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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 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 in counterflow diffusion flames as measured with light extinction, especially for those performed with visible light sources where PAH interference may not be negligible.

Keywords: Light extinction; Soot; Spectral dependence; Counterflow diffusion flame

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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, which requires 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 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 soot formation processes. Soot volume fraction (F_V), as an indication of overall soot mass production/concentration, is a particularly important 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 $E(m) = -\text{Im}[(m^2-1)/(m^2+2)]$ 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 over-estimated.

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 still 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 to 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 $E(m; 532\text{nm})/E(m; 1064\text{nm})$ 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 on 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 N₂-diluted ethylene/air CDFs at elevated pressures. Similarly, Wang and Chung [44] studied the sooting characteristics of ethylene CDFs with CO₂ 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 C₉H₁₂ 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] used multi-color pyrometry to measure soot volume fraction in CDFs with different temperature to investigate the effect of temperature 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, 49]. 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 [50], there is a great need to assess the influence of wavelength on optical soot measurement for 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. Furthermore, the uniformity of soot concentrations in the radial direction, an assumption

frequently made in CDF studies, was also explicitly tested. In addition, as the sooting zone in a typical CDF is rather narrow (order of 1 mm wide) resulting in strong axial F_V gradient, spatial resolution along the axial direction becomes critical for accurate measurement. Therefore, we also tested the effect of beam size on the measured F_V profile and determined the maximum beam diameter allowed. It is hoped that the present work could provide useful information in assessing the uncertainties in quantitative soot measurements for counterflow diffusion flames. Furthermore, by providing parametrized results, it may also be possible to rectify previous CDF measurements with visible light for PAH absorption.

2. Experiment

2.1. Theoretical basis

The intensity of a light beam is attenuated after being passed through a soot-laden flame due to absorption and scattering. Such attenuation can be quantified by determining the ratio of the initial light intensity (I_0) and that after transmitting the flame (I), defined as the transmittance ($\tau = I/I_0$). According to the Beer-Lambert law stating the absorbance of a sample is directly proportional to its optical path length and the concentrations of the attenuating species, τ can be expressed as:

$$\tau = \frac{I}{I_0} = \exp\left(-\int_0^L K_{\text{ext}} ds\right) \quad (1)$$

where L is the length of the light path and K_{ext} is the local extinction coefficient. For a homogeneous system in which K_{ext} is a constant along the optical path, Eq. (1) is reduced to:

$$\tau = \frac{I}{I_0} = \exp(-K_{\text{ext}} L) \quad (2)$$

while for axisymmetric cases such as a counterflow diffusion flame, an inversion algorithm [51, 52] can be used to derive local K_{ext} from line-of-sight averaged data.

In a general case, soot absorption/scattering as well as molecular species absorption can all contribute to the extinction of the incident light and therefore K_{ext} can be written as:

$$K_{\text{ext}} = K_{\text{abs,soot}} + \rho_{\text{sa}} K_{\text{abs,soot}} + \sum_i K_{\text{abs,i}} \quad (3)$$

where $K_{\text{abs,soot}}$ represents the soot absorption coefficient, ρ_{sa} is the ratio of the scattering to absorption coefficient, and $K_{\text{abs},i}$ stands for the absorption coefficient for molecular species i . For soot particles with primary particle sizes significantly less than the laser wavelength (within the Rayleigh limit), the scattering contribution to extinction is rather small and it is a common practice to assume $\rho_{\text{sa}} = 0$ [35, 53]. Note that due partially to a shorter residence time in typical CDFs (see, e.g., [54]), the average particle size is generally smaller than those in coflow diffusion flames, where the assumption of negligible scattering contribution to extinction is also frequently invoked [20, 27]. Theoretical calculations by Simonsson et al. [35] have shown that absorption in the visible to near-infrared regime by major combustion products are dominated by H_2O lines at near 928 nm. However, its contribution to the extinction is in the order of 10^{-4} and can be safely neglected, leaving $K_{\text{abs},i} = 0$ for major molecular combustion products. While PAH species are known to absorb UV-Vis light and those either in the gas phase or condensed on particle surface can contribute to the extinction. In the present study, we did not intend to separate the effects of PAH absorption and therefore the reported K_{ext} should be understood as including both the effects of soot and PAHs. Although this may seem undesirable, it is important to note that such separation is not possible if light extinction is the only experimental technique available. More importantly, as the motivation of this work is to investigate the spectral dependence of light extinction in sooting flames, contribution from PAH species is an integral part and in fact needs to be included if the present data is to be used for assessing previous literature LE data. Nevertheless, it is worthwhile to mention that using the combination of auto-compensating LII and light extinction, it is possible to quantify the contribution of soot precursors to the extinction signal [39]. PAH interference was also suggested to be negligible for near infra-red light with wavelength higher than 700 nm [31, 35].

Assuming negligible scattering, for particles in the Rayleigh limit, the extinction coefficient K_{ext} is related to the particle size distribution function $P(D)$ as:

$$K_{\text{ext}} = \frac{\pi^2}{\lambda} E(m) N \int_0^\infty P(D) D^3 dD \quad (4)$$

where D and N are the size and total number density of primary particles, respectively. Both m and $E(m)$ are expected to vary with λ . Note that by definition the soot volume fraction (F_v) can be expressed as:

$$F_V = \frac{4}{3} \pi N \int_0^\infty P(D) \left(\frac{D}{2}\right)^3 dD = \frac{\pi}{6} N \int_0^\infty P(D) D^3 dD \quad (5)$$

Therefore, the relationship between F_V and the extinction coefficient can be established:

$$F_V = \frac{\lambda}{6\pi} \frac{K_{\text{ext}}}{E(m)} \quad (6)$$

This shows that if the light extinction is solely caused by soot absorption, the spectral dependence of soot optical properties (i.e., $E(m)$) can be determined by performing extinction measurement on a target flame repeatedly with light sources of different wavelengths. As mentioned previously, in this study the influence of PAH absorption was not separated and therefore, instead of directly studying the spectral dependence of $E(m)$, we assumed a constant m value of $1.57-0.56i$ [27] which was proposed for $\lambda = 514$ nm ($E(m)$ becomes 0.259 from $E(m) = -\text{Im}[(m^2-1)/(m^2+2)]$). It should be pointed out that the value of m is expected to depend on soot size, maturity as well as the physicochemical environments in which the soot particles are formed [55]. Nevertheless, the value of $m = 1.57-0.56i$ is used here to be consistent with the previous investigations conducted in diffusion flames [19, 54, 56-61], such that comparisons can be made if needed. Although other m values such as $1.90-0.55i$ [62], and $1.75-1.03i$ [33] have been used in other studies, it is important to note the relative variation of F_V with wavelength or flame conditions would not be affected by the choice of m . The F_V evaluated according to Eq. (6) is termed as “inferred” soot volume fraction, following the work of Simonsson et al. [35], to emphasize the fact that it is different from the real soot volume fraction since: 1) PAH absorption was embedded in K_{ext} and 2) variation of $E(m)$ with λ was not considered. Obviously, the value of the inferred F_V and thus its deviation from the real F_V are dependent on light wavelength. Such deviation in fact gives an indication on how the assumption of constant $E(m)$ and negligible PAH absorption, which are employed in many soot extinction measurements in the literature, affect the measurement accuracy of soot volume fractions.

2.2. Experiment

The experimental apparatus consisted mainly of a counterflow burner with flow control system and a light extinction setup, as schematically shown in Fig. 1. The burner shared a similar design with the one described previously [63]. In brief, two contoured nozzles with inner diameters of 10 mm were arranged to vertically oppose

each other with a separation distance of 8 mm. The oxidizer and the fuel streams were issued from the upper and lower nozzles, respectively, and all the flow rates were set by thermally-based mass flow controllers. To avoid the interference from ambient air movement and the formation of secondary diffusion flames, a curtain flow of nitrogen was provided. The burner was water-cooled to reduce heating by the combustion products.

The counterflow burner was placed on a two-dimensional motorized linear stage. During the measurement, the laser beam was kept fixed in space while the burner was moved relative to the beam such that different locations of the flame can be probed. A dedicated LabVIEW[®] program was developed to control the burner motion and to record the light intensity data automatically.

The light source of the extinction measurement is a diode laser whose emission wavelengths can be varied by interchanging the laser diodes. Ten laser diodes with center wavelengths ranging from 405 to 1064 nm were used and their characteristics are listed in Table S1 of the supplementary material. The diverging laser beams from the laser diode were firstly collimated through an aspherical lens ($f = 4.51$ mm), which was installed in the temperature control unit immediately ahead of the laser diode. The collimated beam then passed a mechanical chopper and two sets of silver mirrors before being focused through a series of concave and convex lenses at the central axis of the burner. The transmitted beam was refocused by an additional convex lens to the entrance of an integrating sphere, where a photodiode was attached serving as the light detector.

Fig. 1. Schematic of the light extinction experimental setup, shown are the burner assembly and the optic/electronic

components; DL: diode laser; CP: mechanical chopper; MR: silver mirror; CCL: concave lens; CVL: convex lens; IS: integrating sphere; PD: photodiode.

It is worthwhile to mention that the sooting zone in our counterflow diffusion flame is rather narrow in the axial direction (~ 1 mm), being confined in a region between the stagnation plane and the fuel side of the flame sheet. This indicates the existence of a steep axial gradient in soot concentrations. Consequently, it is of importance to minimize the beam diameter (D_b) if the axial distribution of soot volume fraction is to be sufficiently resolved, and this was achieved here by employing a Galilean beam expander to expand the collimated beam (thus further reducing the beam divergence) before re-focusing toward the burner axis.

To demonstrate the quantitative effects of D_b on the measured soot profile, axial F_v profiles of typical counterflow flames were measured using laser beams with different focal waist diameters and the results are shown in Fig. 2. Note that the beam diameter was varied by deliberately defocusing the beam and quantified through a knife-edge method [64].

Fig. 2. Effect of beam diameter D_b on the axial distribution of inferred F_V along the centerline (a) and the variation of peak F_V (b) for C_2H_4 SF flames with different X_O , measured with 670 nm laser wavelength.

Both the axial profile and the peak value of inferred F_V (a) were observed to be appreciably influenced by D_b . For measurements with increasingly larger beam size, the soot profiles became progressively broadened, indicating a blurring effect. More importantly, the measured peak F_V , which is in many cases taken as an indication of the soot loading of a flame, tended to decrease as beam diameter increases. Specifically, it decreased by nearly 30% from about 0.7 ppm to 0.5 ppm as the beam diameter increased from 170 to 665 μm . Such drastic dependence on beam size was indeed caused by the steep change of F_V across the particle stagnation plane. As will be detailed in a subsequent section and confirmed by a previous soot modelling study [54], for SF flames located on the oxidizer side from the stagnation plane, the soot particles formed on the fuel side of the flame would continue to grow as they were convected away from the flame toward the stagnation plane, where axial convection velocity became zero and the

particles leaked radially from the stagnation plane. Due to a small diffusivity of particles, they were not likely to penetrate through the stagnation plane, resulting in almost zero soot volume fraction on the immediate opposite side of the stagnation plane. This sudden drop from maximum value to almost zero requires the beam size to be sufficient small to avoid averaging errors.

In this regard, we further plotted the variation of peak F_V of three counterflow flames (differ in X_{O_2}) as a function of beam size in Fig. 2b. As can be seen, the results became reasonably invariant when the beam size is less than about 250 μm . Therefore, in subsequent experiments the beam diameters at the burner axis were all kept in the range from 150 to 200 μm , to minimize the averaging error and the relative difference among experiments as much as possible. Note since the refractive index (thus the focal length) of the lens are dependent on incident beam wavelength, the optical configuration must be adjusted every time when the laser diode was changed, in order to ensure consistent beam diameter.

Counterflow diffusion flames also feature steep temperature/density gradient which may lead to significant beam steering [65]. In the present work, the combined usage of a re-focusing convex lens and an integrating sphere was proven to be effective in minimizing beam-steering. In addition, the mechanical chopper together with the digital lock-in amplifier provided enhanced signal to noise ratio which was especially important for low soot conditions. It may be worthwhile to mention that the optical configuration shown in Fig. 1 was established through trial and error and each component was proven to be essential for reliable measurements.

3. Results and discussion

3.1 Sooting structures in counterflow diffusion flames

Before discussing the spectral dependence of soot light extinction, it is worthwhile to first describe the sooting structures, i.e., the spatial distribution of soot in counterflow diffusion flames. As mentioned previously, sooting counterflow flames can be categorized into two types: soot formation (SF) flames and soot formation/oxidation (SFO) flames, depending on the relative position of the stagnation plane and high-temperature flame sheet [41]. Note that for

a typical counterflow flame with fuel stream consisting of pure fuel ($X_F = 1.0$) and oxidizer stream consisting of air ($X_O = 0.21$), the flame sheet is located on the oxidizer side and the flame is of SF type. Proper dilution in the fuel stream and/or oxygen enrichment in the oxidizer stream can locate the flame on the fuel side if the following criteria is met [66]:

$$\frac{Y_{O,0}/\sqrt{Le_O}}{Y_{F,0}/\sqrt{Le_F}} > \sigma$$

where Y is the mass fraction, Le is the Lewis number, σ is the stoichiometric oxidizer to fuel mass ratio, the subscripts O and F refer to oxidizer and fuel, respectively, and 0 indicates the boundary free stream. The *inferred* soot volume fractions, as determined by light extinction with a 980 nm beam, are shown in Fig. 3a and b for SF ($X_F = 1.0$, $X_O = 0.27$) and SFO ($X_F = 0.26$, $X_O = 1.0$) flames, respectively.

The right panel of Fig. 3a shows schematics of SF and SFO flames. For the SF flame, the reaction zone is located on the oxidizer side of, and spatially separated from the stagnation plane. In this case, the incipient soot particles that forms in the high-temperature fuel-rich region (fuel side of the flame sheet) shall be carried towards the stagnation plane (fuel side) through convection. Meanwhile, the particles will continue to evolve through mass-adding surface reactions and particle-particle coagulation, until they reach the stagnation plane where axial convection velocity reduces to zero and the particles leak out of the particle stagnation plane. Note since the soot growth region is separated from the OH oxidizing zone (blue region in Fig. 3a, as obtained by performing numerical simulations of sooting structures using a detailed gas-phase kinetic mechanism of KAUST PAH Mech 2 (KM2) [67] coupled with a recently proposed PAH-based soot model [54]), soot oxidation is rather limited in this process. It is thus expected that the soot concentrations will reach a maximum value at the stagnation plane, and this is confirmed in the axial profile of soot volume fraction shown in Fig. 3a.

Fig. 3. Inferred F_v distributions along the axial centerline of SF flame: $X_F = 1.0$, $X_O = 0.27$ (a) and SFO flame: $X_F = 0.26$, $X_O = 1.0$ (b), as measured using 980 nm laser beam. Schematics of the sooting structures of typical SF and SFO flames are included at the right. Computed location for particle stagnation plane/flame sheet, as well as the direction for convective soot transport are shown. The light blue shading represents the oxidizing region where OH radicals are present.

For the SFO flame as shown in the right panel of Fig. 3b, the reaction zone was located on the fuel side of the stagnation plane. The soot particles once formed shall be convected towards the stagnation plane (oxidizer side). A significant difference with SF flame is that the particles now must pass the high temperature oxidizing flame zone before reaching the stagnation plane. During this process, intensive oxidative reactions are present which serve to decrease the soot volume fraction. This distinction in soot evolution between SF and SFO flames are reflected in the spatial profiles of soot distribution. As shown in Fig. 3, the soot volume fraction exhibits a skewed profile for the SF flame, indicating the dominance of soot mass growth along the direction of convection. On the other hand, a rather symmetric soot profile can be seen for the SFO flames demonstrating that destructive oxidation overtakes growth as the particles pass the flame sheet.

Following the study of axial soot distribution, we next investigate the variation of soot volume fraction along the

radial direction r . Counterflow diffusion flames are frequently treated to be quasi-one-dimensional [41, 68], meaning that various flow field variables such as axial velocity, temperature and species concentrations vary only in the axial direction while assuming a constant along the radial direction. In the present work, the quasi-one-dimensionality of the soot distribution was directly tested by comparing soot radial distributions, as obtained through tomographic inversion (three-point Abel) on the line-of-sight extinction data [51]. In particular, the radial soot profiles of a CDF ($X_O = 0.30$, $X_F = 1.0$) were measured using a laser wavelength of 670 nm, as shown in Fig. 4. The result shows that the shape and peaks of F_V profiles measured at different radius all coincide well each other up to say $r = 4$ mm, demonstrating the uniform soot distribution along the radial direction in the core region of the flame. Consequently, for the present flames it is reasonable to measure the F_V profile only along the axial centerline assuming quasi-one-dimensionality. Note that according to the local F_V 's measured at different radius, the length of the light path L in Eq. (6) can be determined. Indeed, the curve for $r = 0$ in Fig. 4 was obtained by conducting extinction only along the flame centerline, assuming a one-dimensional soot profile.

Fig. 4. The inferred F_V s profiles at various radial position r of SF flame: $X_F = 1.0$, $X_O = 0.30$.

3.2 Spectral dependence of soot formed in typical SF and SFO flames

So far, we have only discussed the F_V profiles as measured by laser beams at a specific wavelength with Fig. 3 showing the soot evolution process of a typical SF and SFO flames. However, the quantitative value of the inferred F_V may change when lasers with different wavelengths are used. Such wavelength-dependence of the light extinction

measurements will be discussed in the present and subsequent sections. As mentioned previously, the optical properties of soot particles depend on their physicochemical evolutions. In this regard, the spectral dependences of soot at different flame positions (thus different soot evolution stages) were firstly investigated for typical SF ($X_O = 0.27$, $X_F = 1.00$, Fig. 5) and SFO flames ($X_O = 0.90$, $X_F = 0.32$, Fig. 6). Note that the spatial location here was quantified by the distance y from the location of peak F_V ($y = 0$), following the depiction in Fig. 3.

As can be seen in Fig. 5, the inferred F_V 's of SF flame vary noticeably with laser wavelength in the range of 405–780 nm at all flame locations. Quantitatively, F_V at $y = 0$ as measured by the 780 nm light was nearly 35% lower than that measured with the 405 nm laser. On the other hand, the F_V 's were found to be much less sensitive to the choice of wavelength in the near-infrared range. In theory, the inferred F_V should be independent of laser wavelength as the *real* F_V , providing the assumptions of constant $E(m)$ and negligible light absorption by PAHs hold. However, the results shown in Fig. 5a demonstrates the deviation of such assumptions. The obvious decrease of inferred F_V with λ in the 405–780 nm range was most likely caused by the interferences from PAHs [31, 35]. As described in Eq. (2), K_{ext} is proportional to the light attenuation and additional light absorption by PAHs are inevitably embedded, leading to the overestimation of K_{ext} and thus F_V . The light absorption by PAHs is negligible when using laser wavelength in the near infrared spectrum as compared with the visible spectrum [29, 31, 35]. Consequently, the inferred F_V changes little in the near-infrared range and is expected to be closer to the *real* F_V . This result is qualitatively consistent with previous findings in premixed flames [31, 35], although the critical wavelength at which light absorption by PAHs becomes negligible was slightly different due partially to the different flame environments. Specifically, in premixed flames with equivalence ratios $\phi = 2.1$ and 2.3, it was seen that F_V at HAB = 15 mm (the most mature soot) as measured by the 780 nm light was respectively about 27% and 33% lower than that measured with the 405 nm laser [31, 35].

It may be worthwhile to emphasize here that, as also mentioned in the Introduction, an important objective of the present study is to provide quantitative information on how the choice of light wavelength may affect F_V determination in CDFs such that existing literature data that were obtained with visible light extinction can be potentially corrected for wavelength dependence. This is especially relevant considering the strong evidence of PAH

interference for extinction measurement with visible light [31, 35]. In this regard, the data shown in Fig. 5a, as well as those presented in similar

Fig. 5. Spectral dependence of absolute (a) and normalized (b) inferred F_V at different flame location y for C_2H_4 SF flame. The definition of y can be visualized in Fig. 3a.

figures in later sections which cover a wide range of counterflow flame conditions, can be used as a database for uncertainty assessment of existing literature data. To facilitate such usage, the data was normalized in Fig. 5b to the F_V measured at 532 nm, considering 1) it is a typical wavelength utilized for visible light extinction measurements and 2) the refractive index value we used for calculating the F_V were proposed for a nearby wavelength of 514.5nm [27].

Figure 5a further shows that the inferred F_V increases as y decreases toward the stagnation plane regardless of light wavelength, which is expected considering the soot growth process in SF flames described previously. In addition, it is clear from the normalized data (Fig. 5b) that the wavelength-dependence of the inferred F_V also depends on the flame location in that it becomes more significant as y increases. For instance, the normalized F_V decreases by 35% as

wavelength increased from 405 to 780 nm at $y = 0$ mm, while the corresponding value was about 43% for $y = 0.3$ mm. This result is partially understandable considering PAHs are precursors for soot inception and contribute to soot growth. Note that previous sooting structure analysis [54] has shown that in SF flames the PAH concentrations are decreasing along the paths for soot growth (i.e., as y decreases towards the stagnation plane).

Besides PAH absorption, the stronger wavelength-dependence at $y = 0.3$ mm can also be caused by the assumption of constant soot optical property $E(m)$ in deriving the inferred F_V . Since the soot particles are expected to be more mature due to soot mass/size growth through surface reaction and particle coagulation as they are convected from $y = 0.3$ mm towards $y = 0$, it may be regarded that the wavelength-dependence of $E(m)$ is weaker for more mature soot. Considering its practical relevance in determining inferred F_V results, the spectral variation of $E(m)$ will be further discussed in later sections.

Fig. 6. Spectral dependence of absolute (a) and normalized (b) inferred F_V at different location y for C_2H_4 SFO flame. The definition of y can be visualized in Fig. 3b. The inverted U shape arrow near 900 nm demonstrates that as y increases,

normalized F_V in the infrared region first increase and then decrease.

In contrast to the scenario in SF flames, soot particles in a SFO flame are expected to experience both soot formation and oxidation processes as they are convected towards the stagnation plane, and this may lead to different sooting behavior and soot optical properties. In this regard, the wavelength dependence of the inferred F_V in a SFO flame is discussed for both soot growth ($y < 0$) and oxidation regions ($y > 0$). First, we can see from Fig. 6a that for any fixed wavelength, the inferred F_V in the soot growth region always increase with the increase in y , which is expected considering the soot evolution process described previously. In the soot oxidation region of $y > 0$, however, the F_V decrease as y increases, which is due to the destructive oxidation process of soot particles. Compared to the F_V measured with 532nm where PAH absorption may be inevitable, it can be seen in Fig. 6b that in the wavelengths range of 780–1064 nm, where the PAH interferences are expected to be minimal, the normalized F_V increases and then decreases as y increases. With respect to PAH absorption, this is consistent with the growth and oxidation processes of soot in SFO flames in that: 1) PAH concentrations have been shown to decrease with the increase in y in the soot growth region ($y < 0$); 2) PAH concentrations may increase again in the $y > 0$ region due to the destructive particle fragmentation/oxidation process. Indeed, it was previously observed a peak of PAH concentration can be present in the oxidation region of SFO flames [69, 70]. In addition, in terms of the spectral dependence of $E(m)$, the fact that wavelength-dependence becomes weaker as y increases in the soot growth region of $y < 0$ is also similar with the observations in SF flames, where it was also shown that the wavelength-dependence was weaker for more mature soot. In the soot oxidation region of $y > 0$, on the other hand, the wavelength dependence becomes more pronounced as y increases. As oxidizing species tends to react with the outer shell [71] of the soot particles and thus expose the inner core structures, it may be conjectured that light extinction for the inner cores of soot particles may be more sensitive to the choice of wavelength, as compared to the mature graphitized particles. This result again tends to demonstrate that the wavelength-dependence of $E(m)$ is sensitive to the variation of soot maturity or morphology formed at different soot evolution stage. Note in coflow flames [36], it was also observed that at the spectral dependence was stronger for

soot particles at the edge (i.e., oxidized soot) than those at the centerline (i.e., mature soot).

Qualitatively similar with the observation in SF flames, the inferred F_V of SFO flames varied notably as the laser wavelength increased from 405 nm to 670 nm, while the changes were much weaker for the wavelength above 670 nm. However, quantitatively it was noted that the wavelength dependence of maximum F_V in SFO flame is significantly smaller than that in SF flames. For example, Fig. 6b shows that at $y = 0$, the F_V of SFO flames varied less than 6% over the wavelength range of 405–1064 nm, while the corresponding variation in SF flames was more than 35% (Fig. 5b). This may be understood from the fact that the flame temperature in SFO flames were much higher than that in SF flames, resulting in faster soot thermal aging and thus more graphitized/mature particles.

The above results demonstrate the wavelength dependence of soot extinction in counterflow diffusion flames and the drastic differences between SF and SFO flames. In addition, the wavelength dependence of $E(m)$ was shown to be stronger for more nascent soot. For further characterizing the wavelength-dependence behavior, we next move on to a comprehensive study on how the spectral dependences vary under different flame conditions. The flame parameters investigated include most typical variables in counterflow diffusion flames such as fuel/oxidizer dilution level, strain rates and fuel types.

3.3 Effects of oxidizer/fuel dilution level

It has been demonstrated in previous studies [63] and also confirmed later in Fig. 7a that an increase in oxygen concentration (X_O) would enhance soot formation in SF flames, due primarily to its effects on flame temperature. This is understandable as the rates of fuel pyrolysis and other soot growth reactions would be higher in hotter flames. Accompanied with such effects on flame environments is the potential influence of X_O on soot optical properties. To elucidate these influences, the effects of λ on the measured F_V were investigated for SF flames with different X_O and the results are shown in Fig. 7. We focused on the wavelength dependence of the peak F_V , which is frequently used to quantify the soot loading of a CDF. As can be seen, the overall variation of the inferred peak F_V with λ were similar for all X_O cases: they decreased notably in the wavelength range of 405–670 nm while changed only slightly for $\lambda \geq$

780 nm. These results again illustrate that when using laser with 405–670 nm for soot extinction experiments, the F_V could be appreciably overestimated.

Another interesting observation is that the spectral dependence was more significant in cases with lower X_O , even for wavelengths beyond 700 nm. This may be attributed to the different wavelength dependences of $E(m)$ among the various X_O cases as PAH absorption in this near-infrared regime is minimal. Since the inferred F_V s decreases with wavelength beyond 700 nm for all X_O cases, it could be conjectured from Eq. (6) that $E(m)$ also decreases with increasing wavelength, considering the obvious fact that the *real* F_V of a flame is the same regardless of how it was measured. Quantitatively, the rate of decrease of $E(m)$ varies with X_O , as observed in Fig. 7b.

Fig. 7. Spectral dependence of absolute (a) and normalized (b) inferred peak F_V for C_2H_4 SF flames at several X_O .

It is desirable that the relationship of $E(m)$ with wavelength λ are known *a priori* under various flame sooting conditions such that the measurement accuracy of the light extinction method can be improved as compared to typical cases with the constant $E(m)$ assumption. Under the present flame conditions, the variation of K_{ext} with laser

wavelength λ is quantified to obtain the relation of $E(m)$ with λ . As described in Eq. (6), with the assumption of constant $E(m)$, K_{ext} should be proportional to $1/\lambda$. However, since $E(m)$ is wavelength-dependent, the measured K_{ext} will certainly deviate from the theoretical relationship of $K_{\text{ext}} \propto \lambda^{-1}$. A dispersion coefficient α was introduced to quantify such deviation, following the work of Simonsson et al. [35]. The value of α was obtained by fitting the measured K_{ext} with λ in the form of $K_{\text{ext}} \propto \lambda^{-\alpha}$. Note that to avoid the influence of PAH interference, only K_{ext} measured with $\lambda > 670$ nm was used in the data fitting. In this way, the dispersion coefficient α can be extracted to represent the wavelength-dependence of $E(m)$.

Fig. 8. Derived dispersion coefficient as a function of y for C_2H_4 SF flames at various X_O . The evaluation was performed in the wavelength range of 670–1064 nm.

Figure 8 shows the values of α for soot formed at different locations (y) in several SF flames (at various X_O cases). As can be seen, α all deviates from unity, confirming the dependence of $E(m)$ on λ . In addition, for a specified X_O , α increases with the increase in y , while at a specified position, α becomes larger as X_O decreases. As mentioned previously for SF flames, soot particles at larger y are at an earlier evolution stage and thus with a lower soot maturity. The sooting processes in flames with lower X_O are also expected to be slower due to the reduced flame temperature. Therefore, these experimental observations confirm the influence of sooting environments on the wavelength dependence of $E(m)$: the dependence is weaker for more mature soot particles (i.e., at positions with smaller y or flames with higher X_O). The values of α were seen to be larger than unity for the present investigation, consistent with the results of the investigation in premixed flames [35].

Next, the effects of fuel concentration on soot spectral dependences are discussed for SFO flames. Figure 9 shows the inferred peak F_V for SFO flames at several fuel mole fractions ($X_O = 0.90$, $X_F = 0.28$ – 0.32). Soot loading is seen to be enhanced for larger X_F cases, due to the increased concentration of soot precursor species from fuel pyrolysis. A strong spectral dependence of the measured F_V is also observed in the spectral range of 405–670 nm (Fig. 9b). However, a comparison between Figs. 7 and 9 reveals that the wavelength dependences of soot in SFO flames are much less sensitive than those in SF flames. This may partially be explained by the fact that the PAH concentration in typical SFO flames is significantly lower (e.g., about 10 times as reported by Hwang et al. [69]) than in SF flames. As a consequence, the interferences of light absorption by PAHs are weaker for SFO flames. Furthermore, as shown in Fig. S1 of the supplementary material, the dispersion coefficient α decreases with increasing X_F , indicating a weaker wavelength dependence of $E(m)$ at higher X_F case.

Fig. 9. Spectral dependence of absolute (a) and normalized (b) inferred peak F_V for C_2H_4 SFO flames at various X_F .

3.4 Effect of strain rate

It is known that the formation and growth of soot is kinetically controlled such that the sooting characteristics are sensitive to flow residence time [72], which is inversely related to flow strain rate in counterflow diffusion flames. A variation of the nozzle exit velocity (V_0) provides a way to systematically alter the strain rate of CDFs. Figure 10 shows the effects of V_0 (i.e., strain rate) on the measured F_V for typical SF flames with $X_F = 1.00$ and $X_O = 0.25$. It shows that the maximum F_V decreases monotonically as V_0 increases, consistent with the previous findings [18, 57]. As V_0 increases, the residence time available for soot mass/size growth is reduced which eventually leads to the smaller soot size and lower soot concentrations. In addition to the fact that the inferred maximum F_V vary significantly more in the visible than in the infrared spectrum, the spectral dependence is stronger for cases with higher V_0 . The increase of dispersion coefficients α with V_0 , as shown in Fig. S2 of the supplementary material, also indicates the lower soot maturity and thus stronger wavelength-dependence of $E(m)$ at higher V_0 case.

Fig. 10. Spectral dependence of absolute (a) and normalized inferred peak F_V (b) for C_2H_4 SF flames at several V_0 .

3.5 Effect of fuel type

Soot formation starts from fuel pyrolysis such that fuel molecular structure plays a critical role in determining soot properties. It is therefore relevant to investigate the spectral dependence of soot optical properties in flames of different fuels. Although the refractive indices of soot formed in different hydrocarbon fuel flames have been compared in previous studies [61, 62, 73-75], similar work has yet to be tested in counterflow diffusion flames. In this regard, the influences of fuel types on the spectral dependence of the measured F_V in CDFs are investigated. The fuels of C_2H_4 and propene (C_3H_6), with significantly different sooting tendencies, were chosen for comparisons. It can be seen from Fig. 11 that at the same oxygen concentration X_O , the soot loadings of C_3H_6 flames are much higher than C_2H_4 flames, consistent with the higher sooting tendency of C_3H_6 as observed in previous studies [18].

Fig. 11. Comparison of spectral dependence in terms of absolute (a) and normalized (b) inferred F_V for C_2H_4 and C_3H_6 SF flames. Both flames were with $X_O = 0.25$.

A more interesting observation from Fig. 11b is that in spite of higher soot loading of C_3H_6 flames the spectral

dependence of the inferred F_V in C_3H_6 flames is similar with C_2H_4 flames. Quantitatively, as compared to the 405 nm case, the inferred peak F_V of C_3H_6 flame decreases 42.7% at wavelength of 1064 nm while the reduction is 38.6% for C_2H_4 flame. Based on the results of the foregoing, it is expected that higher soot loadings would lead to weaker spectral dependence. However, this is not reflected in Fig. 11. This interesting phenomenon may be attributed to the combined effects of the differences in fuel molecular structures and the fact that the absolute soot loadings between the two flames were drastically different.

In an effort to isolate these effects, the soot loading of the C_3H_6 flame was adjusted to match the level of C_2H_4 flames (both of which was determined with a 980 nm laser) by gradually reducing the X_O of the C_3H_6 flames until the inferred peak F_V decreases to be the same with that in the C_2H_4 flame. It turned out that X_O of the C_3H_6 flame needed to be reduced to 0.19 for the peak F_V to be matched with the C_2H_4 flame with $X_O = 0.25$. As shown in Fig. 12a, the measured F_V of C_3H_6 and C_2H_4 flames have comparable levels in the near-infrared spectrum range. It was now found in Fig. 12b that the inferred peak F_V of C_3H_6 flames is much more sensitive to the variation of wavelength in the range of 405–780 nm. Besides the effects of oxygen concentration shown in Fig. 7, such wavelength dependence of light extinction can be partly attributed to the effects of fuel types.

The larger deviation of the inferred F_V (as measured with visible light) from the real F_V in C_3H_6 sooting flame may be attributed to both stronger PAH interference and the wavelength-dependence of soot optical property $E(m)$. To further identify their respective role, the dispersion coefficient α , as an indicator of deviation of $E(m)$, is evaluated in the wavelength range of 670–1064 nm and derived at the cases where maximum F_V occurs. The result shows that the dispersion coefficient α in C_3H_6 and C_2H_4 flame are in fact comparable (1.26 for C_3H_6 flame and 1.25 for C_2H_4 flame, not shown), indicating that the soot optical property itself seems insensitive to the fuel types, as also observed by Bejaoui et al. [76]. In this regard, it can be concluded that the stronger spectral dependence in C_3H_6 flame is mainly contributed by the light absorption of large PAHs. Indeed, as experimentally reported in the previous work [57], the PAH concentration in C_3H_6 /air flame was much higher than in C_2H_4 /air flame.

It is important to note, however, only two fuels were tested in the present study while more comprehensive

investigations are required for a more general conclusion on the fuel effects on $E(m)$ spectral dependence.

Fig. 12. Comparison of spectral dependence in terms of absolute (a) and normalized (b) inferred F_V for C_2H_4 and C_3H_6 SF flames. The soot production was controlled the same for both flames by using different X_O .

4. Concluding remarks

The dependence of soot light extinction on incident light wavelength was experimentally investigated in counterflow diffusion flames with a diode-laser based setup, capable of providing laser light with wavelengths in the range of 405–1064 nm. The effects of various flame conditions such as oxygen/fuel mole fractions, strain rate and fuel types on the spectral dependence of inferred soot volume fractions were studied. In addition, the relative influences of the assumption of negligible PAH absorption and constant $E(m)$ on the measurement uncertainties were discussed. The major results are summarized as follows:

- (1) For all the investigated flames, the inferred soot volume fraction decreased obviously with wavelength in the

visible range of 405–670 nm, while the change was much slighter in the near-infrared range of 780–1064 nm.

This is mainly attributed to the light absorption of PAHs, which is much more obvious in the wavelength of 405–670 nm as compared to near-infrared spectrum.

- (2) The spectral dependence of the inferred F_V is more obvious at locations further away from the peak F_V location.

Besides, the variation of inferred F_V with wavelength is stronger for SF flames with lower oxygen concentration and larger nozzle exit velocity. As compared to SF flames, the spectral dependence is weaker in investigated SFO flames, which can be attributed to different influence of PAH absorption.

- (3) The soot optical property $E(m)$ was demonstrated to be wavelength-dependent from light extinction results. The

$E(m)$ value tends to decrease with increasing wavelength and, the decreasing ratio was assessed for soot particles formed at different burner distance y and flame conditions, by quantifying the deviation of dispersion coefficient, α from unity within the Rayleigh theory. The results suggested that the wavelength dependence of $E(m)$ is weaker for mature soot at distance $y = 0$, as compared to nascent soot particles at larger distance y .

Besides, it can be concluded that for SF flames with larger oxygen concentration and SFO flames with higher fuel concentration, the dispersion coefficient is closer to unity, indicating the weaker wavelength-dependence of $E(m)$ at such flame condition.

- (4) The soot optical property $E(m)$ was found to be similar between the C_2H_4 and C_3H_6 flames. The stronger spectral dependence of light extinction of C_3H_6 flame (than C_2H_4) was due to stronger light absorption by PAH in C_3H_6 flames.

As an outlook for future research, additional experimental studies investigating the effects of soot morphology on the spectral dependence of light extinction will be valuable. This can be done by analyzing the flame-generated soot samples (i.e., through high resolution electron microscopy) collected from various CDF flame conditions. In addition, the studies on the fuel effects could be expanded.

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Declarations of interest:

None.

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

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Highlights:

- Lights with wavelengths ranging from 405 to 1064 nm used to measure soot volume fraction in counterflow diffusion flames.
- Effects of laser wavelength on soot light extinction depend on the physicochemical environments in which the soot are formed.
- Soot light extinction more sensitive to laser wavelengths in the visible than in the infrared spectrum regime.
- Soot light extinction more sensitive to laser wavelengths in soot formation (SF) flame than in soot formation oxidation (SFO) flames.

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Declarations of interest:

None.