

## ■ ORIGINAL PAPER ■

## Dependences of Thermophoretic Parameters on Gas Properties

MOHD AZAHARI, Bin Razali<sup>1,2\*</sup>, SUZUKI, Masataro<sup>1</sup>, and MASUDA, Wataru<sup>1</sup><sup>1</sup> Nagaoka University of Technology, 1603-1 Kamitomioka, Nagaoka, Niigata, 940-2188, Japan<sup>2</sup> University Tun Hussein Onn Malaysia, Parit Raja, Batu Pahat, 86400, Johor, Malaysia

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**Abstract** : Thermophoretic velocities in surrounding gas of argon, nitrogen, carbon dioxide, methane, or nitrous oxide are measured by means of microgravity experiments. Adopted particles are PMMA spheres with mean diameter of 2.91  $\mu\text{m}$ . The temperature gradient is set at 10 K/mm while the pressure is set at several conditions in the range of 20 kPa to 100 kPa. Terminal velocities of particles suspended in a gas are individually measured. The residue values for all combination of accommodation coefficients are calculated from the obtained experimental results. Studies are made to determine the tangential momentum accommodation coefficient by assuming the thermal accommodation coefficient to be unity. Dependence of the coefficient on gas properties is investigated.

**Key Words** : Microgravity experiment, Thermophoretic velocity, Tangential momentum accommodation coefficient, Thermal accommodation coefficient, Molecular diameter

## 1. Introduction

In a temperature gradient field, a suspended small particle will move towards the lower temperature side. This phenomenon is called thermophoresis, which influences the soot movement in a combustion field. Dobashi et al. [1] have examined a smoke generation process in a flickering pool fire and have inferred that the thermophoretic force might increase the residence time in the soot production region. Choi et al. [2] have conducted experiments to make in-situ observations of the soot particle deposition process within a diffusion flame near a solid wall. A comparison between numerical calculations and experimental results shows that the soot adheres to the wall owing to the thermophoretic effect.

There have been some direct measurements on the thermophoretic velocity of soot particles [1, 3]. Suzuki and Dobashi [3] have revealed that the velocity is dependent not only on the macroscopic size of the soot particle but also on the aggregation condition; experimental results suggest that the velocity is dominated by the size of primary spheres when the aggregation is coarse. Thus, understanding of the phenomenon for a single sphere is important before understanding for an aggregate [4].

The theory of the thermophoretic velocity of a spherical particle

has been carried out by considering the boundary condition on the surface of a single particle suspended in a gas with a temperature gradient [5, 6]. The theory contains two empirical coefficients, i.e., the tangential momentum accommodation coefficient and the thermal accommodation coefficient, which both are usually assumed to be unity [7-10].

Considering the environment of soot, one should understand the accommodation coefficients for both pure gases and gas mixtures. The coefficients for pure gases are needed in order to estimate the coefficients for gas mixtures. In case of pure gases, several experiments have been conducted to investigate the influence of gas species on the thermophoretic velocity [4]. The coefficients have been determined from the experimental results for several pure gases. In case of gas mixtures, experiments have been performed to measure the thermophoretic velocity of several gas mixtures [11]. The appropriate method has been determined in that paper for evaluating mixture's coefficients from values of pure gases.

It would be desirable to specify physical relationship between the coefficients and the physical parameters for all gas species since the combustion gas mixture contains many gas species. Some of the species are radical intermediates, e.g., OH, H, O, CH, etc. It is difficult for conducting experiments using these kinds of gases since these gases are highly reactive.

In this study, an attempt is made to investigate the dependence of thermophoretic parameters on gas properties. The

\* Corresponding author. E-mail: azahari@uthm.edu.my, m-azahari@msl.nagaokaut.ac.jp

thermophoretic parameters for argon and nitrogen are reexamined and compared with those of carbon dioxide, methane, and nitrous oxide.

## 2. Theory of thermophoretic velocity

The theory [4-6] of the thermophoretic velocity is derived from the balance between the thermophoretic force and the drag force, which is given below:

$$V_T = \frac{3\mu C_c}{2\rho T_{F0}} \frac{\left[ (k + C_T Kn) + \frac{4}{3} (C_M C_T Kn^2 + C_M Kn[k-1]) \right]}{\left[ 1 + 3C_M Kn + \frac{9}{2\pi} Pr Kn^2 (1 - \gamma^{-1}) \right] (1 + 2k + 2C_T Kn)} |\nabla T|, \quad (1)$$

where  $\mu$ ,  $|\nabla T|$ ,  $\rho$ ,  $T_{F0}$ ,  $C_c$ ,  $k$ ,  $Kn$ ,  $Pr$ ,  $\gamma$ ,  $C_M$ , and  $C_T$  are the viscosity, the temperature gradient, the density of the gas, the reference temperature, the Cunningham's correction factor, the gas-to-particle thermal conductivity ratio, Knudsen number, Prandtl number, the specific heat ratio, and constants for slip flow and temperature jump, respectively. Here, constants  $C_M$  and  $C_T$  are the ones containing the tangential momentum accommodation coefficient  $\alpha_m$  and the thermal accommodation coefficient  $\alpha_t$ , respectively, which are written as follows:

$$C_M = \frac{2 - \alpha_m}{\alpha_m}, \quad (2)$$

$$C_T = \frac{15}{8} \left( \frac{2 - \alpha_t}{\alpha_t} \right). \quad (3)$$

These accommodation coefficients represent the magnitude of the momentum and energy exchanges in the collision between gas molecules and a particle. Based on the kinetic theory, both coefficients should lie between 0 and 1 [12-14]. The Cercignani-Lampis (CL) model provides a more physical description of the gas-surface interaction [15, 16], which allows  $\alpha_m$  to vary between 0 and 2 while  $\alpha_t$  remains between 0 and 1. In this CL model,  $\alpha_m$  can exceed unity when the surface is rough.

## 3. Experimental

The experimental configuration used in this work is the same as the previous [4]. The gases used in this work are argon, nitrogen, carbon dioxide, methane, and nitrous oxide.

Sample particles are same spherical PMMA particles used in the previous work. The mean diameter and the standard deviation are 2.91  $\mu\text{m}$  and 0.09  $\mu\text{m}$ , respectively. The thermal conductivity of these particles is 0.21 W/(m K). The reference temperature and the temperature gradient of the surrounding gas are 312 K and 10 K/mm, respectively. The pressure is varied from 20 kPa to 100 kPa. The measurement is performed by means of the free-

fall method. The falling distance is 0.6 m, which corresponds to the duration time of the free-fall at 0.3 s. The gravity level is measured by the G-sensor attached on the measuring unit. A variation of the gravity level is measured during the experiment. The range between  $\pm 0.1$  G in gravity level is regarded as the microgravity condition in this work, the duration time of which is about 0.25 s. Drop experiments are conducted repeatedly to accumulate data so that the mean value and its 95% confidence interval of thermophoretic velocity for each experimental condition are statistically obtained. Details are described in the previous paper [4].

The residue  $R$  representing the discrepancy between the experiment and the prediction, which is defined as follows:

$$R \equiv \frac{\sum_{j=1}^n (V_{ej} - V_{pj})^2}{n}, \quad (j = 1, \dots, n) \quad (4)$$

where  $V_e$ ,  $V_p$ , and  $n$  are the thermophoretic velocity of the experiment, the prediction, and the number of the measured particles, respectively. The subscript  $j$  refers to each particle. Values of the residue  $R$  are calculated for all combinations of coefficients. The tangential momentum accommodation coefficient is determined such that the residue  $R$  becomes the lowest by assuming the thermal accommodation coefficient to be unity. The assumption of the coefficient is made on the basis of results in the previous work [4]; the thermophoretic velocity can be predicted quantitatively by modifying the tangential momentum accommodation coefficient while the thermal accommodation coefficient is assumed at unity. The assumption is also consistent with the work by Winkler et al. [17], in which the coefficient is reported to be approximately unity for water vapor, a polyatomic gas having a large collision diameter. Dependence of the tangential momentum accommodation coefficient on gas properties is investigated.

## 4. Results

Figure 1 shows examples of the movement of particles during a free-fall in the surrounding gas of methane at 20 kPa. It is seen that the effect of blowing can be neglected since the particles has almost no movement in the horizontal direction as shown in Fig. 1 (a). Figure 1 (b) shows the movement of particles in the vertical direction. The particles move uniformly to the positive direction of the vertical during the 0.25 s of microgravity period. The velocity of each particle is measured by tracing its movement while it travels within the range of the temperature between 313  $\pm$  2 K. The measurement of the velocity should be taken at a fixed temperature since the thermophoretic velocity is dependent not only the temperature gradient but also the temperature itself. It is seen that the velocity of each particle can be considered as

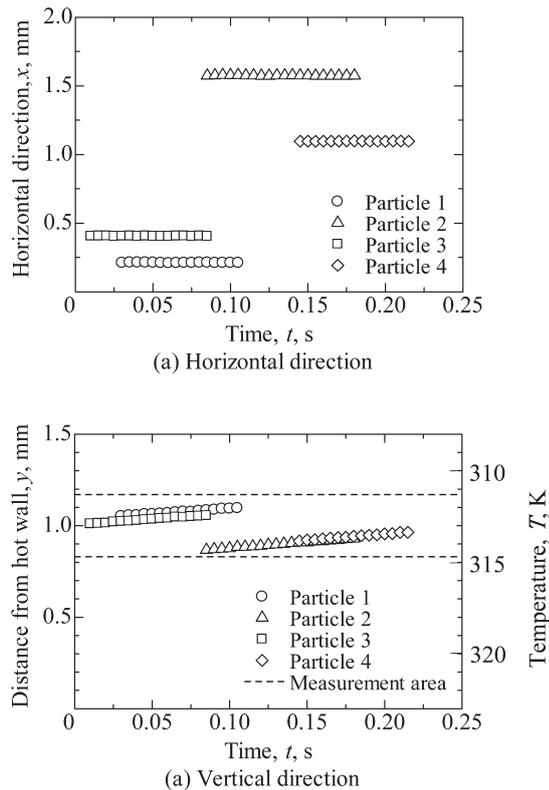


Fig. 1. Movement of particles in surrounding gas of methane.

constant in the range. The velocity is constant also in other gases.

Figure 2 shows the reduced thermophoretic velocity for each gas species. The dimensionless reduced thermophoretic velocity is calculated as follows:

$$V_{TR} \equiv \frac{V_T T_{F0}}{\nu |\nabla T|}, \quad (5)$$

where  $\nu$  is the kinematic viscosity of the gas. This parameter is often used when making comparison between different conditions. The black and the white symbols represent the obtained results from the previous work [4] and this work, respectively. Lines represent predictions calculated from different combinations of the coefficients, which will be explained later. Error bars in the figure indicate the 95% confidence interval for the mean. It is noted that the pressure, the reference temperature, and the temperature gradient of those additional data from this work for argon, nitrogen, and carbon dioxide are 70 kPa, 338 K, and 60 K/mm, respectively. These results are added for increasing the number of experimental data such that the measurement accuracy increases. It is explained beforehand that the residue values for all combinations of the coefficients are calculated from the discrepancy between the experiment results and the prediction from eq. (5).

Figure 3 shows contours of residues for each gas species. Solid lines represent the contour of the residue. The residue values are

calculated within the range of  $0 \leq \alpha_t \leq 1$  and  $0 \leq \alpha_m \leq 2$ , which are the limitations of both coefficients. It is seen that the contours have positive orientations for all tested gases. The dotted line represents the best approximation of the thermal accommodation coefficient for each tangential momentum accommodation coefficient. The thermal accommodation coefficient is determined such that the residue  $R$  becomes the lowest.

There is a problem for estimating the coefficients since the fitting gives several possible combinations. The problem for making rigorous calculation of both the coefficients is caused by the complexity of the equation of thermophoretic velocity or force [18]. Figure 4 shows the minimum value of the residue for each tangential momentum accommodation coefficient for argon. It is seen that there are several coefficients exhibit approximately the same values of the residues. A small increase up to only 4% is noticeable even when the coefficient is reduced from 1.000 to 0.600.

Comparisons of predictions between different combinations of the coefficients for each gas species are shown in Fig. 2. It is seen that for all gases, both predictions are in good agreement with the experiments within the range of the tested conditions.

In order to investigate the dependence of the coefficients on gas properties, the tangential momentum accommodation coefficient is estimated by assuming the thermal accommodation coefficient to be unity. The black arrow in Fig. 3 represents the estimated tangential momentum accommodation coefficient for each gas species. The coefficients are approximately the same for argon and nitrogen, while a significant difference is seen between the coefficients for the other three gases.

## 5. Discussion

It is interesting that the tangential momentum accommodation coefficient differs among the tested gases. Different coefficients between gases can be explained by gas properties. Several researches have been done to investigate the dependence of the coefficient on gas properties. Arya et al. [19] have conducted a molecular simulation for a wall-slip phenomenon in rarefied gases flowing through micro- and nano-channels and have found that the coefficient decreases as the collision diameter of the molecule increases. Gronych et al. [20] have performed experiments to determine the coefficient by means of a viscosity vacuum gauge with a vibrating metal ribbon. The measurements show that the coefficient is dependent on the molecular weight; the coefficient of the lighter gas molecule is greater than that of heavier ones. Dadzie and Meolens [21] have proposed a new model of scattering kernels by considering more possible reflection types of molecules at a wall by employing a partial coefficient concept of various modes, i.e., translation, rotation, and vibration. The coefficient has been suggested to be influenced

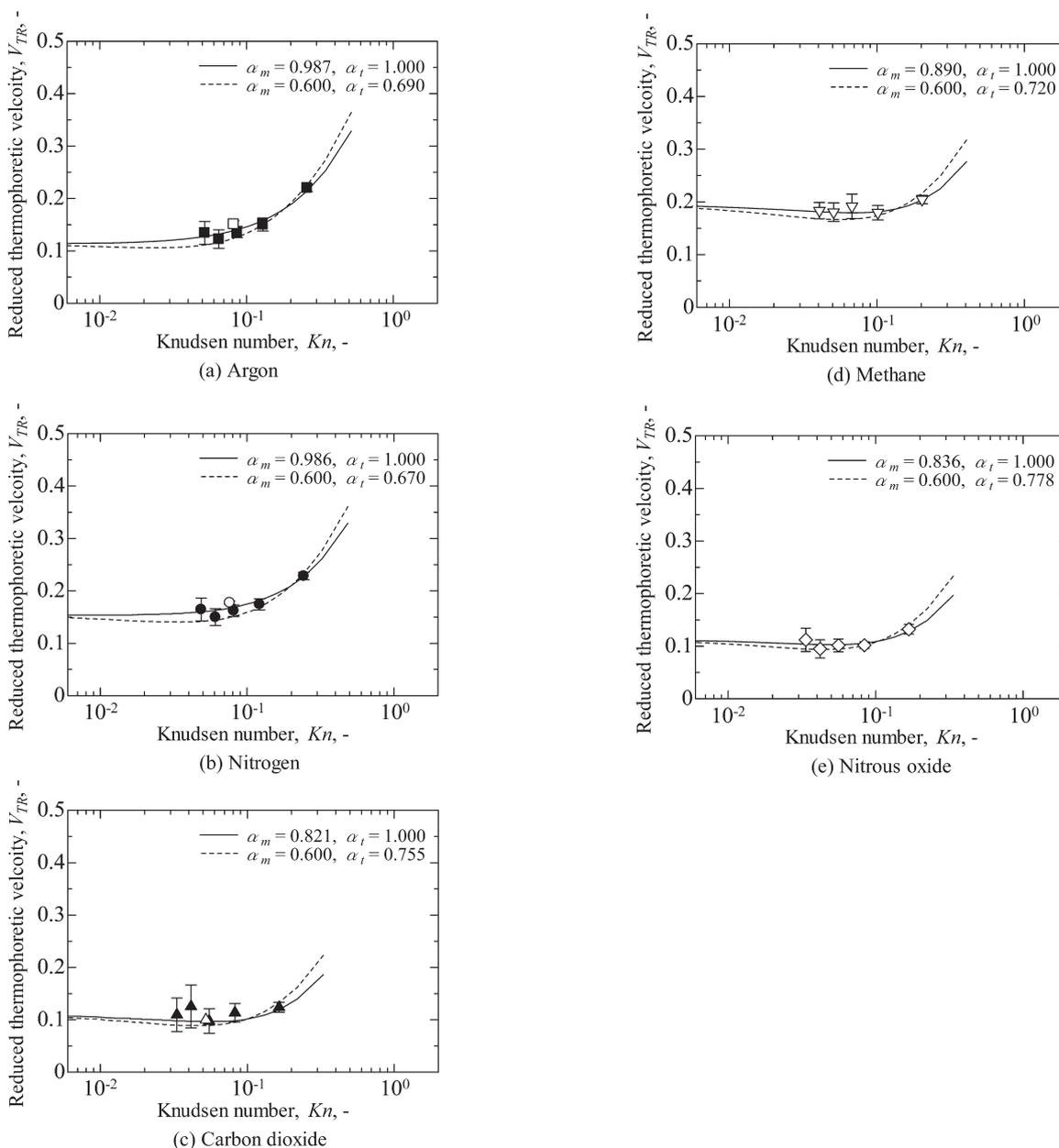


Fig.2. Reduced thermophoretic velocity for each pure gas. Error bars represent the confidence interval (95%) for the mean.

by the degree of freedom. Table 1 shows the estimated tangential momentum accommodation coefficient and the gas properties for each tested gases. It is seen that carbon dioxide has almost the same properties as nitrous oxide, while differences are found between other three gases.

Figure 5 shows the dependence of the tangential momentum accommodation coefficient on the molecular weight of gases. It is suggested that the coefficient decreases as the molecular weight increases [20]. Two disagreements are seen between the results in this work and the hypothesis given in the reference. In cases of methane, nitrogen, and argon, it is seen that the coefficient increases as molecular weight increases. In cases of carbon dioxide and nitrous oxide, the coefficients for both gases are

smaller than the coefficient for argon even the molecular weight are approximately the same for those gases.

Figure 6 shows the dependence of the coefficient on the degree of freedom of gases. The results in this work are inconsistent with the hypothesis that the coefficient is influenced by the degree of freedom [21]. It is seen that the results cannot be explained by a simple relationship between the coefficient and the degree of freedom. Two curious relationships are found from the results. First, the coefficients for argon and nitrogen are almost identical even both gases have different degree of freedom. Second, the coefficient for nitrogen is significantly greater than the coefficients for carbon dioxide and nitrous oxide, even nitrogen has the same degree of freedom with the other two gases.

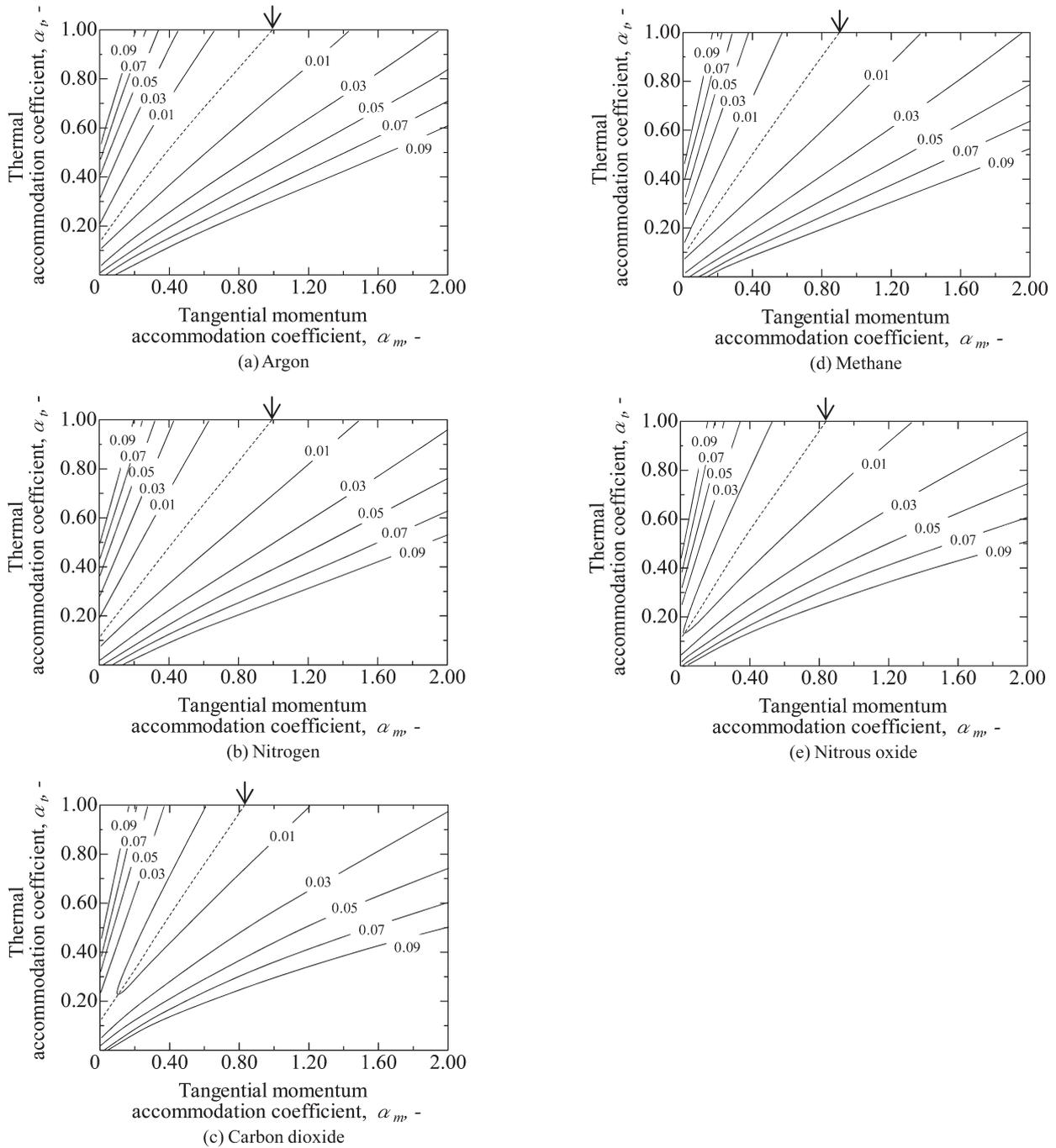


Fig.3. Residue value of each combination of coefficients for each gas. The broken line represents center line of the contour.

Figure 7 shows the dependence of the coefficient on the molecular diameter of gas molecule. The results in this work can be explained qualitatively based on the effect of molecular diameter; the coefficient decreases as the diameter increases. These results are consistent with the work done by Arya et al. [19]. The value of coefficient is presumably dependent upon the interaction characteristic between gas molecules and the surface. The results infer that when a small gas molecule collides with the surface, it loses the tangential momentum more than a large gas molecule.

There is a possibility that the coefficient is also dependent on the material of the solid; Thomas and Lord [22] have measured coefficients for polished and rough surfaces of steel spheres to find some influence of surface condition of the solid wall. However, this problem can be neglected in the present work since all experimental data are obtained from the same kind of particles.

Considering combustion fields, the soot is aggregated in a diffusion flame. It is noted that the gas concentration varies depending on positions in the flame. At some position, the flame

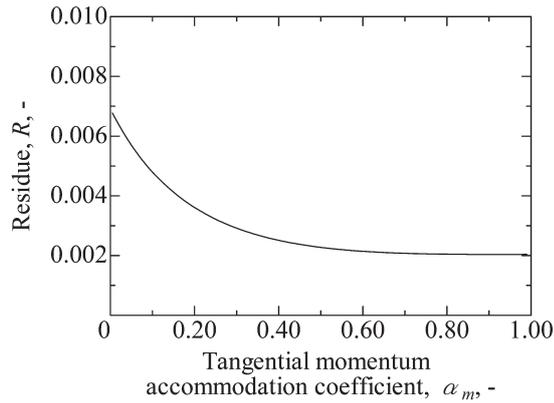


Fig.4. Minimum value of residue of each tangential momentum accommodation coefficient for argon.

Table 1. Estimated tangential momentum accommodation coefficient and gas properties for each gas.

Gas	$\alpha_m$ , -	Molecular weight, g/mol	Degree of freedom, -	Molecular diameter, Å
Argon	0.987	39.944	3	3.659
Nitrogen	0.986	28.02	5	3.784
Carbon dioxide	0.821	44.01	5	4.643
Methane	0.890	16.04	6	4.158
Nitrous oxide	0.836	44.02	5	4.662

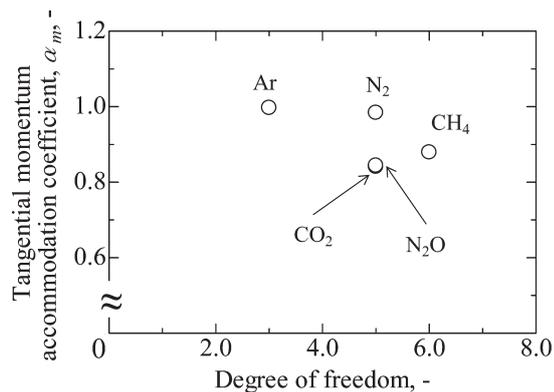


Fig.5. Relation between coefficient and molecular weight.

contains gases having a large molecular diameter. Results in this work show that the tangential momentum accommodation coefficient decreases as the diameter increases such that the thermophoretic velocity decreases. The soot aggregation is dependent on the thermophoretic effect [1, 2]; the aggregation decreases as the thermophoretic velocity decreases. The results in this work inferred the possible reason influences the soot

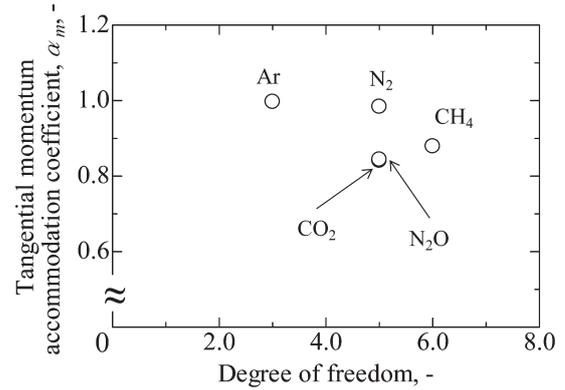


Fig.6. Relation between coefficient and degree of freedom.

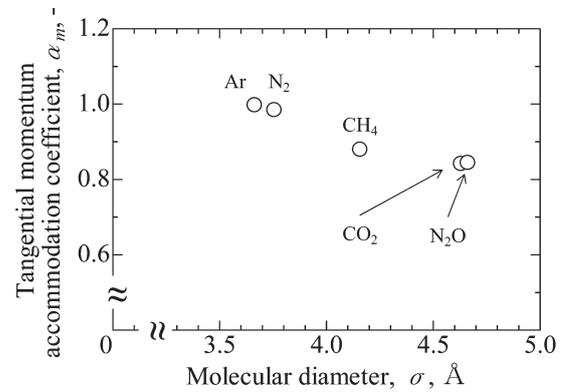


Fig.7. Relation between coefficient and molecular diameter.

aggregation in the diffusion flame.

There is a difference in size between particles used in this work and soot particles. Comparing to the size of aggregated soot particles, the size of the current work is not so much of different since the size of tested particles is  $3 \mu\text{m}$  and that of aggregated soot particles are distributed between  $0.5 \mu\text{m}$  and  $10 \mu\text{m}$ . On the other hand, comparing to the size of primary soot particles, the size itself is quite different; the size of primary soot particles are distributed between  $10 \text{ nm}$  and  $100 \text{ nm}$ . However, the Knudsen number is in the same regime at some condition, e.g.,  $Kn = 0.6$  when the temperature is at  $700 \text{ K}$  and the pressure at  $0.5 \text{ MPa}$  even there is a significant difference in size. This condition would be realistic for some combustion systems.

## 6. Conclusions

In this study, the thermophoretic parameters of several gases are measured from the experimental results, and following results are obtained:

1. The value of the residue  $R$  is calculated from the discrepancy between the experiment results and the prediction; several combinations of the coefficients are possible to fit the

prediction with the experimental results.

2. The tangential momentum accommodation coefficient is estimated by assuming the thermal accommodation coefficient to be unity; the tangential momentum accommodation coefficient are approximately the same for argon and nitrogen, while significant differences are seen for methane, nitrous oxide, and carbon dioxide.
3. A relation is shown between the tangential momentum accommodation coefficient and the molecular diameter; the coefficient decreases as the diameter increases.

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