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Effect of buoyancy on soot formation in gas-jet diffusion flame †

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Abstract

In order to investigate the effect of buoyancy on soot formation in gas-jet diffusion flame, we conducted one set of experiments with the High-Temperature Air Combustion Technology (HiCOT) system and another set under partial gravity conditions. Ethylene (C_2H_4) was used as fuel, and soot volume fractions for the flame were observed as shadow graph images with backlight. In the experiment with the HiCOT, the oxygen concentrations were O_2 = 15 %, 17 %, and 23 %, with constant flame temperature and surrounding air temperatures of 1100 K, 900 K, and 300 K, respectively. We found that the soot volume fraction in the flames increased with the increase of the oxidizer temperature. In the partial gravity experiment meant to identify the buoyant effect, the results showed that the soot volume fraction depended on the gravity level. These results imply that soot formation in a gas-jet diffusion flame with the HiCOT is strongly affected by the buoyant flow due to oxidizer flow into the soot formation field.

Keywords: Diffusion flame; Soot; High-temperature air combustion technology; Buoyancy; Partial gravity

1. Introduction

Soot particles produced in flames play an important role as a heat-radiating medium. Soot radiation can be desirable because it improves thermal efficiency in large-scale boiler furnaces. However, soot is also a common air pollutant and soot deposition on heat exchangers may cause significant deterioration of thermal efficiency [1-4].

Recently, High-Temperature Air Combustion Technology (HiCOT) was introduced into industrial furnaces such as boiler furnaces and steam reforming processes. High-Temperature Air Combustion Technology (HiCOT) has several advantages over conventional combustion systems and has attracted great interest due to its practical significance [5-7]. The HiCOT can achieve higher combustion efficiency [8] with lower oxygen concentration [9] because of its high air temperature. Thus, the HiCOT can reduce the amount of soot produced in the system [10]. Moreover, the HiCOT can improve the performance of an industrial furnace because its uniform temperature distribution around the flame lowers NO_x emissions [11, 12]. Therefore, the soot formation in a gas-jet diffusion flame with high air temperature is an important subject in combustion science.

In the study of high air temperature combustion under normal gravity conditions [13], it was shown that soot formation under high air temperature conditions fairly increased with increase in surrounding air temperature, under which the buoyancy induced flow caused by the difference of gas densities of flame and surrounding air was reduced. Therefore, the authors considered that effect of buoyancy-induced flow could be one of the important factors to determine soot formation in high temperature air combustion, which is the motivation of this research. While extensive research [14-19] has been done on soot formation under microgravity conditions, research concerning partial gravity conditions is very limited. Kaplan et al. [19] showed that the oxygen gradients in reduced gravity flames are considerably less steep compared to those of normal gravity. This result indicated that the flow rate of air to the nonbuoyant flames is reduced. Although it is important to understand the phenomenon in partial gravity from the view of high air temperature combustion, this field has never been investigated systematically. One of the reasons is that it is difficult to change the effect of buoyancy without changing the temperature conditions. One such method for changing the buoyancy effect is to change the gravity conditions, namely partial gravity conditions.

To investigate the effect of buoyancy on soot formation in gas-jet diffusion flames, this study utilized experimental set-

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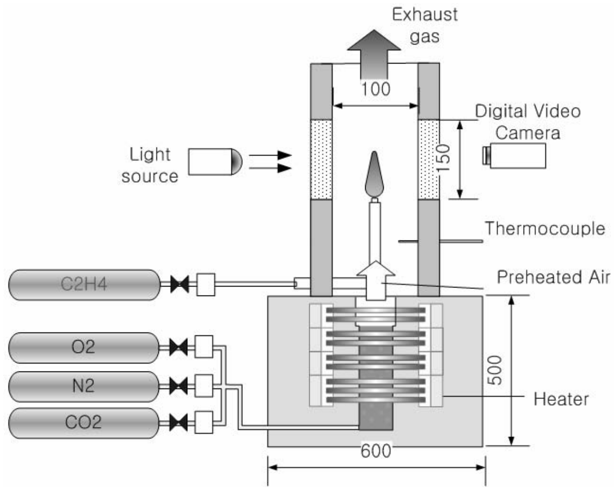


Fig. 1. Schematic of experimental setup with HiCOT.

ups for the HiCOT and the partial gravity conditions. The experimental results can be applicable to understanding the soot formation in a flame with a high surrounding air temperature.

In the HiCOT experiments, the flame temperatures were calculated for each air temperature by CHEMKIN Oppdif code [20], which sets them as the same temperature. The air consisted of oxygen, nitrogen, and carbon dioxide, and ethylene was supplied as fuel. The air conditions for the HiCOT experiments were as follows: (a) 23%O_2-300 K, (b) 17%O_2-900 K, and (c) 15%O_2- 1100 K. In the partial gravity experiments, the oxygen concentration of air (N_2 balance) and the gravity level were varied as the parameters. The soot volume fraction was obtained as a shadow graph image taken by the backlight technique.

2. Experimental setup of hicot

Fig. 1 shows a schematic of an experimental setup with the HiCOT. A stainless steel cylindrical burner with 3 mm inner diameter and 4 mm outer diameter is located in the center of the combustion duct (100 mm × 100 mm), which is in turn surrounded by a 50 mm thick insulator. Using an electric heater, an air mixture consisting of oxygen, nitrogen, and carbon dioxide is preheated up to 1100 K. The temperature of the air is monitored by an R-type thermocouple inserted upstream of the air flow in the burner. The fuel flow rate and the air flow rate are kept at 2.1mL/s (0.3 m/s) and 1.5L/s (0.15 m/s), respectively. The combustion duct has a fused silica glass window for observing the image of the flame. To determine the soot volume fraction in the flame, a 150 W metal halide lamp casts light on the flame and a shadow graph image is taken by a digital video camera with a 532 ± 5 nm band-pass filter.

Shadow graph images were taken by video camera and attenuation ratio to the original shadow graph brightness was determined two-dimensionally. The Bouguer-Lambert-Beer formula used to estimate the light extinction phenomena is:

$$-\ln\frac{I}{I_0} = Q_{ext}\frac{\pi}{4}D^2NL \tag{1}$$

where I_0 is the initial light intensity, I is the transmitted light intensity, Q_{ext} is the attenuation coefficient D is the particle diameter, N is the number density, and L is the optical path length. Eq. (2) shows coefficient of light extinction, $Q_{ext,}$ which is applicable to the soot particle aggregate in the flame.

$$Q_{ext} = Q_{abs} + Q_{sca} \tag{2}$$

where Q_{abs} is coefficient of absorption and Q_{sca} is coefficient of scattering. Light extinction of soot aggregate by scattering is negligible in comparison with the light extinction by absorption. Thus, Q_{ext} can be assumed to be $Q_{abs.}$. The coefficient of absorption, Q_{abs}, by particles can be expressed as follows from the formula of Rayleigh's Law [21].

$$Q_{abs} = 12\frac{\pi D}{\lambda}\frac{2nk}{(n^2+k^2)^2+4\left\{1+(n^2-k^2)\right\}} \tag{3}$$

where λ is wavelength of selected light wavelength (Q_{abs}). For soot particle, n = 1.56 and k = 0.5 from empirical data [22]. Then the concentration of soot density is shown as below.

$$C_s = -\frac{2}{3}\frac{\rho_s D}{LQ_{abs}}\ln(\frac{I}{I_0}) \quad (\text{g/mm}^3) \tag{4}$$

where $\boldsymbol{\rho}_s$ is the density of soot particle, C_s is the concentration of soot particle. Partial volume fraction of soot is $F_v = C_s \times \frac{1}{\rho_s}$, and can be re-written as Eq. (5) [23] from Eq. (4).

$$F_v = \frac{2}{3\times 2.95}\frac{\lambda}{L}(-\ln\frac{I}{I_0}) \quad (\text{mm}^3/\text{mm}^3) \tag{5}$$

From Eq. (5), soot volume fraction in the flame can be calculated by obtaining incident light intensity, I_0, intensity of extinction light, I, and physical length of extinction, L. The selected wavelength of the incident light (λ) is designated by the band pass filter described below. An interference filter (532nm center, 10nm band width for half value) was used to select the wavelength for extinction image and also to eliminate light emission from the luminous flame. Since the attenuation measurement of backlight is a kind of in-line method, two-dimensional image should be reconstructed into concentric distribution by Abel-transformation method assuming axis-symmetric distribution.

The experimental conditions, i.e. the combination of the temperature and the oxygen concentration of the oxidizer, were determined by calculation with CHEMKIN [20] code in order to keep the same flame temperature. The conditions chosen were as follows: (a) 23%O_2-300 K, (b) 17%O_2-900 K, and (c) 15%O_2-1100 K.

Fig. 2 shows calculated flame temperatures under the three air conditions versus the mole fraction of CO_2 in the oxidizer.

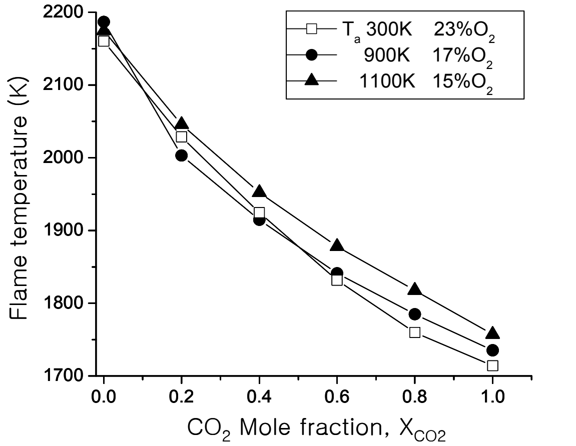


Fig. 2. Calculated flame temperatures for three air conditions versus mole fraction of CO_2 ($X_{CO2}=P_{CO2}/(P_{CO2}+P_{N2})$).

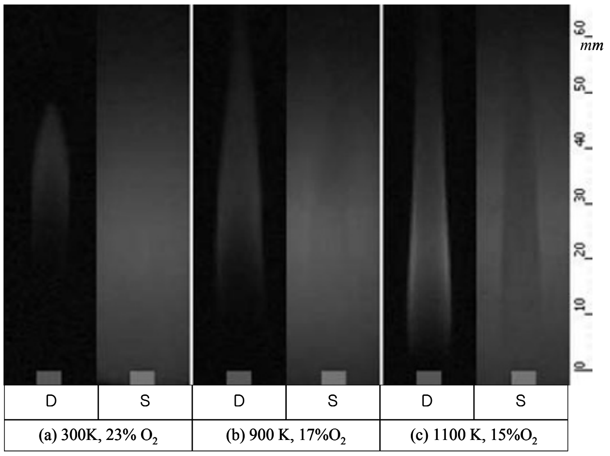


Fig. 3. Direct images (D) and Shadow graph images (S) of the flames in HiCOT (CO_2 = 0 %, 1G).

In the figure, the mole fraction of CO_2 is a ratio of CO_2 to CO_2 plus N_2. The calculation results show that the calculated temperatures for the three air conditions decrease as the mole fractions of CO_2 increase, and that the differences of temperature for the three different conditions is small. The mole fraction of carbon dioxide can be obtained from

$$X_{CO_2} = \frac{P_{CO_2}}{P_{CO_2} + P_{N_2}} \tag{6}$$

3. Results and discussion

3.1 Soot formation in HiCOT system

Fig. 3 shows the direct images and the shadow graph images of the flames for (a) 23%O_2-300 K, (b) 17%O_2-900 K, and (c)15%O_2-1100 K (N_2 is supplied as a balance gas). In Fig. 3, it is found that the flame luminosity with high air temperature (condition (c)) is the highest and the flame luminosity with low air temperature (condition (a)) is the lowest. It can also be observed that the attenuation rate of back light (shadow) exhibits the same trend as the luminosity of the flames.

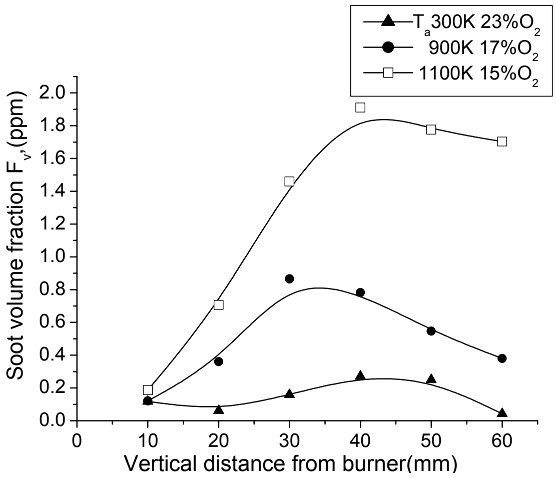


Fig. 4. Local soot volume fractions for three conditions in HiCOT (1G).

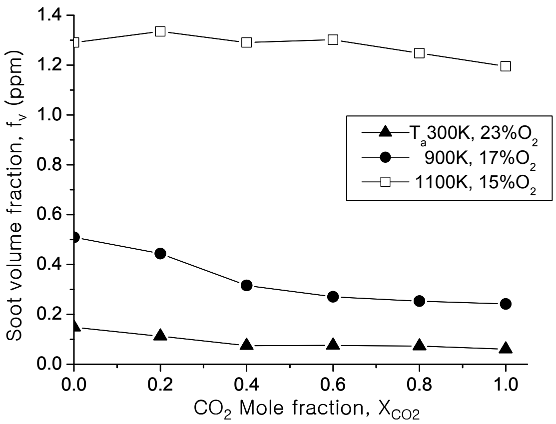


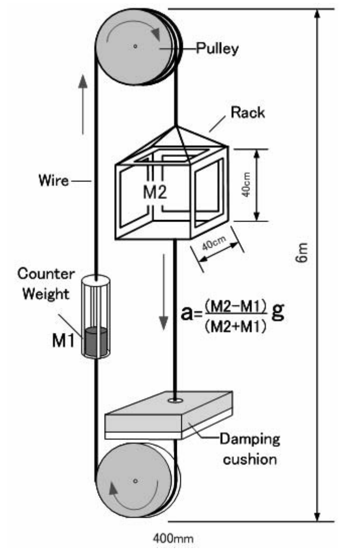
Fig. 5. Soot volume fractions for three air conditions versus mole fraction of CO_2 in HiCOT (1G).

The length of the flame also increases with increasing air temperature.

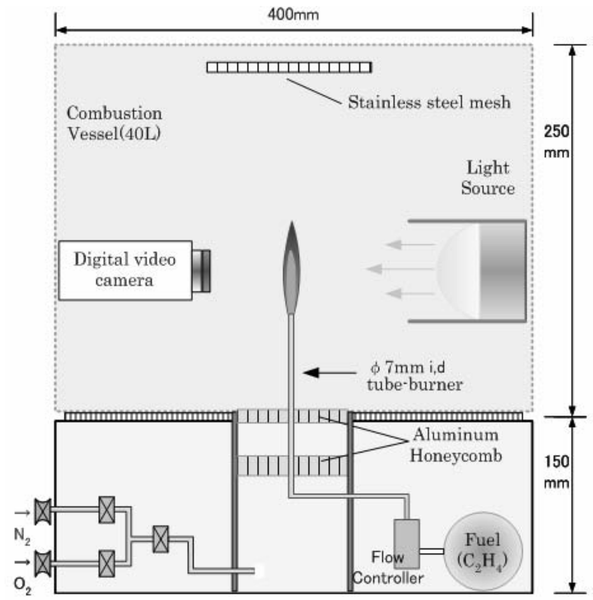
Fig. 4 shows the local soot volume fraction at distances 10 mm to 60 mm above the burner exit, for each of the three conditions. Under high air temperature and low oxygen concentration, a relatively large amount of soot is produced. This is because of the long residence time caused by low flow velocity, which in turn is induced by small buoyancy effect. Moreover, soot oxidation can be expected to suppress because the amount of air flowing into the flame is decreased. When air temperature is 1100 K with 15%O_2, the soot volume fraction at a height of 60 mm is 1.8×10^6, 4-9 times higher than those of other flames. These results show that soot oxidation under high air temperature conditions is delayed because the effect of air flowing into the soot formation region is limited.

Next, the effect of mixing CO_2 on soot formation was investigated, and the differences in soot volume fraction were measured by the back light technique.

Fig. 5 shows the soot volume fractions under three air conditions versus various mole fractions of CO_2 in normal gravity, 1G. The soot volume fractions are averaged at 10, 20, 30, 40,



(a)



(b)

Fig. 6. Drop facility for partial gravity (a) and Experimental setup for partial gravity experiment (b).

50, and 60 mm above the burner exit. Lee et al. [14] indicated that the soot formation in the flame is affected by (a) residence time of the soot in the soot formation field, (b) gas composition, and (c) temperature of the soot formation field. Nevertheless, it is worth noting that although the difference of flame temperature for three air conditions is small, the amount of soot produced in the flame under each condition is quite different. The soot formation fluctuation, occurring in spite of keeping the same flame temperature and the same total calorific value (i.e. total fuel rate) in each set of conditions, is caused by the difference in residence time and the deviation of gas composition in the flame. Generally speaking, the residence time can be changed by changing flow conditions, including the volume flow rates of fuel and air and the buoyant flow. The gas composition can be changed by altering the initial gas concentration and flow conditions influenced by the buoyant flow.

As shown in Fig. 5, the soot volume fraction decreases as the mole fraction of CO_2 increases. When the mole fraction of CO_2 is 0.0, the soot volume fractions for the three flames are 1.29 ppm, 0.5 ppm, and 0.15 ppm, respectively. When the mole fraction of CO_2 is 1.0, the soot volume fractions for the three flames are 1.19 ppm, 0.24 ppm, and 0.06 ppm. The amount of soot for the three flames decreases about 8 %, 50 %, and 60 %, respectively. It is thought that the oxidation due to OH radicals, produced by the CO_2+H=CO+ OH reaction, plays a significant role in the decrease of soot formation [24, 25].

Although the correlation between soot volume fraction decrement and CO_2 increment is supposed to be consistent, the actual decrement ratio for each flame reveals a large discrepancy. The effect of CO_2 appears to be minimal under high-temperature conditions. This can hardly be explained without considering the difference in the amount of oxidizer supplied to the flame. Therefore, the effect of oxidizer supply to the flame on the soot volume fraction was investigated under the assumption that the amount of the supplied oxidizer had differed. There are two ways to change the amount of air that flows into the flame: changing the surrounding air velocity in microgravity [26] and varying the gravity level. In this study, the gravity level was varied to change the effects of buoyant flow.

3.2 Soot formation in partial gravity conditions

Experimental results for the HiCOT combustion system showed that the soot volume fraction increases with the increase of surrounding air temperature. This implies that soot formation is affected by the buoyant flow caused by natural convection, which is in turn induced by the temperature difference between the flame and the surrounding air. Thus, to identify the effects of buoyant flow, partial gravity experiments were carried out without changing the thermal conditions (i.e. fuel and air conditions).

Figs. 6(a) and 6(b) show the drop facility and experimental setup for the partial gravity experiments. An experimental rack that can support the setup in Fig. 6(b) is hung using steel wire and connected to the counterweight. Gravity in the experimental rack is controlled by changing the difference in weight between the experimental set-up and the counterweight. The gravity is adjustable from 0.3 G to 1.0 G. The gravity used in this study was 1.0 G (no fall), 0.7 G, 0.5 G and 0.3 G. The gravity level is determined by an equation as follows:

$$A = \frac{(M_2 - M_1)}{(M_2 + M_1)} g \tag{7}$$

where A is the required gravity level , M_1 and M_2 are rig weight and balance weight, respectively, and g is earth gravity.

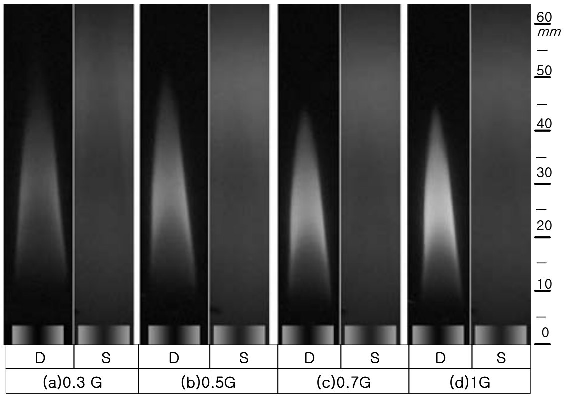


Fig. 7. Direct (D) and Shadow graph images (S) of the flames (O_2 = 21 %) in partial gravity conditions.

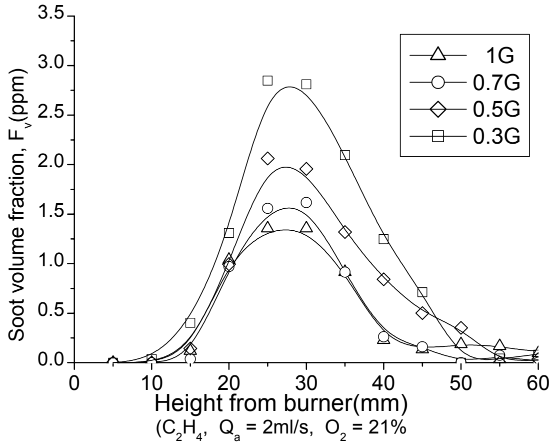


Fig. 8. Soot volume fraction in partial gravity conditions (C_2H_4, 2.0 mL/s, 21%O_2).

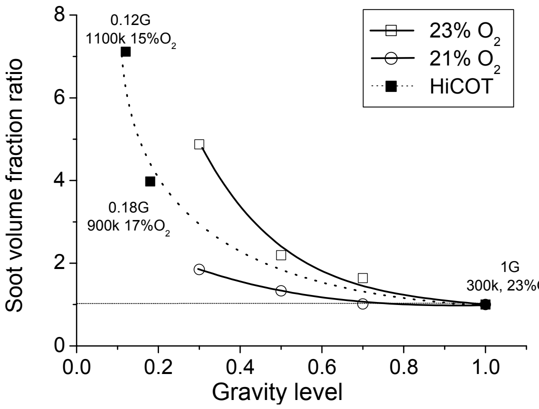


Fig. 9. Effects of gravity level on soot volume fraction ratio.

The accelerations of Eq. (7) were verified using accelerometers.

If the height of fall is about 4.7 m, about 1.2 s of 0.3 G conditions can be obtained. In Fig. 6(b), a cylindrical burner of 7 mm inner diameter and 8 mm outer diameter is located in the center of the enclosed combustion chamber (400 mm (B) × 400 mm (W) × 250 mm (H)) and the ethylene diffusion flame is formed above the burner exit (fuel flow rate = 2.0mL/s). The surrounding air can be arbitrarily pre-set as a composition of O_2 and N_2. The direct images and the shadow graph images are obtained in a similar way to that used in the HiCOT system.

Fig. 7 shows the direct and shadow graph images of the flame in gravity conditions of 0.3, 0.5, 0.7, and 1.0G. The surrounding air temperature is 300 K and the oxygen concentration is 21%. Because the buoyancy-induced convection is significantly reduced, diffusion effect becomes the dominant mechanism in transport as the gravity level decreased. In Fig. 7, the attenuation of light intensity in low-gravity conditions is higher than that in high-gravity conditions, while the luminosity of the flame in low-gravity conditions is lower than that in high-gravity conditions. The length and width of the flame increase with decreasing gravity level due to the reduction in axial velocity and the thicker diffusion layer. The flicker of flames in gravity conditions of 0.3, 0.5, 0.7 G is small, as the buoyancy-induced instability that caused flicker in the 1G flame almost disappears in the 0.3, 0.5, 0.7 G environment.

Fig. 8 shows the local soot volume fractions under four gravity conditions versus height from the burner. The local soot volume fraction increases and then decreases as height above the burner increases under each gravity condition. The maximum values of soot volume fraction for different kinds of flames are 2.84×10^{-6} for a 0.3 G flame, 2.0×10^{-6} for a 0.5 G flame, 1.6×10^{-6} for a 0.7 G flame, and 1.36×10^{-6} for a 1 G flame. The fraction of the 0.3 G flame has increased by 2.1 times as compared to that of the 1 G flame. The maxima of soot volume fractions appear between 25 mm and 30 mm from the burner, and the maximum positions for the flames are moved downstream as the gravity level decreases. As mentioned in Fig. 4, in the HiCOT experiments, the amount of air flowing into the flame was small under high air temperatures. As a result, the effect of CO_2 was weak. The results in Fig. 8 show good agreement with the results in Fig. 4. In other words, the relative velocity between the flame and the surrounding air in the HiCOT experiment is decreased. Thus, the amount of soot decreases due to the increase in the amount of air that flows in the flame. We can conclude that the high concentration of soot at 0.3 G is caused by a decrease in the oxidation of soot, which is due to the decrease in the amount of air that flows into the flame.

Fig. 9 shows the integrated soot volume fraction ratios for various gravity levels. Each soot volume fraction was normalized to 1G condition. Two oxygen concentrations, 21 % and 23 %, were tested. The HiCOT data were converted into the gravity level by calculating the difference in air density between the flame temperature and the air temperature. Then the data were plotted, as shown in Fig. 9. An interesting feature is that the soot volume fraction increases with decreasing gravity level. This implies that the soot produced in the flame is increased by decreasing buoyant flow around the flame. Because the oxidation rate of soot in reduced gravity flames decreases due to the decrease of air entrainment in nonbuoyant flames, the soot volume fraction ratio for each of the three cases increase as the gravity level decreases. These results are a good agreement with the results of the HiCOT experiment.

4. Conclusions

In this study, we investigated the influence of surrounding air temperature on soot formation for gas-jet ethylene diffusion flames. A few experiments were conducted using the High-Temperature Air Combustion Technology (HiCOT) system, and others have been tested various partial gravity conditions.

In the HiCOT experimental system, the soot volume fraction in the flame, in spite of a constant flame temperature, increased as air temperature increased. It is inferred that the buoyant flow, which is induced around the flame due to the temperature difference between the flame and the air, is able to influence the soot formation and oxidation in the flame. To identify the buoyancy effect on soot formation, partial gravity experiments with 0.3G, 0.5 G, and 0.7G were carried out. The soot volume fraction was found to increase with decreasing gravity level. It was also found that the buoyant flow induced by gravity changes the residence time, and that the gas composition of the air that flows into the flame has a significant effect on the soot formation in the gas-jet diffusion flame.

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