Effect of particle morphology on the thermophoretic behavior of soot particles

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Introduction
Soot is one of the main pollutants emitted from combustion devises. It is important for appropriate emission control of combustion devises to understand behavior of soot particles in combustion field. If the size of particle is very small, smaller than the mean free path of the ambient gas, the particle in a field with a temperature gradient experiences a force toward colder region. This phenomenon is called thermophoresis, which is mass transfer phenomenon induced by temperature gradient. Soot is very small particle and formed near combustion field, where very steep temperature gradient exists. In order to appropriately understand the soot formation process in combustion field, the effect of thermophoresis must be taken into account. Some researchers proposed thermophoresis has significant effect on soot formation process [1-3]. However, the effect cannot be analyzed in detail because of the lack of basic information and data concerning the thermophoretic effect on soot particles. Only little studies [4-9] have been done to investigate the quantitative effects of thermophoresis. In our previous study, Ono et al. measured the thermophoretic velocities of soot in microgravity conditions [9]. It was found that the measured velocities of soot particles were much more rapid than those expected by the sizes of aggregated soot particles. It was also found that the measured velocities of soot particles are almost equal to those evaluated by the formula of Waldmann. In the study, the thermophoretic velocities and sizes of aggregated soot particles could not be measured simultaneously. The measured velocities for the soot of various aggregated sizes under the same temperature gradient are almost same, and then it was concluded that the thermophoretic velocities are almost independent of the size of the aggregated soot particles used in the experiments. Therefore, the effect of the sizes is still unclear in detail. In the present study, the velocity and size of each particle are measured simultaneously. Using the experimental results, the effect of particle morphology on the thermophoretic behavior of aggregated soot particles was examined.

Experiments
Soot is aggregated particle consist of very small primary carbon particle of nano scale size. Because thermophoresis is the phenomenon between gas molecule and particles, many elements have effect on thermophoresis. In this study, the effect of the aggregated size of the soot particles was examined to understand the effect of particle morphology on thermophoresis. Carbon black particles are used as the sample of soot particles, which are specially manufactured to have uniform size (about 24 nm) of primary particles. Dispersing
the carbon black particles into air, aggregated carbon particles of variety of aggregated sizes can be made, which consist of the primary particles of almost uniform size.

The thermophoretic velocity was measured by the experimental setting shown in Fig. 1 [9]. In the experiments, each particle was observed individually between two aluminum plates (90 mm × 90 mm, distance 2 mm); each plate was set to be different temperature. A heater was set in the hotter plate of upper side. The temperature gradient of each experiment can be determined by measuring the temperatures by thermocouples at two positions of different heights from the plate. The temperature gradient was varied in the range from 0 to 20 K/mm. The natural convection can be well suppressed by this experimental setting in this range of temperature gradient [10].

In the measurements, the carbon black particles on the mesh were introduced into the observed field between the two plates by carrying with blowing air into the field. The carbon black particles were observed by a backlight using a microscope (resolution: about 3-4 µm) and recorded by a digital video recorder. This microscopic observation can realize the simultaneous measurement of the size and moving behavior of aggregated carbon particles. Observed area was the center of the field between the plates. Just after the carbon black particle is carried into the observed field with a temperature gradient, it starts to move toward the lower plate (colder plate). The velocity of the moving soot particle reaches a constant terminal velocity within sufficiently short time. This constant terminal velocity corresponds to the summed velocity of the thermophoretic velocity and the terminal velocity of the measured particle. Therefore, the thermophoretic velocity has to be calculated by subtracting the terminal velocity in the field without temperature gradient from the measured terminal velocity. At first, experiments in the field without temperature gradient were performed. The measured terminal velocities in the field without temperature gradient for the particles of aggregate size up to 350 µm were shown in Fig. 2. It is found that the relation between the measured terminal velocities in the field without temperature gradient and the size of aggregated particles is almost one-to-one correspondence. Therefore, the terminal velocity in the field without temperature gradient can be estimated by the measured size of the aggregated particle.

Results and Discussions

Fig. 3 shows the measured thermophoretic velocities with the temperature gradient \( \nabla T \). It is
found that the thermophoretic velocity is increasing as the temperature gradient increases. The thermophoretic velocity reaches 6.2 mm/s at $\nabla T = 18.4$ K/mm.

It is known that the thermophoretic velocity of solid spherical particle is proportional to $\nu \nabla T / T$ and the measured thermophoretic velocities were plotted on the graph against $\nu \nabla T / T$ (Fig.4). Fig. 4 represents that the measured thermophoretic velocity is almost proportional to $\nu \nabla T / T$ even for the aggregated particles, whose aggregate seize are in the range from 10 to 300µm. In our previous study [9], it was found that the thermophoretic velocities of aggregates with open structure almost agree with those evaluated by the formula proposed by Waldmann [6] for the free-molecular regime,

$$U_T = -\frac{3\nu}{4(1 + \frac{\pi}{8} \alpha_m)} \frac{\nabla T}{T}$$

where $\nu$ is the kinetic viscosity of surrounding gas, $T$ is the temperature of surrounding gas, and $\alpha_m$ is the accommodation factor. This equation is indicated as a solid line on the graph. It is shown that the measured thermophoretic velocities in this study also agree with those evaluated by the formula of Waldmann even if the size of aggregated particle is far larger than the size corresponds to free-molecular regime. To examine the effect of the size of aggregated particles, $U_T / (\nu \nabla T / T)$, which corresponds to the slope of the line on Fig.4, was plotted against the size of aggregated particles (Fig.5).

Thermophoresis becomes effective in rarefied gas atmosphere. The rarefied condition is characterized by using Knudsen number Kn, which is the ratio of the mean free path of ambient gas to the characteristic size of the particle. When $Kn > 10$, the regime is called the free-molecular regime, the thermophoretic velocity can be estimated by eq. (1) and does not depend on the particle size. When $Kn < 0.01$, the thermophoretic effect becomes negligible (continuum regime). Between these two regimes, some formulae were proposed to estimate the thermophoretic velocity for solid spherical particles [4, 5]. The value of $U_T / (\nu \nabla T / T)$ calculated by the formulae by Waldmann, Derjaguin
and Brock are indicated in Fig. 5. It is seen that the measured thermophoretic velocities for the aggregated carbon particles are much faster than expected and almost same as the velocity for the particle of free-molecular regime, even if the aggregate size is so large as 300 µm. This result suggests that thermophoretic velocities of aggregated particles must be governed by not the aggregate size but rather the primary particle size, because the primary particle size of the carbon black is about 24 nm (Kn = 2.8) and this size almost corresponds to free-molecular regime.

In Fig. 5, thermophoresis is still effective even if the size of aggregated particle is in the range from 100 to 300 µm, where generally thermophoretic effect becomes negligibly small. No distinct dependence on the size of aggregated particles is found in the range of the size from 10 to 300 µm. The measured data of thermophoretic velocity spread widely. The reason of this data spread is considered to be the other morphological factors than the sizes of aggregate and primary particle, as the primary particle size is almost uniform in the experiments.

Conclusions

In this study, simultaneous measurements of the velocity and the size of aggregated soot particles were performed to examine the effect of particle morphology on the thermophoretic velocity. As the sample particle of soot, carbon black was used, which consists of the primary particles of almost uniform size (about 24 nm). The measured thermophoretic velocities were much larger than expected by their aggregate size. The measured thermophoretic velocities are about 0.01-0.14mm/s at the temperature gradient from 2 to 20 K/mm and almost same as the velocity evaluated by the formula for the free-molecular regime proposed by Waldmann even if the size of aggregated particles is very large (300 µm). Thermophoresis on soot particle is still effective even if the size of aggregated particle is in the range from 100 to 300 µm, where generally thermophoretic effect becomes negligibly small. No distinct dependence on the size of aggregated particles is found in this study. The data spread of measured thermophoretic velocity suggests that there should be other morphological factors than the sizes of aggregate and primary particle.

References