

Transmission Electron Microscopy of Soot Particles Directly Sampled in Diesel Spray Flame - A Comparison between US#2 and Biodiesel Soot

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ABSTRACT

For a better understanding of soot formation and oxidation processes in conventional diesel and biodiesel spray flames, the morphology, microstructure and sizes of soot particles directly sampled in spray flames fuelled with US#2 diesel and soy-methyl ester were investigated using transmission electron microscopy (TEM). The soot samples were taken at 50mm from the injector nozzle, which corresponds to the peak soot location in the spray flames. The spray flames were generated in a constant-volume combustion chamber under a diesel-like high pressure and high temperature condition (6.7MPa, 1000K). Direct sampling permits a more direct assessment of soot as it is formed and oxidized in the flame, as opposed to exhaust PM measurements. Density of sampled soot particles, diameter of primary particles, size (gyration radius) and compactness (fractal dimension) of soot aggregates were analyzed and compared. No analysis of the soot micro-structure was made. The overall morphology of the biodiesel soot bears similarity to that of #2 diesel, but the soot density, primary particle size, and fractal dimension are smaller for biodiesel.

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INTRODUCTION

For further and effective reduction of the particulate matter (PM) in the exhaust of a diesel engine, it is important to understand the properties and the microstructure of soot particles which reflect their formation and oxidation processes. There have been many studies on diesel soot morphology that include observations of soot particles sampled from the exhaust with electron microscopy [1, 2, 3, 4, 5]. However, in order to better understand the formation and oxidation processes of soot particles in the engine

cylinder, it is necessary to investigate the morphology of soot particles inside a diesel spray flame. Recently, we have developed a technique to directly sample soot particles inside a diesel spray flame [6]. The benefits of this direct sampling compared to exhaust measurements include (1) analysis of soot particles formed in different regions in diesel spray flame, (2) back-to-back comparison with in-situ optical measurements, (3) avoidance of complicated condensation issues, dilution, SOF, or mixing with lube oil, in the exhaust, and (4) avoidance of possible continued aggregation as particles collide in the exhaust. In this work, we extend this

soot particle sampling and analysis by Transmission Electron Microscopy (TEM) to include Soy-Methyl Ester biodiesel, aiming at a direct comparison of soot particle properties between the cases fuelled with the bio-diesel and conventional diesel. It has been reported that exhaust soot particles originating from bio-fuels exhibit oxidation rates and structure notably different from the ones from conventional diesel fuel [7, 8, 9, 10]. This study aims to compare the morphology of soot particles directly sampled in the soy-methyl ester biodiesel and US#2 conventional diesel spray flames under an identical experimental condition. The greatest possible care was taken to acquire statistically reliable datasets of diameter of primary particles, size (gyration radius) and compactness (fractal dimension) of soot aggregates through a labor-intensive, manual processing of the TEM images of thousands of sampled soot particles.

EXPERIMENTAL SETUP

SOOT SAMPLING

Diesel combustion and soot sampling experiments were conducted using the optically-accessible constant volume combustion vessel at Combustion Research Facility of Sandia National Laboratories shown in Fig. 1. Numerous studies on fundamental diesel spray combustion processes have been conducted using this facility via multiple optical diagnostic techniques. Ref. [11, 12] provide a detailed description of the vessel design, operating characteristics and experimental considerations. The vessel is cubic-shaped, 108mm on a side, and four sapphire windows are mounted on right, left, bottom, and front walls. A common-rail fuel injector with a single-hole nozzle is mounted on the top center and a fuel spray is injected into the center of the combustion vessel. More details on the injection hardware and rate of injection may be found in [11, 13]. A soot sampling probe is fixed on the opposed side wall to the injector. Fig. 2 shows example shadowgraphs of a diesel spray flame and the soot sampling probe taken in setup with (a) uniform ambient temperature of 450K and (b) 2380 μ s after the start of fuel injection when at a reacting condition at an elevated temperature of 900 K. A 3mm diameter carbon coated copper grid for TEM observation is mounted in the sampling probe. The probe is placed parallel to the spray axis to skim the gas containing soot from the spray flame while attempting to minimize flow disturbance at least to the upper half of the spray flame. Through a 1-mm diameter hole, reacting flows containing soot enter the probe and soot particles are deposited on the cooler TEM grid via thermophoresis. This restrictive passage ensures quenching of combustion and also protects the grid from excessive heating. Therefore, the sampled soot particles should correspond to soot present at that position. The grid and the sampling probe were heated to 450K together with the combustion vessel. The grid temperature increases from 450 K because of heat transfer with the reacting jet at temperatures as high as 2300K, but because of the thermal inertia, the temperature remains much lower than that of the

reacting jet. Dobbins and Megaridis [14] argue that the cold grid surface freezes heterogeneous reactions of the captured soot particles and prevents changes in the soot morphology after impacted upon the grid surface. Measurements of the grid temperature and the investigation of grid temperature effects on the sampled soot will be one of our future tasks. Experimental conditions are summarized in Table 1. In the present study, low-sulfur diesel (US#2) and soy-methyl ester bio-diesel (BD) fuels were used as test fuels. The biodiesel fuel is a neat (100%) soy-derived biodiesel composed of five methyl esters; 11% methyl palmitate, 4% methyl stearate, 25% methyl oleate, 53% methyl linoleate and 7% methyl linolenate. More details of fuel properties have been reported elsewhere [15]. The injection duration was set to 6ms. Experience shows that the soot particles deposited onto the grid surface during the single-shot spray injection is small enough to avoid overlapping of multiple particles. It has also been checked that the soot load on the grid changes with injection duration, but the soot morphology does not [6, 16].

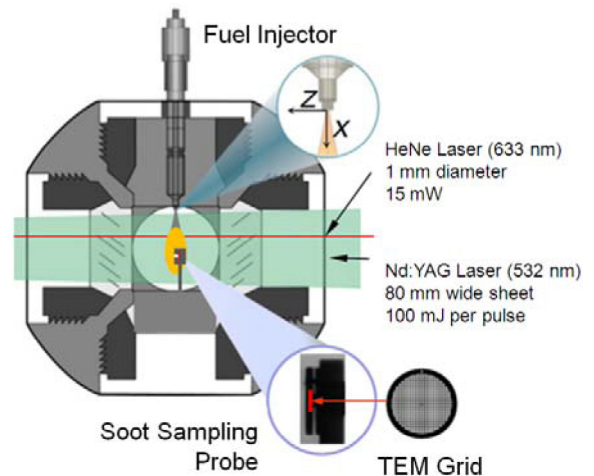


Figure 1. Constant volume combustion vessel

Table 1. Experimental conditions-

Surrounding conditions	
Gas density: ρ_a [kg/m ³]	22.8
Temperature: T_a [K]	1000
Pressure: P_a [MPa]	6.7
O ₂ concentration: X_{O_2} [%]	15
Injection conditions	
Nozzle	0.09mm $\times$ 1
Injection pressure: P_{inj} [MPa]	150
Injection duration: Δt_{inj} [ms]	6
Fuel	Low sulfur diesel (US#2) & Soy-Methyl Ester (BD)
Sampling conditions	
Distance from orifice: z [mm]	50
TEM grid type	carbon coated copper grid

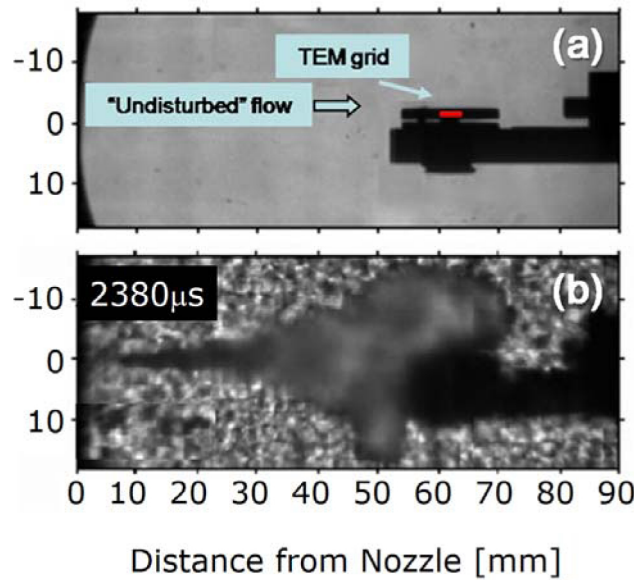


Figure 2. Shadowgraphs of diesel spray flame and soot sampling probe-

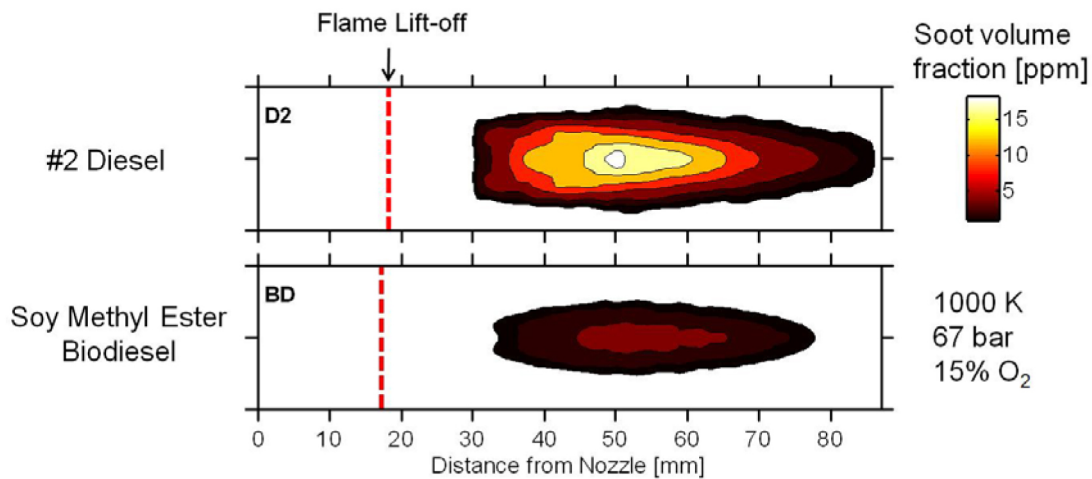


Figure 3. Soot volume fraction distribution in quasi-steady diesel spray flames fuelled with US#2 and BD fuels

In the present study, the soot sampled onto the TEM grid at 50mm from the nozzle for US#2 and bio-diesel cases were compared. The reason for the choice of the sampling location at 50mm from the nozzle is as follows. Figure 3 shows soot volume fraction measured in diesel spray flames fuelled with the US#2 and the BD fuels obtained by combining Planer Laser Induced Incandescence (PLII) and light extinction techniques. As shown in the experimental setup in Fig. 1, PLII was performed using a 80-mm-wide, 532-nm Nd: YAG laser sheet and simultaneously Light (Laser) Extinction was conducted using a helium-neon (HeNe) laser, which enabled the computation of the two-dimensional soot volume fraction. Details of the optical measurements are explained in references [6, 13]. Fig.3 shows that the soot volume fraction in the bio-diesel case is significantly lower than in the US#2

case. However, the relative profiles of soot distribution in the flame, showing a peak soot region around 50mm from the nozzle, are similar between the US#2 and BD cases. The flame lift-off locations, shown by dashed lines in Fig.3, are also similar between the two cases. Our previous observations on variation of soot morphology along different axial locations in spray flame have shown that the sizes of primary particles and aggregates of soot in spray flames exhibit the maximum around the peak soot region [17, 18]. Therefore, the present study compares the most grown-up soot directly sampled at the same location in the spray flame with similar flame structure using two different fuels.