Evaluation of morphological effect on behavior of aggregated particles by thermophoresis

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1 Introduction

Soot is a main pollutant emitted from combustion devises. It is important for appropriate emission control of combustion devises to understand the behavior of soot particles in combustion fields. If the size of a particle is very small, that is, as small as the mean free path of the ambient gas, the 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. Soot is a very small particle formed near a combustion field, where a very steep temperature gradient exists. In order to appropriately understand the soot formation process in a combustion field, the effect of thermophoresis must be taken into account. Some researchers proposed that the thermophoretic effect on soot formation was significant [1-3]. Only few studies [4-9] have been conducted to quantitatively evaluate the thermophoretic effect. Since soot particles are aggregates of smaller primary carbon particles of nano-scale sizes, the thermophoretic behavior of soot particles was found to be different from that of solid spherical particles [9]. However, the factors that affect the thermophoretic effect of soot particles have not been analyzed in detail.

In our previous studies [10,11], Suzuki et al measured thermophoretic velocities of five different carbon black samples and examined the effects of bulk density and DBP absorption on the magnitude of thermophoresis. Although some qualitative relationships were found between these parameters and thermophoretic velocity, more detailed observations are needed in order to evaluate the morphological effect on thermophoresis of an aggregated particle quantitatively.

In this study, we examine the effect of particle morphology on the thermophoretic behavior of an aggregated carbon particle in detail. In the measurements, fourteen different types of carbon black samples are used, whose morphological characteristics vary from each other. A new parameter, dimensionless density defined as the ratio of the bulk density to the true density of each sample, is proposed to correlate measured thermophoretic velocities.

2 **Experiments**

Fourteen different samples of carbon black particles were used. Each carbon black sample was specially produced to have primary particles of a uniform size. The characteristics of them are shown in Table 1. Each sample is different in morphological characteristics. Table 1 includes the newly proposed morphological factor, dimensionless density ([bulk density]/[true density]). It will be shown

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that the dimensionless density is a morphological factor that directly influences the thermophoretic behaviors of aggregated particles, regardless of their type.

In the experiments, each particle was observed individually between two aluminum plates (Fig.1); each plate was set to be at a different temperature. The temperature gradient of each experiment, varied from 0 to 20 K/mm, was determined by measuring the temperatures by thermocouples at two different heights from the plate. The natural convection can be well suppressed by this experimental setting in this range of temperature gradient [12]. Carbon black particles on a screen mesh were introduced into the field of



Fig.1 Schematic of measurement field

observation between the two plates by carrying them with an air stream. The carbon black particles were observed by a backlight using a microscope (resolution: about 3-4 μ m) and recorded by a digital video recorder, realizing the simultaneous measurements of the moving behavior and the size of each aggregated particle. Immediately after a carbon black particle is carried into the observation area, it starts moving downward. The velocity of the moving particle reaches a constant terminal velocity within a short period of time. This constant terminal velocity is the summation of the thermophoretic velocity can be calculated by subtracting the gravitational falling velocity form the measured terminal velocity.

At first, experiments with no temperature gradient were performed. It was found that, for each sample, the relation between the gravitational falling velocity and the size of an aggregated particle, the whole size of aggregated particle, was almost one-to-one correspondence. The particle is almost round on the screen and its diameter is used as 'the size of aggregated particle' Therefore, the gravitational falling velocity can be estimated by the measured size of the aggregated particle. The estimated gravitational falling velocity was adjusted by considering the change in air viscosity due to the temperature difference.

	Size of primary particle (nm)	Bulk density (g/cm ³)	True density (g/cm ³)	Dimensionless density	Kn of primary particle
А	16	0.15	1.77	0.0846	4.25
В	16	0.20	1.69	0.1184	4.25
С	20	0.11	1.79	0.0614	3.40
D	21	0.17	1.81	0.0941	3.24
Е	24	0.14	1.79	0.0783	2.83
F	24	0.18	1.81	0.0996	2.83
G	24	0.19	1.81	0.1048	2.83
Н	26	0.13	1.81	0.0718	2.62
Ι	28	0.10	1.80	0.0557	2.43
J	30	0.13	1.81	0.0719	2.27
K	30	0.16	1.81	0.0884	2.27
L	42	0.10	1.84	0.0545	1.62
М	78	0.26	1.87	0.1389	0.87
Ν	80	0.28	1.87	0.1497	0.85

Table 1: Characteristics of samples

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3 Results and Discussion

It is known that the thermophoretic velocity of a solid spherical particle is proportional to $v\nabla T/T$ as follows:

$$U_T = K_{th} \frac{v \nabla T}{T} \tag{1}$$

where K_{th} is thermophoretic coefficient, v is the kinematic viscosity of surrounding gas. Waldmann [6] theoretically showed that K_{th} ≈ 0.55 for solid spherical particles in the free-molecular regime. In the transient regime between the free-molecular and the continuum regimes, K_{th} is a function of the Knudsen number (mean free path/particle diameter) [4, 5]. As for aggregated particles, Tsai et al [13] found that Eq. (1) holds and $K_{th} \approx 0.55$ for soot particles of aggregate sizes in the free-molecular regime. Ono et al [9] tested soot particles with opener structures (lower packing degrees of aggregation) of aggregate sizes in the transient and continuum regimes, and obtained similar results to [13]. Our target is similar to [9], but we test particles of different packing degrees of aggregation. Goals of this study are to validate Eq. (1) and to find major parameters on which K_{th} depends for these particles.

The measured thermophoretic velocities are plotted against $v \nabla T/T$ in Fig. 2. Similarly to [13] and [9], the thermophoretic velocities of each sample are proportional to $v \nabla T/T$.





Unlike these previous studies [9, 13], however, K_{th} (slope of the best-fitting line in Fig. 2 for each sample) is lower than the free-molecular-regime value, a difference due to the particle morphology.

Here we explain the newly proposed morphological factor of aggregates, that is, dimensionless density. The bulk density represents the overall packing degree of aggregation, whereas the true density represents the density of the material itself. To eliminate the effect of different true densities of tested samples (see Table 1), the bulk density was normalized by the true density, yielding the dimensionless density. Based on SEM and TEM observations, a relationship is found that the structure of particles becomes closer as the dimensionless density increases.

Fig. 3 shows that the dimensionless density can well correlate the thermophoretic velocities of different samples; the greater the dimensionless density is, the smaller the thermophoretic coefficient is. As the dimensionless density decreases, the thermophoretic coefficient seems to approach the value for the free-molecular-regime, $K_{th} = 0.55$, suggesting that the smaller dimensionless density leads the behavior of an aggregated particle which is even in the continuum regime (based on the aggregate size) to that of the small, primary particle which is in the free-molecular regime. As the dimensionless density increases, K_{th} approaches zero. This means that aggregated particles of large dimensionless densities behave similarly to spherical solid particles of the diameter equal to the aggregate size. These relations indicate that the dimensionless density can be used to evaluate the morphological effect on thermophoresis of aggregated particles, regardless of the particle type.

4 Conclusions

The velocities and the sizes of aggregated carbon particles were simultaneously measured to examine the effect of particle morphology on the thermophoretic velocity. Fourteen different carbon black samples with well-defined aggregation characteristics were used to represent soot particles. The measured velocities were almost proportional to $v\nabla T/T$. However, K_{th} is different among samples. This difference is attributed to the particle morphology of each sample. A new factor, dimensionless density ([bulk density]/[true density]) is proposed as a morphological factor of an aggregated particle. It increases when the structure of particles becomes closer and can be used to evaluate the morphological effect on thermophoresis of an aggregated particle, regardless of the particle type; the higher the dimensionless density is, the smaller the thermophoretic coefficient is (i.e., the slower the thermophoretic velocity is). As the dimensionless density decreases, the thermophoretic coefficient seems to approach the value for the free-molecular-regime, K_{th} =0.55. The smaller dimensionless density leads the behavior of an aggregated particle of aggregate size in the continuum regime to be similar to that of the small, primary particle, which is in the free-molecular regime.

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