conditions led Epstein<sup>4</sup> to the formula for the velocity of thermophoresis

$$\mathbf{v}_{T\Phi} = -\frac{3}{4} \frac{\nu}{T} \frac{\nabla T_{\infty}}{1 + (-\frac{\varkappa_{i}}{2\varkappa_{o}})} .$$
(3)

Experiments showed that Eq. (3) yields a somewhat depressed value (by about a factor of 1.5) of  $\varkappa_{\rm TP}$  for  $\varkappa_i/\varkappa_e \leq 1$ . This, and also the insensitivity of  $v_{\rm TP}$  to the character of the interphase interaction, involve the simplifications adopted in calculating the coefficient of thermal slip. But Eq. (3) works especially poorly in the case in which  $\varkappa_i/\varkappa_e \geq 1$ . The velocity of thermophoresis, as shown experimentally, though declining somewhat, does not do so to the significant degree predicted by the Epstein formula.

The further refinement of the theory was developed upon taking account of precisely these two circumstances.

# 2. CALCULATION OF THE COEFFICIENT OF THERMAL SLIP $\mathcal{K}_{\mathrm{TS}}$

The principal simplifying assumption adopted by Maxwell in calculating the coefficient of thermal slip consisted in the idea that the flux of molecules moving toward the interphase boundary  $(J^-)$  and from it  $(J^+)$  are considered to be independent at any distances from the boundary, while the molecules  $J^+$  have the temperature of the wall and  $J^$ have the bulk temperature of the gas.

However, despite the lack of rigor and even the contradiction of this assumption, from the physical standpoint Maxwell's result possesses only one substantial flaw: the value that he obtained for the coefficient of thermal slip does not depend on the character of the interaction of the gas molecules with the interphase surface. The first attempt to improve Maxwell's result was undertaken by Deryagin and Bakanov.<sup>5</sup> For this purpose they used the method of halfspace expansions of Gross and Ziering,<sup>6</sup> which was proposed for solving the Boltzmann equation near the boundary of a gas and a solid surface. Moreover, a new approach was used to solving problems of this type that was based on using the principles of the thermodynamics of irreversible processes (TIP). Although an error was made in deriving the finite formula in Ref. 5, on the whole this approch proved fruitful and subsequently led to solving the problem.

The erroneous result did not remain unnoted and was subjected to criticism.<sup>7</sup> However, the sources of error were pointed out wrongly. Deryagin, Yalamov, and Ivchenko<sup>8</sup> in essence agreed with the position of Brock and perceived the failure of the study in the fact that "the applicability of the reciprocity principle of Onsager in isolation to the Knudsen layer cannot be rigorously grounded." Moreover, they expressed doubt on the possibility of application to the Knudsen layer of a "differential form of the isothermal heat flux." As an alternative the authors solved the direct problem of thermal slip by applying for this purpose the kinetic equation in the form of Bhatnager, Gross, and Krook (BGK)<sup>9</sup> with specularly diffuse conditions of reflection of molecules from the phase boundary. The result of calculating the coefficient of thermal slip agreed with Maxwell's value. It also proved to be insensitive to the degree of diffuseness of reflection. The authors ascribed this to the crudeness of the BGK model.

Later Yalamov, Ivchenko, and Deryagin<sup>10</sup> calculated the velocity of thermal slip, this time solving it by the method of half-space expansions of the Boltzmann equation. They found a weak dependence on the degree of diffuseness  $\varepsilon$ of reflection of the molecules: for  $\varepsilon = 1$  (diffuse reflection)  $K_{\rm TS}$  proved to be  $\approx 0.891$  while for  $\varepsilon = 0$  (specular reflection),  $K \approx 0.865$ .

Sone<sup>11</sup> solved this problem within the framework of the BGK model with diffuse reflection of the gas molecules from the boundary surface. When analyzing the results of this study, and also studies of similar type, one must bear in mind the fact that, when one uses the BGK model, the Prandtl number equals unity, in contrast to the true value 2/3 for monatomic gases. The results of all the studies performed on the basis of the BGK model are recalculated below with account taken of this factor. The author obtained the value 1.149 for the coefficient of thermal slip, i.e., 1.532 times larger than the Maxwell value (see also Refs. 33 and 34). Sone and Yamamoto<sup>12</sup> repeated this result. It was confirmed also

in the study of Loyalka,<sup>13</sup> where the thermal slip of a gas in a cylindrical tube was treated on the basis of the BGK equation (an insignificant difference arose in the fourth digit after the decimal point).

Yalamov, Ivchenko, and Deryagin<sup>14</sup> solved the problem of thermal slip by two methods: in one of them they used the equation in the BGK form, and in the other the Boltzmann equation for Maxwellian molecules. Although the result here should turn out to be the same, the calculation led to different values of  $K_{TS}$ .

In the two limiting cases of specular ( $K_{\rm TS} = 3/4$ ) and diffuse scattering ( $K_{\rm TS} = 1.149$ ). Abramov and Gladush<sup>15</sup> calculated the thermal slip of the gas along the inhomogeneously heated boundary within the framework of the equationin the BGK form. Abramov<sup>16</sup> calculated the thermal slip for an arbitrary specular-diffuse scattering by the same method. The author represented the result in the form

$$K_{\rm TS} = \frac{3}{4} \left( 1 + \frac{\varepsilon}{2} \right) \,. \tag{4}$$

Sone<sup>17</sup> obtained Maxwell's result by solving the BGK equation for almost specular reflection. Kogan and Makashev<sup>18</sup> obtained  $K_{TS} = 1.14$  within the framework of the same model for the case of diffuse scattering. Loyalka<sup>19</sup> solved the problem of slip of a Maxwellian gas on the basis of the Boltzmann equation by a variational method. The value of  $K_{TS}$  proved to be 1.125, which is close to the value obtained earlier by the author within the framework of the BGK model, although somewhat differing from it.

Ivchenko and Yalamov<sup>20</sup> used the ellipsoidal model of Holway<sup>79</sup> to calculate the velocity of thermal slip. They sought the solution in the form of an expansion in a half-space. They obtained the values 1.169 ( $\varepsilon = 1$ ) and 3/4 ( $\varepsilon = 0$ ).

Loyalka and Cipolla<sup>21</sup> obtained the following value for  $\varepsilon \approx 1$  within the framework of the BGK model and a specular-diffuse scheme:

$$K_{\rm TS} = \frac{3}{4} \left[ 1,5324 - 0,5(1 - \varepsilon) \right],$$

while as  $\varepsilon \rightarrow 0$ 

$$K_{\rm TS} = \frac{3}{4} (1 + 0.5756 \varepsilon).$$

Further, by solving this problem by a variational method, Loyalka<sup>22</sup> obtained

$$K_{\rm TS} = \frac{3}{4} \left( 1 + \frac{\varepsilon}{2} \right),$$

repeating the result of Abramov.<sup>16</sup>

Throughout the region of variation of  $\varepsilon$ , the authors of Ref. 21 proposed the interpolation formula

$$K_{\rm TS} = \frac{3}{4} (1 + 0.532 \varepsilon)$$

Onishi<sup>23,26</sup> confirmed this result within the framework of the BGK model. Skakun, Suetin, and Chernyak<sup>24</sup> calculated the coefficient of thermal slip in the diffuse scattering of molecules by using two models: BGK ( $K_{TS} = 1.146$ ) and ellipsoidal ( $K_{TS} = 1.194$ ). Chernyak, Margilevskiĭ, Porodnov, and Suetin<sup>25</sup> (see also Ref. 32) performed calculations for the BGK equation with specular-diffuse boundary conditions by the Bubnov–Galerkin method. The result that they obtained was represented in the form

$$K_{\rm TS} = \frac{3}{4} \left( 1 + \frac{\varepsilon}{2} \right) \,.$$

That is, it also matches the reseult of Abramov.<sup>16</sup> Loyalka, Petrellis, and Storvick<sup>26</sup> undertook a numerical calculation of the velocity of thermal slip for the BGK model (see also Ref. 35) and an arbitrary accommodation of the gas molecules at the surface. The result agrees with Eq. (4) to an accuracy of  $\sim 2\%$ .

Bakanov, Derjaguin, and Roldughin returned in Ref. 27 to using the TIP method for calculating the velocity of thermal slip. The isothermal heat of transport was calculated on the basis of the linearized Boltzmann equation for a model of hard spheres by using a modified method of half-space expansions for an arbitrary specular-diffuse scheme of interaction of the molecules with the phase boundary. They obtained the value  $K_{\rm TS} = 1.18$  for diffuse scattering.

In the framework of the same model, Bakanov<sup>28</sup> calculated the thermal slip of a gas by solving the direct problem of finding the distribution function of the gas molecules near an inhomogeneously heated surface. In the two limiting cases of large and small diffuseness of the scattering at the surface, he obtained

$$K_{\text{TS}} = \frac{3}{4} (1,08 + 0,49 \varepsilon) \quad \text{for} \quad \varepsilon \approx 1,$$
  
$$K_{\text{TS}} = \frac{3}{4} (1 + 0,655 \varepsilon) \quad \text{for} \quad \varepsilon \to 0.$$

Moreover, the same study treated thermal slip over a rather broad range of pressures, including large values of the Knudsen number as calculated with respect to the width of the distance between the plates of the plane-parallel channel.

To summarize what has been said, we can conclude that, for a gas that is not too rarefied, the coefficient of thermal slip for  $\varepsilon = 0$  can be expressed in the most general form by the formula

$$K_{\rm TS} = \frac{3}{4} (1 + A(\varepsilon)). \tag{5}$$

When  $\varepsilon \approx 1$  it acquires the form

$$K_{\rm TS}(\varepsilon) = \frac{3}{4}(a+b\varepsilon),$$

while when  $\varepsilon \rightarrow 0$  we have

$$K_{\rm TS}(\varepsilon) = \frac{3}{4}(1+c\varepsilon)\,.$$

Table I presents the values of the constants *a*, *b*, and *c* and also  $K_{\rm TS}$  ( $\varepsilon = 1$ ) obtained by different authors. In analyzing these data, we should remember that they were calculated for different models of the intermolecular interaction, which cannot fail to affect the magnitude of the numerical values of the coefficients. However, it is important to stress (as will be essential below) that, when  $\varepsilon \approx 1$ —the most widespread case of diffuse scattering—almost all the calculated values of  $K_{\rm TS}$  lie in the range 1.1–1.2. For specular reflection we have  $K_{\rm TS} = 0.75$ . Thus the theoretically possible range of values of  $K_{\rm TS}$  amounts to 0.75–1.2.

The material stated above pertains to the case of slip of the gas along a plane surface. Yet if the gas slips along a curved surface, then generally one must take account of the influence on the slip of the curvature of this surface. This probem was solved independently by two authors,<sup>29,30</sup> who obtained close-lying results.<sup>1)</sup> We should note that this refinement proved inessential to the problem of thermophoresis.

#### 3. THE EFFECT OF THERMAL CONDUCTIVITY

Analysis of the phenomenon of thermophoresis of objects of high heat conductivity shows that the concepts of thermal slip of the gas as the sole cause of TP are evidently insufficient. Actually, if the heat conductivity of the object is large, then its temperature is practically homogeneous. That is, the temperature gradient along the surface is infinitesimally small. Nonetheless, the TP of such objects is observed experimentally, while the effect is almost the same as for objects of low heat conductivity.

In seeking the factors on which the mechanism of TP of objects of high heat conductivity is based, an important con-

TABLE I.

$K_{\rm TS}(\varepsilon = 1)$	a	Ь	c	References	
1,1493	1,0324	0,5	0,5756	[21]	
1,1493	1,0	0,532	0,532	[21,23]	
1,125	1,0	0,5	0,5	[16,22,23,25, 26,32]	
1,1775	1,08	0,49	0,655	[28]	
1,18	1,0	0,573	-	[27]	
1,149	-	-	-	[11,12,15,33, 34]	
0,75	-	-	-	[2,8]	
0,891	-	-	-	[10]	
1,14	-	-	-	[10,18]	
1,146	-	-	-	[24,35]	
1,194	-	-	-	[24]	
0,924	-	-	-	[35]	
1,125	-	-	-	[19]	
1,1493	-	- 1	-	[13]	
1,1494	-	-	-	[36]	
1,169	-	-	-	[20]	

tribution was made by Brock,<sup>37</sup> who proposed taking account in the boundary conditions, along with thermal slip, of viscous slip (which was also first discovered theoretically and calculated by Maxwell) and of the temperature jump at the surface of the object. The latter is proportional to the derivative of the temperature of the gas normal to the surface. It has been studied experimentally and theoretically by Lazarev<sup>38</sup> and Smoluchowsky.<sup>39</sup>

Brock sought the solution in the form of a power series in the Knudsen number  $\text{Kn} = \lambda / R$  (R is the radius of curvature of the object suspended in the gas, and  $\lambda$  is the mean free path of the gas molecules) and obtained a formula which, when  $\text{Kn} \rightarrow 0$ , goes over into the result of Epstein. For objects of high heat conductivity the terms proportional to Kn come to dominate. Brock's formula describes the thermophoresis of objects of high heat conductivity qualitatively better. Later Jacobsen<sup>40</sup> obtained this formula by performing calculations in the third approximation of the Chapman–Enskog method.<sup>41</sup>

Another approach to solving this problem was proposed by Deryagin and Bakanov.<sup>42</sup> It is based on applying the principles of TIP. The formulation of the problem differs from the traditional ones in that one does not fix the temperature gradient in the gas and the slip velocity, as in the direct hydrodynamic problem, but the velocity of relative motion of the object and the gas, the temperature of which at an infinite distance from the object is constant. The relationship between this effect and the velocity of thermophoresis is then found by using the Onsager principle.

The authors treated the nature of these two interconnected phenomena in the presence of isothermal heat transport in a gas caused by inhomogeneity of the pressure p. This effect is known<sup>41</sup> in the kinetic theory of gases. The boundary conditions in this formulation differed from Brock's conditions: the classical conditions of continuity were fixed for the velocity and the temperature, while a jump was assumed for the normal heat fluxes:

$$\varkappa_{i} \frac{\partial T_{i}}{\partial r} = \varkappa_{e} \frac{\partial T_{e}}{\partial r} - \frac{3}{2} \nu \frac{\partial p}{\partial r} .$$

This approach aroused a negative reaction of some specialists.<sup>43</sup> Moreover, the calculations themselves proved not to be free from errors, which were later corrected. Subsequently Derjaguin and Yalamov,<sup>44</sup>who remained within the framework of this concept, supplemented the boundary conditions with a temperature jump as per Smoluchowsky and viscous slip as per Maxwell. As a result they obtained the previous formula with the addition of terms proportional to the Knudsen number, which ultimately proved to be small corrections.

A new impetus to the development of the theory of thermophoresis came from the study of Dwyer.<sup>45</sup> He proposed solving the problem of hydrogasdynamics by using relationships derived from the Grad equation. It is essential that in this approximation not only the heat fluxes depend on the pressure gradient, as was assumed in the study of Deryagin and Bakanov,<sup>42</sup> but also the stresses in the gas are related to the temperature inhomogeneity. Dwyer obtained the boundary conditions by solving the Boltzmann equation and calculating the components of the velocity, temperature, and radial heat flux at the boundary in a 13-moment approximation. The expression given in this study for the TP force, though rather unwieldy, does not differ strongly from the Brock expression in form. One can see this especially well when both expressions are written in the form of a power series in Kn restricted in the first term. However, there is a substantial difference. When  $\kappa_i / \kappa_e \to \infty$ , the velocity of TP according to Dywer acquires the form

$$\mathbf{v}_{\mathrm{TP}} = -\nu \frac{\nabla T_{\infty}}{T} \operatorname{Kn} \left( \frac{3}{2} \beta_1 - \frac{6}{\alpha_3} \right).$$

Here  $\beta_1$  ( $C_i$  in the Brock formula) is a function of the energy accommodation coefficient  $\alpha$ :  $1/\alpha_3$  ( $C_m$  in the Brock formula) is a function of the momentum accommodation coefficient  $\varepsilon$ . We can easily see that, for certain values of  $\alpha$  and E, the velocity of thermophoresis can change sign. Analysis shows that the physical nature of the sign change in the velocity arises from taking account of the thermal stresses in the gas involving the inhomogeneity of the temperature gradient. Such an unusual result aroused objections from a number of specialists<sup>46,76</sup> (see also Ref. 47).

Also Sone<sup>48</sup> arrived at the conclusion of possible existence of negative thermophoresis. He calculated the force acting on an object of high thermal conductivity immersed in a gas, while a small constant temperature gradient was maintained in the gas. In this case the thermal slip in the zero-order approximation in Kn, as was noted above, equals zero owing to the absence in this approximation of a tangential temperature gradient at the surface of the object. However, in the first approximation in Kn it no longer vanishes. Moreover, the mixed derivative of the temperature is taken into account. As a result a phenomenon was discovered theoretically, which the authors called second-order slip. In the absence of other forces this effect leads to motion of the object in the direction of increasing temperature, i.e., to negative thermophoresis. The problem was solved within the framework of the Navier-Stokes-Laplace equations with boundary conditions that were derived on the kinetic level. Namely, the distribution function of the molecules of the gas in the Knudsen layer was found on the basis of using the kinetic equation in the BGK form, and then the velocity and temperature of the gas were calculated in the standard way as functions of the distance from the phase boundary.

Vestner, Kubel, and Waldmann<sup>49</sup> developed another method of obtaining the boundary conditions. They used an approach developed by Waldmann,<sup>50</sup> in which the derivative of the entropy is calculated at the phase boundary. The linear relations between the generalized "fluxes" and "forces," supplemented by the obvious conservation laws, constitute the system of phenomenological boundary conditions for the equations of hydrogasdynamics. They include also corrections generalized by allowing for the rarefaction of the gas in the expressions for the fluxes of heat, momentum, interdiffusion, etc., in the bulk of the gas far from the phase boundary. Thus the boundary conditions automatically take account of viscous and thermal slip, the temperature jumps, and the thermal fluxes at the boundary. The expression obtained by the authors for the thermophoretic force has the form of a ratio of power polynomials in the Knudsen number up to terms containing Kn<sup>5</sup> (with undetermined coefficients,

which were fitted by comparison with the experimental data). The authors paid attention to the importance in principle of taking account of Onsager symmetry in making this choice. In particular, they noted the inadequacy of the scheme of Grad (13-moment approximation) and the Maxwell scheme, in which Onsager symmetry is not obeyed. In this study, for objects of high heat conductivity at small values of the Knudsen number, they also predicted negative thermophoresis.

References 49 and 50 not only returned to using the method of TIP for solving the problem of thermophoresis, but also demonstrated the broad potentialities of applying it to many problems of this type, where the important role is played by the behavior of the gas near the interphase boundary. To be sure, keeping the terms containing high powers of the Knudsen number in the final result arouses objections, and all the more in that at the same time the authors lost sign of an effect involving the jump in the normal derivative of the velocity of the gas (it was taken into account in the study of Sone<sup>48</sup>) that is proportional to the first power of the Knudsen number. These ideas were taken into account in a study by Bakanov and Roldugin,<sup>51</sup> where the boundary conditions were obtained by the TIP method, and also the constants (kinetic coefficients) calculated at the kinetic level were used. The authors a priori restricted the treatment in the calculations to first-order terms with respect to the Knudsen number, while they solved the Navier-Stokes-Laplace equations both within the framework of the classical hydrogasdynamic approach and by the TIP method. Both methods led to identical results for the velocity of TP in the approximation linear in the Knudsen number  $Kn = \lambda / R$ . We emphasize that the Epstein formula is treated in these studies as the zero-order approximation formula in  $\lambda / L$ , where L is the characteristic scale of variation of the gas temperature far from the object. This theory also predicts negative thermophoresis for objects of high heat conductivity for small values of the Knudsen number.<sup>2)</sup>

Sone and Aoki<sup>33</sup> solve the problem of TP by an asymptotic method developed by Sone for an arbitrary mechanism of reflection of the gas molecules from the interphase boundary. In addition to the mentioned effects, they also took account of effects of curvature of the object. The formula derived for the velocity agrees qualitatively with the result of Bakanov and Roldugin<sup>51</sup> (without taking account of the curvature). There is a certain difference in the numerical coefficients—owing to using different models for calculating the kinetic coefficients—for Sone and the Boltzmann equation and the hard-sphere model—for Bakanov and Roldugin.

Negative thermophoresis for  $\varkappa_i / \varkappa_e \ge 1$  and  $Kn \le 1$  was found also in a calculation by Gorelov,<sup>53</sup> where the Boltzmann equation (an integral form) for a hard-sphere model and a diffuse law of reflection from the interphase boundary was solved numerically by the Monte Carlo method.

A numerical calculation of TP by the method of Bubnov and Galerkin based on using linearized kinetic equations with intermolecular-collision operators in the BGK form and an ellipsoidal model was undertaken by Beresnev and Chernyak.<sup>54</sup> In the collisions of the gas molecules with the surface they assumed an arbitrary accommodation of the energy and the tangential momentum. An approximation for the TP force was selected for Kn  $\leq 1$  by processing the numerical results for the S-model with  $\varepsilon = \alpha = 1$ . The calculation also confirmed the possibility of negative TP of objects of high heat conductivity.

Poddoskin, Yuushkanov, and Yalamov<sup>55</sup> found the distribution function of the gas molecules with an arbitrary accommodation law of the energy and momentum by a hydrodynamic method based on using the S-model and the method of half-space expansions. This result was then used to obtain the boundary conditions at the surface of the object. They contain temperature jumps of the object normal to theh phase boundary, and also slip, including second-order slip. The velocity of thermophoresis was calculated.

To summarize the review of the theoretical studies, we can state the following.

The contemporary state of the theory of thermophoresis in gases at small Knudsen numbers describes the velocity of thermophoresis with an expression of the form

$$v_{\rm TP} = -K_{\rm fS}(\epsilon) \frac{\nu}{T_0} \frac{\nabla T_{\infty}}{1 + \frac{\kappa_{\rm i}}{2\kappa_{\rm e}}} (1 + {\rm Kn} \cdot \Phi)$$
(6)

Here  $\Phi$  is some function of the accommodation coefficients of the tangential momentum  $\varepsilon$  and energy  $\alpha$ , as well as the ratio  $\varkappa_i / \varkappa_e$ . In the most general form the results of the calculations of the function  $\Phi(\varepsilon, \alpha, \varkappa_i / \varkappa_e)$  in the different studies can be represented by the formula

$$v^* = -v_{\mathrm{T}\Phi} \frac{2 + (\kappa_i/\kappa_e)}{2\nu \nabla T_{\infty}} T_{\mathrm{o}} = K_{\mathrm{TS}} (1 + \mathrm{Kn} \cdot \Phi).$$

The fundamental property of the function  $\Phi$  in which we are interested consists in the fact that when  $\varkappa_i / \varkappa_e \gg 1$  the first term is the principal one, and the function  $\Phi$  is directly proprotional to  $x_i / x_e$ . The differences in the proportionality coefficients are due to their origin or to the differing hypotheses on the intermolecular and interphase interactions, or to errors in the calculations themselves. Table II gives the numerical values of the stated coefficients for  $\varepsilon = 1$  and  $\alpha = 1$ obtained by different authors. Figure 1 shows the variation of  $\Phi(\alpha)$  (Ref. 51) for  $\varepsilon = 0.8$  (1'-3') and  $\varepsilon = 1$  (1"-3") and for different values of  $\kappa_i / \kappa_e$ : 1 (1', 1"), 10 (2', 2"), and 100 (3', 3"). We see that when  $\alpha > 0.3$  for  $\kappa_i / \kappa_e \approx 1$ , the absolute value of  $\Phi$  is small and it changes sign when  $\alpha \sim 0.5$  $(>0 \text{ when } \alpha < 0.5 \text{ and } < 0 \text{ when } \alpha > 0.5)$ . With increasing  $x_i/x_e$  the variation of  $\Phi(\alpha)$  becomes sharper, and sign change occurs at large values of  $\alpha$ .

When  $\alpha \sim 1$ , according to the theory, the velocity of TP of particles of high heat conductivity should decline with increasing Knudsen number (an exception is the conclusion of Ref. 55), while it should increase when  $\alpha < 0.9$ . Thus a possibility arises in principle of deciding on the magnitude of the energy acommodation coefficient of the gas molecules as they collide with the surface of the particle from the variation of the velocity of thermophoresis at small Kn ( $\rightarrow 0$ ).

### 4. EXPERIMENTAL RESULTS

All the methods known today of measuring the velocity of thermophoresis are rather complex in the practical respect and contain a large number of sources of errors. Moreover, as analysis has shown, the standard approach to proTABLE II.

A	B	С	Ď	Φ			References
				$x_i/x_e = 1$	$\star_i/\star_e = 10$	* <sub>i</sub> /* <sub>e</sub> =100	
-0,45	0,3	-2,07	-1,43	3,06	-9,33	-50,5	[51]
2,26	0,55	-2,18	-0,15	1,023	18,9	221,6	[55]
-0,805	0,368	-1,441	-0,773	-2,29	-11,16	-84,1	[33]
-0,1	4,52	-0,19	-4,91	-2,12	-5,47	-15,19	[34]

cessing the obtained results, as a rule, is inapplicable when one is speaking of comparison with theory. As an example one can cite the comparison in Ref. 40 of the calculated and experimental results. Figure 2, which we have taken from Ref. 40, shows their rather good agreement. However, we shall demonstrate below that it turns out upon more detailed anlaysis that these results diverge considerably more significantly than it seems in Fig. 2.

Let us turn to Eq. (6). We shall rewrite it in the form

$$\Phi(\varepsilon, \alpha, \varkappa_{1}/\varkappa_{e}) = A(\varepsilon, \alpha) \frac{\varkappa_{1}}{\varkappa_{e}} + \frac{B(\varepsilon, \alpha) + C(\varepsilon, \alpha) \frac{\varkappa_{1}}{\varkappa_{e}}}{1 + \frac{\varkappa_{1}}{2\varkappa_{e}}} + D(\varepsilon, \alpha) .$$
(6a)

The graph of the  $v^*(Kn)$  dependence is a straight line whose ordinate for Kn = 0 equals  $K_{TS}$ , while the slope is  $K_{TS}\Phi$ . Now it remains to express from the existing experiments the function  $v^*(Kn)$  and compare the experimental values of  $K_{TS}$  and  $\Phi$  with the theoretical values shown in Tables I and II.

Before we proceed to describe the experiments, it is a propos to make some preliminary remarks. Since the reliable theoretical results were obtained mainly in the region  $Kn \ll 1$ , it is natural to make the comparison with experiment

only at these values of the Knudsen number. Moreover, as is implied by what we have said above, it is proposed to conduct the comparison with experiment primarily by taking the limit of the value of the velocity of TP corresponding to Kn = 0. This defines the decisive role of those data of the experiment that were obtained in the region of values of Kn corresponding to the condition  $Kn\Phi \leq 1$ . For large values of the ratio  $\varkappa_i/\varkappa_e$  this is almost always equivalent to the requirement ( $\varkappa_i/\varkappa_e$ )  $Kn \leq 1$ . Unfortunately the overwhelming number of experimental studies has been performed without taking account of these circumstances and are therefore unsuitable for our analysis. Naturally such data have not been subjected to analysis.

Apparently the first experimental study in which an attempt was undertaken to start a systematic study of TP is that of Fredlund.<sup>56</sup> He measured the effect of the action of a temperature field on a disk suspended on a balance arm. The experiments were performed in different gases: hydrogen, argon, nitrogen, oxygen, and air, in the pressure range from 1 to 2000 dyne/cm<sup>2</sup>. It was established that, for a fixed distance between the plates at different temperatures, the force of TP increases linearly with the pressure, passes through a maximum, and further on declines as  $1/p^2$ .

Eight years later Rosenblatt and La Mer<sup>57</sup> used a Milliken condenser to measure the TP force acting on a drop of tricresyl phosphate in air. The radius of the particle lay in the range 0.4–1.6  $\mu$ m, while the air pressure in the chamber was varied from 45 to 760 Torr, which corresponds to Knudsen numbers starting at 0.035 and on up. We should note that the inequality ( $\chi_i / \chi_c$ ) Kn  $\leq 1$  is not obeyed here.



FIG. 1. Dependence of the function  $\Phi$  on  $\alpha$ .<sup>51</sup>

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FIG. 2. Comparison of the experimental (1) and calculated (2) results.40

Schadt and Cadle studied the thermophoresis of aerosol particles made of a number of substances in air: stearic acid, NaCl, carbonyl iron,<sup>58</sup> and also tricresylphosphate and mercury.<sup>59</sup> The first of these studies used the method of precipitation in a thermoprecipitator at atmospheric pressure. The dimensions of the particles lay in the range from 0.1 to 4.5  $\mu$ m. For the first time a considerable excess (by a factor of 20–40) was found in the measured value of the TP force for NaCl and carbonyl iron over that calculated by the Epstein formula. In the second study the authors used a Milliken condenser. Here the conclusion of the previous study was confirmed for NaCl (30-fold excess). The result for mercury proved to be even higher (50-fold as compared with the calculated value).

Unfortunately in both studies a value of the Knudsen number < 0.1 was not attained even in a single experiment.

The painstaking studies, described in great detail, of Schmitt<sup>60</sup> with a Milliken condenser and particles of various silicone oils and paraffin in argon, nitrogen, CO<sub>2</sub>, and hydrogen ( $x_i/x_e = 1-10$ ) showed good agreement, in the opinion of the author, with the formula for TP of small particles.<sup>61,62</sup> For large oil particles in nitrogen, argon, and CO<sub>2</sub>, the minimum value of the Knudsen number at which measurements were made amounted to 0.05; the number of points corresponding to Kn <0.1 is also rather large. However, since the ratio  $x_i/x_e$  was of the order of 10, the necessary condition for correct comparison with the theory,  $(x_i/x_e)$ Kn <1, was not fulfilled in even one of the experiments. For the oil-hydrogen pair all the measurements were performed at Kn > 0.1.

Thus also these results offer no opportunity to make a correct comparison with the theory.

A Milliken condenser was used also in the study of Jacobsen and Brock.<sup>40</sup> The thermophoretic force acting on an NaCl particle in argon was measured. The gas pressure was varied over the range 200–800 Torr. The radius of the particles amounted to 0.4–1.0  $\mu$ m. Although the Knudsen numbers were small (the minimum value was 0.06), however, owing to the large ratio  $\varkappa_i/\varkappa_e = 250$ , the condition  $(\varkappa_i/\varkappa_e)$ Kn  $\ll$  1 was not satisfied, and one cannot use the results for a reliable comparison with theory.

The merit in these experiments lies in the isothermal measurements. By using them the value was determined of the coefficient of viscous slip. The values of the rest of the parameters entering into the formula for the TP force,<sup>40</sup> which the authors found by fitting to the experimental result, demand serious correction.

Saxton and Ranz<sup>65</sup> used a Milliken condenser to study aerosols of castor oil and paraffin in air at atmospheric pressure. The diameter of the particles lay in the interval from 0.481 to 2.16  $\mu$ m. The minimum value of the Knudsen number amounted to ~0.07. Nevertheless, one cannot use these data for comparison with theory, since the condition  $(\varkappa_i/\varkappa_e)$ Kn  $\leq 1$  is not fulfilled.

A Milliken condenser was also used in the study of Kousaka, Okuyama, Nishio, and Yoshida.<sup>77</sup> They used as the object of study aerosols of tobacco smoke, stearin, and dioctylphthalate in air at atmospheric pressure. The ratios  $x_i/x_e$  amounted to 8.47, 5.08, and 5.08. The Knudsen numbers for each type of aerosol particles were not varied in the course of the experiment and were of the order of 0.1–0.05. Measurements of the TP velocity were performed at

different values of the temperature gradient. The absolute value of the velocity for fixed Kn exceeded by about twofold that calculated by the Brock formula,<sup>37</sup> and was closest of all, in the opinion of the authors, to the result of Deryagin and Yalamov.<sup>78</sup>

The obtained result allows us to consider the apparatus developed by the authors for measuring the TP of aerosols to be promising. One can draw a final conclusion of its productivity after the data of measurements of the  $v_{\rm TP}$  (Kn) have been published.

Tong<sup>67</sup> analyzed the method of measurements using a Milliken condenser. He noted the following defects of this method: errors in determining the dimensions of the particles, their shape, density, charge, and also the presence of convection, and practical restrictions on the minimum dimensions of the particles and the applied pressures. In turn, the author proposed using the method of deflection of a sphere of known dimension, material, and weight, suspended on a thin filament in a vacuum chamber, under the influence of a horizontal temperature gradient to measure the TP force. In Tong's experiments the spheres had dimensions from 1 to 5.1 mm, length of filament 93 cm, and diameter 1  $\mu$ m. The pressure in the chamber was varied from  $10^{-3}$  to 100 Torr. The distance between the plates having different temperatures was 5 and 1.3 cm. The materials of the spheres were: teflon, polyethylene, aluminum, cork, and silicone resin. Special barriers were set up in the chamber to eliminate convection. Existence of convection was not observed at all pressures less than 100 Torr.

The results of measurements for aluminum and cork in helium and air were used in the study. The method applied by the author enabled performing measurements in the range of values of the Knudsen numbers from 0.016 up. However, this proved not to suffice for aluminum. At a ratio  $x_i/x_e = 1580$  (in helium) the value of the parameter  $x_i/x_e Kn$  is considerably greater than unity.

The results of the experiments of Ref. 67 with spheres of cork in helium correspond to all the necessary criteria where our method of comparing theory and experiment can be carried out without any doubts. Figure 3 shows a graph of the  $v^*(Kn)$  relationship. Extrapolation to  $Kn \rightarrow 0$  yields  $v^*(0) = 4.18$ . This result unequivocally indicates the measured magnitude of the effect to be too high. The reasons for this are not clear. Apparently the flaw resides in the very method of measurement or the design of the apparatus.

Davis and Adair<sup>68</sup> performed a series of analogous measurements of the TP force acting on a sphere made of cork in He, HD, Ne, Ar, and N<sub>2</sub>. Unfortunately, in the overwhelming number of these experiments in the region Kn < 0.1, only 2–3 points were obtained. And only in helium  $\varkappa_i/\varkappa_e = 0.3$ ) was it possible to perform a more or less reliable processing of the results of the measurements. The results are presented in Fig. 3. As in the previous study, the value  $v^*(0) = 4.38$ proved to be too high. Moreover, the slope angle of the curve of  $v^*(Kn)$  at the point Kn = 0 proved to be anomalously large<sup>3)</sup> in absolute magnitude ( - 26, 55). All this confirms our assumption of error in the very design of the experiment.

In a special cuvette—model of a channel where motion in the direction of a temperature drop was imposed on the gravitational sedimentation of particles—Deryagin and Rabinovich (see Ref. 63) measured the velocity of TP of particles of tobacco smoke, NaCl, and drops of liquid paraffin



FIG. 3. Dependence of  $v^*$  on Kn. 1—oil-air,  $x_i/x_e = 8, 13, K_{TS} = 1.22;^{57}$  3—cork-helium,  $x_i/x_e = 0.3, K_{TS} = 4.53;^{68}$  5—cork-helium,  $x_i/x_e = 0.3, K_{TS} = 4.38;^{67}$  7—polystyrene-helium,  $x_i/x_e = 0.8, K_{TS} = 2.63;^{72}$  9—polystyrene-neon,  $x_i/x_e = 2.43, K_{TS} = 2.51;^{72}$  2, 4, 6, 8, and 10 are the rectilinear approximation of the results of the measurements.

 $(R = 0.3-0.6 \ \mu\text{m})$  in air at pressures of 30-200 and 760 Torr. The Knudsen numbers layer in the range 0.15-0.4. For the reasons cited above we did not process these data.

Keng and  $Orr^{64}$  studied thermophoresis in a disk thermoprecipitator of original construction. The aerosol particles had about the same diameter of ~1  $\mu$ m. The experiments were performed with air at atmospheric pressure. The Knudsen number amounted to about 0.15. Certain qualitative results are of interest. Thus, it was found that the TP force of practically all the studied aerosols did not depend on the heat conductivity of the particles. Here the latter varied by almost 100-fold, depending on the material of the particles. For our purposes the results of this study cannot be used since the dependence of the TP force on Kn was not obtained there.

Storozhilova and Schcherbina<sup>66</sup> used the method of freely falling drops of transformer oil  $(R = 10 \,\mu\text{m})$  in a vertical slit (H/L = 80), where H is the height, and L is the distance between the walls), with a horizontal temperature gradient, to measure the velocity of TP. Although the measured value of the velocity of TP was close for Kn = 0.006 to the calculated value for Kn = 0, however, this result can be a random coincidence. The point is that no measurements were made in this study at other values of Kn that could substantially increase the reliability of these data.

The jet method proposed by Deryagin and Storozhilova was used to study<sup>69</sup> the TP of aerosol particles of liquid paraffin and table salt in air at a pressure of 160–170 Torr, and also particles of MgO,  $NH_4Cl$ , and tobacco smoke. The results that were obtained are characterized by a very large scatter. The measurements were performed in the region of Knudsen numbers where comparison with the existing theories is not valid.

F. Prodi, Santachiara, and V. Prodi applied an analogous method.<sup>70</sup> They studied an aerosol of NaCl in air. Comparison of these data with the existing theories is also impossible owing to the insufficient number of points for Kn < 0.1and noncompliance with the condition  $(\kappa_i / \kappa_e) Kn \leq 1$ .

Talbot, Cheng, Schefer, and Willis<sup>71</sup> undertook an at-

tempt to study the thermophoresis of particles by measuring their concentration in a flow of aerosol near a heated wall. A flow with velocity of 4 m/s emerged from a nozzle and blew onto a plate heated to 1300 K. Measurements of the velocity and concentration of the particles were performed with an argon laser and an optical system with a photomultiplier. The particles amounted to spheres of aluminum oxide of diameter  $\sim 2 \ \mu m$ . Measurements were performed for Kn = 0.15. The experiments yield no information on the dependence of the effect on the Knudsen number. Hence they cannot be used for our purposes.

Kanki and Juchi<sup>72</sup> performed a large number of measurements by using a highly sensitive torsion balance to which the spherical particles being studied were attached. The materials of the particles used were polystyrene (d = 0.25), glass (d = 0.226 cm), aluminum (d = 0.24 cm), and the gases were argon, neon, and helium at pressures from 0.1 to 100 Torr. The distance between the plates at different temperatures was  $\approx 2 \text{ cm}$ .

Experimentally the appearance of a TP force was established at Kn = 0.005. At Kn ~ 0.5 the force passed through a maximum, and then showed a tendency to decrease. A weak dependence of the TP force on  $\varkappa_i / \varkappa_e$  was noted. Negative thermophoresis was not found.<sup>4)</sup> We have processed the data of the measurements of thermophoresis of polystyrene in helium  $\varkappa_i / \varkappa_e = 0.8$ ) and neon ( $\varkappa_i / \varkappa_e = 2.43$ ) (see Fig. 3). In the former case we have  $v^*(0) = 2.63$ , and in the latter  $v^*(0) = 2.51$ . We should acknowledge both results to be too high. The data on polystyrene in argon, as well as glass in helium, for which the condition ( $\varkappa_i / \varkappa_e$ )Kn  $\ll 1$  is satisfied, contain only 2–3 points. The rest of the measurements, although made at low Kn, have no points at all corresponding to the condition  $\varkappa_i / \varkappa_e$ Kn  $\ll 1$ .

Thus not one of the experiments among these published in the literature up to now, strictly speaking, can either confirm or reject any of the existing theories of thermophoresis linear in Kn. Rather one can state the converse: the theory indicates the inadequacy of the experimental studies. Bakanov, Deryagin, and Roldugin<sup>74</sup> proposed a method of studying TP based on experiments on the thermal polarization of objects in a gas flow. For the case of oil-argon<sup>75</sup> the values  $K_{\rm TS} = 1.2$ ,  $\varepsilon = 0.95$ , and  $\alpha = 0.81$  were obtained by calculation within the framework of the model of Ref. 51. If we purely formally carry out the procedure that we have developed of finding  $v^*(0) = K_{\rm TS}$ , as well as  $\varepsilon$  and  $\alpha$  [from the slope angle of  $v^*({\rm Kn})$ ] from the measurements of Rosenblatt and La Mer<sup>57</sup> (oil-air), we obtain a rather close result:  $K_{\rm TS} = 1.22$ ,  $\varepsilon = 1$ ,  $\alpha = 0.83$ .

We note in passing, if we also formally apply the developed method of comparison with theory to the experiments of Jacobsen and Brock<sup>40</sup> (NaCl-argon,  $\varkappa_i/\varkappa_e = 250$ , Kn = 0.06 and up), which N. A. Fuks considered standard,<sup>76</sup> the result proves highly unsatisfactory:  $v^*(0) = -$  (minus!)3.78. This undoubtedly indicates that either the data of the measurements are far too low, or as we see it, more likely, they are completely unsuitable for comparison with any of the contemporary analytical theories, including Ref. 40, the region of applicability of which is restricted to values Kn <  $\varkappa_e/\varkappa_i$ .

### 5. CONCLUSION

Although the phenomenon of thermophoresis was first observed more than 100 years ago, only in 1929 was Epstein able to establish one of the physical causes of the phenomenon-the thermal slip of the gas along the nonuniformly heated surface of the object. The systematic investigation, which started in 1946 with the study of Rosenblatt and La Mer, soon revealed the imperfection of this theory, especially for objects of high heat conductivity of relatively large dimensions. Brock, Dwyer, Waldman, Deryagin, Bakanov, Roldugin, Sone, and others have contributed to the understanding of the physical nature of thermophoresis. In particular, they have establishd that, along with thermal slip, an important role is played, especially in the case of TP of objects of high heat conductivity and large dimensions, by the temperature jump at the gas-object boundary, temperature stresses, and isothermal heat transport in the gas. In this regard we must not fail to note Refs. 42, 49, 50, and 51 on application of the TIP methods. It was established that the decisive factor in assessing the reliability of any particular theory of thermophoresis is the need of observance of Onsager symmetry. Among the entire spectrum of theoretical studies on thermophoresis, only a few<sup>33,49,51,55</sup> satisfy this condition. In our opinion, it makes sense to subject only these studies to further "triage" by comparison with experiment.

On the other hand, since all the formulas obtained analytically for the case of large particles are suitable only in the first order in Kn  $\leq 1$ , comparison with experiment is justified only for small deviations from the formula of the zero-order approximation (we recall that we adopt as the zero-order approximation of the TP theory the result obtained in the first order in  $\lambda / L$  where L is the characteristic scale of variation of the temperature of the gas, i.e., the dimensions of the measurement chamber). Unfortunately, not so many experimental data have been obtained in this region of Knudsen numbers. Many measurements, although made at low enough Kn, were performed at only one point. This detracts to a considerable degree from their value as an instrument for testing the theory. The situation is especially unsatisfactory with the experimental study of the thermophoresis of large objects of high heat conductivity. In this case the requirement of smallness of deviation from the zero-order approximation reduces to the far more rigorous requirement  $Kn \ll \kappa_e/\kappa_i$ . This condition for objects of high heat conductivity was not satisfied in even one experimental study, the results of which, thus, are only estimates of the quantities being investigated. Naturally they cannot claim a serious role in solving the problem of the reliability of any particular theory.

In the light of the above, we can state that experiments with aerosols, especially those of high heat conductivity, are apparently unpromising for purposes of rigorous quantitative comparison with theory. Great optimism is inspired by the method of measuring the TP force acting on an object of considerable dimensions. However the situation here is as yet unsatisfactory. Analysis of the results of these experiments unambiguously indicates methodological errors in designing them. An appreciable role in distorting the results is played by the influence of the walls of the chamber as a consequence of their finite dimensions. Attempts to take account of or to eliminate this influence have been undertaken. However, a  $v_{\rm TP}$  (Kn) relationship having an explicitly marked maximum undoubtedly indicates precisely this effect.

We should note for the sake of fairness that the elimination of the influence of the walls by simply increasing the dimensions of the chamber involves serious difficulties in compensating for the thermal convection of the gas. Of course, one can avoid this difficulty, if the experiments are performed under conditions of weightlessness.

Another pathway, more realistic today, is to study the thermal polarization of objects in a gas flow.<sup>74,75</sup> The fundamental problem in these experiments consists in measuring small temperature differences. For objects of not too great heat conductivity, such measurements have already been performed.<sup>75</sup> As regards objects of high heat conductivity, further refinement of the method is necessary. In particular, one can attempt to use a battery of thermocouples instead of a single one. In turn, this requires increasing the dimensions of the object of measurement, and as a consequence, using chambers of larger dimensions, pumps of high throughput, etc.

<sup>21</sup> These two methods have also been used by Yalamov and Gaĭdukov.<sup>52</sup> Coincidence of the results of the two calculations was obtained within the framework of the zero-order (Epstein) approximation.

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<sup>&</sup>lt;sup>1)</sup> The calculation of this effect in Ref. 31 proved to be faulty.

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<sup>&</sup>lt;sup>4)</sup> Sutugin and Petryanov-Sokolov report<sup>73</sup> a confirmation of the conclusions of the theory with respect to the possibility of existence of negative thermophoresis. They arrived at this conclusion upon subjecting to analysis the data on the degree of dispersion of deposits of NaCl in a thermoprecipitator and on ultrathin fibers. They established that there is an upper bound of dimensions of the particles that enter the deposit of the thermoprecipitator. Particles of larger dimensions (Kn < 0.3–0.6) do not deposit on the cold walls of the thermoprecipitator, as the authors conjecture, owing to a change in the sign of the thermophoretic force.

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