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Measurement of soot optical properties in the near-infrared spectrum

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Abstract

The dimensionless extinction constant, K_e, was measured using the NIST Large Agglomerate Optics Facility (LAOF) for soot produced from acetylene and ethene flames. Measurements were performed simultaneously using light sources at 632.8 and 856 nm. The experiments at 856 nm represent the longest wavelength for which accurate extinction measurements have been performed for soot. The mean values of present measurements of K_e at 632.8 nm for the acetylene and ethene soot are 8.12 ± 0.59 and 9.65 ± 0.54, respectively. For acetylene, the mean value of K_e measured at 856 nm was 8.83 ± 0.69, whereas the mean value for ethene at the same wavelength was 9.35 ± 0.51. The reduction in discrepancy for the fuels between 632.8 and 856 nm may be related to beam shielding effects. As in the case of 632.8 nm, the measured K_e values for 856 nm are significantly larger than values calculated using traditional methods. The present measurements provide a more reliable value of K_e for use in optical-based soot diagnostics. © 2000 Elsevier Science Ltd. All rights reserved.

1. Introduction

Accurate soot optical properties are essential for correct interpretation of laser-based diagnostic measurements of soot. For light extinction measurements, the dimensionless soot extinction constant, K_e, and the mass specific extinction coefficients, σ_s, are used to determine concentrations using the familiar Bouguer's Law:

$$\frac{I}{I_0} = \exp\left(- K_e \frac{f_v}{\lambda} L \right) = \exp(- \sigma_s M_s L) \tag{1}$$

Bouguer's Law relates the ratio of the transmitted (I) and incident (I_0) intensities to the concentration of soot (f_v, volume fraction or M_s, mass/volume), the pathlength through the soot, L, and soot optical property (K_e or σ_s). Determination of f_v and M_s from light extinction measurements requires accurate values K_e and σ_s. Often the values of K_e and σ_s are calculated using the Rayleigh-limit solution of the Mie analysis Eq. (2) with complex refractive index, m, obtained from the literature [1–4]:

$$K_e = \frac{36\pi n_\lambda k_\lambda}{\left(n_\lambda^2 - k_\lambda^2 + 2\right)^2 + 4n_\lambda^2 k_\lambda^2}; \quad \sigma_s = \frac{K_e}{\rho_s \lambda} \tag{2}$$

where n_λ and k_λ are the real and imaginary parts of the complex refractive index, and ρ_s is the density of soot.

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Nomenclature

d_p	diameter of the soot primary particle
f_v	soot volume fraction
f_{vg}	soot volume fraction measured using gravimetric technique
I	transmitted laser intensity
I_0	incident laser intensity
K_e	dimensionless extinction constant
k_λ	imaginary part of the refractive index of soot
L	pathlength of light extinction
m_s	mass of filter collected soot
M_s	mass concentration of soot
n_λ	real part of the refractive index of soot
x_p	optical size parameter

Greek symbols

λ	wavelength of light
σ_s	mass specific extinction coefficient
ρ_s	density of soot

There are concerns regarding this method of determining soot optical properties. Using simultaneous gravimetric sampling and light extinction techniques, Choi and co-workers [5] measured K_e at 632.8 nm for soot produced from a rich premixed acetylene flame and found that it was nearly a factor of two larger than the K_e calculated using Eq. (2) (with $m = 1.57 - 0.56$ [1,6], which is the most commonly used set of refractive indices in the visible spectrum). Additional experiments performed to determine the influence of fuel and burner type [7] indicated only small variations in K_e. The large disparities between measurements and calculations of K_e using Eq. (2) (with available refractive index data) indicate that caution is necessary when interpreting light extinction data for soot concentration measurements [8].

The motivation of the present study is to accurately measure K_e in the near-infrared spectrum. There are several important reasons for obtaining soot optical properties in the near-infrared spectrum in general. First, lower levels of light scattering are expected compared to visible wavelengths. Second, there is likely to be lower levels of attenuation, which can extend the pathlength/concentration range of the light extinction measurements. Third, soot optical properties in the near-infrared and infrared ranges are required for accurately characterizing radiant transport in flames and fires [9–11]. Fourth, soot optical properties in the near-IR are commonly used for two wavelength pyrometry applications to measure soot temperature in flames [12,13] and fires [11,14]. Furthermore, efforts to simultaneously measure gas species and soot concentrations in flames using near-IR tunable diode laser (TDL) spectroscopy [15] require knowledge of soot optical properties.

2. Experimental descriptions

Experiments were performed using the NIST Large Agglomerate Optics Facility (LAOF) to accurately measure optical properties of soot from laminar acetylene and ethene flames. A detailed description of the apparatus can be found in Mulholland and Choi [7], thus only a brief discussion is provided here. Fig. 1 displays a schematic of the experimental apparatus including the LAOF and laminar burner systems. The laminar burner fuel nozzle has an outer diameter (o.d.) of 12.7 mm and the outer brass tube has an o.d. of 10.8 cm. A thread of smoke emitted by a laminar flame is mixed with dilution air as it flows through a tripper plate. The mixture is further diluted with air prior to its entrance into the transmission cell.

Simultaneous light extinction measurements were performed using a 10 mW He–Ne laser operating at 632.8 nm and a 30 mW diode laser operating at 856 nm. The motivation for simultaneous measurement using two lasers was to eliminate the influence of experiment-to-experiment variations in the determination of K_e dependence on wavelength. A pellicle beamsplitter was used (see Fig. 1) to produce co-linear beams through the transmission cell. The beams were directed through the cell to the silicon photodiode detector using gold-coated mirrors. Rotating beam blocks were attached in front of both light sources to selectively monitor the wavelength of interest. Once a steadily-burning flame was established, the incident intensity of the transmitted beam at 856 was monitored for approximately 60 s (by blocking the beam at 632.8 nm, denoted by A in Fig. 2). The incident transmitted intensity at 632.8 nm was then measured for 60 s (by blocking the 856 nm beam, denoted by B). After the background measurement was acquired, soot was introduced into the transmission cell. When the intensity ratio stabilized, the exhaust flow was directed through the filter to collect the soot (denoted by C). Light extinction information was recorded as soot was simultaneously collected on the filter. Extinction measurements were performed in alternating 60 s intervals for 856 nm (denoted by D) and 632.8 nm (denoted

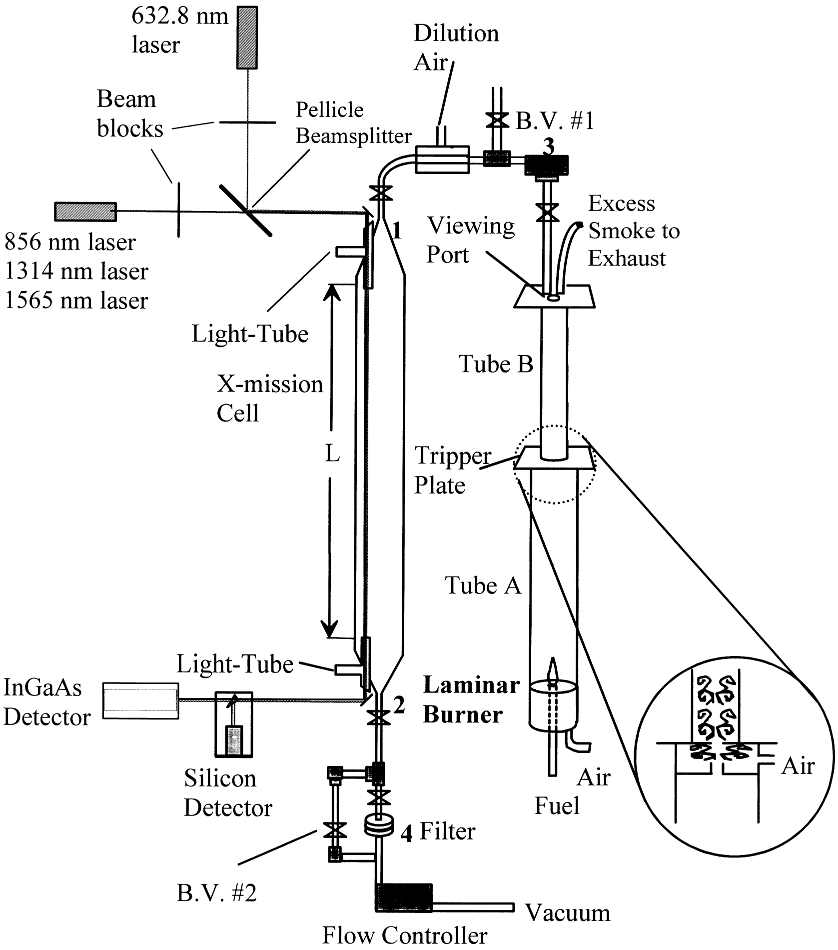

Fig. 1. Schematic of the large agglomerate optics facility and the laminar burner system.

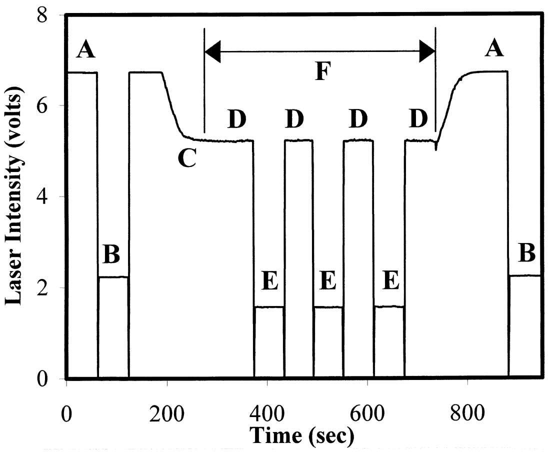

Fig. 2. Laser transmittance through the LAOF cell using a 856 nm and a 632.8 nm laser.

by E) wavelengths. The soot collection and simultaneous light extinction monitoring period was approximately 7 min. After completing the soot collection, clean air was passed through the cell (denoted by the end of F). The intensity measurements for both light sources were again recorded to ascertain whether soot was deposited on the optical windows during the collection period. The mass of sampled soot, m_s, collected on glass fiber filter was then weighed using a microbalance (2–3 μg sensitivity) with an electrostatic neutralizer consisting of a small α-emitter (500 mC, Po^{210}) to neutralize the charged filters. The mass concentration of smoke, M_s, was determined from the ratio of the mass of the deposited smoke, m_s, to the total volume of air flow through the filter.

The dimensionless extinction constant, K_e, was determined for each experiment using the measured intensity ratio, the volume of the sampled gas, V, and

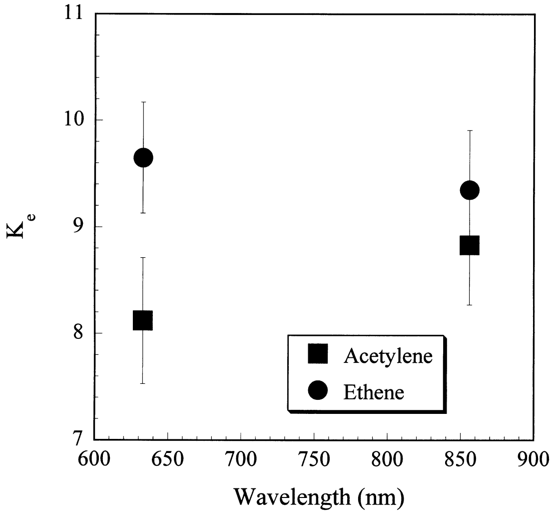

Fig. 3. K_e measurement for ethene and acetylene at 632.8 and 856 nm.

the measured mass of the soot, m_s using the following equation:

$$K_e = \frac{-\ln\left(\frac{I}{I_0}\right)\lambda}{f_{vg}L} = \frac{-\ln\left(\frac{I}{I_0}\right)\lambda\rho_s}{m_s L}V \qquad (3)$$

Soot density of 1.74 g/cc (measured in a previous study for acetylene [5]) was assumed to be constant for both fuels[1].

Previous experiments [7] using the same apparatus indicated that a combined standard uncertainty (68% confidence level), excluding the variation in the density, was 2.7%. In addition, the LAOF-measured and calculated dimensionless extinction constant of 0.5 μm polystyrene spheres (for which the refractive index is accurately known) agree within 4%, thus providing a convincing test of the accuracy of the present measurement technique.

3. Results and discussions

Fig. 3 displays the measured K_e for acetylene and ethene soot. The discrepancy between the present and previous measurements for the same fuels using the identical apparatus and the same soot density [7] was less than 3%. The mean values of present measurements of K_e at 632.8 nm for the acetylene and ethene soot are 8.12 ± 0.59 and 9.65 ± 0.52, respectively. For acetylene, the mean value of K_e measured at 856 nm was 8.83 ± 0.56, whereas the mean value for ethene at the same wavelength was 9.35 ± 0.51. The uncertainty limits correspond to the estimated total expanded uncertainty (95% confidence level) based on total Type B and total Type A uncertainties [17]. The total Type B uncertainties are based on scientific judgement rather than statistical means and equal 2.7% of the mean value [7]. The total Type A uncertainties (which are evaluated by statistical methods) based on typically five repeat measurements on each of three days equal about 0.5% of the mean for ethene and about 2% of the mean for acetylene. The measured K_e values at 856 nm are also significantly larger than the value of 5.2 calculated using Eq. (2) with index of refraction reported by Dalzell and Sarofim [1]. Therefore, these data indicate, similar to the 632.8 nm case, that use of calculated K_e at 856 nm can lead to potentially large errors in soot concentration.

The acetylene K_e is nearly 16% smaller than the ethene K_e at 632.8 nm while for the 856 nm case, the difference in these values is only 6%. This reduction in the difference between acetylene and ethene K_e is most likely caused by the beam shielding effect. Beam shielding is caused by attenuation of light by the primary spheres on the front side of the agglomerate reducing the intensity reaching the spheres on the rear side of the agglomerate. Numerical modeling [18] of this behavior indicates that this phenomenon can cause a measurable variation in soot optical properties. For the same aggregate structure (i.e. fractal dimension) the effect of beam shielding is dependent on the optical size parameter, x_p ($x_p = \pi d_p/\lambda$, d_p = primary particle size) and the number of particles that constitute the aggregate [19]. The primary particle size for overfire acetylene soot produced in a 5 cm turbulent burner is nearly 35% larger than that for ethene, and the presence of very large agglomerates for acetylene smoke is evident from the grainy appearance of the laser beam going through acetylene smoke compared to the more uniform appearance for ethene smoke. These differences suggest that acetylene is more susceptible to beam shielding effects. The decrease in the optical size parameter due to the increase in wavelength may therefore be responsible for the increase of K_e as observed in the acetylene data presented in Fig. 3. The reduction in K_e for ethene at the higher wavelength is thought to be caused by a decrease in the scattering cross section with increasing wavelength. Additional experiments and modeling efforts [18] are required to elucidate these effects.

The measurements of K_e at 632.8 and 856 nm in this study provide accurate data for a more reliable analysis of soot concentration and temperature using light extinction and two-wavelength pyrometry techniques, respectively. Additional experiments extending the

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[1] Previous measurements by Wu and co-workers [16] indicated only a 3% difference between ethene and acetylene soot density.

range of wavelengths considered along with simultaneous scattering measurements and measurements at elevated temperatures will be required to gain a broader understanding of the influence of wavelength on soot optical properties and soot optical property effects on heat transfer.

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