2710 Technical Notes

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Experimental study on the relation between thermophoresis and size of aerosol particles

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1. INTRODUCTION

In a field with temperature gradient, it is known that masstransfer is also induced by the temperature gradient. When a small particle is suspended in such a field, it will be driven to move toward a colder region. This phenomenon is known as thermophoresis. The study on thermophoresis, which was started at the beginning of this century, has been performed actively by many researchers because of its practical importance $[1-4]$ and theoretical interest $[5-8]$.

In order to elucidate the thermophoretic effect, the basic data are indispensable. However, the measurement of the thermophoretic effect is not so easy. Small particles may be easily moved by various kinds of effects. Distinguishing only the thermophoretic effect from other effects quantitatively needs a rather complicated process which is likely to cause errors. Therefore, in order to get reliable data, it is desirable to perform careful experiments in simple fields where other effects are negligible.

In normal gravity, however, we can hardly eliminate the effects of gravity and natural convection. The gravitational effect changes with the diameter and/or density of the particles and prevents us from simplifying the experiment. Especially, the gravitational effect on large and heavy particles exceeds the relatively small thermophoretic effect and makes the accurate measurement difficult.

The effect of natural convection is a more serious problem in the measurement. The effects of natural convection and thermophoresis are induced simultaneously by the same temperature gradient, and determining them separately and quantitatively by the experiments is quite difficult. It has been the main issue of the experimental studies how the effect of natural convection should be estimated. Because of the reason described above, accumulation of data in the past has been not enough to verify various theoretical results or to predict accurately the behavior of particles.

We successfully avoided these difficulties by establishing an experimental method utilizing microgravity environment, where the effects of gravity and natural convection became negligible [9]. It was found that our experimental method and microgravity environment gave us ideal fields for the measurement. The experimental fields were found to be quasi-steady with monotonous temperature gradient, and the movement of individual particles induced only by the thermophoretic effect could be observed. By performing experiments in microgravity, we are now able to determine the temperature gradient and the corresponding particle vel-

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ocity induced by the thermophoretic effect without the disturbance due to gravitation on particles and natural *convec*tion.

Thermophoresis is thought to be an inherent phenomenon of the interaction between particles and gaseous molecules. Therefore, it is presumed that the effect depends on the Knudsen number (Kn) , which means how the size of particles is close to the mean free path of molecules. The *Kn* for a gasparticle system is a function of radius *R* of the particle and the mean free path λ of surrounding gas and is defined as $Kn \equiv \lambda/R$. The particles in various practical systems have a spread in the size. Therefore, in order to design systems in which the effect is adequately estimated and the behavior of particles is well controlled, it is indispensable to understand the relation between the diameter of particles and the thermophoretic effect. In this study the velocities induced by the thermophoretic effect were measured in microgravity changing the diameter of the particles from 1 to 30 μ m, and the dependence of the thermophoretic effect on the diameter was examined.

2. EXPERIMENTS

2.1. *Experimental method*

The method of the experiments was the same as described in Ref. [9]. Its general reliability was also discussed in detail in the same paper. The microgravity environment was realized by using the drop tower facility of JAMIC (Japan Microgravity Center). This facility provides a microgravity level of 10^{-5} g and duration of 10 s. The measurement field with temperature gradient was established between two metal plates installed horizontally. The upper plate was heated and the lower was left to be the room temperature. The plates were separated at first and the upper plate was heated to the predetermined temperature under normal gravity condition. Then it was forced to approach the cold plate quickly after the microgravity environment was established. The distance between the hot and cold plates during the measurement was set to be 2 mm. The gas in the field was air of atmospheric pressure.

The time duration needed for the temperature distribution between the plates to become steady and that for the initial movement of the gas to decrease to be negligible are approximately equal to δ^2/α and δ^2/ν , respectively. By setting the experimental field to be $\delta = 2 \times 10^{-3}$ m, the time duration for reaching a steady condition is evaluated less than 0.1 s, which is negligible compared to the experimental time duration.

The temperature distribution of the experimental field between the plates was measured by using a Mach-Zehnder interferometer and two sets of thermocouples. The locations of interference fringes were recorded every l/30 s by a CCD camera. The outputs of the thermocouples were processed by a computer every l/40 s. To evaluate the temperature distribution accurately, the interferometer was adjusted so that fringes coincided with isotherms.

The particles were mixed with air flow and introduced to

the experimental field between the two plates. They were illuminated by a He-Ne laser beam, and Mie scattering light from the individual particles was also recorded by the CCD camera. The particle velocity was determined exactly by a slope of the time-location relation obtained from the series of the recorded images.

The test section was very narrow. In order to measure the locations accurately, it was magnified by 1.2 on the CCD surface by a convex lens. Owing to this magnification, the reading error became less than 10 μ m. The experiments were performed confirming that the temperature fields were quasisteady with monotonous gradient and the particles moved with constant speed in the direction opposite to the temperature gradient [9].

2.2 *The particles used in the experiments*

The particles used in the experiments were of $SiO₂$ and PMMA, which are provided by Liquid Gas Co. Ltd and Sekisui Plastics Co. Ltd, respectively. The diameter examined was 1.0, 2.7, 10, and 30 μ m for the SiO₂ particles, and 5.0, 12, and 30 μ m for the PMMA particles. The two materials are different in thermal conductivity. The values are 1.4 and 0.15 W m^{-1} K⁻¹, respectively when the temperature is 300 K. In order to prevent agglutination, dried air was used and the tank in which the particles were stored was vibrated when they were mixed with air flow. The particles are spherical and almost uniform in size.

3. **RESULTS AND DISCUSSIONS**

The results of particle velocity measurements are shown in Table 1. In the table, the material and diameter of the particles which could be known beforehand, and the measured temperature gradient are presented also. In order to make the effect of diameter clear, the velocities corresponding to the temperature gradient of 40 K mm⁻¹ are compared. In the case when the data obtained at that temperature gradient were not available, the values were evaluated by interpolation on the basis of the data obtained at other temperature gradients close to 40 K mm-' and presented in the table.

Figures 1 and 2 show the relations between the diameter and velocity of the $SiO₂$ and PMMA particles, respectively. The solid lines express the present results. The amount of the scattering in measured values is also shown by the error bars. These results indicate that the size distribution of the used particles, which causes the scattering in the results, is narrow especially for $SiO₂$ particles enough to discuss the relation between diameter of particles and thermophoretic effect.

From these figures it is found that the particle velocity induced by the temperature gradient tends to become large as the particle diameter becomes small. It should be noted that smaller particles, on which the effect of inertia, gravity, etc., appear not so conspicuously, receive more remarkably the thermophoretic effect than larger ones.

Those results are compared with the predictions obtained from the Brock equation [5, 10], which is usually quoted in

Material	Particles Diameter D (μm)	Temperature gradient ∇T $(K \, \text{mm}^{-1})$	Particle velocity measured in the experiments U_T $(mm s^{-1})$
SiO ₂	1.0	40.0	1.68
SiO ₂	2.7	22.0	0.58
		80.0	3.00
		$40.0*$	1.03
SiO ₂	10	40.0	0.69
SiO ₂	30	40.0	0.53
PMMA	5.0	31.4	0.55
		49.5	1.14
		$40.0*$	0.80
PMMA	12	31.4	0.40
		42.0	0.73
		$40.0*$	0.66
PMMA	30	31.4	0.35
		49.5	0.78
		$40.0*$	0.53

Table 1. The measured temperature gradient and thermophoretic effect

*These values are obtained by interpolation using above two data.

Fig. 1. The relation between the diameter and velocity of the SiO₂ particles ($\nabla T = 40$ K mm⁻¹). Solid line: measured in the present study. Dotted line : predicted based on the Brock equation [S, IO].

the study of thermophoresis. The particle velocities predicted by the Brock equation are expressed with the dotted lines in the same figures. For SiO₂ particles, as was pointed out in our previous paper [9], the measured particle velocities under the thermophoretic effect were two or three times larger than the predicted ones.

As for the relation between the velocity and diameter, which is of the main concern of this paper, there are differences between the present results and predicted ones, although the tendencies seem to be similar. When the diameter changes from 30 μ m to 5 μ m, for an example, the increase in velocity is 0.23 mm s⁻¹ in the present results and 0.10 mm s^{-1} in the predicted ones. In this case the difference is about twice, and the difference between them becomes larger as the particle becomes smaller.

As for PMMA particles that are of lower thermal conductivity, the measured and predicted velocities are close

Fig. 2. The relation between the diameter and velocity of the PMMA particles ($\nabla T = 40$ K mm⁻¹). Solid line: measured in the present study. Dotted line: predicted based on the Brock equation [5, IO].

compared to those of the SiO₂ particles. However, the dependence of the particle velocity on diameter is quite different. Considering the same example as described above, i.e. when the diameter changes from 30 μ m to 5 μ m, the increase in velocity is 0.27 mm s^{-1} in the present results and 0.06 mm s⁻¹ in the predicted ones. The difference is more than four times and serious compared to the case for $SiO₂$ particles. According to the Brock equation, the dependence becomes small as the thermal conductivity of the particles reduces. This is quite different from the results presented in Figs 1 and 2. Our present results indicate that the particle velocity induced by the thermophoretic effect changes more sensitively with the diameter than ever thought especially for particles of small diameter and/or low thermal conductivity. The results obtained through this study would give us a good reason for restarting theoretical studies on thermophoresis.

4. CONCLUSIONS

The relation between the diameter of particles and thermophoretic effect was examined accurately by getting rid of the particle movement induced by the effects of natural convection and gravity. The particle velocities induced by the thermophoretic effect were measured changing the diameter of particles from 1 to 30 μ m.

It is found that in the extent of experimental conditions in this study the thermophoretic effect depends on the particle diameter. The particle velocity becomes large as the particle diameter becomes small. It should be noted that smaller particles, on which the effect of inertia, gravity, etc. appear not so conspicuously, receive more remarkably the thermophoretic effect than larger ones. Comparing the measured and predicted particle velocities induced by the thermophoretic effect, it is indicated that the particle velocity changes more sensitively with the diameter than ever thought especially for particles of small diameter and/or low thermal conductivity.

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