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Optical measurement of volume fraction and organic mass fraction of ultra-fine soot particles emitted from inverse diffusion flames

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ABSTRACT

Soot particles of various organic carbon (OC) contents were generated with a triple co-flow burner. TEM images showed that the primary particle boundaries were more ambiguous when OC contents were high. A set of diode lasers of various wavelengths and a white light source were used to measure light extinction over a wide range of wavelengths (450–850 nm), and the logarithm of the light intensity ratio was plotted as a function of the inverse of the various wavelengths. The slopes of the plot were taken from the both ends of the wavelength range, and their ratio was calculated. The results show that when the OC contents are high, the ratio increases to approximately 10, whereas the ratio remains nearly constant at a value of 1 when the OC contents are minimal. The analysis of the slope ratios shows that the wavelength-dependent absorption behavior of soot does not depend on the soot volume fraction or the soot particle size; rather, the slope ratio depends only on the soot's refractive index, which in turn is related to its OC contents. In addition, the soot volume fraction and OC contents can be simultaneously identified without any chemical analysis based on the pre-determined relationship between the slope ratio and the organic fraction of soot.

1. Introduction

Soot particles from combustion of hydrocarbon fuels cause atmospheric pollution, and these particles are known to have adverse health effects on humans [1–3]. In addition, atmospheric soot particles absorb sunlight and therefore contribute to global warming [4]. To address these concerns, various regional and international regulations have been implemented on soot emissions from automotive engines and industrial combustors. As the environmental regulations have become increasingly restrictive, many studies have been conducted to characterize and control soot emissions from combustors [5–9].

Typical soot particles consist of elemental carbons (ECs), organic carbons (OCs), sulfates and minerals, and the sizes and compositions of soot particles are highly dependent on the fuel type and the combustion process from which they originate [10]. For example, soot from automotive diesel engines consists mainly of ECs and OCs, and their size ranges from some tens of nanometers to a few hundred nanometers. In contrast, soot from coal power plants may contain minerals and sulfates, depending on the coal fuel sources [11].

The toxicity induced by exposure to or inhalation of soot particles highly depends on the size and contents of the soot particles. For example, OC contents, which are more abundant in soot at the initial stage of formation, have been found to be more toxic in *in vitro* tests [12]. It has been suggested based on animal experiments that small soot particles of approximately 100 nm or less, known as ultra-fine soot particles (UFPs), are more likely to cause respiratory and circulatory diseases [13]. Light absorption by soot particles is also strongly correlated with the particles' sizes, as well as their refractive indices, which are determined by the soot's chemical composition [14]. It has therefore been suggested that characterizations of soot particles and assessments of their health and environmental effects should be conducted based not only on their concentration in the atmosphere or the total inhaled mass, but also on the particles' size distribution and chemical composition.

Soot undergoes various formation and oxidation processes inside flames [15–20], and these processes determine the size, morphology, and chemical composition of the emitted soot particles. Optical techniques are mainly used to investigate soot in flames because these techniques interfere minimally with the flame and allow instantaneous measurements [21–26]. In particular, characteristics such as soot concentration and the extinction coefficient are determined by the refractive index values of the soot particles. Accordingly, there have been

Abbreviations: EC, elemental carbon; LII, laser-induced incandescence; OC, organic carbon; PA, particulate analyzer; PSA, particle size analyzer; SCCM, standard cubic centimeter per minute; SLPM, standard litter per minute; TEM, transmission electron microscopy; UFP, ultra-fine soot particle

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many studies on predicting the refractive index of soot for more accurate soot measurements. Charalampopoulos et al. [27] conducted an experimental study using pre-mixed propane flames with an equivalence ratio of 1.8 and presented the refractive index of soot particles in flames as a function of the incident light wavelength. Therssen et al. [28] measured laser-induced incandescence (LII) signals from soot particles in flames using lasers with wavelengths of 532 nm and 1064 nm, and then calculated the ratio of the soot refractive index function at each wavelength. The results showed that methane diffusion flames had a value close to 1, whereas pre-mixed acetylene/air flames showed a value close to 1.1. Based on LII signals measured using lasers with various wavelengths, Yon et al. [29] reported that the refractive index of soot particles depended on the incident light wavelength in the range of 300–1100 nm. Bescond et al. [30] used ethylene diffusion flames and a commercial soot generator to investigate the light extinction of soot within a wavelength range of 200–1100 nm, reporting that light absorption effects increased as OC contents increased, especially with shorter wavelengths.

In addition to optical techniques, transmission electron microscopy (TEM) has been utilized to acquire morphological changes of soot in flames and to investigate soot formation mechanisms [31,32]. Kholghy et al. [33] examined the differences in soot morphology in the height and axial directions of co-flow diffusion flames. Kempema et al. [34] analyzed the changes in the gyration radius of soot caused by the aggregation of soot particles. In addition, Bambha et al. [35] reported that the aggregate size and morphologies of soot particles significantly impact LII and scattering signals. The light scattering effect becomes negligible compared to the absorption effect when the particle size is less than a few hundred nanometers [36].

Despite the accuracy and instantaneous nature of the optical diagnostic techniques, current optical methods have not been satisfactorily applied to measuring the chemical composition of soot. Real-time measurements of soot concentration in exhaust gas can be accomplished optically, for example, by light extinction methods, but these methods cannot be applied to analyze the soot's chemical composition. Currently, soot's OC contents are measured with high-precision chemical analyzers that require soot deposition on a sampling filter. The purpose of this study is to present an optical method that can be utilized to determine soot composition, especially its OC contents. The proposed method offers the advantages of optical diagnostics and avoids sample collection and chemical analysis procedures.

In this study, an inverse diffusion flame was used to continuously emit a sufficient amount of soot particles. An inverse diffusion flame was formed by switching the positions of the fuel and the oxidizer of the normal diffusion flame. In this case, the fuel is located outside the flame so that the soot-generating region is also located outside the flame surface [37]. Therefore, the generated soot is emitted from the flame without an oxidation process, since there is no oxidizer around the soot particles. Owing to these characteristics, inverse diffusion flames are commonly used in studies on initial soot formation and growth in the absence of soot oxidation processes [38,39]. Accordingly, a triple co-flow burner [40] was used in this work. Fig. 1 shows a schematic of the burner. Air was supplied to Nozzles 1 and 3, and the fuel was supplied to Nozzle 2. When the airflow rate at Nozzle 3 is much greater than that at Nozzle 1, the flame is mainly formed by the flows from Nozzles 2 and 3. Although it is possible to generate a limited amount of soot without the airflow at Nozzle 1, a significant amount of soot can be emitted from the flame by supplying some air through Nozzle 1. Given with the airflow at Nozzle 1, a small inverse diffusion flame is formed inside the main flame. The temperature near the nozzle tip increases due to the inner flame, and thus, the degree of fuel pyrolysis increases as well. This phenomenon is known to promote soot generation [37]. In this configuration, the degree of carbonization of the emitted soot particles was controlled by adjusting the airflow rate at Nozzle 3.

The light extinction characteristics of the emitted soot particles were measured, using both white light with a continuous spectrum and

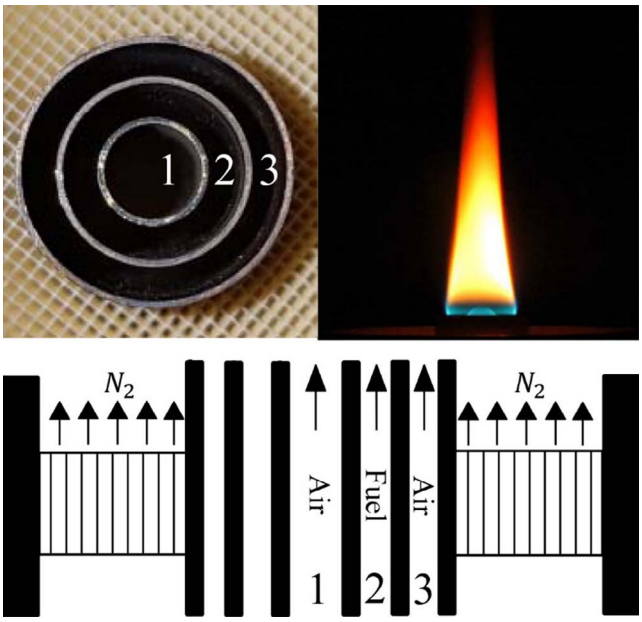

Fig. 1. Schematic diagram and photographs of the triple co-flow burner.

diode lasers of various discrete wavelengths as light sources to examine the wavelength dependence of the light extinction. Photodiodes and a spectrophotometer were used to measure the light intensity over a wide range of wavelengths. In addition, TEM images of the soot particles were acquired, and the aerodynamic sizes of the particles were measured by a particle size analyzer (PSA). Moreover, a particulate analyzer (PA) was utilized to chemically analyze the OC contents in the emitted soot under various burner flow rate conditions, and these results were compared with the light extinction results.

2. Light extinction theory

Soot particles absorb and scatter incident light. The intensity of light that is extinguished as it passes through a region containing soot particles exist can be expressed as Eq. (1), which is based on the Lambert-Beer law [41].

$$\ln\left(\frac{I_o}{I}\right) = K \cdot L \tag{1}$$

Here, I_o represents the original intensity of incident light; I is the light intensity after passing through the sooty region; K is the light extinction coefficient; and L is the light path length. With C, Q_{abs}, and Q_{sca}, representing the number concentration (#/volume), absorption efficiency, and scattering efficiency of soot particles with a radius of R, respectively, then the light extinction coefficient K is expressed as follows:

$$K = C \cdot \pi R^2 \cdot (Q_{abs} + Q_{sca}) \tag{2}$$

If the sizes of the particles present on the path are sufficiently small relative to the incident light wavelength (λ), then Q_{abs} and Q_{sca} are as Eqs. (3) and (4), respectively, assuming the Rayleigh limit (R≪ λ) [36].

$$Q_{abs} \cong 4\left(\frac{2\pi R}{\lambda}\right) Im\left(\frac{m^2-1}{m^2+2}\right) \tag{3}$$

$$Q_{sca} \cong \frac{8}{3}\left(\frac{2\pi R}{\lambda}\right)^4 \left|\frac{m^2-1}{m^2+2}\right| \tag{4}$$

Here, m is the refractive index of the soot particles, expressed as a complex number in the form of $m = n + ik$. Under the Rayleigh limit condition, $(R/\lambda)^4$ in Q_{sca} converges to zero much faster than R/λ in Q_{abs}, and therefore Q_{sca} in Eq. (2) can be considered negligible compared to