

Sizing of Soot Particles in Diesel Spray Flame -A Qualitative Comparison between TEM Analysis and LII/Scattering Laser Measurements

Masaki Kuribayashi, Yutaro Ishizuka, and Tetsuya Aizawa
Meiji University

ABSTRACT

For better understanding of soot formation and oxidation processes in a diesel spray flame, two kinds of planar soot imaging techniques, Laser-Induced Incandescence (LII) and Laser Scattering (LS) techniques, were applied simultaneously to a diesel spray flame in a constant-volume combustion vessel under a diesel-like condition (2.5MPa, 940K). An analysis of LII and LS images yielded 2-dimensional distribution images of concentration, size and number density of soot particles in the spray flame, based on an assumption that LII and LS signals are proportional to the soot particle size to the power of 3 and 6, respectively. In order to obtain clearer variation trend in the soot concentration, size and number density distribution in significantly fluctuating single-shot diesel spray flames, spontaneous and time-integrated ensemble averaging of the laser-measured images were employed. The resulting spontaneous and time-integrated images exhibited different trends in which the spontaneous images showed larger particles more concentrated in the spray head than in the steady-state-like time-integrated one. The time-integrated laser measurement results of soot particle size distribution in the spray flame were qualitatively compared with Transmission Electron Microscope (TEM) analysis results of soot particles directly sampled in a diesel spray flame under the identical experimental condition. The laser-and TEM-based results showed a similar trend that young small soot particles formed in the upstream region increase their size up to the midstream due to surface growth and agglomeration, and then decrease their size in the downstream due to oxidation.

CITATION: Kuribayashi, M., Ishizuka, Y., and Aizawa, T., "Sizing of Soot Particles in Diesel Spray Flame -A Qualitative Comparison between TEM Analysis and LII/Scattering Laser Measurements," *SAE Int. J. Fuels Lubr.* 6(3):2013, doi: 10.4271/2013-01-2576.

INTRODUCTION

For further reduction of diesel particulate matter emission, detailed understanding of soot processes in diesel spray flame including both formation and oxidation is important. Owing to the progress in fundamental soot formation dynamics [1] and also to rapid evolution in computing technologies, the numerical analysis of soot processes in diesel flame is making a remarkable progress employing detailed chemical reaction schemes and becoming capable of predicting soot particle properties at least in a qualitative manner [2, 3, 4, 5]. However, comprehensive experimental data for validation of these increasingly detailed numerical results are lacking.

There have been numerous experimental efforts on both in-situ and ex-situ investigations of soot processes in diesel spray flame. As for the in-situ investigation, which is mainly based on optical diagnostic techniques, majority of studies

have been focusing on soot concentration, ranging from readily measurable KL factor to more quantitative volume fraction. However, more detailed properties of soot particles such as the size and the number density are needed for further understanding of soot processes. Pinson [6] and Dec [7, 8] proposed a technique to obtain a qualitative 2-dimensional distribution of soot particle size from the ratio of Laser Scattering (LS) to Laser-Induced Incandescence (LII) signal intensities and applied it to diesel spray flame. Kosaka et al. [9] extended this technique with practical assumptions on size dependency of signal intensities to simultaneous 2-dimensional imaging of relative soot concentration, particle size and number density distributions in diesel spray flame. Recently developed time-resolved LII technique has also been applied to 2-dimensional soot size imaging in diesel spray flame and qualitative comparison has been made with multidimensional CFD results of diesel flame using a detailed

kinetic soot model [10]. A fundamental question is, however, what kind of soot size are measured by these optical techniques. It should also be kept in mind that the optically measured soot size is basically a relative, qualitative and spatially averaged value, for example within a pixel resolution of 2-dimensional imaging, based upon very unrealistic assumptions on soot morphology such as perfect spheres.

As for the ex-situ investigation of in-flame soot processes, Transmission Electron Microscopy (TEM) analysis of soot particles directly sampled in diesel spray flame is emerging as a powerful and quantitative technique for soot sizing. Kook and Pickett pioneered the morphological analysis of soot particles thermophoretically sampled onto a TEM grid directly exposed to diesel spray flame in a constant volume vessel [11] and more recently in a diesel engine [12]. Authors extended this technique to detailed examination along different in-flame axial locations [13, 14], fuel comparison among conventional diesel, biodiesel and Fischer-Tropsch Diesel (FTD) [15, 16], nanostructure analysis of primary particles [17], and relevant uncertainty analysis [18]. However, a major limitation of this direct soot sampling and TEM analysis compared to the highly spontaneous optical techniques explained above is the time resolution. Since the TEM grid is constantly exposed to diesel flame, the sampled soot is considered to be a time-integrated mixture of soot particles throughout the injection and combustion duration, while a time-resolved soot sampling technique with a high-speed shutter covering the grid is under development by the authors.

Since the ex-situ sampling/TEM analysis are becoming applicable for sizing of soot particles in diesel spray flame in addition to the in-situ optical measurement, it is natural to make a crosscheck and a mutual comparison between these two techniques. The comparison between the optically measured qualitative and spontaneous soot size and the sampling/TEM-based quantitative and time-integrated soot size should be complementary and valuable for better understanding of the soot processes both in transient and steady-state regimes of diesel spray flame. In the present study, the spatial distributions of soot size in diesel spray flame, based on the simultaneous LII/LS 2-D imaging technique and the sampling/TEM analysis along different in-flame axial locations, were compared under the identical experimental condition.

EXPERIMENTAL METHOD

Optical Diagnostics for Soot

The cross-sectional 2-D distributions of relative soot volume fraction, relative soot size and relative soot number density in diesel spray flame were measured via the simultaneous LII/LS 2-D imaging technique. The details of the measurement principles can be found in a paper by Kosaka et al. [9]. In this technique, practical assumptions on

the size dependency of LII and LS signal intensities are employed as follows;

$$\text{Relative soot concentration} \quad f_v \propto \text{LII} \quad (1)$$

$$\text{Relative soot size} \quad D \propto (\text{LS}/\text{LII})^{1/3} \quad (2)$$

$$\text{Relative soot number density} \quad N \propto \text{LII}^2/\text{LS} \quad (3)$$

where f_v , D , N , LII and LS denote soot volume fraction, soot size, soot number density, LII and LS signal intensities, respectively.

The shape of soot particles is assumed to be single spheres. As for the LII signal intensity, Melton [19] conducted a numerical simulation of thermal radiation from incandescent soot heated and vaporized by pulsed laser absorption and showed that the LII signal intensity is proportional to the soot size to the power of 3.3. The assumption in the present study, to the power of 3, results in the reduction of measured soot size by 55%. However, the general trend in the relative soot size distribution within the flame and thus the conclusion of the present study explained in the later section is not affected by this assumption. The light scattering measured in the present study is assumed to be in the Rayleigh regime, considering that the gyration radius of in-flame soot aggregates via TEM analysis are mainly distributed below 50nm (corresponding to gyration diameter of 100nm). The scattering intensity from soot particles in Rayleigh regime is, at least theoretically, known to be proportional to the soot size to the power of 6. The effects of aggregate shapes and the fusion of multiple primary particles within aggregates indeed have effects on absorption, radiation and scattering of light by soot particles [20, 21], but these effects are not considered in the present study. Therefore, the soot size and number density measured in the present study are inherently qualitative.

Optical Setup and Constant Volume Combustion Vessel

Figure 1 shows an overview of the optical setup for simultaneous 2-D imaging of soot via the LII and LS techniques. As the excitation light source for the simultaneous LII and LS imaging, a frequency doubled pulsed Nd:YAG laser (Solar TII LS-2137U, 250mJ at 532nm,) was used. The laser beam passed through a set of cylindrical lenses to produce a horizontal laser sheet with a thickness of 0.5mm and a width of 30mm. The laser sheet penetrated the central cross section of a diesel spray flame in a constant volume combustion vessel through a pair of sapphire windows of 35mm inner diameter as shown in Fig. 1.

The laser pulse energy was selected to ensure that the LII from soot particles in diesel spray flame are detected over the whole cross section of the flame and also measured in the

saturation regime. The laser energy density in the laser sheet is estimated to be 1.7 J/cm^2 , which is equivalent to or slightly higher than the ones employed in the LII measurements of diesel in-flame soot by other researchers[6, 7, 8, 9, 10]. The measured soot size distribution did not exhibit notably smaller size in the laser incident side, which ensures that the measured soot size is not affected by soot evaporation due to excessively high laser energy.

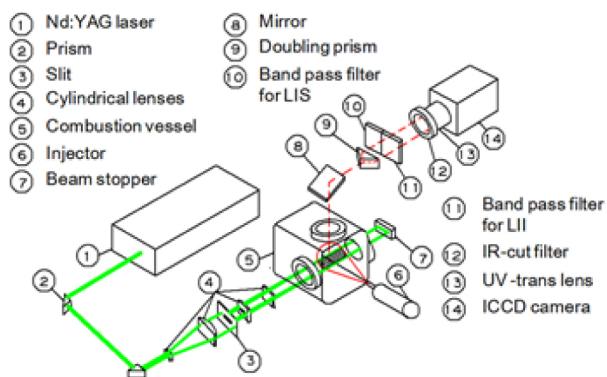


Fig. 1. An overview of optical setup for simultaneous 2-D imaging via LII and LS techniques

The LII and LS images were taken through another sapphire window on the top of the combustion vessel. Since the imaging region in the spray flame is limited by the window diameter and the laser sheet width, the distance from the injector tip to the measurement location was varied by inserting different spacers between the combustion vessel and the head mounted with the injector. The imaging setup is composed of band pass filters, a doubling prism, ND filters, UV lens (Micro-Nikkor 105mm, F4.5) and an ICCD camera (Andor iStar DH734). The spectral characteristics of the band pass filters are: central wavelength of 532nm with 10nm FWHM for LS imaging, and 400nm with 65nm FWHM for LII imaging, respectively. By using a doubling prism, simultaneous 2-D images of LII and LS from soot particles in the spray flame were obtained as shown in [Figure 2](#). The gate width of ICCD camera was set at 40ns to minimize background radiation from the luminous flame. Background correction of the natural soot luminosity was not performed, but the background signal level was less than 2% of the LII signal intensity. There was non-negligible background, up to 7%, in the LS signal mainly due to secondary scattering from the combustion chamber wall especially in the downstream sooty region in the flame. The image analysis was then only performed in the regions where sufficient LII signal from soot was detected, which effectively excluded the scattering signal from the chamber wall. Synchronization among a single-shot diesel spray flame, laser pulse and ICCD camera gating was controlled by a timing controller (LabSmith, LC880) and a digital delay generator (SRS DG535).

The combustion vessel has an inner volume of 560 cm^3 and is equipped with a common-rail fuel injector with a single-hole nozzle mounted on its head. Diesel-like high-

pressure and high-temperature conditions (2.5MPa, 940K) were simulated by spark-ignited combustion of a premixture of acetylene (C_2H_2), oxygen (O_2) and nitrogen (N_2). [Table 1](#) shows the experimental conditions. The test fuel used in the present study is Fischer-Tropsch Diesel (FTD) fuel. Authors have been investigating soot formation and oxidation processes in FTD spray flame via direct in-flame soot sampling/TEM analysis and also various optical techniques including EEM spectroscopy with a multi-wavelength laser source. The present study was conducted in the series of these FTD fuelled experiments.

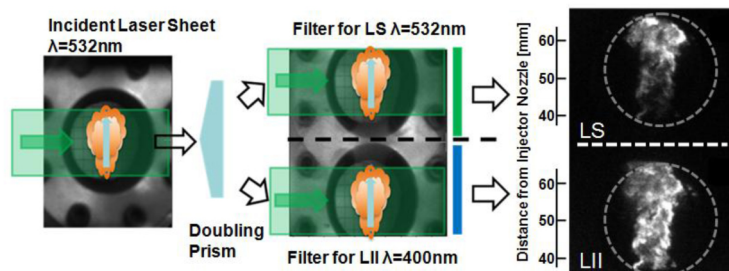


Fig. 2. Example images of LII and LS taken through a doubling prism

Table.1. Experimental conditions

Temperature : Ta	940K
Pressure : Pa	2.5MPa
O ₂ concentration : X _{O₂}	21%
Injection Pressure : ΔP _{inj}	80MPa
Injection Duration : Δt _{inj}	2.3ms
Fuel	Fischer-Tropsch Diesel
Observation Timings	1.5, 2.1, 2.7, 3.3ms
Observation Locations(center)	50, 80, 110mm

Selection of Measurement Timings for LII/LS Imaging

[Figure 3](#) shows high-speed laser shadowgraphs of a single-shot diesel spray flame, which was taken under the same experimental condition but with different setup as the one shown in [Fig.1](#), using a CW blue laser, a laser line filter and a high-speed camera. The black thick shadow seen in the shadowgraphs mainly at the center and the head of the spray flame is the soot cloud. Soot seems to appear around 50mm downstream from the nozzle. [Figure 4](#) shows the total