EXPERIMENTAL STUDY OF THERMOPHORESIS OF AEROSOL PARTICLES

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The velocity of thermophoresis of aerosol particles in the slip flow region, about which no conclusion has yet been drawn from the many results of experimental and theoretical work, was studied from the experimental point of view. A new experimental method using an ultramicroscope was developed to meet most of the necessary conditions to obtain reliable data on thermophoresis, such as accurate observation of velocity under an accurately known temperature gradient and prevention of the action of any forces except thermal force. The experimental results were compared with some of the most representative theories, and were found in good agreement with Derjaguin's theory.

Introduction

Because of the practical interest in particle deposition on heat exchanger tubes and in particle collection by scrubbers and thermal precipitators, as well as the theoretical interest in evaluating the phenomenon, extensive experimental investigations of thermophoresis have been made[5].

In the large-Knudsen number region $Kn \gg 1$, theory and experiment are found to be in satisfactory agreement. In the smaller region or the slip flow region, $Kn \leq 1$, though various theories have been proposed, sufficient reliable experimental data to verify them have not been obtained because of the difficulty in accurate measurement of the velocity of thermophoresis.

This paper presents experimental data on the velocity of thermophoresis in the slip flow region obtained by a new technique developed to determine the accurate velocity of thermophoresis. The data are then compared with some representative theories of thermophoresis[1–3,6,8–10].

Experimental Method

Several experimental methods to determine the velocity of thermophoresis have been developed. Derjaguin examined the available experimental methods which had been reported and classified them into four types[5]. Each of them, however, seems to have some unavoidable faults. In accurate determination of the velocity of thermophoresis, the following conditions should be essentially satisfied: 1) to know the accurate temperature gradient where the velocity of particles is just observed; 2) to prevent the action of any non-thermal forces, such as fluid drag due to fluid flow, photophoretic and electric forces; 3) to avoid convective flow of aerosol induced by the temperature difference[7]; 4) to know accurately the diameter of spherical particles; and 5) to observe the velocity itself directly under the above conditions. Considerable spread in experimental data obtained by different authors is thought to be caused by lack of some of the above conditions. The experimental method presented in this paper was developed so that the above conditions were satisfied as much as possible.

The experimental technique applied in this study is in principle much the same as that previously developed by the authors for size analysis of aerosol particles[11]. The only difference between them lies in the observation cells.

Fig. 1 shows the experimental apparatus. The observation cell fixed on the stage of an ultramicroscope has a water jacket into which cooling water controlled in temperature ranging 0°C to room temperature is circulated to cool the bottom wall of the cell. The bottom wall was made from brass plate which has a large heat capacity. The upper wall of the observation cell consists of a glass plate through which the particles suspended in the cell were observed by the ultramicroscope. The side walls of the cell were made from polyvinyl chloride for thermal insulation. A temperature gradient was formed between the upper glass wall and the bottom one, and its extent was controlled by changing the temperature of the

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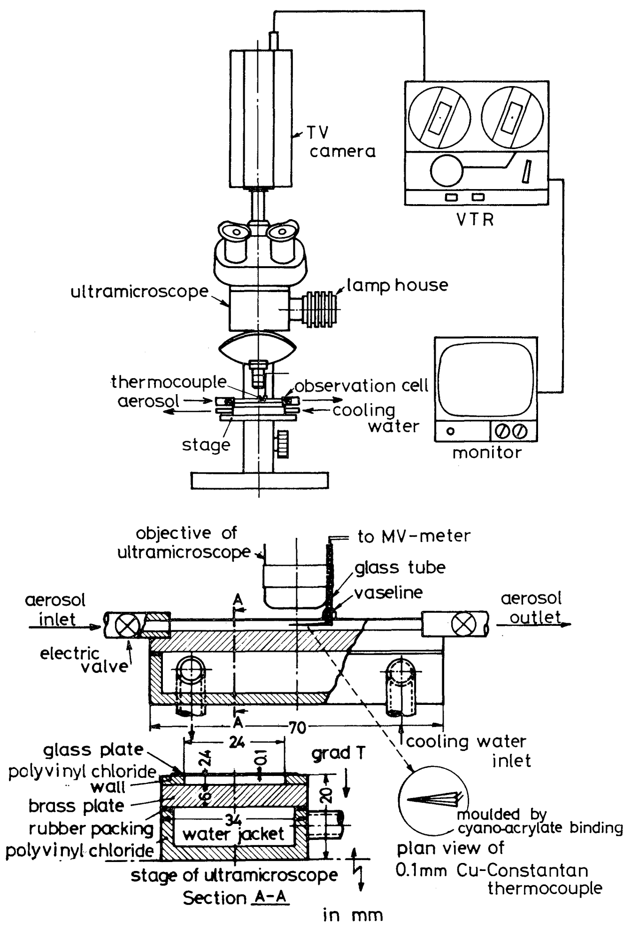

Fig. 1 Experimental apparatus

bottom wall, the upper wall being left at room temperature. The ratio of cell width to height was selected as ten so that convective flow induced by temperature gradient in the cell could be avoided[7].

The aerosol to be observed is cooled by a heat exchanger to the mean temperature in the cell and is introduced into the cell. After several seconds of admitting the aerosol, the flow is instantaneously stopped by closing the electric valves shown in Fig. 1. Then a linear temperature field is formed within a short time throughout the cell except in the vicinity of the side walls. The value of the time interval, τ, needed for the aerosol in the cell to warm up is given by[5]

$$\tau = h_d^2 \rho_f C_p / K_e \tag{1}$$

where C_p is the specific heat of the aerosol, ρ_f density, K_e thermal conductivity of the aerosol and h_d half of the cell depth. In the present case τ comes to 0.05 seconds, which is negligibly small compared with the observation period $t_{1/2}$ described later. Exact solution of this problem can be obtained by an analogical method in solving the establishment of Couette flow, and it gives a still smaller value. Thus the field may be regarded as at steady state.

The focus of the ultramicroscope was set at a given depth h from the inner surface of the upper glass wall by adjusting the height of the stage, on which the cell was fixed, up and down. Thus the particle numbers at various depths was observed. The temperature gradient in the cell, on the other hand, was also preliminarily measured by a small thermocouple in the cell shown in Fig. 1, the depth of which was also adjusted by displacement of the stage. The stage displacement in these measurements was determined by the height gauge installed in the microscope.

Aerosol particles in the cell start to settle just after closing the valves of the cell under the influence of gravity and thermophoresis. The particles appearing in sight of the microscope, which is focused at a certain depth of the cell, h, are recorded by a video recorder as sedimentation progresses until the particle disappears from sight. Knowing the depth h and the time $t_{1/2}$ at which half of the initial particles disappear from sight of the microscope, the settling velocity of a particle having median diameter $D_{p_{50}}$, though it is resultant velocity shown below, is determined:

$$U_G(D_{p50}) + U_T(D_{p50}) = h/t_{1/2} \tag{2}$$

$U_G(D_{p_{50}})$ represents the gravitational settling velocity and is easily obtainable by measurement where no temperature gradient is formed in the cell. Furthermore, $U_G(D_{p_{50}})$ can be converted into the median diameter of the particles, $D_{p_{50}}$, by using the Stokes-Cunningham equation. Thus the values of both $U_G(D_{p_{50}})$ and $D_{p_{50}}$ are accurately evaluated. In consequence the velocity of thermophoresis of a particle of $D_{p_{50}}$ in diameter, $U_T(D_{p_{50}})$, can be determined by observing $t_{1/2}$ under existence of temperature gradient. Eq. (2) is valid when resultant velocity of U_G and U_T increases monotonously with particle diameter. As it is well known that the dependence of particle size on the velocity of thermophoresis is small this condition will be satisfied in most cases unless there exists an extremely large temperature gradient.

Aerosol particles used in this study were tobacco smoke, stearic acid and DOP. Aerosols of both stearic acid and DOP were generated by a La Mer-Sinclair type generator and tobacco smoke was generated by a simple smoking apparatus[11]. Aerosols thus generated were cooled by a heat exchanger and were then observed.

The size distributions of aerosol particles are shown in **Fig. 2**. They were obtained by the ultramicroscopic method[11] using the same cell as shown in Fig. 1 but having no temperature gradient in it.

Experimental Results and Discussion

Figs. 3 and **4** show the experimental results. The temperature gradients measured in the cell are shown at the right side of the figures. They seem to be

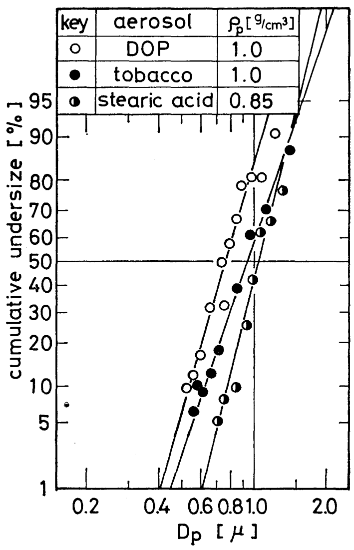

Fig. 2 Size distribution of aerosol particles

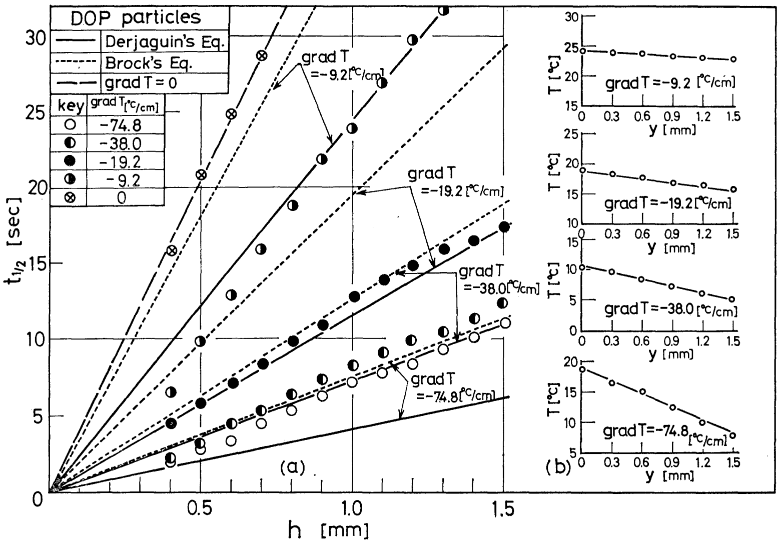

Fig. 3 (a) Relation between $t_{1/2}$ and h (b) Temperature gradient

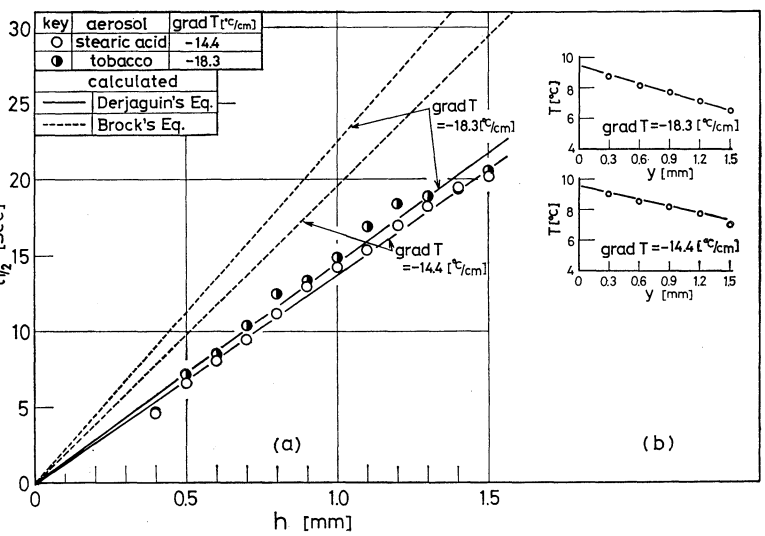

Fig. 4 (a) Relation between $t_{1/2}$ and h (b) Temperature gradient

linear. The abscissa of the figures, y, indicates the depth from an arbitrary position which roughly corresponds to 0.15 mm in actual depth h from the upper wall. In the left-side figure, the relation between $t_{1/2}$ in Eq. (2), at which half of the initial particles disappear from sight of the microscope, and h, the depth from the inner surface of the upper wall of the cell, were plotted. To compare these experimental values with theoretical ones, the following two representative theoretical equations proposed by Derjaguin[3] and Brock[1] were adopted.

$$U_T^D(D_p) = -3\frac{\mu}{\rho T}\left\{\frac{K_e + C_t K_i(2\lambda/D_p)}{2K_e + K_i + 2C_t K_i(2\lambda/D_p)}\right\} \times \left\{\frac{\text{grad } T}{1 + 2C_m(2\lambda/D_p)}\right\} \quad (3)$$

$$U_T^B(D_p) = \frac{1}{2} \times U_T^D(D_p) \quad (4)$$

Since the gravitational settling velocity, $U_G(D_{p50})$, and accordingly the median diameter of the particles, D_{p50}, have been already determined by experiment, substitution of $U_G(D_{p50})$ and $U_T(D_{p50})$ which can be calculated by the above equations knowing D_{p50} and other experimental constants into Eq. (2) gives the theoretical relation between $t_{1/2}$ and h. The solid and dotted lines in Figs. 3 and 4 are those thus predicted by Derjaguin and Brock, respectively. It can be found that good agreement exists between the solid lines calculated on the basis of Derjaguin's theory and the experimental values, except only the case of large temperature gradient, grad $T = -74.8$°C/cm. The experimental data of grad $T = -74.8$°C/cm were obtained under an undesirable condition where hot air was blown onto the surface of the upper wall of the cell to obtain a large temperature gradient. The deviation from Derjaguin's theory in this case is

Table 1 Comparison of experimental results with theories

Aerosol	grad T [°C/cm]	$U_T(D_{p50})$ [cm/sec] Exp.	Eq. (3)	Eq. (4)	notice
DOP	-9.2	2.72×10^{-3}	2.64×10^{-3}	1.33×10^{-3}	$K_e=5.9\times10^{-5}$ [cal/cm·sec·°K][5)]
	-19.2	5.83×10^{-3}	5.51×10^{-3}	2.75×10^{-3}	$K_i=3.0\times10^{-4}$ [cal/cm·sec·°K][4)]
	-38.0	9.90×10^{-3}	1.09×10^{-2}	5.45×10^{-3}	$D_{p50}=0.6\sim0.8$ [μ]
	-74.8	1.16×10^{-2}	2.14×10^{-2}	1.07×10^{-2}	convective flow occurs
Stearic acid	-13.5	3.39×10^{-3}	3.83×10^{-3}	1.91×10^{-3}	$K_i=3.0\times10^{-4}$ [5)]
	-14.4	3.86×10^{-3}	4.08×10^{-3}	2.04×10^{-3}	$D_{p50}=1.04$
Tobacco smoke	-18.3	4.93×10^{-3}	4.82×10^{-3}	2.41×10^{-3}	$K_i=5.0\times10^{-4}$ [5)], $D_{p50}=0.94$

caused by poor temperature control of the upper wall of the cell and also by convective flow of aerosol in the cell owing to the large temperature gradient.

The velocities of thermophoresis in various experimental conditions were determined by the slope of Figs. 3 and 4, subtracting those at zero temperature gradient. The results are shown in **Table 1**. Good agreement is also found between the experimental results and the values calculated by Eq. (3).

In ultramicroscopic observation of particle numbers at a certain depth of the cell, h, it was noteworthy that the particles disappeared suddenly from sight of the microscope at the time $t_{1/2}$ while they disappeared gradually under zero temperature gradient. This sudden disappearance shows the small dependence of particle diameter on the velocity of thermophoresis, as expected from Eqs. (3) and (4).

The particle number concentration of aerosols in experiment was about $4\times10^6\sim8\times10^6$, which corresponds to about $50\sim100$ particles in sight of the microscope. At these concentrations almost no effect of Brownian coagulation on the change in particle number concentration occurs[12)]. The effect of photophoresis by illumination of the ultramicroscope on settling velocity was completely avoided by intermittent lighting.

Conclusion

The velocity of thermophoresis in the slip flow region was studied experimentally. The experimental method presented herein was developed to meet most of the necessary conditions for accurate measurement of thermophoresis, and it gives very reliable data on thermophoresis compared with those so far reported. The results were compared with the theories proposed by Derjaguin and by Brock, and were found in good agreement with Derjaguin's theory rather than Brock's.

Acknowledgment

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Nomenclature

C_m	= tangential momentum first-order slip coefficient[1)] ≈ 1.23	[—]
C_t	= temperature jump first-order slip coefficient[1)] ≈ 2.16	[—]
D_p	= diameter of particle	[cm], [μ]
h	= depth from inner surface of the upper wall of a cell	[cm], [mm]
K_e, K_i	= thermal conductivity of gas and particle, respectively	[cal/cm·sec·°K]
T	= temperature of gas	[°C], [°K]
$t_{1/2}$	= the time when half of the initial number disappear	[sec]
$U_G(D_p)$	= velocity of gravitational settling	[cm/sec]
$U_T(D_p)$	= velocity of thermophoresis	[cm/sec]
y	= depth from an arbitrary position in a cell	[mm]
λ	= mean free path of gas molecules	[cm]
μ	= viscosity of fluid	[g/cm·sec]
ρ	= density of fluid	[g/cm^3]
ρ_p	= density of particle	[g/cm^3]

<Subscript>

50	= for median diameter	

<Superscripts>

D	= for Derjaguin	
B	= for Brock	

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