

Effects of Ambient Oxygen Concentration on Soot Processes in Diesel Spray Flame - A Qualitative Comparison between TEM Analysis and LII/Scattering Laser Measurements

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ABSTRACT

For better understanding of soot formation and oxidation processes in diesel combustion, effects of ambient oxygen concentration on in-flame diesel soot particle properties including concentration, size, number density and morphology were investigated in a constant volume combustion vessel via simultaneous LII (Laser-Induced Incandescence) / LS (Laser Scattering) imaging techniques and TEM (Transmission Electron Microscopy) analysis. An analysis of LII and LS images yielded 2-dimensional distribution images of concentration, size and number density of soot particles in diesel spray flame, based on a practical assumption that LII and LS signals are proportional to the soot particle size to the power of 3 and 6, respectively. The laser measurements and TEM analysis results of soot particles directly sampled in the diesel spray flame showed a consistent general trend that in the case of 21% ambient O_2 concentration soot is formed earlier in the upstream region and disappears earlier due to faster oxidation, while in the case of 15% ambient O_2 concentration, soot is formed later in more downstream region and disappears more slowly. In the case of 21% ambient O_2 concentration, locational differences in in-flame soot processes were clearly observed in which many small young soot particles formed in the upstream grow into larger particles and the number density decreases in the downstream due to agglomeration. The lowered 15% ambient oxygen concentration slowed oxidation and disappearance of soot particles in the flame downstream and thus made continued agglomeration in the flame apparent, especially in the TEM analysis results. A comparison between the laser-measured and TEM-based sizes of soot particles in the 15% ambient O_2 case showed that laser-measured soot size and TEM-based gyration radius of soot aggregates exhibit over threefold similar increase in the flame towards downstream. On the other hand, the TEM-based primary particle diameter increased in the flame towards downstream only by up to 30%. These results indicate that the optically measured soot size based on the simultaneous LII/LS technique in the present study represents more of the aggregate size than the primary particle size.

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INTRODUCTION

As a powertrain for passenger vehicles, modern turbo-charged direct-injection diesel engines are superior to gasoline engines in fuel efficiency and thus expected to play an important role as effective and practical measures against global climate change. However, it is challenging to simultaneously achieve the ultra low particulate (mainly soot) emissions meeting ever stringent regulations and higher thermal efficiency with reasonable cost of the engine system. In order to streamline the development of such advanced diesel engines, detailed and quantitative understanding of soot processes such as soot particle formation, oxidation and particle morphology in diesel spray flame is very important. Owing to the progress of computing technologies and fundamental soot formation

dynamics [1], the numerical analysis of soot processes in diesel spray flame was enabled [2, 3, 4, 5]. However, a wide range of experimental data for validation of these detailed numerical results is lacking. To compensate this lack of experimental data, researchers have been making an effort via in-situ and ex-situ diagnostics of in-flame soot processes. The representative in-situ diagnostics is mainly based on optical diagnostic technique, which gives useful information such as soot concentration ranging from readily measurable KL factor to more quantitative volume fraction. Pinson [6] and Dec [7, 8] proposed a technique to obtain 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 [9] extended this technique with practical assumptions on size dependency of

signal intensities for simultaneous 2-dimensional imaging of relative soot concentration, particle size and number density distribution in diesel spray flame. Recently, time-resolved LII has also been applied to obtain soot particle size distribution in diesel spray flame and qualitative comparison has been made with multidimensional CFD results of diesel flame using a detailed kinetics soot model [10]. However, in these techniques, it should 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 unrealistic assumptions on soot morphology such as perfect spheres. Therefore, there remains a fundamental question that what kind of soot size is measured via these optical techniques. Dobbins [11] and Megaridis [12] proposed a soot aggregates theory that soot is composed of diameter of primary particles d_p and gyration radius of soot aggregates R_g . Recently, Kamimoto [13] proposed a theoretical formula presenting the emissivity of diesel spray flames using soot aggregate scattering theory. However, it has not been well examined and explained which of diameter of primary particles d_p and the gyration radius of soot aggregates R_g resembles optically measured soot size.

The representative ex-situ investigation of soot processes is mainly based on Transmission Electron Microscopy (TEM) analysis of soot particles directly sampled in diesel spray flame. In-flame soot particles are directly sampled onto a TEM grid which is exposed to a diesel spray flame throughout the injection and combustion duration [14, 15, 16, 17, 18]. Therefore, TEM analysis data of soot particles sampled in diesel spray flame does not have time resolution and is considered to be a time-integrated representation of the soot morphology during the combustion duration. Although the spatial/time resolutions of the TEM analysis is inferior to that of optical diagnostics, the TEM analysis is powerful tool to investigate quantitative data of soot particles such as detailed morphology information including primary and aggregate sizes, aggregate structure, and their distributions. In this study, a qualitative comparison between the LII/Scattering laser measurements and the TEM analysis were made under different ambient oxygen concentration relevant to EGR conditions for better understanding of the effects of ambient oxygen concentration on soot processes in diesel spray flame.

EXPERIMENTAL METHOD

Optical Diagnostics for Soot

Simultaneous LII/LS 2-D imaging techniques were applied to a diesel spray flame to obtain the cross-sectional 2-D distribution of relative soot volume fraction, soot size and number density. The details of the measurement principles can be found in a paper by Kosaka [9]. In this technique, practical assumptions of LII and LS signal intensities are introduced as follows;

Relative soot concentration

$$fv \propto LII$$

(1)

Relative soot size

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

Relative soot number density

$$N \propto LII^2/LS \quad (3)$$

where fv , D , N , LII and LS denote soot volume fraction, soot size, soot number density, LII and LS signal intensities, respectively. In this technique, soot particles are assumed to be perfect spheres. It should also be noted that the optically measured soot size is a spatially averaged value for millions of soot particles within a pixel resolution of 2-dimensional laser imaging, which might have been dominated and biased by strong signals from small number of large aggregates. Therefore, the comparison between the laser-measured and the TEM-analysis-bases soot sizes made in the later section is 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 LII and LS techniques. A frequency doubled pulsed Nd:YAG laser (Solar TII LS-2137U, 250mJ at 532nm) was used for LII and LS techniques. With the increasing recent understanding as for the LII that the excitation by 532nm may give rise to interference from larger PAH's fluorescence, many investigators would opt for 1064nm these days. The 532nm was, however, selected mainly due to its significant simplicity in the experimental setup. The authors also had previously conducted spectral investigations of PAH fluorescence and LII under the same experimental conditions with multiple excitation wavelengths. The results showed that the PAH fluorescence excited around 500nm was negligibly weak compared with the LII signal intensity [19]. Shot-by-shot laser pulse energy correction was not performed in the present study, while the fluctuation of the laser pulse energy was less than 2%. The laser beam is converted into a laser sheet with a thickness of 0.5mm and a width of 30mm through four cylindrical lenses. The laser sheet penetrated into a constant volume combustion vessel and 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. The laser energy density in the laser sheet is estimated to be $250\text{mJ}/(0.5\text{mm} \times 30\text{mm}) = 1.7\text{J}/\text{cm}^2$, 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]. However, 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.

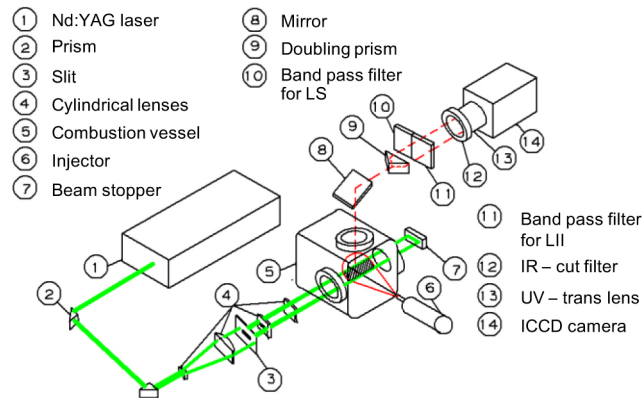


Figure 1. An overview of optical setup for simultaneous 2-D imaging via LII and LS techniques

The LII and LS images were taken through another sapphire window on the top of the combustion vessel. Since the imaging region is limited due to the sapphire window size, the distance from the injector tip to the measurement location was varied by inserting different spacer between the combustion vessel and the head mounted with 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 filter 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 LS signal intensity was attenuated by 1/32 using ND filters to roughly level the LII and LS signal intensities. The gate width of ICCD camera was set at 40ns to minimize background radiation from the luminous flame. As for the LII signal, background correction of the natural soot luminosity (less than 2%) was not performed. In the LS signal, there was non-negligible background, up to 7%, 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.

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 2.5MPa ambient pressure is on the low side of diesel-like pressures for modern turbo-charged diesel engines, which was selected mainly due to experimental limitations and comparison with other previously accumulated experimental results.

Table 1. Experimental conditions

Surrounding Conditions		
Ambient gas density: ρ_a	9.5kg/m3	
Temperature: T_a	940K	
Pressure: P_a	2.5MPa	
O_2 concentration: X_{O_2}	21%	15%
Injection conditions		
Nozzle	$\phi 0.14\text{mm} \times 1$	
Injection pressure: P_{inj}	80MPa	
Injection duration: Δt	2.3ms	
Fuel	Fischer-Tropsh Diesel	
Fuel amount: M_f	8.84mg	
Imaging conditions		
Observation Timing	1.5, 2.1, 2.7, 3.3ms	1.8, 2.7, 3.6, 4.5ms
Observation Location(center)	50, 80, 110mm	

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 an also various optical technique including EEM spectroscopy with a multi-wavelength laser source [19]. The present study was conducted in the series of these FTD fuelled experiments [18].

Selection of Measurement Timing for LII/LS Imaging

Figure 3 shows high-speed laser-shadowgraphs of a single-shot diesel spray flame in the case of 21% and 15% ambient oxygen concentrations, which were taken under the same experimental conditions as the LII/LS experiments using instead a CW blue laser, a laser line filter and a high-speed camera. The black thick shadow observed mainly at the midstream and downstream regions is the soot cloud. Soot seems to appear around 50mm and 60mm from the nozzle in the 21% and the 15% cases, respectively. The red square

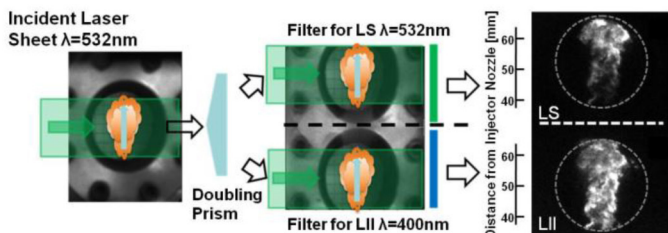


Figure 2. Example images of LII and LS taken through a doubling prism

The combustion vessel has an inner volume of 560cm³ and is equipped with a common-rail fuel injector with a single-hole nozzle mounted on its head. Diesel-like high-pressure and

locations and timings were selected as the LII/LS imaging timings, representing soot formation, growth and oxidation processes in diesel spray flames.

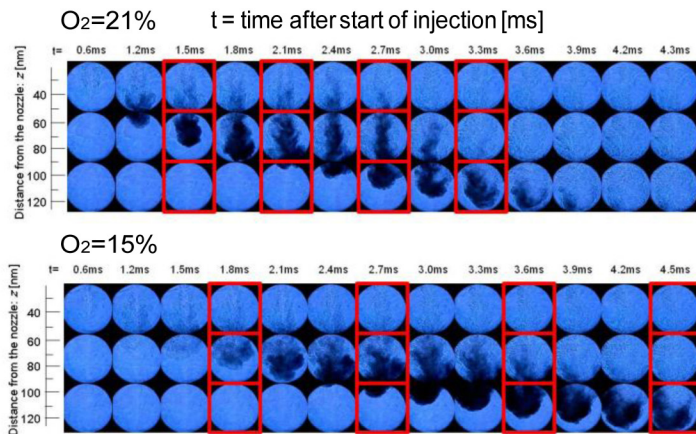


Figure 3. High-speed laser shadowgraphs of diesel spray flame at different timings after start of injection in the cases of 21% ambient oxygen concentration (top) and 15% ambient oxygen concentration (bottom)

Sampling and TEM Analysis of In-Flame Soot Particles

Diesel combustion experiments for in-flame soot sampling were conducted using the same combustion vessel as the one used for the optical measurements. Details of the sampling and TEM analysis techniques have been explained elsewhere [20, 21]. In brief, an intrusive soot sampler with a 3.1mm-diameter cavity on the tip of the top horizontal surface was placed in the combustion vessel as shown in Figure 4. An amorphous-carbon-coated 3mm-diameter copper TEM grid was placed in the cavity and directly exposed to a single-shot diesel spray flame so that the in-flame soot particles are thermophoretically sampled onto the grid surface. As briefly mentioned in the Introduction, the soot particles on the TEM grids are temporally integrated over the injection duration and considered to be a mixture of particles both at transient (head and tail of jet) and quasi-steady phases. Authors previously checked the morphology variation of the sampled soot with different injection durations. The variation was not significant and thus the soot samples in the present study is considered somewhat representing quasi-steady soot. More details of the transient morphology variation of the in-flame diesel soot is under examination with a newly developed high-speed shuttered soot sampler.

The sampled soot particles were observed using a High-Resolution Transmission Electron Microscope (JEM2100F, 200keV, 0.19nm point resolution) equipped with a CCD camera (Gatan Ultrascan 1000/FIRST LIGHT, 2048×2048 pixels). Figure 5 shows an example TEM image of soot particles directly sampled in a diesel spray flame. The diameter of primary particles d_p and the gyration radius of soot aggregates R_g were acquired from the TEM images, which were largely manual processes and supported by a MATLAB-based software developed at CRF of Sandia National Laboratories

and revised by the authors. It should be noted that the primary diameter measurement is not a simple “image analysis” process, but it is based on the recognition, by the operator, of a complicated 3-D shape and structure of soot aggregate only from its projected TEM image. For the dataset referred in the present study, total of 3000 soot particles, observed at 5 different on-grid locations on 50 TEM images obtained at 5 different axial locations in diesel spray flame, were analyzed.

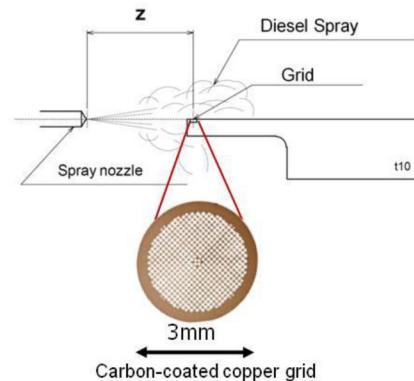


Figure 4. Side view of soot sampler placed in diesel spray flame

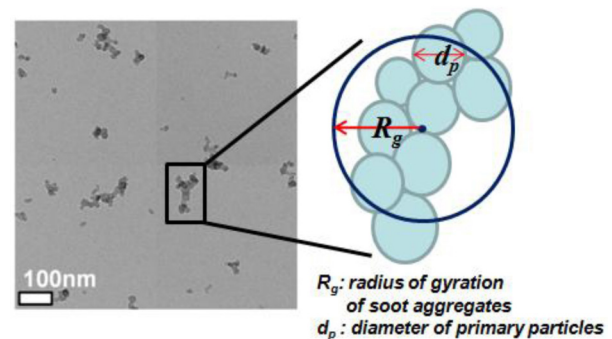


Figure 5. Example TEM images of soot particles directly sampled in diesel spray flame

RESULTS

LII/LS-Based 2-D Distributions of Relative Soot Concentration, Size and Number Density in Diesel Spray Flame

LII and LS images of soot in diesel spray flame, as shown in the examples in Figure 2, were taken at different timings after start of injection in the case of 21% and 15% ambient oxygen concentrations. The relative soot concentration images at the top correspond to the LII intensity images. The images of relative soot particle size and relative soot number density were obtained by the equations (1), (2), (3). The whole spray flame at each timing is actually a combination of 3-images separately taken at upstream, midstream and downstream regions for different single-shot spray flames under the identical condition. Discontinuous boundaries seen in the images around 65mm and 95mm from the injector nozzle correspond to the edge of the laser sheet or imaging area in each experiment. These discontinuities, however, do not obstruct qualitative interpretation of general tendency in the