International Journal of Heat and Mass Transfer 52 (2009) 4695–4700

Contents lists available at ScienceDirect

International Journal of Heat and Mass Transfer

journal homepage: www.elsevier.com/locate/ijhmt

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 mm. 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.

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.

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

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

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],

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

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doi:10.1016/j.ijheatmasstransfer.2009.05.017

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

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

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, cylinder 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}

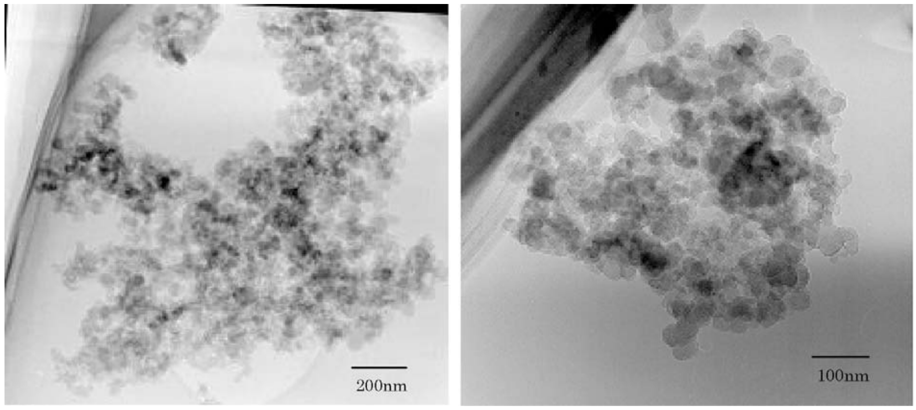

Fig. 1. TEM pictures of samples (sample #44).