

Two-Color Diffused Back-Illumination Imaging as a Diagnostic for Time-Resolved Soot Measurements in Reacting Sprays

Julien Manin, Lyle M. Pickett, and Scott A. Skeen
Sandia National Laboratories

ABSTRACT

Despite ongoing research efforts directed at reducing engine-out emissions, diesel engines are known to be one of the largest sources of atmospheric particulate matter (i.e., soot). Quantitative measurements are of primary importance to address soot production during the combustion process in the cylinder of diesel engines. This study presents the capabilities of an extinction-based diagnostic developed to quantitatively measure the soot volume fraction in n-dodecane sprays injected in a high-pressure, high-temperature vessel. Coupled with high-speed imaging, the technique yields time-resolved measurements of the soot field by relying on a diffused back-illumination scheme to improve extinction quantification in the midst of intense beam steering. The experiments performed in this work used two wavelengths, which, when combined with the Rayleigh-Debye-Gans theory, provide information about the optical and physical properties of soot. For validation of the extinction imaging technique and comparison with prior work, the path-averaged soot volume fraction has been simultaneously measured using laser extinction.

We found the sensitivity of this diagnostic to soot volume fraction to be below 1 ppm despite beam-steering influences in the time-resolved data, and we were able to quantify higher soot concentrations in engine-relevant spray flames than previous studies by our group that used laser-induced incandescence (LII) in conjunction with path-averaged laser extinction. This work demonstrated a preference for shorter visible wavelengths, as soot-generated extinction is higher while broadband flame emission is lower at these wavelengths. Consequently, shorter visible wavelengths resulted in a greater signal-to-noise ratio. Analysis of two-color extinction indicated that the refractive index and morphological properties of soot change during the formation and oxidation processes.

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INTRODUCTION

Direct injection diesel engines have become the first choice for ground transportation systems seeking high efficiency; however, this mode of combustion suffers from particulate matter (mainly soot) in the exhaust. Recent studies indicate that climate change forcing due to soot has been underestimated by a factor of two for the last decade [1] and concerns of small particulate matter (PM) on health are intensifying. As a result, the pollutant emission regulations with respect to soot/PM are becoming progressively stricter. With the ongoing research on fuels and combustion strategies to reduce soot emissions, more quantitative and reliable diagnostics are needed to understand the in-cylinder soot processes.

Over the years, many diagnostics have been implemented in order to study soot particle formation, oxidation, and

particle morphology. Laser extinction measurement (LEM) and laser-induced incandescence (LII) represent the most commonly used in-situ optical diagnostics for the measurement of soot volume fraction (f_v) in flames. In some cases, laser scattering measurements have been coupled to LEM to obtain additional information about primary particles and aggregates [2]. While single-color LEM alone can only provide information about f_v , LII measurements in conjunction with an accurate physical model can yield information about primary particle size in addition to the soot concentration thanks to the simultaneous LEM calibration. Two-color LII, which involves measuring LII at two laser excitation wavelengths, eliminates the need for LEM calibration [3] because the temperature of the probed region can be obtained from the ratio of the two LII signal intensities. In all cases, knowledge of the particles' optical

properties (i.e., refractive index) is required. The uncertainty in the refractive index of soot is well documented and is generally accepted to be large [4], thus affecting the measurements [5]. In addition, the vast majority of soot measurements in general have been made at atmospheric pressure and under steady, laminar flame conditions. Thus, detailed data at high-pressure are scarce in the literature.

Another drawback when measuring soot concentration in combustor engine sprays using 2-D planar LII in conjunction with LEM is the single-shot aspect of the measurement [6, 7]. Because the duration of the injection and combustion event for these studies is less than 10 milliseconds, the 10 Hz Nd:YAG lasers commonly used for LII only provide a single image per injection thus, the temporal evolution of soot during the event is not captured. The use of high-power burst lasers for high repetition-rate LII can be a solution [8]; but, this is recent technology and the cost associated to developing such installation will prohibit the widespread use of such systems in the near-term.

Another weakness of 2-D planar LII is exposed when soot concentrations are high enough to cause significant laser attenuation. As an example, for an optical thickness (commonly identified as KL in soot measurements) of 1, less than 40 % of the original laser energy will pass through the soot cloud. When KL is 2, which is a realistic value for diesel sprays, almost 90 % of the laser energy has been absorbed (or scattered). Laser attenuation is often accompanied by signal trapping, which in this case indicates that the LII signal has been attenuated by the soot cloud on its way to the detection system. Compensating for these effects can be difficult and, in general, creates major difficulties for soot level quantification [9].

Soot concentration measurements in flames have also been made using line-of-sight attenuation imaging techniques. Greeves and Meehan [10] imaged the sooting region of a flame using back-illumination; however, their results were negatively affected by density gradients in the flame because the optical system was based on shadowgraph imaging. Similar schlieren-based or shadowgraph systems were used by Chang et al. [11] and Miyamoto et al. [12] to measure soot. Nakakita et al. [13] compared different optical setups to image soot extinction with back-illumination and showed definite improvements when using diffuse lighting over collimated light due to density gradients bending the light away from the detector (beam steering). Works from Greenberg and Ku [14] and Snelling et al. [15] used a two-dimensional transmission approach to measure the soot field in 2-D. They used large optics to increase the collection angle rather than diffuse lighting. The technique proved to provide accurate information on soot concentration with low noise, but the efficacy of the setup for turbulent flames under high-pressure conditions has not been demonstrated. Thomson et al. [16] further improved the optical arrangement from Snelling et al. [15] by using a diffuse illumination source in order to reduce the effects of beam steering and improve sensitivity without amplifying the uncertainties. Forward-

illuminated light extinction is another implementation of the line-of-sight attenuation technique [17], which has proven to be useful when only one optical access is available.

To investigate the temporal evolution of f_v , in high-pressure, high-temperature spray flames while avoiding the aforementioned limitations or complexities of the other techniques, we have developed a high-speed back-illumination extinction imaging technique. This new diagnostic was validated by concurrently measuring the path-averaged optical thickness, KL , using LEM. By combining imaging experiments done simultaneously at two wavelengths to measure soot-generated extinction, we find that the optical properties of soot vary with time and space. A tomographic reconstruction of the soot field has been used to provide quantitative 3-D information of the evolution of the soot volume fraction in diesel-like sprays.

Following the present introduction, this document is composed of five additional sections. The next section discusses the optical properties of soot in diesel flames to justify the use of the diagnostic presented in this paper. The third section describes the diagnostic itself in detail, as well as the image processing required to obtain KL . The fourth section presents the results, from optical extinction to 3-D soot volume fraction as measured in sprays injected into high-pressure and high-temperature environments. Information about soot physical properties determined by the two-color technique are presented in the fifth section. The last section of the paper summarizes the capabilities and performance of this extinction-based diagnostic to measure time-resolved soot volume fraction as well as the main findings after applying the technique to combustor diesel sprays.

OPTICAL PROPERTIES OF SOOT

This section describes the steps necessary to obtain the soot volume fraction as measured by extinction imaging. The effects of primary particle size, number of primary particles per aggregate, and the influence of the refractive index on the soot volume fraction are investigated.

Rayleigh-Debye-Gans Approximation for Particle Aggregates

The predictions of soot optical properties have been based on the Rayleigh-Debye-Gans (RDG) theory for fractal aggregates in a way similar to that described by Dobbins and Megaridis [18]. The use of the RDG theory requires the following assumptions concerning the primary particles and aggregate structure: (1) the particle aggregates must follow a log-normal size distribution, (2) the primary particles must be spherical, (3) the primary particles make contact only at the surface, (4) the primary particles must be organized in a mass fractal-like structural arrangement with a fractal dimension less than two, and (5) the refractive index must be uniform across the population. The mass fractal approximation implies a fixed relationship between the number N_p of

primary particles per aggregate, the primary particle diameter d_p , and the radius of gyration of the aggregate R_g , as follows:

$$N_p = k_f \cdot \left(\frac{R_g}{d_p}\right)^{D_f}, \quad (1)$$

where k_f and D_f are the fractal pre-factor and fractal dimension, respectively. The parameters of Eq. (1) maybe obtained through direct sampling of the soot particles from the flame followed by Transmission Electron Microscope (TEM) [19, 20]. With this information, one can then follow the work of Köylü and Faeth [21] to treat either mono- or poly-disperse aggregate populations using the RDG approximation. From the RDG approximation the angular scattering function can be computed to yield the scattering, absorption, and extinction cross-sections. These parameters require knowledge of the (potentially wavelength dependent) complex refractive index of the soot particles and the wavelength at which the calculations are to be performed. It must be noted, however, that the RDG approximation and the calculations of the cross-sections are considered valid for the single scattering regime only. An evaluation of the potential for multiple scattering of light within the soot cloud is given later in the analysis of the diagnostic's uncertainty.

Extinction and Soot Volume Fraction

As an extinction-based diagnostic, the soot volume fraction is related to the amount of light that has been absorbed or scattered by the soot cloud. The two-dimensional extinction measurements in this work were converted to optical thickness, KL , using the well-known Beer-Lambert law as presented in Eq. 2:

$$\frac{I}{I_0} = \exp(-KL). \quad (2)$$

In Eq. 2, I is the measured transmitted intensity, I_0 is the incident intensity, K is the dimensional extinction coefficient and L is the path length through the soot cloud.

The soot volume fraction, f_v , from the measurement of KL extinction follows from small particle Mie theory [6]:

$$f_v = \frac{K\lambda}{k_e}, \quad (3)$$

where λ is the wavelength of the incident illumination and k_e is the dimensionless extinction coefficient. The dimensionless extinction coefficient can be obtained from the following relationship [6]:

$$k_e = (1 + \alpha_{sa}) \cdot 6\pi \cdot E(m). \quad (4)$$

The refractive index function $E(m)$ in Eq. 4 represents the imaginary part of $(m^2-1)/(m^2+2)$, in which m is the complex refractive index of the soot particles. The scattering-to-

absorption ratio α_{sa} corresponds to the ratio of the scattering and absorption cross-sections. Both the absorption component and scattering component can depend on the wavelength of the incident light. The absorption component can depend on the physicochemical structure of the soot particles, while the scattering component can be sensitive to the primary particle size, the number of primary particles per aggregate, and the physical arrangement of particles in each aggregate (morphology).

Wavelength Dependence

Based on the current knowledge about soot formation and growth, the morphological properties, and especially the number of primary particles per aggregate, are expected to change with time and position during a spray combustion event. The optical properties of soot are dependent upon several parameters, as briefly mentioned earlier in this paper or detailed by Shaddix et al. [22]. Interesting observations made by Skeen et al. [23] showed that the difference in f_v measured at two wavelengths was not uniform throughout the sooting region, especially at high ambient temperature. They noted that the two-color extinction measurements revealed two distinct regions: one closer to the lift-off length and a larger region further downstream. They associated this change in optical properties to a rapid decrease in the hydrogen-to-carbon ratio of the particles as they transition from nascent to mature soot. This transition can also be associated with the growth of the soot particles, which in its turn means that the number of primary particles per aggregate is increasing as soot particles are growing and becoming more mature.

In theory, simultaneous extinction measurements using two wavelengths could potentially yield information about d_p , N_p , and/or m . In practice, however, such details may be elusive since identical and nearly ideal illumination and acquisition systems would be required for both colors. By substituting f_v in Eq. 3 with another wavelength, we obtain the following equality:

$$\frac{K_1\lambda_1}{k_{e1}} = \frac{K_2\lambda_2}{k_{e2}}. \quad (5)$$

The parameters used in Eq. 5 are those described for Eq. 3, only the subscript indices 1 and 2 indicate whether the parameter corresponds to wavelength 1 or wavelength 2. By recombination of Eq. 5, we obtain:

$$\frac{K_1}{K_2} = \frac{k_{e1}\lambda_2}{k_{e2}\lambda_1}, \quad (6)$$

In Eq. 6, the left-hand side represents the experimentally obtained ratio of the KL images at the two different colors and L cancels since the path lengths are equivalent. The right-hand side of Eq. 6 includes the wavelengths of the two

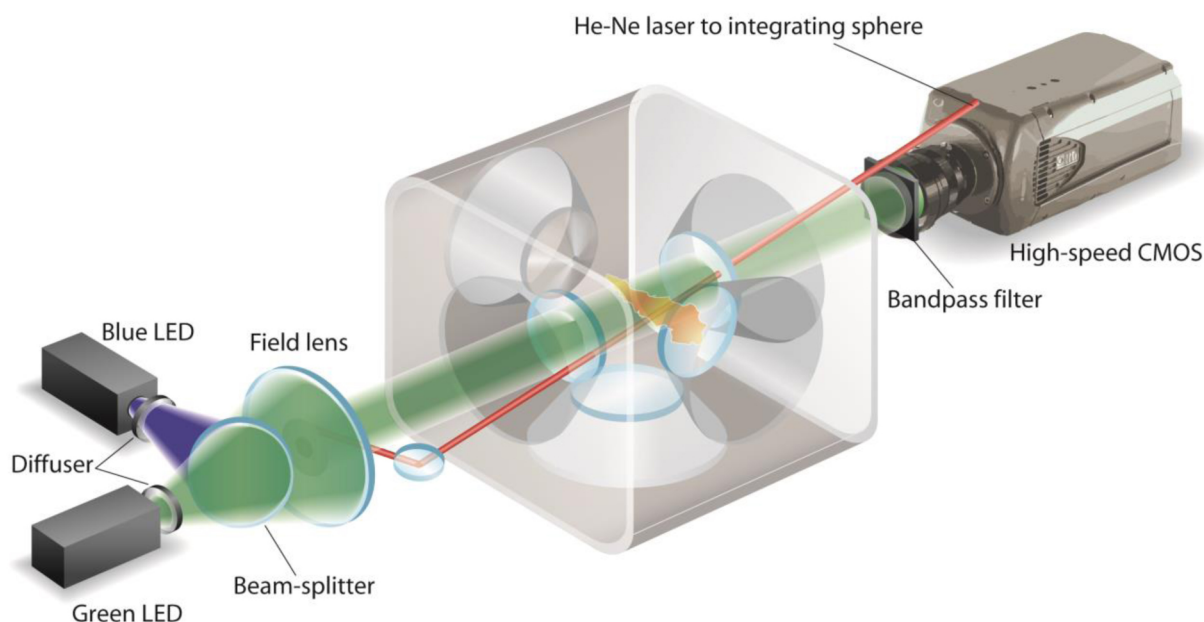


Figure 1. Optically accessible combustion vessel in the center. The 2-color diffused back-illumination optical arrangement with LED lighting (left) and high-speed camera (right). Path-averaged laser extinction layout (red beam through flame).

illumination sources and the dimensionless extinction coefficients.

The pursuit of additional information beyond f_v , via this two-color technique involves comparing measurements of the KL ratio in the flame (L.H.S. of Eq. 6) with predicted $k\lambda$ ratios (R.H.S. of Eq. 6) from the RDG theory and Eq. 4 while independently varying the different RDG input parameters (e.g., refractive index, d_p and N_p).

EXPERIMENTAL ARRANGEMENTS

The experimental installation used to generate the sprays and inject liquid fuels into a high-temperature, reactive environment has been documented in previous publications [24, 25]. A brief summary of the details pertinent to the current study is provided in this section. More attention is given to a description of the optical diagnostic presented in this paper together with the image processing and the methodology to measure and quantify the soot volume fraction in diesel flames.

Combustion Vessel and Injection System

An optically accessible high-pressure and high-temperature combustion vessel has been used for these experiments [24]. This vessel is a pre-burn type combustion chamber, which means that a combustible mixture of gases is ignited to reach the desired thermodynamic conditions at the time of injection. Prior to and during operation, the temperature of the entire vessel is maintained at 188°C by electric heaters. Spark plugs ignite the combustible gas mixture to rapidly elevate pressure and temperature within the vessel. After ignition and combustion, the pressure and temperature decrease during a short cool-down period until

the target conditions are reached and the spray is injected. At the time scales of the event (a few milliseconds), the conditions inside the chamber are considered constant and temperature measurements in the vessel have confirmed a fairly uniform temperature distribution across the testing section.

This chamber is capable of simulating charge gas conditions over a wide range of operating pressures and temperatures (up to 35 MPa and 1400 K), including that of diesel engines at top-dead-center. The combustion chamber can be operated with different oxygen partial pressures to simulate the addition of exhaust gas into the intake port of an engine (EGR). Different ambient conditions have been performed for this test campaign: ambient temperatures from 700 to 1200 K, ambient densities from 7.6 to 22.8 kg/m³ and oxygen concentrations from 13 to 21 % (by volume). However, for the purpose of this paper, only a few operating points have been selected as detailed later. To get a more complete view on the influence of the different ambient conditions, Skeen et al. [23] measured soot volume fraction in diesel-like flames while varying temperature, pressure and oxygen concentration using the same diagnostic. The nearly cubical chamber is approximately 1-liter in volume and each side can be equipped with a window for optical access. Two spark plugs are mounted on one side of the cube to ignite the premixed gases. The fuel injector is mounted on another side of the chamber, leaving four sides for optical access to the event. Figure 1 provides visual information about the lay-out of the vessel and its optical accesses.

A commercial, common-rail, solenoid-actuated injector equipped with a single-hole nozzle of 0.090 mm diameter has been used in this study. The axially drilled orifice has a

converging conical shape (KS 1.5) and hydro-grinding has been performed to minimize cavitation. This injector (serial # 210370) is part of the ECN family of injectors (www.sandia.gov/ecn). The nozzle tip of the fuel injector is held at a constant temperature of 363 K during operation. The rest of the injection system is composed of a common-rail, high-pressure lines, and an air-driven fuel pump. Even though injection pressure has been varied to study the influence of this parameter on soot formation [23], the results shown hereafter only correspond to an injection pressure of 150 MPa. High-grade n-dodecane has been used as a fuel to meet the standards of the ECN working group.

Extinction-Based Two-Color Diagnostic

The two-color, diffused, back-illumination diagnostic setup implemented in this work consists of a dual wavelength illumination source and a high-speed camera on opposing sides of the spray in a line-of-sight arrangement. High-output, ultra-fast, blue (406 nm, 17 nm FWHM) and green (519 nm, 34 nm FWHM) LEDs have been developed in-house to make this experiment possible. Although the illumination spectra for both LEDs are somewhat broad, the wavelength dependence should be easily noticeable in the RDG calculations as the wavelength difference is substantial (more than 100 nm). Both LEDs are capable of short (below 20 ns), high-power (in excess of 5 W peak power) pulses of light at repetition rates up to several megahertz. An LED pulse duration of 2 μ s was chosen as a compromise between illumination intensity and speed sufficient to “freeze” the soot fields within high velocity sections of the fuel jet. Each LED passed through a 50° circular pattern engineered diffuser with the intent of creating Lambertian intensity profiles. A dichroic beam splitter with a transition centered on 485 nm efficiently combined the two illumination sources onto one virtually identical beam of light. A 150-mm diameter, 150-mm focal length Fresnel lens focused the light onto the testing section (i.e., the spray) to create a relatively large (more than 60 mm in diameter) uniformly illuminated background. This arrangement followed the guidelines proposed by Ghandhi and Heim [26] to efficiently illuminate sprays in environments subjects to large density gradients (causing beam steering) with minimal light intensity losses (more than 90 % transmission).

On the other side of the sprays, the high-speed camera running at 82 kHz for these tests was equipped with a 50 mm f/1.2 Nikkor lens and a 500 D close-up lens to maximize collection angle by placing the camera closer to the spray. The exposure time was set to 1.3 μ s in order to acquire the highest intensity period of the LED illuminations. To effectively image the dual-wavelength extinction signals, a special combination of filters was placed in front of the camera objective. This dual-bandpass filter transmits from 375 nm to 475 nm and from 500 nm to 565 nm (both measured at FWHM), passing the full spectrum of the blue LED and the majority of the spectrum from the green LED as shown in Fig. 2. A neutral density filter of OD 1.3 was added

to reject most of the broadband luminosity from the flame. Figure 2 plots the transmission as a function of wavelength for the combination of filters used in this study. The optical setup of the two-color diffused back-illumination diagnostic is also shown in Fig. 1.

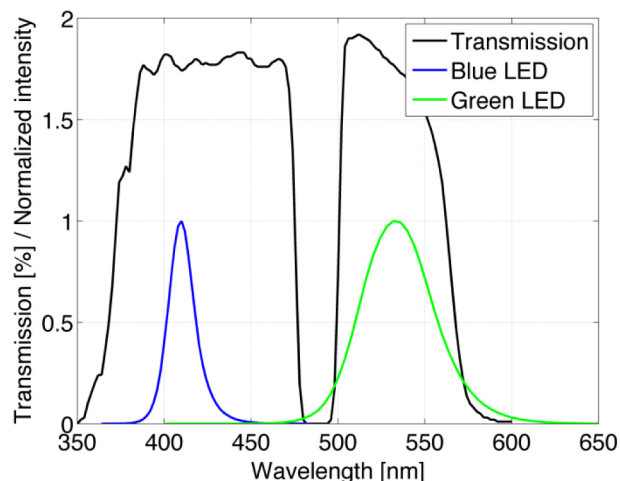


Figure 2. Spectral transmission of the filter association placed in front of the high-speed camera. The relatively low transmission is due to the neutral density filter used to reject flame luminosity. The Gaussian-like curves plot the illumination spectra for both LEDs.

Procedures and Calibration

The parameters used to calculate the optical properties of soot through the RDG approximation and obtain soot volume fraction are grouped in Table 1.

Table 1. Principle parameters used for the calculations of the RDG approximation and the soot volume fraction.

Parameters	Values
Fractal prefactor k_f	8.36
Fractal dimension D_f	1.77
Refractive index m	1.75-1.03i
Extinction coefficient k_e	7.46 (green), 7.76 (blue)
Aggregate size N_p	150
Particle diameter d_p [nm]	16 nm
Collection angle [mrad]	162 mrad
Wavelength λ [nm]	519 (green), 406 (blue)

The values for k_f , D_f , N_p and d_p have been assigned based on the results of TEM soot sampling under similar conditions [19]. These values present good agreement with other works reporting on soot sampled in diesel-like flames [27, 28]. For simplicity, the dimensionless extinction coefficients k_e were first calculated based on a constant refractive index. The value of the refractive index reported by Williams et al. [29] is considered more appropriate for the present work because they derived the refractive index by measuring the extinction