



Effect of particle morphology on thermophoretic velocity of aggregated soot particles

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ARTICLE INFO

Article history:

Received 30 September 2008
Received in revised form 24 April 2009
Accepted 18 May 2009
Available online 23 July 2009

Keywords:

Thermophoresis
Thermophoretic velocity
Particle morphology
Soot
Carbon black
Dimensionless density
Aggregate

ABSTRACT

A particle under a temperature gradient experiences a force toward the colder region if a particle is very small. This phenomenon is called thermophoresis, a mass transfer phenomenon induced by the temperature gradient. It is well known that the magnitude of the thermophoretic force depends on the size of particle and the temperature gradient. In addition, the magnitude of the thermophoretic force depends on various factors such as the ambient gas's kinematic viscosity and thermal conductivity and the morphology of the particle. To understand thermophoresis in detail, the effects of these factors need to be evaluated. In this study, we accurately measured the thermophoretic velocity of aggregated particles in order to understand the effect of particle morphology. We used carbon black particles of well-defined aggregation parameters to systematically understand the effect of morphology. In addition, we introduced a new optical system to measure the velocity and the size of each particle simultaneously. Five different samples of carbon black particles with different aggregation parameters were used to systematically understand the effect of morphology. The measured thermophoretic velocities were almost proportional to $v\nabla T/T$. The measured dimensionless thermophoretic velocities, $U_T/(v\nabla T/T)$, were much larger than those expected based on the size of aggregates and rather close to those expected based on the primary particle size even when the size of aggregates are larger than 100 nm. This result infers that thermophoretic velocity of an aggregated particle is governed by the primary particle size. The dimensionless density, the ratio of the bulk density to the true density, which represents the overall packing degree of aggregate, is also found to have a significant effect on the thermophoretic behavior.

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1. Introduction

A particle under a temperature gradient experiences a force toward the colder region. This phenomenon is called thermophoresis, a mass transfer phenomenon induced by the temperature gradient. The thermophoretic force on a particle is not negligible when the particle is small and under a large temperature gradient. It is well known that the magnitude of the thermophoretic force depends on the size of particle and the temperature gradient. In addition, the magnitude of the thermophoretic force depends on various factors such as the ambient gas's kinematic viscosity and thermal conductivity and the morphology of the particle. To understand thermophoresis in detail, the effects of these factors need to be evaluated.

As to the particle morphology, spherical solid particles have been used in most previous studies. Here we briefly review results of these studies. Thermophoresis becomes effective in a rarefied gas atmosphere. The rarefied condition is characterized by the

Knudsen number Kn , which is the ratio of the mean free path of ambient gas to the characteristic size of the particle. In previous studies, several researchers proposed theoretical and semi-empirical models to describe thermophoretic effects. When $Kn > 10$, the regime is called the free-molecular regime and the thermophoretic velocity U_T can be estimated by Eq. (1), derived, for example, by Waldmann [1]; U_T is independent of the size of particle.

$$U_T = \frac{-3v}{4\left(1 + \frac{5}{8}\alpha_m\right)} \cdot \frac{\nabla T}{T} \quad (1)$$

where α_m is the accommodation factor ($0 \leq \alpha_m \leq 1$).

When $Kn < 0.01$, on the other hand, the thermophoretic effect becomes negligible (continuum regime). For the transient regime between the two conditions mentioned above, two different equations were proposed by Brock [2] and by Derjaguin and Yalamov [3] to estimate thermophoretic force. Of these, Eq. (2) is the formula proposed by Brock [2],

$$U_T = - \frac{2C_s v \left(\frac{k_g}{k_p} + C_t Kn \right) \frac{\nabla T}{T}}{\left(1 + 2C_m Kn \right) \left(1 + 2 \frac{k_g}{k_p} + 2C_t Kn \right)} \quad (2)$$

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where k_g and k_p are the thermal conductivities of surrounding gas and particle, respectively, C_s , C_m , and C_t are model parameters and 0.75, 1.14, and 2.18 are, respectively, reported as their proper values. The equation proposed by Derjaguin and Yalamov [3] is

$$U_T = - \frac{3\nu \left(\frac{k_g}{k_p} + C_t Kn \right) \frac{\nabla T}{T}}{\left(1 + 2 \frac{k_g}{k_p} + 2C_t Kn \right)} \quad (3)$$

Eqs. (2) and (3) show that the thermophoretic velocities depend on Kn , i.e. the thermophoretic velocities depend on the size of (spherical) particle in the transition regime. Eqs. (2) and (3) contain parameters (C_s , C_m , and C_t). These coefficients can be different for different atmospheres [2] and the choice of values for a particular system requires further investigation. For example, Toda et al. [4] conducted accurate thermophoresis experiments under well-controlled microgravity conditions and reported that the thermophoretic velocities evaluated by Eqs. (2) and (3) are much smaller than the measured values, suggesting the difficulty in predicting thermophoretic velocity.

In addition to the difficulty mentioned above, particles that are affected by thermophoresis are often not spherical but have irregular shapes. For example, soot is aggregate of nano-scale primary particles and each soot particle has an irregular shape. In order to appropriately understand the soot formation and growth processes, the effect of thermophoresis must be taken into account, because these processes usually occur in a combustion field with a steep temperature gradient. Some researchers proposed that the thermophoretic effect on soot formation was significant [5–7]; however, the effect cannot be evaluated in detail because of the lack of basic information and data concerning the thermophoretic effect on particles of irregular shapes. Only few studies [8–16] have been conducted to quantitatively evaluate the thermophoretic effect on irregular-shaped particles.

Theoretical research on the thermophoresis of soot particles by Rosner et al. [8] examined such specific aggregates as chain-like, cyl-

inder particles. They compared their results with experimental studies using the deposition rates of soot on a cooled target. However, measuring deposition rates is an indirect method to determine the thermophoretic effects, as many factors other than thermophoresis affect the deposition rate. Therefore, direct measurement of thermophoretic effects is desirable for adequate evaluation.

In our previous study, direct measurements of the thermophoretic velocities of soot particles were conducted under microgravity conditions [10]. Interestingly, it was found that the measured thermophoretic velocities were much greater than those expected based on the sizes of the aggregated soot particles; with a given temperature gradient the measured velocities were almost constant (independent of the size of particles) and almost equal to that evaluated by a theory for the free-molecule regime (in which the size of particle is much smaller than the mean free path of ambient gas molecules). It was also found that the measured thermophoretic velocities varied among soot samples of different aggregation morphology. These results suggested that the thermophoretic behavior of soot particles was different from that of solid spherical particles and the effect of particle morphology needed to be considered for estimating the thermophoretic velocities. However, the effect of particle morphology of aggregated particles on thermophoretic behavior has not been examined in detail. Furthermore, the thermophoretic velocities and the sizes of aggregated soot particles were not measured simultaneously in the previous studies.

In this study, we accurately measured the thermophoretic velocity of aggregated particles in order to understand the effect of particle morphology. We used carbon black particles of well-defined aggregation parameters to systematically understand the effect of morphology. In addition, we introduced a new optical system to measure the velocity and the size of each particle simultaneously. Using the experimental results, the effect of particle morphology on the thermophoretic behavior of aggregated soot particles is discussed.

Table 1
Properties of carbon black samples.

	Size of primary particle (nm)	Bulk density (g/cm ³)	True density (g/cm ³)	Dimensionless density (bulk density/true density)	Kn of primary particle	Kn of aggregated particles
#40	24	0.14	1.79	0.0783	2.8	7.7×10^{-4} – 6.8×10^{-3}
#44	24	0.18	1.81	0.0996	2.8	4.0×10^{-4} – 1.4×10^{-2}
#45	24	0.19	1.81	0.1048	2.8	4.5×10^{-4} – 1.4×10^{-2}
#30	30	0.13	1.81	0.0719	2.2	3.4×10^{-4} – 6.8×10^{-3}
#33	30	0.16	1.81	0.0884	2.2	4.0×10^{-4} – 6.8×10^{-3}

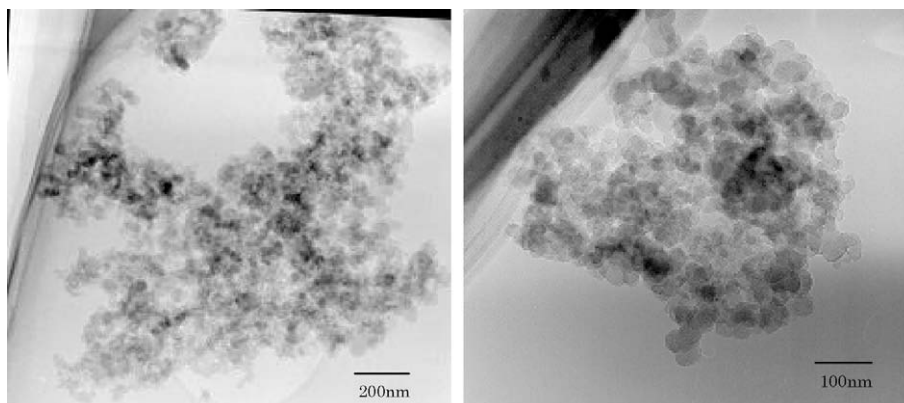


Fig. 1. TEM pictures of samples (sample #44).

2. Experiments

In the experiments, the behavior of each particle of well-defined morphology was observed individually in a field with a temperature gradient, and thermophoretic velocities were systematically measured as basic data of thermophoresis of soot particles. Thermophoretic velocity, one of the basic characters of thermophoresis is the velocity of the moving particle when the thermophoretic force is balanced with the drag force.

2.1. Particles

A soot particle is an aggregated particle of smaller primary carbon particles of nano-scale sizes. In this study, the effect of the packing degree of aggregation and the effect of the size of aggregated particles were examined to understand the effect of particle morphology on thermophoresis. Five different samples of carbon black particles were used. Each carbon black sample was specially produced to have primary particles of a uniform size by oil-furnace methods. The properties of these samples are shown in Table 1. Samples are commercial products made by Mitsubishi Chemical Corporation. Each sample is different in the packing degree of aggregation. The size of primary particles of each sample is almost uniform; 24 nm for three samples and 30 nm for other two samples. Note that each sample contains particles of different aggregation sizes, although aggregated particles of each sample consist of primary particles of a uniform size (TEM pictures shown in Fig. 1). The aggregates have different aggregation sizes but the primary particle sizes of both aggregates are nearly same as shown in Fig. 1. The aggregation size typically ranges from 5 to 180 μm and Knudsen number of aggregates is from 3.0×10^{-4} to 1.4×10^{-2} (Table 1). In this study, the dimensionless density, which is the ratio of the bulk density of the samples to the true density of carbon materials, is introduced to characterize the overall packing degree of aggregation. The calculated values of the dimensionless density of the samples are about 1/10 (Table 1). The bulk density and the true density are measured by a method predetermined by Japanese Industrial Standard (JIS). The dimensionless density, which represents the overall packing degree of aggregation, is used as one of the morphological characters. The size of the aggregates is defined as the average diameter of the aggregates since the overall shape of the aggregates that are observed is almost globular. Then, carbon black particles were dispersed into the air in the measurement field, and aggregated carbon particles of various aggregation sizes can be prepared, consisting of primary particles of an almost uniform size.

2.2. Experimental setting

Thermophoretic velocity was measured by the experimental setting shown in Fig. 2. In the experiments, each particle was observed individually between two aluminum plates (90 mm \times 90 mm, gap distance 2 mm); each plate was set to be at a different temperature. A heater was used to heat up the upper

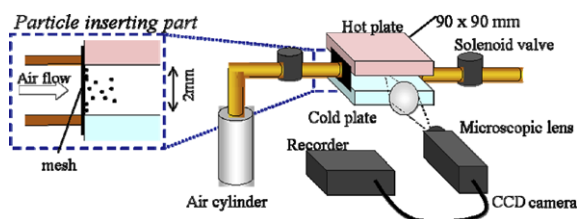


Fig. 2. Experimental setting.

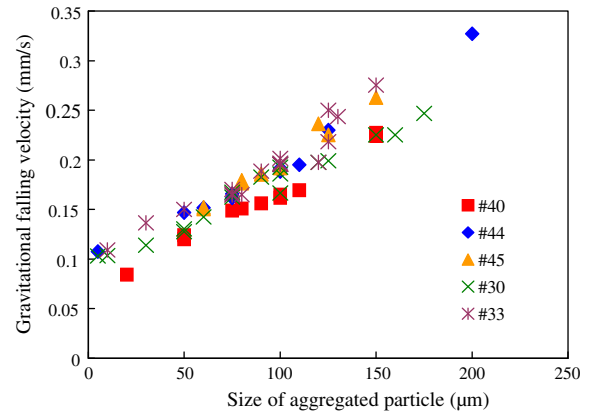


Fig. 3. The relation between the size of aggregated particle and the gravitational falling velocity.

plate, and the lower plate at room temperature was colder. The temperature gradient of each experiment was measured by two thermocouples at two locations of different heights from the plate. Toda et al. [4] and Dobashi et al. [17] investigated temperature profile in this experimental setting using an interferometer optical system. It was found that temperature gradient is constant within the measurement field and therefore it is enough to measure temperatures at two positions to calculate temperature gradient in this measurement field. The temperature gradient was varied from 0 to about 30 K/mm. In a field with a temperature gradient, natural convection must be generated, and the induced convective flow prevents the accurate measurements of thermophoretic velocities. However, the natural convection can be well suppressed in this range of temperature gradient by this experimental setting (hotter plate located at the upper side; narrow gap between two plates; air-tight chamber) [11]. The Grashof number of measurement field is on the order of 10^{-4} , suggesting that the effect of buoyancy is negligibly small and could be ignored.

In the measurements, the carbon black particles were first held on a small mesh plate. To deposit particles on the mesh, it was inserted into a container filled with carbon black particles. Then the carbon black particles were introduced into the field of observation between the two aluminum plates by carrying them with an air stream blown into the field. The pressure of air tank used in measurements is about 3 kgf/cm² and the volume of air tank is 1 L; the flow velocity of air passing through the test section is then estimated to be about 5 cm/s. The carbon black particles were observed by a microscope (resolution: 3–4 μm) using a backlight and recorded by a digital video recorder. This microscopic observation system can realize the simultaneous measurement of the size

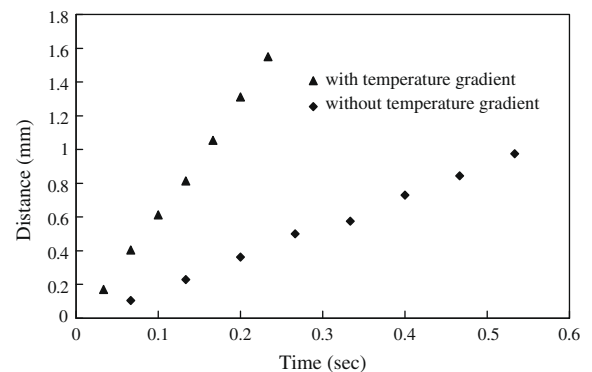


Fig. 4. The measured particle behavior (moving distance versus time), sample #40.

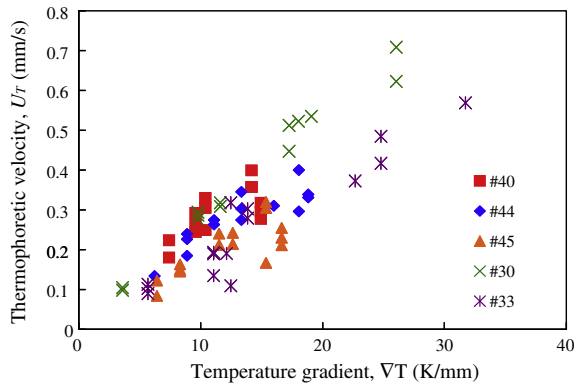


Fig. 5. The relation between thermophoretic velocity and the temperature gradient.

and the moving behavior of aggregated carbon particles. The magnification of microscope and the time of observation were changed due to the size of aggregates; a lower magnification rate was used to increase observation time for observing relatively large particles. The observation area was at the center of the field between the plates. Immediately after a carbon black particle is carried into the observation area with a temperature gradient, it starts to move toward the lower plate (colder plate). The velocity of the moving particle reaches a constant terminal velocity within a sufficiently short period of time. The shock-absorbing time of the particles is around 0.02 s [4,9], which is sufficiently small. The observed constant terminal velocity is the summation of the thermophoretic velocity and the gravitational falling velocity of the particle. Therefore, the thermophoretic velocity can be calculated by subtracting the gravitational falling velocity from the measured terminal velocity. This calculation method was used by Ohi et al. [11], who confirmed that the method reproduced the data obtained under a microgravity condition when the temperature gradient was less than 60 mm/K.

2.3. Measurement of the gravitational falling velocity

It is necessary to measure the gravitational falling velocities to obtain the thermophoretic velocities. At first, experiments with no temperature gradient were performed using the five samples. Fig. 3 plots the gravitational falling velocity as a function of the measured size of the aggregated particles. To obtain the falling velocity, the positions of an individual particle at different times are plotted

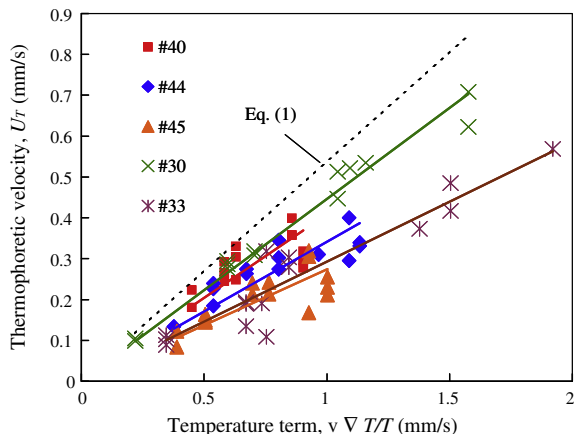


Fig. 6. The measured thermophoretic velocity plotted against the temperature term $v∇T/T$.

in Fig. 4. The gravitational falling velocity is then determined as the steady-state terminal velocity. It was found that, for each sample, the relation between the gravitational falling velocity and the size of aggregated particle was almost one-to-one correspondence. Therefore, the gravitational falling velocity of an aggregated particle can be estimated by its measured size. The estimated gravitational falling velocity was adjusted by considering the change in air viscosity due to the temperature difference. Then, the thermophoretic velocity can be calculated by subtracting the adjusted gravitational falling velocity from the measured terminal velocity corresponding to the size of each aggregated particle.

3. Results and discussions

Fig. 5 shows the measured thermophoretic velocities of five different samples at various temperature gradients $∇T$. The measured thermophoretic velocities are determined when the moving velocity reaches a constant value as shown in Fig. 4. It is found that the thermophoretic velocity increases as the temperature gradient increases. By heating the upper plate up to 318 K, the range of temperature gradient tested in this study is 3.6–31.7 K/mm; and the observed thermophoretic velocity ranges from less than 0.1 mm/s to about 0.7 mm/s. A particle of sample #30, which has the smallest bulk density (Table 1), achieves the highest thermophoretic velocity in this study, 0.71 mm/s, when the temperature gradient of 26.0 K/mm is applied. According to the analysis of the measured data, the error on velocity measurement of this system is about 2–15%. And then the error on the thermophoretic velocity, which is calculated by subtracting the measured gravitational falling velocity from the measured terminal velocity, is estimated as 4–20%, also considering the influence of cancellation of significant digits. It is known that the thermophoretic velocity is influenced not only by temperature gradient but also by temperature itself as well as by the change in gas viscosity associated with the temperature change. During our experiments, the temperature of the measuring position slightly increases as the temperature gradient increases. To clarify the effect of temperature gradient, therefore, it is necessary to adjust the effects of changes in temperature and gas viscosity. Since it is known that the magnitude of thermophoresis depends on the value of $∇T/T$ and the kinematic viscosity $ν$ of ambient atmosphere (air), the measured thermophoretic velocities were plotted against $ν∇T/T$ to compensate the effect of temperature change (Fig. 6). In Fig. 6, the dependence of the thermophoretic velocity on temperature can be analyzed accurately. Fig. 6 shows that the thermophoretic velocity of each sample is nearly propor-

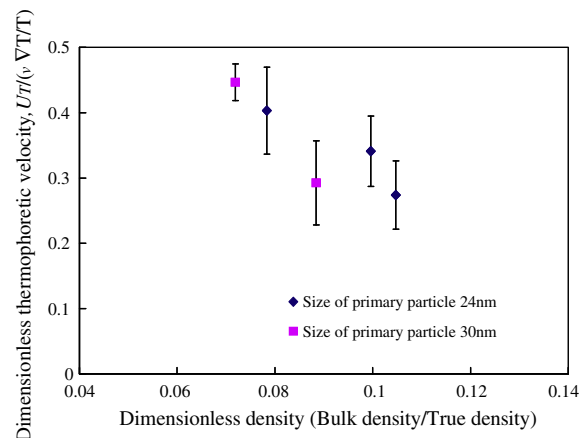


Fig. 7. The relation between the bulk density and the dimensionless thermophoretic velocity (value of the slopes in Fig. 4).

tional to $v\nabla T/T$ with the standard error less than 6.6%. Since each carbon black sample contains particles of different aggregation sizes, Fig. 6 suggests that the aggregation size has a minor influence on thermophoretic velocity. In Fig. 6, the thermophoretic velocity evaluated by the free-molecular-regime theory (Eq. (1)) is shown by dashed line. The thermophoretic velocities of some of samples (#30, and #40) are close to the free-molecular-regime velocity, a result similar to our previous studies [9,10], which showed that the thermophoretic velocities of aggregated particles with open structures (corresponding to low bulk densities) almost agreed with the free-molecular-regime velocity. We then suggested that the thermophoretic velocity of aggregated particles was determined by the size of primary particles. Fig. 6, however, shows that the thermophoretic velocities of other samples (#33, and #45) are considerably lower than the free-molecular-regime velocity, indicating that the thermophoretic velocity of aggregated particles is affected not only by the size of primary particles but also by the morphological characteristics of each aggregated particle, for example, bulk density.

The slopes of the lines shown in Fig. 6, $U_T/(v\nabla T/T)$, dimensionless thermophoretic velocity, are different for different samples. It is thought that the dimensionless thermophoretic velocities depend on the morphological characters of each aggregated particle. To examine the effect of the morphological character on thermophoresis, the measured (dimensionless) thermophoretic velocities are plotted against the dimensionless density in Fig. 7. Error bar

in Fig. 7 indicates the standard deviation of each sample. Fig. 7 shows that the higher the dimensionless density is, the slower the dimensionless thermophoretic velocity is. The dimensionless density represents the overall packing degree of aggregation. It is thus found that the overall packing degree of aggregation affects the thermophoretic velocity of the aggregated particles. An increase in dimensionless density means that the aggregated particle becomes of a closer structure. Therefore, the thermophoretic velocity becomes slower as the aggregated particles are of a closed structure. It indicates that the packing degree of aggregation is an important factor for the thermophoretic behavior of aggregated particles. This graph shows the qualitative relationship between the thermophoretic velocity and the overall packing degree of aggregation. More quantitative evaluation of the relationship would involve the geometrical characterization of individual particles, and such results will be reported in a later work. It is thought in Fig. 7 that the effect of primary particle size (24 nm and 30 nm) is minor, possibly because the primary particle sizes are in the free-molecular regime, in which the thermophoretic velocity is independent of particle size (see Eq. (1)).

To examine the effect of the size of aggregated particles, the dimensionless thermophoretic velocity, $U_T/(v\nabla T/T)$, was plotted against the size of aggregated particles (Fig. 8(a)). In Fig. 8(a), thermophoretic effect on carbon black particles is shown in the range of the size of aggregated particles from 10 to 180 μm . For the purpose of reference, the data of spherical solid carbon particle [18] are also shown in Fig. 8. Fig. 8 includes Eqs. (1)–(3). Here the values of k_g and k_p are 0.0241 and 129 W/m K, respectively. As is well known (see Eqs. (2) and (3)), the dimensionless thermophoretic velocity of spherical solid particle decreases as the particle size increases and is far smaller than that of aggregated particle at the same size. On the other hand, no clear dependence of the thermophoretic velocity on the size of aggregated particles is found in this range (though a slight increase in thermophoretic velocity can be seen with an increase in aggregate size, brief discussion on which is given below). It seems that the dimensionless thermophoretic velocity of each sample is at an almost same level even though the aggregation size widely varies. Moreover, it is also found that the thermophoretic velocity does not decrease even if the size becomes more than 100 μm . When a particle is spherical and solid and its size is 100 μm , the particle is in the continuum regime and the thermophoretic effect on it is usually negligible. Figs. 5 and 6 indicate that the thermophoretic velocity depends only weakly on the size of aggregated particles but significantly on their morphological characteristics.

All data obtained in this study are plotted in Fig. 8(b) with the curves estimated by Eqs. (1)–(3), in which the horizontal axis is Kn and log scale. The representative length scale used when calculating Kn is the size of the aggregated particles. The size of aggregated particles is distributed from the transition regime to continuum regime. It is found that the measured thermophoretic velocities for the aggregated carbon particles are much faster than expected by transition-regime equations (Eqs. (2) and (3)), and close to the velocity for particles in the free-molecular regime. Even if the size of aggregated particle is more than 100 μm or $Kn < 0.0015$, which corresponds to the continuum regime, the measured thermophoretic velocities are close to the velocity for particles in the free-molecular regime. This is a notable result because thermophoretic effect is usually negligible for spherical solid particles larger than 100 μm . This result infers that the thermophoretic velocity of an aggregated particle is governed not by the size of aggregated particle but by the size of primary particle within this range of the size of aggregated particles, as the sizes of primary particle of the carbon black samples are about 24 nm and 30 nm (Table 1) and almost correspond to the free-molecular regime. In addition to the size of primary particles, the dimensionless density,

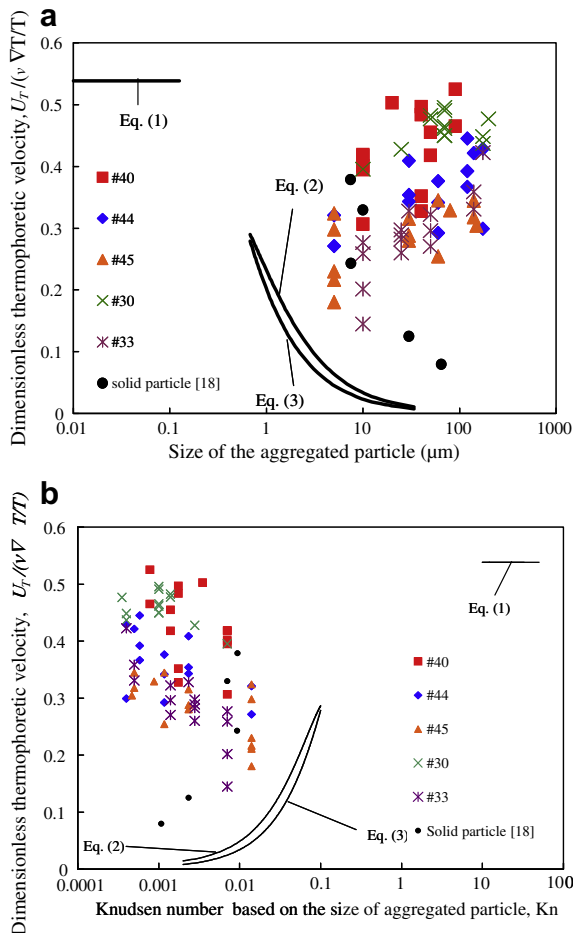


Fig. 8. (a) The relation between the size of the aggregated particle and the dimensionless thermophoretic velocity. (b) The relation between the size of the aggregated particle and the dimensionless thermophoretic velocity; the horizontal axis is log scale.

the ratio of bulk density, which represents the overall packing degree of aggregate, to the true density, has a significant effect on the thermophoretic behavior as discussed above.

Fig. 8(b) also shows that dimensionless thermophoretic velocity slightly decreases with an increase in the Knudsen number. This result suggests that the dependence of thermophoretic velocity of aggregates on aggregate size is different from that of spherical particles (Eqs. (2) and (3)). This phenomenon may be related to the theoretical finding by Rosner et al. [8], who studied the thermophoretic velocity of chain-like particles and showed that the thermophoretic velocity increases with an increase in chain length, a similar result to the present study. The present paper focuses on the effect of the dimensionless density (bulk density/true density), and the effect of aggregate size (minor as compared with the dimensionless density) will be a topic of future study.

4. Conclusions

In this study, an experimental method was developed to simultaneously measure the velocity and the size of aggregated soot particles. Experiments were performed to examine the effect of particle morphology on the thermophoretic velocity. Five different samples of carbon black particles with different aggregation parameters were used to systematically understand the effect of morphology. The measured thermophoretic velocities were almost proportional to $v\nabla T/T$. The measured dimensionless thermophoretic velocities, $U_T/(v\nabla T/T)$, were much larger than those expected based on the size of aggregates and rather close to the velocity for particles in the free-molecular regime, even if the size of aggregate is more than 100 μm . This result infers that thermophoretic velocity of an aggregated particle is governed by the primary particle size (corresponding to the free-molecular regime). The dimensionless density (the ratio of the bulk density to the true density), which represents the overall packing degree of aggregate, is also found to have a significant effect on the thermophoretic behavior.

Acknowledgments

This work was supported by the Global COE Program for Chemistry Innovation, MEXT, Japan. A part of this work was conducted in Center for Nano Lithography & Analysis, The University of Tokyo, supported by MEXT, Japan.

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