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An experimental study on the spectral dependence of light extinction in sooting ethylene counterflow diffusion flames

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ABSTRACT

A light extinction technique is widely-adopted for quantitative measurement of soot volume fractions. The measurement accuracy is dependent on the optical properties of soot, which are expected to vary with the wavelength of incident light and the physicochemical environments in which soot is formed. In the present study, a diode laser–based light extinction setup, capable of providing light with variable wavelengths ranging from 405 to 1064 nm, was utilized to investigate the in-situ spectral dependence of light absorption for soot formed in counterflow diffusion flames. Soot volume fractions (F_V) were inferred from the extinction level of these laser beams for a series of flames parameterized by oxygen/fuel mole fractions, nozzle exit velocities, and fuel types. Special attention was given to distinguish between the soot formation (SF) and soot formation/oxidation (SFO) flames, considering their notable differences in soot evolutions. It was found that the inferred F_V as measured with visible light (405–670 nm) was always significantly higher than those measured with near-infrared light (> 780 nm). In addition, the quantitative decrease of F_V with the increase in light wavelength (λ) was found to be different for soot particles formed at different flame locations and/or flame conditions, even in the spectral range above 780 nm for which polycyclic aromatic hydrocarbon (PAH) interferences are expected to be minimal. This confirms the wavelength dependence of the soot optical property $E(m)$. In particular, the value of $E(m)$ tends to decrease with increasing wavelength and the rate of decrease is lower for more mature soot particles. Furthermore, by fitting the extinction coefficient with wavelength in the near-infrared range, the quantitative relation of $E(m)$ with λ was derived and compared among various flame conditions. The present study demonstrates that soot formed at different conditions have different optical properties. The results are also expected to provide essential information for uncertainty evaluation in literature F_V data as measured in counterflow diffusion flames with light extinction, especially for those performed with visible light sources where PAH interference may not be negligible.

1. Introduction

Soot emissions from incomplete combustion of hydrocarbon fuels are known to have negative impacts on both the atmospheric environment and human health [1–3]. Its presence also indicates a loss in thermal efficiency for practical combustion devices [4]. Therefore, it is of importance to develop innovative technologies to control soot emission, necessitating a fundamental understanding of its formation mechanisms.

Soot formation has been known to be one of the most complex phenomena in combustion, involving complicated interactions among combustion chemistry, fluid mechanics, mass/heat transport, and particle dynamics. As such, it may not be surprising to find that many aspects of soot formation remain poorly understood [5,6]. Such a lack of fundamental knowledge motivates researchers to study in detail the sooting characteristics of laboratory-scale reactors and laminar flames, in which a simple flow configuration makes the investigation on soot mechanisms more tractable. For instance, shock tubes [7,8], flat premixed flames [9–12], axisymmetric coflow diffusion flames [13–16], and quasi-one-dimensional counterflow diffusion flames [17–19] have been used. In these investigations, accurate measurement of various soot properties is a prerequisite for meaningful understanding of the soot formation processes. Soot volume fraction (F_V), as an indication of overall soot mass production/concentration, is a particularly important

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variable for accurate experimental determination.

Due to their non-intrusive nature, optical techniques are favored for F_V measurements in small laboratory flames. These include soot spectral emission (SSE) [20,21], laser-induced incandescence (LII) [22–24], and light extinction (LE) [16,25–27]. Both the SSE and LII techniques are based on the analyses of the spectral radiation of soot particles and their differences lie partially in the temperature at which soot radiates. In the LII measurements, soot particles are heated by a high-power laser beam to a temperature up to 4000 K [28,29], while for the SSE technique soot radiation at local flame temperature are analyzed. The LE method relies on the fact that soot particles absorb light in a broad spectral range and the extent of the absorption is related to F_V, quantifiable through the Raleigh-Debye-Gans theory [30]. Note that an essential prerequisite for accurate F_V measurements with these optical techniques is the knowledge on how soot particles interacts with light, which is typically described by the soot complex refractive index m. Unfortunately, the value of the absorption function $E(m) = -\mathrm{Im}[(m^2-1)/(m^2+2)]$ and its dependence on light wavelength has been shown to vary with the evolution and environments in which soot are formed [31–33], such that it has become a typical source for uncertainties in F_V measurements. For the LE technique, in addition to soot particles, molecular species such as polycyclic aromatic hydrocarbons (PAHs) can also contribute to the extinction [34], especially for light in the UV–Visible range such that the measured F_V could be overestimated.

A number of investigations have been conducted previously to identify the effects of incident/detection light wavelength and flame conditions on soot optical properties. For example, Zerbs et al. [29] conducted soot extinction measurements for two premixed ethylene-air burner-stabilized flames with equivalence ratios of 2.1 and 2.3. Three lasers with wavelengths at $\lambda = 532$, 632.8 and 1064 nm were used, and different absorbing behaviors were observed. When the height above burner is small (HAB $<$ 4 mm) where no soot luminosity was present, notable absorption was still detected for the 532 and 632.8 nm beams, while it was transparent to the 1064 nm beam. In general, the level of light extinction for the 1064 nm beam was significantly less as compared to 532 and 632.8 nm across the whole flame zone. They further argued that such differences cannot be explained solely by the dependence of soot $E(m)$ on wavelength, instead the absorption by PAHs must also play a role. Simonsson et al. [35] extended the work of Zerbs et al. [29] by performing extinction measurements using 12 different wavelengths in the range of 405–1064 nm and found the dependence of $E(m)$ on light wavelength was strongest for nascent soot and decreased for soot located at increasingly larger HAB. Migliorini et al. [31] conducted spectrally-resolved extinction measurement for both rich premixed and coflow diffusion flames using broadband lamps as the light source. The attenuated light signals were spectrally separated by a spectrometer before being recorded with a CCD camera. Significant variations of $E(m)$ with wavelength (in the range of 450–700 nm) were noticed although the dependence was generally weaker for the diffusion flame. Furthermore, $E(m)$ was seen to stabilize above 700 nm for both flames, based on which the authors recommended to use light sources above 700 nm in soot extinction experiments. On the other hand, Coderre et al. [36] stated that for cool soot extracted from a methane coflow diffusion flame, the variation of $E(m)$ with wavelength in the range of 450–750 nm could be ignored. Using the two-excitation wavelength LII method, Cleon et al. [37] measured the ratios of the soot absorption function $E(m)$ at 532 and 1064 nm along a rich premixed methane flame. The results showed that the value of $E(m;\ 532\,\mathrm{nm})/E(m;\ 1064\,\mathrm{nm})$ decreased significantly with HAB in the nascent soot region while it tended to level out at higher HAB, indicating that soot at different flame locations have different optical properties. Michelsen et al. [38], by conducting LII experiments with 532 and 1064 nm beams, determined the absorption cross-section of mature soot from coflow ethylene diffusion flames. Lechowski et al. [39] measured the F_V of a coflow diffusion ethylene flame using both 2D light extinction and two-color auto-compensation LII techniques. The apparent F_V measured with the extinction method showed a dependence on wavelength and are always higher than those measured by LII, especially at low HAB.

It can be noted that the above studies were conducted either on premixed or coflow diffusion flames, while no similar work focusing on the spectral dependence of light extinction of soot, to the best of the authors' knowledge, has been done in counterflow diffusion flames. It is rather obvious that the flame conditions and thus the soot-forming environments are drastically different between premixed and diffusion flames. Indeed, Migliorini et al. [31] have observed differences in optical properties between soot particles formed in these two types of flames. What is equally true is that there also exist major differences in terms of soot evolution between coflow and counterflow diffusion flames. For instance, soot formed in the fuel-rich region of coflow flames will always be convected downstream towards the high temperature flame front where fuel and oxidizer are mixed stoichiometrically. As a result, the oxidation of soot by oxygen and hydroxyl radical is inevitable. In fact, many studies have observed the morphology change of soot after being oxidized in coflow flames [13,40]. For counterflow diffusion flames, however, the evolution of soot particles is dependent on the relative position between the stagnation plane and the flame front. It is possible, by adjusting the dilution levels of fuel and oxidizer streams and thus the stoichiometric mixture fraction, to establish a soot formation (SF) flame where soot particles, once formed, will be convected away from the flame without much oxidation [41]. Furthermore, the residence time for soot particles in coflow flames is typically much longer than in counterflow flames [42], which may lead to their different behaviors in light extinction due to different soot aging time.

A counterflow diffusion flame (CDF) is a canonical flame configuration which has a close relation with the laminar flamelet model for turbulent combustion. This fact together with CDF's unique sooting characteristics has motivated various research groups to perform detailed CDF-based soot investigations, mostly using optical measurement techniques. Among these, Amin and Roberts [43] combined extinction and two-angle scattering with a 514.5 nm beam to measure both soot volume fraction and morphology in N_2-diluted ethylene/air CDFs at elevated pressures. Similarly, Wang and Chung [44] studied the sooting characteristics of ethylene CDFs with CO_2 dilution using light extinction at 514.5 nm. Sung and coworkers used LII with 532 nm excitation to measure soot volume fraction in CDFs of butane and butanol isomers [19] and jet fuels [45] and relied on the extinction with a 632.8 nm beam for LII signal calibration. Conturso et al. [46] performed LII with 266 nm excitation to study the effects of C_9H_{12} alkylbenzenes isomers on soot formation in ethylene CDFs and the results were reported as relative LII intensities. Feng et al. [47] investigated soot formation of model biodiesel fuels in CDF using extinction with a 488 nm beam and Gleason et al. [48,49] used multi-color pyrometry to measure soot volume fraction in CDFs to investigate the effects of temperature and pressure on soot inception.

Based on the increasing evidence that PAHs can contribute to visible light extinction and thus cause interference with soot measurement, researchers have advocated to perform soot LE experiments with $\lambda > 700$ nm [29,31,50]. Nevertheless, as mentioned before, many contemporary studies are still using extinction with visible light to probe soot formation in CDFs. Considering the optical properties of soot may be different depending on its sources [51], there is a great need to assess the influence of wavelength on optical soot measurement in CDFs.

In this regard, the present study utilized diode lasers with 10 different wavelengths in the range between 405 and 1064 nm to investigate the spectral dependence of soot extinction in CDFs. Distinction were made between soot formation (SF) and soot formation/oxidation (SFO) flames and various flame condition parameters such as fuel type, strain rate, level of fuel dilution (fuel mole fraction X_F) and level of oxidizer dilution (oxygen mole fraction X_O) were systematically varied.