

Direct Sampling, TEM Analysis and Optical Measurement of Soot Particles at Different Axial Locations in a Transient Spray Flame

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ABSTRACT

In order for better understanding of soot formation and oxidation processes in diesel spray flame, the morphology, microstructure and sizes of soot particles directly sampled in a transient spray flame were analyzed via high-resolution transmission electron microscopy (HRTEM). The soot distribution in the spray flame was simultaneously observed by high-speed shadowgraphy. The transient spray combustion was achieved in a constant volume combustion chamber under a high pressure and high temperature condition (940 K, 2.5 MPa). The soot samples were taken at different axial locations in the spray flame, 40 - 90 mm from the injector nozzle, using an intrusive soot sampler. Pressure histories, heat release rates and laser shadowgraphs of spray combustion were compared between the cases with and without the soot sampler in order to check the intrusion effect of soot sampler on combustion. Density of soot particles directly sampled onto TEM grids in the spray flame increased from 40 to 70 mm, and then decreased from 70 to 90 mm from the injector nozzle tip, which correspond to formation and oxidation regions in the spray flame observed by laser shadowgraphs. Diameter of primary particles, radius of gyration of soot aggregates and fractal dimension of soot particles were also analyzed.

INTRODUCTION

In order to reduce emission of diesel particulate matter, it is important to understand the soot formation and oxidation processes in a diesel spray flame. It is known that microstructure of soot particles reflects soot formation processes and affects on oxidation processes of soot particles. Electron microscope is known as one of the most powerful tools to observe and understand the morphology and microstructure of soot particles. Therefore high-resolution transmission electron microscopy (HRTEM) of soot particles sampled

from the exhaust gas stream has extensively been performed [1–6]. However, in order for better understanding of soot formation and oxidation processes in diesel spray flame, the properties of soot particles directly sampled in diesel spray flame are required.

On the other hand, although the investigation of soot behavior in diesel spray flame via laser-induced incandescence (LII) and laser-induced fluorescence (LIF) have been reported [7–10], understanding of the relationship between the results of such optical measurements and properties of soot particles obtained through TEM observation is lacking. Recently, Kook and Pickett have attempted direct sampling, TEM observation and optical volume fraction measurement of soot in spray flames fuelled with conventional and surrogate jet fuels [11]. The present study takes a similar approach to their pioneering work and aims to examine the soot processes in transient spray flames fuelled with diesel fuels. Our previous papers reported on TEM observation and morphological analysis of soot particles directly sampled in diesel spray flame fuelled with JIS#2 conventional diesel fuel [12] and soy-methyl ester bio diesel fuel [13] in constant volume combustion chambers. The sampling locations of soot particles in these studies were, however, limited to 3 axial locations in the spray flame, 40, 50 and 70 mm from the injector nozzle.

The present paper reports on the soot behavior in a transient spray flame fuelled with JIS#2 standard diesel fuel, investigated via high-speed laser shadowgraphy of diesel combustion in addition to the TEM observation of simultaneously and directly sampled soot particles. The soot sampling locations were also extended to 6 different axial locations from 40 mm to 90 mm from the injector nozzle orifice, aiming for more comprehensive understanding of soot processes in diesel spray flame.

EXPERIMENTAL SETUP

CONSTANT VOLUME COMBUSTION CHAMBER

Diesel combustion was achieved in a constant volume combustion chamber shown in Fig. 1. A common-rail fuel injector with a single-hole nozzle is mounted on its head. The chamber is equipped with 3 quartz windows and its internal volume is 560 cm³. A high-pressure and high-temperature condition (2.5 MPa, 940 K) was simulated by spark-ignited premixed combustion of a pre-mixture of acetylene, oxygen and nitrogen. The pressure was set below a typical modern diesel in-cylinder condition mainly due to safety limitation of the experimental devices used in the present study. However, ongoing analysis of soot samples acquired at a higher pressure condition (6.7 MPa) is not showing significant differences in the general trend of soot formation and oxidation in the flame and the properties of soot particles compared with those of the present study. Therefore, the results of the present study should be useful for discussion of soot formation and oxidation in diesel combustion. Soot sampling and optical measurement location can be varied by changing the spacer between the head and the chamber.

SOOT SAMPLER

Figure 2 shows the “soot sampler” which was designed for direct soot sampling in diesel spray flame. The sampler is 10 mm-thick and equipped with a 3.1 mm-diameter and 1.5 mm-deep cavity on the top horizontal surface to hold carbon-coated copper grid for TEM observation. The TEM grid is a 3 mm-diameter copper mesh covered with a 30 nm thick amorphous carbon film. The grid was directly exposed to the diesel spray flame and the soot particles were thermophoretically sampled onto the grid surface. Distance between the injector nozzle tip and the center of TEM grid can be adjusted by selecting different spacers for the combustion chamber. The sampler was electronically heated to 373 K together with the

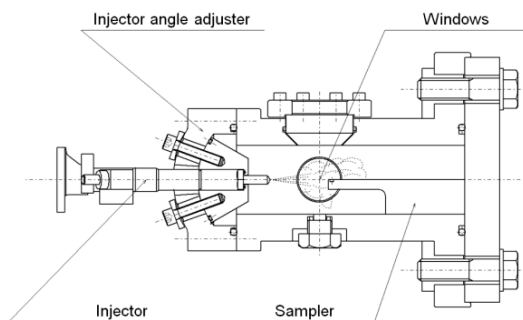


Fig. 1 Constant volume combustion chamber

combustion chamber wall to avoid condensation.

OPTICAL SETUP

In order to investigate the intrusion effect of soot sampler on diesel combustion and behavior of soot particles in diesel spray flame, high-speed laser shadowgraphy of the spray flame was conducted. Figure 3 shows the experimental setup for the high-speed laser shadowgraphy. A CW blue laser beam (460 nm, 800 mW) of 40 mm in diameter penetrates the diesel spray flame in the combustion chamber through a pair of quartz windows and the sequential shadow images of diesel spray flame (10,000 fps) are captured by a high-speed video camera (NAC GX-1) equipped with a band-pass filter.

TEM OBSERVATION OF SOOT PARTICLES

Soot particles directly sampled in a diesel spray flame onto the grid were observed using a high-resolution transmission electron microscope (JEOL, JEM-2100F) with a specified maximum point resolution of 0.19 nm. The acceleration voltage is 200 keV. All images were digitally recorded with a CCD camera (Gatan Ultrascan 1000/FIRST LIGHT). Image resolution is 2048 x 2048 and physical pixel resolution is 14 μm. The grid directly exposed to the diesel spray flame was observed without any thermal or chemical pre-treatments to maintain the original properties of soot particles in diesel spray flame as much as

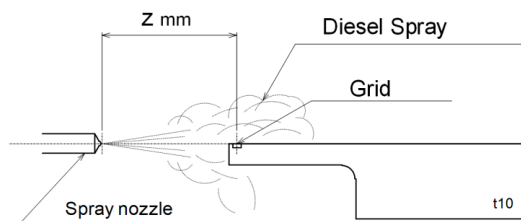


Fig. 2 Side view of soot sampler

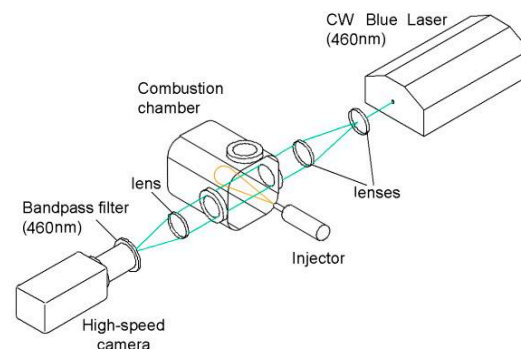


Fig. 3 Optical setup for high-speed laser shadowgraphy of diesel spray flame

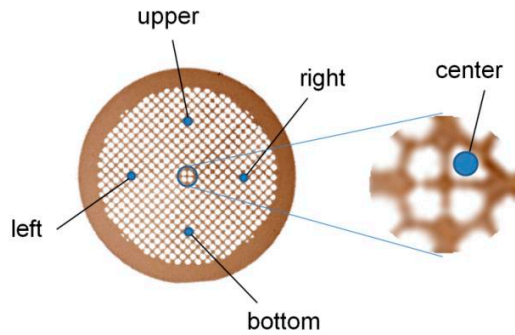


Fig. 4 Observation locations on TEM grids

possible. The TEM images of soot particles were taken at 5 different locations on the grid as shown in Fig. 4 and more than 5 images were captured at each location. Soot concentration and size of primary particles were analyzed from TEM images at magnifications of x6,000 and x20,000 respectively. The x6,000 magnification is suitable for obtaining statistically reliable soot concentration and aggregate size data, while the x20,000 magnification is required to size measurements and shape recognition of primary particles.

TEM IMAGE ANALYSIS OF SOOT PARTICLES

Primary particle size, soot aggregate size and fractal dimension of soot particles are essential properties that can help us to characterize the formation of diesel particulates and understand particulate growth process. Diameter of primary particle d_p , radius of gyration of soot aggregate R_g and projected area of soot aggregate A were acquired from TEM images using a MATLAB-based software. This image analysis software was developed at CRF of Sandia National Laboratories and revised by the authors. The number of primary particles within an aggregate, n , was calculated from substituting the cross section of soot aggregates and primary particles $A_p = \pi d_p^2 / 4$ as shown in equation (1). Fractal dimension of soot particles was obtained from the logarithmic plots of R_g/d_p and n following the equation (2) [14]. Radius of gyration of soot aggregate, R_g , was calculated using projected shape of each soot particle on binarized TEM images following pixel-based equation (3).

$$n = \left(\frac{A}{A_p} \right)^\alpha \quad (1)$$

$$n = k_f \left(\frac{R_g}{d_p} \right)^{D_f} \quad (2)$$

$$R_g^2 = \frac{1}{m} \sum r_i^2 \quad (3)$$

n : number of primary particles per aggregate
 A : projected area of soot aggregate [nm^2]
 A_p : primary particle cross section [nm^2]
 α : exponential factor for particle overlap (=1.09) [15]
 k_f : pre-factor
 D_f : fractal dimension of soot particle
 R_g : radius of gyration of soot aggregate [nm]
 d_p : averaged diameter of primary particle [nm]
 m : number of pixels per aggregate
 r_i : distance between each pixel and centroid of soot aggregate [nm]

EXPERIMENTAL CONDITIONS

Table 1 summarizes the experimental conditions. The high pressure and high temperature condition (2.5 MPa, 940 K) was achieved by premixed combustion of a premixture of acetylene, oxygen and nitrogen. A common-rail fuel injector with a single-hole nozzle was used for single-shot injection of JIS#2 conventional diesel fuel. Soot particles were sampled at 6 different axial locations (40 mm to 90 mm from the injector nozzle) in the diesel spray flame for understanding of soot formation and oxidation processes. One sample grid was obtained at each location.

RESULTS

INTRUSION EFFECT OF SOOT SAMPLER

Intrusion effect of soot sampler on diesel combustion was checked by comparing pressure histories, heat release rates and high-speed laser

Table 1 Experimental conditions

Surrounding conditions	
Ambient gas density: ρ_a [kg/m^3]	9.5
Temperature: T_a [K]	940
Pressure: P_a [MPa]	2.5
O ₂ concentration: X_{O_2} [%]	21
Injection conditions	
Nozzle	$\phi 0.14 \text{ mm} \times 1$
Injection pressure: P_{inj} [MPa]	80
Injection duration: Δt_{inj} [ms]	2.5
Fuel	JIS#2
Injection amount: M_f [mg]	10.3
Sampling conditions	
Distance from orifice: z [mm]	40, 50, 60, 70, 80 and 90
Grid type	Carbon coated copper grid
Temperature of sampler: $T_{sampler}$ [K]	373

shadowgraphs of diesel combustion with and without the soot sampler. It is indeed difficult to examine the intrusive effect of the sampler on diesel combustion and soot formation solely from the heat release. The volume of the soot sampler, approximately 22 cm^3 , is very small compared to the chamber volume and thus the effect of its volume is inherently small. However, the heat release rate is one of the most fundamental measures to understand the combustion process and it is important to verify that the spray impingement onto the sampler do not cause, for example, noticeably reduced heat release and/or elongated ignition delay compared to the case

without sampler.

Figure 5 shows the comparison of pressure histories at the top and heat release rate at the bottom between the cases with the sampler placed 40 mm downstream from the nozzle tip, corresponding to the nearest to the nozzle, and without the soot sampler. Ignition delay detected by the pressure rise was about 0.6 ms with and without the sampler. The maximum pressure was about 2.66 MPa at about 5 ms after start of injection in both cases. Pressure traces do not show noticeable differences between these cases. Heat release rates show noticeable vibration, which is due to pressure oscillation in the combustion chamber. Slightly earlier rise of the heat release compared to the one in the pressure is due to smoothing. Ignition delay, general trend of heat release and combustion period of diesel combustion do not show significant differences between these cases with and without the soot sampler. Noticeable differences were not observed also among cases with different distance from the nozzle tip to the sampler.

Figure 6 shows high-speed laser shadowgraphs of diesel spray flame with and without the soot sampler. Although the view field through the optical window of the combustion chamber used in the present study is limited to 35 mm in diameter, behavior of soot clouds in the spray flame around the soot sampler can clearly be observed. Dark shadow due to thick soot cloud is not seen until 0.7 ms after start of injection as shown in Fig. 6. Soot cloud is only seen in the fuel-rich central region in the spray flame and not seen in the fuel-lean spray periphery where soot particles are oxidized by surrounding air. Spray tip velocity was about 50 m/s in both cases with and without the sampler and significant difference was not seen. The shadowgraphs clearly show that the grid is directly exposed to the soot cloud.

The impingement of the spray flame onto the sampler surface facing upstream (to the left in the

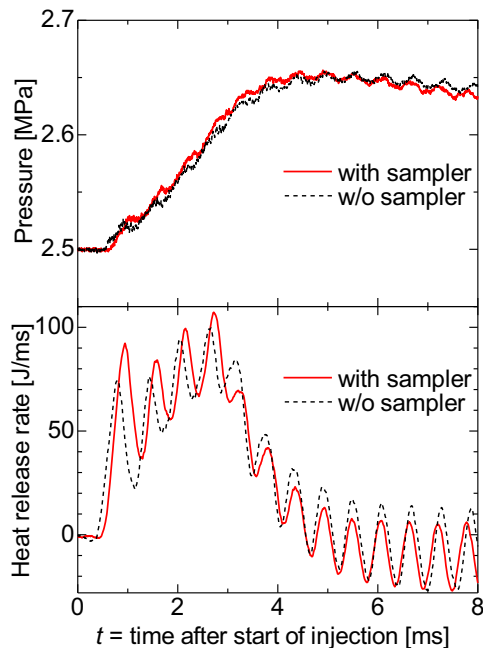


Fig. 5 Pressure histories and heat release rates of diesel spray flame with and without the soot sampler

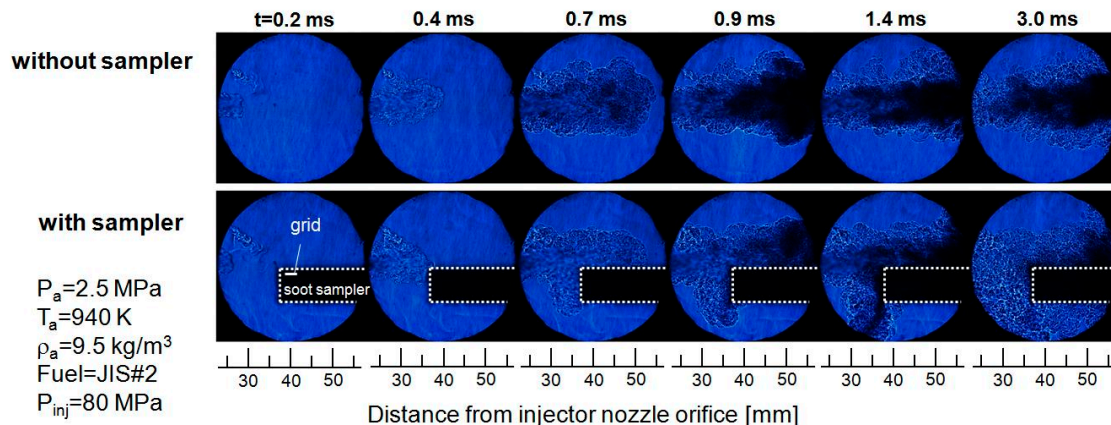


Fig. 6 High-speed laser shadowgraphs of diesel spray flame with and without the soot sampler