

Nanostructure Analysis of In-flame Soot Particles under the Influence of Jet-Jet Interactions in a Light-Duty Diesel Engine

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ABSTRACT

Some soot particles emitted from common-rail diesel engines are so small that can penetrate deep into the human pulmonary system, causing serious health issues. The analysis of nano-scale internal structure of these soot particles sampled from the engine tailpipe has provided useful information about their reactivity and toxicity. However, the variations of carbon fringe structures during complex soot formation/oxidation processes occurring inside the engine cylinder are not fully understood. To fill this gap, this paper presents experimental methods for direct sampling and nanostructure analysis of in-flame soot particles in a working diesel engine. The soot particles are collected onto a lacey carbon-coated grid and then imaged in a high-resolution transmission electron microscope (HR-TEM). The HR-TEM images are post-processed using a Matlab-based code to obtain key nanostructure parameters such as carbon fringe length, fringe-to-fringe separation distance, and fringe tortuosity. Of particular interest is how jet-jet interactions impact the soot nanostructures because a wall-jet head merging with a neighbouring jet head is well known to cause high soot formation due to rich mixtures. The soot sampling was conducted for three different jet configurations including two single jets (Jet A and Jet B) and a double jet (Jet A&B). Results show that soot primary particles from all the jet configurations are comprised of two distinctively different structures of multiple amorphous cores and concentrically-oriented carbon-layer shells. From about 5000 carbon fringes processed for each jet configuration, the Jet A and Jet B samples show nearly identical mean values for the fringe length, separation distance, and fringe tortuosity of 0.955~0.962 nm, 0.398~0.399 nm, and 1.22, respectively. This suggests that jet-to-jet (or hole-to-hole) variations make a minimal impact on the soot nanostructures. The Jet A&B sample also displays similar tortuosity; however, 4% higher fringe separation distance (0.415 nm), which is well outside of the error range, and the increased proportion of highly reactive short carbon fringes suggest that soot particles formed in the jet-jet interaction region are more pre-mature and reactive than those formed in the single-jet head region.

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INTRODUCTION

The environmental and health-related issues of soot particles and ever more stringent emission regulations have long been a major challenge to diesel engine developers [1, 2]. High-pressure common-rail fuel injection systems have been a primary tool to address these issues, which achieve a significant reduction in soot yield [3] for the decreased total mass and particle size [4, 5, 6]. However, recent studies suggested that these small soot particles emitted from modern diesel engines could cause higher damage to human lung cells than large particles from black-smoking engines due to their defective internal structures and highly reactive surface patterns [7, 8]. Addressing this issue is calling for a thorough understanding of diesel soot nanostructures during key formation and oxidation processes inside the engine cylinder, which is the focus of this study.

Numerous studies have reported that the nanostructure of soot primary particles can reveal critical information of soot reactivity [9, 10, 11, 12, 13, 14], which refers to the structural characteristics of

carbon lamella (carbon layer plane segments) within a soot primary particle [15]. From high-resolution transmission electron microscope (HR-TEM) imaging of sampled soot particles, the internal carbon layer structures of soot primaries show a mix of disordered or oriented fringes [16, 17, 18]. Figure 1 shows an example HR-TEM image of a selected soot aggregate (top-left) and primary particle (top-right). Shown at the bottom are the physical properties dependent upon dimensions of carbon fringes within the soot primary particle. Two widely used nanostructure parameters are illustrated including fringe length (planar extent of carbon layers) and fringe separation (the spacing between two adjacent carbon layers). By taking a ratio of the measured fringe length to the distance between the fringe end-points, fringe tortuosity is also calculated. It is well known that shorter fringe length, larger fringe separation, and higher tortuosity represent higher reactivity of soot particles [10, 11, 14]. More reactive soot particles might be easily oxidised before exit through the exhaust; however, any remaining particles can be more harmful due to higher possibility of carrying toxic chemicals and causing biological interactions [7, 8, 12].



There are fine papers reporting the nanostructure of diesel soot particles sampled not only from the engine exhaust [14] but also from the cylinder of a diesel engine [19, 20]. From these studies, the nanostructure of soot particles in later combustion stages is found to show more graphitic structures than those in earlier combustion stages, with increased fringe length, decreased fringe tortuosity and narrower fringe separation. This suggests that the particles in the later combustion stages are more resistant to soot oxidation. Detailed studies conducted in a constant-volume combustion vessel simulating diesel-engine-like ambient and injection conditions [18, 21] also provided useful information about soot nanostructures. For example, the decreased fringe separation was reported for soot particles sampled further downstream of the reacting jet where high soot oxidation is expected.

Previous studies, however, do not fully address important questions on the soot nanostructure variations with developing diesel flames inside the engine cylinder. For instance, a diesel jet impinging on the bowl wall (jet-wall interaction) and then merging with a neighbouring jet head (jet-jet interaction) in realistic engine conditions are well known to produce a region of rich mixtures for high soot formation [3, 22, 23, 24]. In particular, jet-jet interactions can induce the re-entrainment of hot combustion products, which will lead to the decreased lift-off length and in turn increased soot formation [25, 26, 27, 28]. However, little is known about how the nanostructure of soot particles formed under the strong influence of jet-jet interactions is different to that in a single jet.

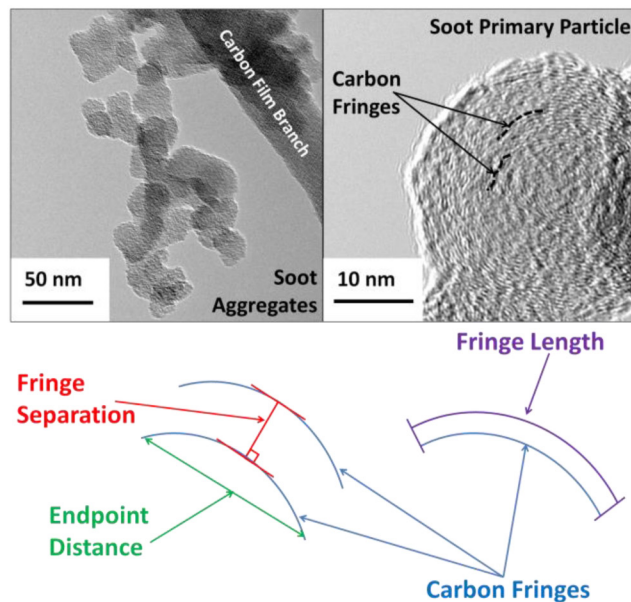


Figure 1. High-resolution TEM images of a diesel soot aggregate (top-left) and a primary particle comprised of many carbon fringes (top-right). Shown at the bottom are definitions of nano-scale carbon fringe parameters.

In this study, we perform the nanostructure analysis of soot primary particles sampled directly from a reacting diesel jet in a working engine, based on our previous work on the thermophoretic soot particles sampling for micro-scale analysis (i.e. soot aggregate size and primary particle size) [29, 30, 31]. The soot particles sampling conducted in a diesel engine provides more realistic conditions than the constant-volume combustion chamber [18, 21]. Also, this

in-flame soot sampling technique grants better targeting and sample preservation than the bulk gas sampling method [19, 20], which allows us to sample soot particles from specific locations of interest (e.g. jet-jet interaction region). We investigated three different jet configurations including two single jets (Jet A and Jet B) and a double jet (Jet A&B) to address issues associated with jet-to-jet (or hole-to-hole) variations as well as jet-jet interactions. The soot primary particles sampled onto a lacey carbon grid were imaged using a HR-TEM, which was post-processed to obtain aforementioned nanostructure parameters, i.e. the fringe length, fringe separation, and tortuosity.

EXPERIMENTS

Diesel Engine Specifications and Operating Conditions

Table 1. Engine specifications and operating conditions

Displacement volume [Ⓢ]	498 cc (single cylinder) [Ⓢ]
Bore [Ⓢ]	83 mm [Ⓢ]
Stroke [Ⓢ]	92 mm [Ⓢ]
Compression ratio [Ⓢ]	17.7 (original) 15.5 (modified) [Ⓢ]
Swirl ratio [Ⓢ]	1.4 [Ⓢ]
Coolant temperature [Ⓢ]	90°C [Ⓢ]
Intake air temperature [Ⓢ]	30°C [Ⓢ]
Engine speed [Ⓢ]	1200 rpm [Ⓢ]
Fuel [Ⓢ]	Ultra low sulphur diesel [Ⓢ]
Cetane number [Ⓢ]	51 [Ⓢ]
Common-rail [Ⓢ]	Second-generation Bosch [Ⓢ]
Nozzle [Ⓢ]	Minisac, hydro-grounded, K1.5/0.86 [Ⓢ]
Number of holes [Ⓢ]	7 holes (original) [Ⓢ]
	1 hole (Jet A) [Ⓢ]
	1 hole (Jet B) [Ⓢ]
	2 holes (Jet A&B) [Ⓢ]
Hole diameter [Ⓢ]	134 μm (nominal) [Ⓢ]
Included angle [Ⓢ]	150° [Ⓢ]
Rail pressure [Ⓢ]	70 MPa [Ⓢ]
Injection duration [Ⓢ]	2.34 ms [Ⓢ]
Injection timing [Ⓢ]	-6°C A TDC (Jet A) [Ⓢ]
	-7°C A TDC (Jet B) [Ⓢ]
	-7°C A TDC (Jet A&B) [Ⓢ]

The in-flame soot sampling was performed in an automotive-size, single-cylinder optical diesel engine. Detailed specifications and operating conditions of the engine are listed in Table 1. The engine has a displacement volume of 498 cm³ with 80-mm bore and 92-mm stroke. The original combustion chamber had a compression ratio of 17.7. As shown in Fig. 2, however, a portion of the piston bowl-rim was removed to insert a soot sampling probe into the diesel flame trajectory without the risk of collisions with moving piston and valves. The tribology design for lubricating piston movement against cylinder linear is to apply piston rings made of rubber-based material that are able to self-lubricate, which is avoid the oil film formation on optical windows and the possible contamination of a lacey TEM

grid. The geometric compression ratio was therefore decreased to 15.5, which is still within the range of diesel engines available in the market (e.g. Mazda Skyactiv-D). Details about the piston modification and its impact on sampled soot particles are found in our previous work [29]. The swirl ratio (mean flow speed over mean piston speed) was fixed at 1.4. Heated water at 90°C was flown through engine head and cylinder wall to simulate thermally stable and warmed-up engine operating conditions. The temperature of naturally aspirated intake air was measured using a thermocouple installed in the intake port, which indicated 30°C throughout the experiments. The engine speed was held constant at 1200 revolutions per minute (rpm) using an electric motor. A piezoelectric pressure transducer (Kistler 6056A) was used to record in-cylinder pressure traces and to calculate the apparent heat release rate (aHRR), using a simple energy balance assuming ideal gas and steady conditions.

The in-cylinder pressure and aHRR are useful to understand overall combustion conditions of the soot sampling experiments. Figure 2 also shows two high speed cameras (VisionResearch Phantom v7.3) used for simultaneous imaging of hot soot luminosity: one through the cylinder liner window (side-view image) and the other through the piston window and 45° reflex mirror placed beneath the piston (bottom-view image). Although this direct imaging of sooting diesel flames visualises only the strong radiation signals of soot particles at high temperature (i.e. hot soot luminosity), it is useful to understand the temporal and spatial development of diesel flames and the TEM grid position with respect to the developing diesel flame.

The fuel used in the present study was a conventional ultra-low sulphur diesel with a minimum cetane number of 51. A second generation Bosch common-rail system was used for fuel injection, which is controlled by a universal injection and timing controller (Zenobalti 5100). The original solenoid injectors used in the current study had 7 nozzle holes with a nominal diameter of 134 μm and an included angle of 150°. In this study, the sampling experiments were repeated for Jet A, Jet B and Jet A&B configurations as illustrated in Fig. 3.

Three different nozzles were manufactured by blocking some of the original seven holes using a laser-welding technique. For instance, the Jet A nozzle left only a hole directing 4 o'clock open while the other six holes were laser welded. Two more nozzles were manufactured using the same technique for Jet B and Jet A&B configurations, which enables the study of hole-to-hole variations and jet-jet interactions. For all the three jet configurations, the common-

rail pressure and fuel injection duration were fixed at 70 MPa and 2.34 ms, respectively, which corresponds to 9 mg per injection per hole [29]. The injection timing was set at -6 crank angle degrees after top dead centre ($^{\circ}\text{CA aTDC}$) for Jet A case and was slightly adjusted for the other two hole configurations to match the combustion phasing. This was to isolate the effect of ambient gas conditions at the start of fuel injection on soot development. However, it was noted that the fixed injection pressure and duration approach would lead to higher amount of fuel injection for Jet A&B than that for Jet A or Jet B experiments. How different injection mass affects the soot nanostructure analysis of the present study will be discussed later in the Results and Discussion section.

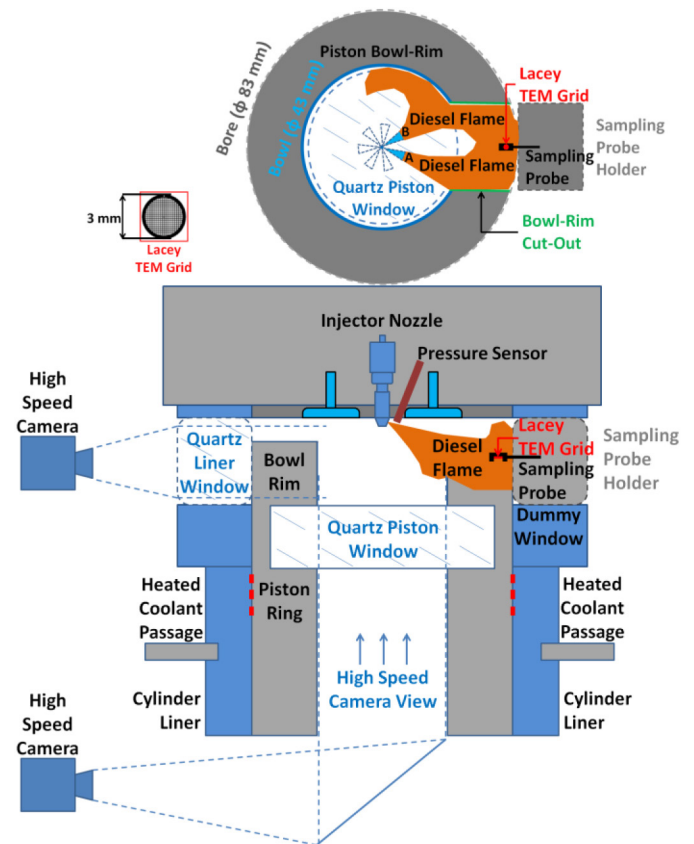


Figure 2. Illustration of the optical diesel engine and soot sampling system in the top-view (top) and side-view (bottom) orientations. Also shown are two high-speed cameras setup for side-view (through liner window) and bottom-view (through piston window) imaging.

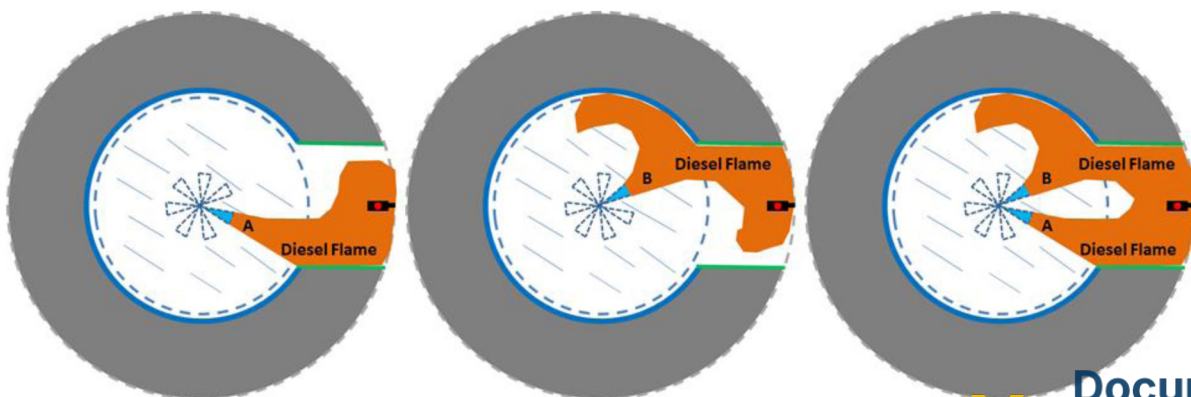


Figure 3. Illustration of diesel flame trajectories of Jet A (left), Jet B (middle) and Jet A&B (right) for soot sampling experiments.

Soot Particles Sampling

Figure 2 shows the schematics of the combustion chamber and soot sampling system used in the present study. One of four quartz liner windows of the optical diesel engine was replaced with a metal dummy window to hold the soot sampling probe. At the tip, a 3-mm diameter, 300-mesh, 150- μm -thick lacey carbon coated copper TEM grid (Emgrid LC300-Cu-150) was accommodated. The carbon film coated on the grid had breached holes to eliminate the background interferences in HR-TEM imaging. The sampling probe had a 1.5-mm hole providing a small passage for a direct exposure of the lacey TEM grid to the soot laden flame. The restricted passage protected the TEM grid from excessive heat and allowed reaction quenching for the deposition of soot particles on the lacey carbon film via thermophoresis, which is the result of high temperature gradient between hot soot particles and cold carbon film surface [32].

Prior to the start of diesel injection, the engine was motored for about 90 s until the in-cylinder gas reaches stable and repeatable thermal conditions. The fuel injection was executed for every 10th cycle (i.e. 10 skip-firing) to expel the residual gases from the previous firing cycle as well as to protect the quartz window from high thermal loading, which was repeated for 10 times per each sampling experiment (i.e. 10 injections onto the TEM grid). The 10-injection approach was used to ensure enough number of soot particles deposited on the lacey carbon film for statistically meaningful data analysis while avoiding soot overloading issues, which was discussed in detail in our previous work [29].

HR-TEM Imaging and Post-Processing

The sampled soot particles were imaged using a HR-TEM (Philips CM200) with a point resolution of 0.25 nm and an accelerating voltage of 200 kV. A CCD camera with pixel resolution of 2688 by 2672 was used to digitise the soot particle images (see Fig. 1 for example). Considering the in-homogenous nature of a diesel flame and soot particles within it, a total of 10 HR-TEM images were obtained from 10 different on-grid locations for each sample. The associated issue of image-to-image variations and its influence on the soot nanostructure analysis will be discussed later in the Results and Discussion section.

A Matlab-based image processing and analysis code was developed by the Aizawa group at Meiji University [18], based on the algorithm proposed by Yehliu et al. [16]. The image post-processing begins with the extraction of carbon fringe skeletons from HR-TEM images, as shown in Fig. 4. The code implements six steps including the manual selection of region of interest and the image inversion to highlight fringes, the contrast enhancement using histogram equalisation, Gaussian low-pass filtering to reduce noises, a top-hat transformation for correcting uneven illumination of image (i.e. flat correction), the image binarisation using the threshold-based Otsu method, and the morphological closing and opening process to reconnect the fringes broken during the filtering and binarisation steps. The used image processing parameter values for each step are summarised in Table 2. The final result of carbon fringe skeletons shown on the bottom-right corner of Fig. 4 is used to obtain nanostructure parameters.

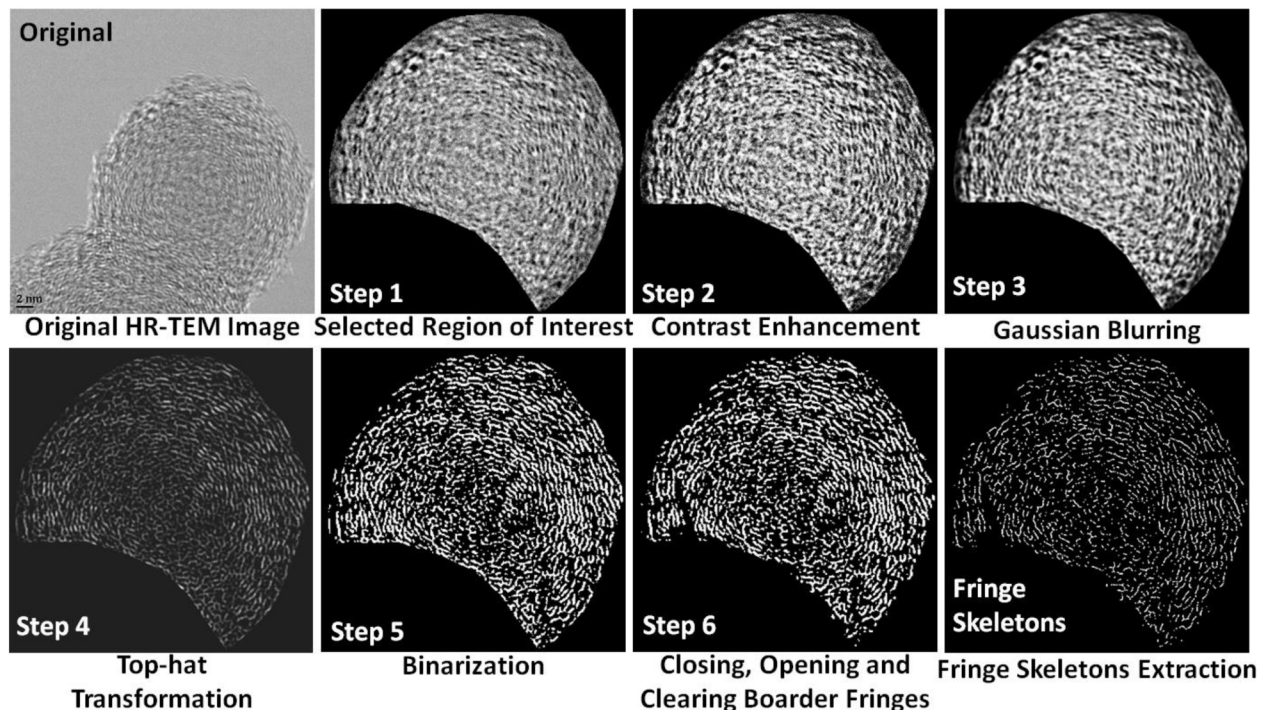


Figure 4. Post-processing procedures of HR-TEM images to obtain carbon fringe skeletons - adapted from Ref. [18]

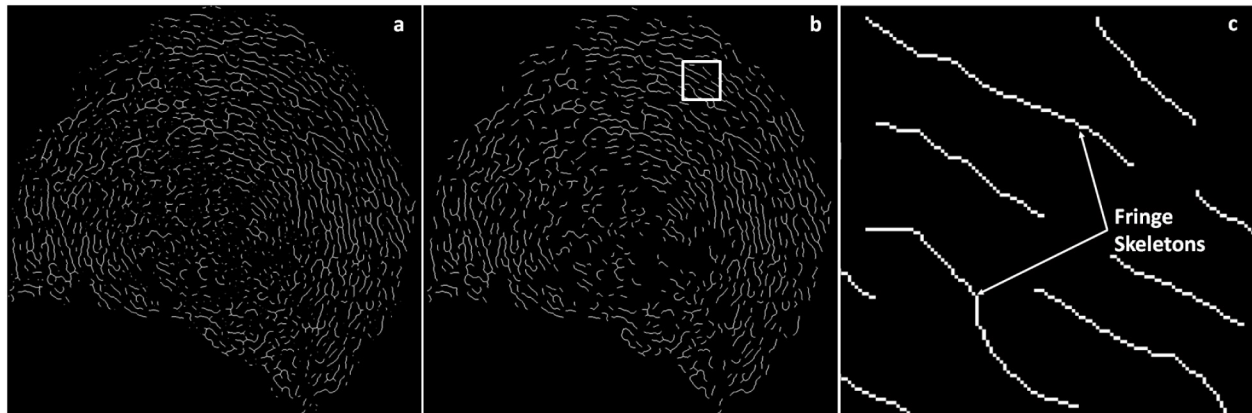


Figure 5. Raw carbon fringe skeleton images (a), Removal of short fringes (b), and a close-up view of fringe skeletons (c)

The nanostructure analysis code implements the elimination of artifacts. For instance, the fringes with less than the size of a single aromatic ring (e.g. 0.246 nm [33]) were ignored as shown in Fig. 5 (left and middle). In the rightmost image of Fig. 5, a close-up view of the selected region of the fringe skeletons is shown. From these fringe skeletons, the fringe length and tortuosity are automatically generated using a known calibration scale. However, the fringe separation distance, i.e. the mean distance between two adjacent fringe skeletons, requires a manual selection of fringe pairs to avoid faulty and multiple pairings of fringes [18]. In the present study, the total number of processed fringes and fringe pairs were 5368 and 1832 for Jet A, 5265 and 2868 for Jet B, and 4832 and 2218 for Jet A&B, respectively.

Table 2. Image processing parameters

Processing parameter	Value
Gaussian filter size	10 pixels
Gaussian filter deviation	3 1/pixel
Top-hat transformation structural element	Disk shape (radius of 9 pixels)
Binarization threshold	0.13
Morphological opening structural element	3 by 3 pixel square
Morphological closing structural element	4 by 4 pixel square
Spatial resolution	0.016 nm/pixel

RESULTS AND DISCUSSION

In-Cylinder Pressure and aHRR Traces

The ensemble-averaged traces of in-cylinder pressure and apparent heat release rate (aHRR) of all the jet configurations of the present study are plotted for various crank angle locations in Fig. 6. The “motored” in-cylinder pressure (black solid line) is also plotted. Regardless of the jet configuration, the aHRR traces show negative values right after the start of injections at $-6 \sim -7^\circ\text{CA aTDC}$, suggesting evaporative cooling of diesel fuel. This is also evident in the in-cylinder pressure traces showing lower “fired” pressure than the motored pressure. Pre-combustion mixing of fuel and air occurs

until the start of combustion at around 7°CA aTDC . Afterwards, the high-temperature reaction leads to a drastic increase in aHRR and causes the fired in-cylinder pressure to exceed the motored pressure.

A noticeable trend in Fig. 6 is two-fold higher peak in-cylinder pressure and aHRR of Jet A&B than that of Jet A or Jet B configurations. This difference is markedly higher than the difference between two single jets. The latter was related to the spatial evolution of flames such that the Jet A flame develops mostly within the bowl-rim cut-out region but for Jet B, half of the flame travels along the bowl wall and the other half penetrates into the cut-out region (see Fig. 3). Potentially, richer mixtures are formed for Jet A, which could lead to faster reaction and thus higher peak aHRR. For much higher peak in-cylinder pressure and aHRR of Jet A&B, it was noted that the injection duration was fixed for all the jet configurations and thus twice higher amounts of fuel (or more) was injected for the double jet. This means the bulk gas pressure and temperature was much higher for Jet A&B during the main heat release, which would impact soot formation processes significantly. Indeed, previous studies reported that higher pressure and temperature causes the increased soot formation [34] and more mature particle nanostructures (e.g. longer fringe length and lower fringe tortuosity) [35]. Nonetheless, the fixed injection duration was required to maintain the same jet dynamics of a single jet and to allow for long jet-jet interaction period, which are also important in soot processes. For example, if the injected mass were fixed, the injection duration would be reduced for Jet A&B configuration, meaning significantly increased pre-combustion mixing and very short time for jet-jet interactions. One idea to achieve comparable peak in-cylinder pressure and aHRR of the Jet A&B configuration would be to open another nozzle hole that will not interact directly with single Jet A or Jet B (e.g. diesel jet pointing to 9 o'clock). However, our previous work [36] showed that soot particles from a single jet opposite to the bowl-rim cut-out region can be sampled as the sooting jet travels along wall and eventually highly oxidised soot particles are exposed to the TEM grid. As the soot particle structures at this late combustion stage is largely different to those sampler much earlier, the nanostructure analysis would be negatively impacted and thus we proceeded with the original single-jet idea.