

In-Flame Soot Sampling and Particle Analysis in a Diesel Engine

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ABSTRACT

In-flame soot sampling based on the thermophoresis of particles and subsequent transmission electron microscope (TEM) imaging has been conducted in a diesel engine to study size, shape and structure of soot particles within the reacting diesel jet. A direct TEM sampling is pursued, as opposed to exhaust sampling, to gain fundamental insight about the structure of soot during key formation and oxidation stages. The size and shape of soot particles aggregate structure with stretched chains of spherical-like primary particles is currently an unknown for engine soot modelling approaches. However, the in-flame sampling of soot particles in the engine poses significant challenges in order to extract meaningful data. In this paper, the engine modification to address the challenges of high-pressure sealing and avoiding interference with moving valves and piston are discussed in detail. Of particular interest is the uncertainty caused by a selection of the on-grid locations for transmission electron microscope imaging and cycle-to-cycle fluctuations of the engine combustion. Marked variations are observed in the number and projected area of soot particles depending on these variations; however, their impacts on the size of aggregates and primary particles is found to be minor. Also, the morphology of soot particles appears not to be sensitive to the exposure duration of the grid to the sooting flame; however, the duration is ultimately limited by soot over-loading. Two different injection pressures are selected to test the usefulness of the in-flame soot sampling in a diesel engine and the results show that a decrease in the size of soot aggregates and primary particles with increasing injection pressure exceeds the uncertainty.

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INTRODUCTION

Modern diesel engines used in a car, SUV and light-duty truck have better fuel economy than petrol engines due to a higher compression ratio. However, soot particles from diesel engines have a negative impact on human health and the environment. For instance, small soot particles with a diameter of less than 2.5 μm can reach vital organs such as heart, liver, brain and lung cells and cause a number of cardiovascular and respiratory diseases [1]. Black carbon particles are also suspected to contribute to global warming with about 60% of the warming effect of carbon dioxide [2]. Increased pre-combustion mixing of fuel and air using small nozzle holes and high injection pressure might achieve non-sooting combustion [3]; however, with elevated flame

temperatures oxides of nitrogen (NO_x) form via the thermal (Zeldovich) mechanism, resulting in the well-known soot- NO_x trade-off. Simultaneous reduction of soot and NO_x emissions might be possible if advanced combustion regimes using a high rate of exhaust gas recirculation [4-5] or locally lean fuel-air mixture throughout the entire cylinder [6, 7, 8] are implemented to decrease the flame temperature below formation thresholds. Still, how to suppress unburnt hydrocarbon (uHC) and carbon monoxide (CO) emissions is in question [9-10]. Aftertreatment of soot particles using diesel particulate filters can be implemented, permitting some in-cylinder soot production, but this technology adds cost for production and maintenance and lowers fuel economy.

Despite its significance, little is understood about soot formation and oxidation process in diesel engine environments. Conventional methodologies such as exhaust opacity measurement, filtered smoke number (FSN) measurement, and scattering-based soot mass measurement [11] provide limited information about how soot formation and oxidation is varied in response to a change in engine operating conditions. As mentioned earlier, the size of soot particles is an important parameter and therefore many instruments are commercially available such as electrical low pressure impactor (ELPI) and scanning mobility particle sizer (SMPS). These instruments, however, measure the aerodynamic (or mobility) diameter assuming a spherical shape of soot particles. This approximation can be improved by conducting soot particles sampling and subsequent imaging using a transmission electron microscope (TEM) [12, 13, 14, 15, 16]. In this technique, a fast-moving actuator places a TEM grid within the high-sooting exhaust gases for a very short period of time. The high temperature gradient created between hot soot particles and the cool TEM grid induces soot deposition on the grid surface (*i.e.* thermophoresis) where further reaction of soot formation, growth or oxidation ceases.

From TEM images, soot particles appear as single primaries or aggregate structures consisting of many spherical (primary) particles. The aggregates appear to change in shape from bead to stretched chain as well as sizes ranging from several nanometres to hundreds of nanometres. Variations associated with sampling location and sampling time (grid exposure to the sooting gas) pose another challenge. Despite these issues, one would agree that the thermophoretic soot sampling is useful technique to get an understanding of soot shape, structure and morphological details, which cannot be obtained from other methods. It should be noted that, however, these morphological understandings are from the exhaust soot. Soot particles in the diesel flame would be better represented if the soot particles were sampled directly from the reacting diesel jet. Such in-flame soot sampling is widely used in an open flame burner [17] to provide essential information of soot precursor carbonisation [18, 19, 20], particle oxidation [21, 22] and extinction coefficient of soot particles [23].

Recently, we applied the direct soot sampling technique to a reacting diesel jet in a constant-volume combustion chamber simulating diesel-engine-like, high-pressure and high-temperature ambient conditions [24, 25, 26, 27, 28]. An example of TEM images from this in-flame soot sampling is shown in Fig. 1 [26]. The soot sampling has been successfully implemented for various conditions such as sampling locations relative to the reacting jet [24, 25, 26] and fuel types [26, 27]. At the same time, the TEM image post-processing technique has been advanced for boundary detection of soot aggregates, primary particle selection, fractal dimension calculation [27] and analysis of soot nanostructure [28].

In this study, we apply the thermophoretic in-flame soot sampling in a diesel engine. Ambient and injection conditions used in the constant-volume chamber experiments were typical of diesel engines. However, the conditions were fixed during the combustion event and isolated from complex flow and jet-wall interaction, which significantly impact soot formation [29, 30, 31]. The soot sampling in more practical and realistic engine environments was conducted by Li *et al.* [32]. They used a fast sampling valve that extracts soot-laden gases from the combustion chamber of a diesel engine, and then these gases are exposed to the TEM grid. This method provides morphological information of in-cylinder soot aggregates in a diesel engine, which cannot be obtained from a point measurement of laser-induced incandescence for particle sizing [33, 34, 35]. However, this “in-cylinder sampling” approach raises a critical question whether or not the extracted soot particles represent the soot particles within the diesel flame, since the residence time, temperature, pressure and fuel-ambient mixture all change during the sampling process. If a TEM grid can be placed inside the combustion chamber of a diesel engine so that it is directly exposed to the diesel flame, the same level of soot details can be obtained as in the constant-volume combustion experiments without worrying about potential issues associated with the in-cylinder sampling. To our best knowledge, a direct TEM sampling of soot particles in a diesel engine has never been attempted.

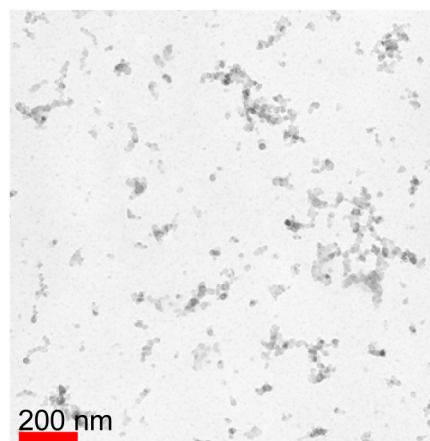


Figure 1. Transmission electron microscope image of soot particles sampled directly from the reacting diesel jet in a constant-volume combustion chamber [26]

This paper presents the design and setup of in-flame soot sampling probe and its application in an automotive-size optical diesel engine. Required engine modification and its impact on diesel flame development are introduced before TEM images of soot particles are discussed. Of particular interest is the uncertainty associated with TEM image-to-image and engine cycle-to-cycle variations. Also, soot loading is discussed in great details to find how exposure duration impacts the number and size of soot particles. The findings from this study will serve as a basis to conduct

further investigation of soot particle shape and structure for various engine operating conditions.

EXPERIMENTS

Diesel Engine and Operating Conditions

A single-cylinder optical diesel engine was used to perform thermophoretic soot sampling directly from the diesel flame. Specification of the engine and its operating conditions are listed in [Table 1](#). The engine has a displacement volume of 498 cc with a small 83-mm bore and 92-mm stroke, typical of an automotive-size diesel. The original combustion chamber had a high compression ratio of 17.7 (geometric). When it was modified for the soot sampling experiment, the compression ratio was reduced to 15.2. The engine has dual intake ports with a throttle control to vary a swirl ratio depending on the engine load conditions. The throttle was left fully open, which set a swirl ratio of 1.4. The wall temperature was kept at a constant 90°C by delivering heated water to the engine head, cylinder liner and engine block. The temperature of naturally-aspirated intake air during the engine run was measured at 30°C. All experiments were conducted at a fixed engine speed of 1200 revolutions per minute (rpm) controlled by an electric motor.

The used fuel was a conventional ultra-low sulfur diesel with cetane number of 46. The fuel injection system was composed of a Bosch second-generation common-rail and 7-hole solenoid injector connected to an injection driver (Zenobalti 5100). For the soot sampling experiments, however, the nozzle was modified so that only one hole was opened. At a fixed rail pressure of 70 MPa, the injection duration for this single-hole injector was 2.34 ms (actual), with which 9 mg per injection was measured. The injection timing (electronic) was fixed at -10 crank angle degrees after top dead centre (°CA aTDC) for all experiments. Due to the injection delay, the actual injection began at -7°CA aTDC.

The used engine is optically accessible for various optical/laser-based imaging diagnostics. In this study, a simple high-speed imaging of soot luminosity (*i.e.* hot soot radiation) was performed to understand the temporal development of sooting diesel flame. [Figure 2](#) (top-left) shows the field of view (ϕ 40 mm) from below, illustrated by a dashed line, and bowl (ϕ 43 mm) and bore (ϕ 83 mm) outlined by a blue solid line and grey dashed line, respectively. The fuel injector was positioned at the centre of the bore. Side-view imaging via a quartz liner window was also performed to visualise the squish area that was not shown in the bottom-view images.

A high-speed camera (VisionResearch Phantom v7.3) was used to capture natural luminosity of combustion. The frame rate was set at 36,000 frames/s with an exposure time of 10 μ s. The image resolution was 256 by 256 pixels and the maximum image count was 16,384 (from 0 to 16,383 for a 14-bit image). The lens attached to the camera was a Nikon Prime Micro 105-mm lens with f/11 that allowed a sufficient depth-of-field and image sharpness.

The optical engine was run by an electric motor at a fixed speed of 1200 rpm and “skip-fired” at every 10th cycle. This skip-firing minimised the cyclic thermal stresses in quartz windows and reduced residual gases from previous firing cycle, resulting in decreased cycle-to-cycle variations. The total number of skip-fired cycles was adjusted to control soot loading on the TEM grid. During the motored and fired cycles, in-cylinder pressure data were acquired using a piezoelectric transducer (Kistler 6056A with a glow plug adapter) and was ensemble averaged, which was used to calculate apparent heat release rate (aHRR).

Table 1. Engine specification and operating conditions

Displaced volume	498 cc (single cylinder)
Bore	83 mm
Stroke	92 mm
Compression ratio	17.7 (original) 15.2 (modified)
Swirl ratio	1.4
Coolant temperature	90°C
Intake air temperature	30°C
Engine speed	1200 rpm
Fuel	Conventional diesel
Cetane number	46
Common-rail	Second-generation Bosch
Nozzle	Minisac, hydro-grounded, K1.5/0.86
Number of holes	7 hole (original) 1 hole (modified)
Hydraulic fuelling rate for 7-hole nozzle	400 cc/30s at 10 MPa
Hole diameter	134 μ m (nominal)
Included angle	150°
Rail pressure	70 MPa
Injection duration	1.2 ms (electronic) 2.34 ms (actual)
Injected mass per injection	9 mg
Injection timing	-10°CA aTDC (electronic) -7°CA aTDC (actual)

Engine Modification for Soot Sampling

The engine combustion chamber geometry and soot sampling system is illustrated in [Fig. 2](#) (top and middle). Shown at the bottom are photos of the piston and installed soot sampling probe (right below a valve at the left). To enable in-flame soot sampling, the original combustion chamber required several modifications. Firstly, a single-hole injector was used instead of a conventional multi-hole injector. The original injector had seven holes, each with 134 μ m in diameter (see [Table 1](#) for details). In the present study, six of them were laser-welded so that only one hole (jet 7 in [Fig. 2](#)) was left open. This single-hole injector was purposely

used to avoid deposition of soot particles from other reacting jets. Also, the single-hole allows long injection duration of 2.34 ms (or injected mass of 9 mg) within the safety limit of optical windows. This long injection is equivalent to over 60 mg per injection for the original seven-hole injector and therefore covers upper mid to high load conditions where soot emission is the most problematic.

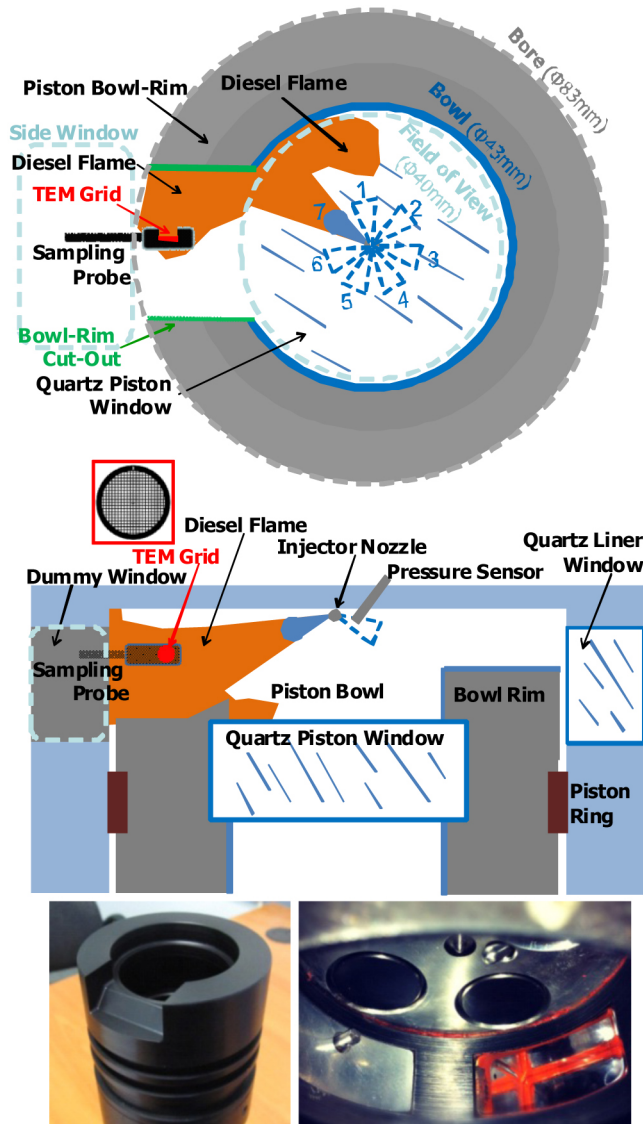


Figure 2. Schematics of the combustion chamber of the used optical diesel engine. Engine modifications for soot sampling and sooting diesel flame development relative to the sampling probe are illustrated for the bottom-view and side-view orientations. Shown below the schematics are photos of the cut-out piston and the sampling probe inside the combustion chamber.

In Fig. 2, it is depicted that the single reacting jet is not symmetric in the bottom-view image. This is because half of the jet penetrates into the bowl-rim cut-out region while the other half impinges on the piston bowl-wall. To place the

sampling probe holding a TEM grid within the diesel flame even when the piston was near top dead centre, a 30 mm wide section of the piston-bowl rim was removed. As a consequence, a half of the diesel jet penetrates farther without impinging on the piston bowl-wall while the other half impinges on the bowl wall and then penetrates in a radial direction along the curvature. As the injection continues, a jet-wall vortex is formed and the jet head moves back towards the nozzle [30]. Another vortex would be created within the bowl-rim cut-out region after the other half of the jet impinges on the cylinder liner wall. Therefore the TEM grid is supposedly exposed to the head region of this liner-wall impinging jet during the main combustion event. Other sooting jet regions such as impingement point and jet centre are also of interest. For example, the highest soot concentration is measured at the jet impingement point due to the limited air entrainment [29, 30, 31]. On the other hand, if the grid is placed at the jet centre and closer to the nozzle, soot particles at early formation stage might be captured. These are in our future plan together with other engine operating condition variations; however, in the present study we fix the diesel jet and sampling position as illustrated in Fig. 2.

How the in-cylinder pressure and development of sooting flame are affected by the aforementioned engine modifications is demonstrated in Fig. 3. Shown at the top-left are ensemble-averaged in-cylinder pressure traces of 25 motored cycles as well as 25 skip-fired cycles for three different combinations of multi-hole/single-hole nozzle and original/cut-out piston. The first notable trend from the plot is that the in-cylinder pressure is much higher for the 7-hole injector than the 1-hole injector. This was due to a higher fuelling rate for the 7-hole injector with a fixed injected mass of 9 mg. As shown in the fuel injection rate (top-right plot) measured by a Bosch-type injection rate meter [36], the injection duration of the 1-hole injector is nearly 3-fold longer than the 7-hole injector. The higher rate of fuel delivery would increase the rate of heat release, which is reflected in the higher in-cylinder pressure. By contrast, a low, extended rate of heat release is expected for the 1-hole injector with a low and long injection rate. Another notable trend is that the motored pressure is lower for the cut-out piston resulting in lower fired in-cylinder pressure. For instance, the TDC pressure is 3.2 MPa and the peak fired pressure is 3.6 MPa, both about 1 MPa lower than those of the original piston. Also, the phasing is retarded such that the initial rise of fired in-cylinder pressure occurs about 7°CA later.

The circles on the traces of in-cylinder pressure and fuel injection rate illustrate imaging timings of the soot luminosity images shown at the bottom of Fig. 3. The leftmost image for the 7-hole injector shows seven distinct soot luminosity pockets at about 20 mm from the nozzle near the bowl wall. Also shown are single-hole flames for the original piston and cut-out piston. It appears that the soot luminosity is symmetric with respect to the jet axis for the original piston

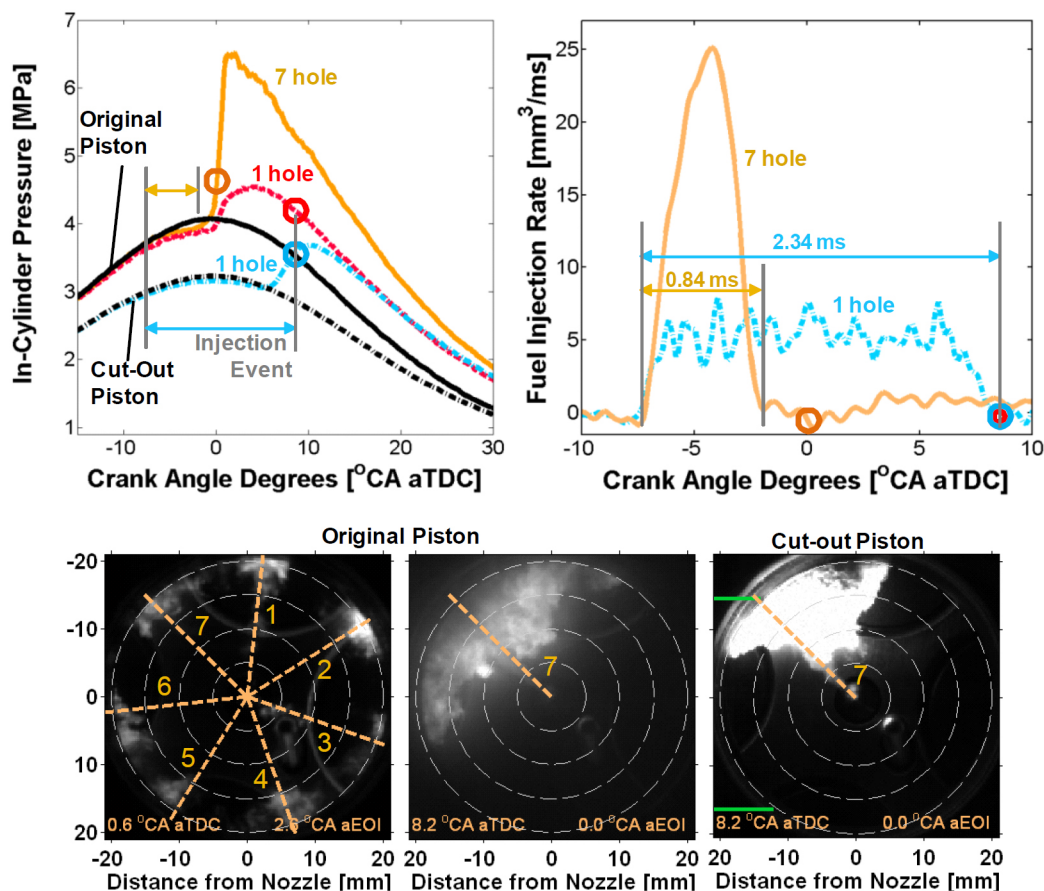


Figure 3. Traces of in-cylinder pressure and fuel injection rate for the 7-hole and 1-hole injectors. Soot luminosity images are shown at the bottom to help understand differences in sooting flame development for 7-hole/original piston, 1-hole/original piston and 1-hole/cut-out piston from left to right. Jet centre lines are annotated by orange dashed lines. Shown at the image bottom-left and bottom-right are imaging timing in °CA aTDC and °CA aEOI, respectively. Horizontal green lines are drawn for the rightmost image to show the bowl-rim cut-out.

case. In other words, the left and right halves of the wall-impinging jet nearly mirror each other. By contrast, a piston with the bowl-rim cut-out annotated by green horizontal lines allows the left half of the jet to penetrate farther without impinging on the bowl wall. This causes an asymmetric jet. The impacts of the bowl-rim cut-out and single-hole arrangement on diesel combustion are significant and should be considered to interpret the results in the following sections.

Soot Sampler Design

In experiments conducted using a constant-volume combustion chamber [24, 26], the soot sampler was stationary and exposed to reacting diesel jet for a long period of time (8 ms at 150 MPa) throughout the varying combustion stages. Quick insertion-type soot sampling, as in open flame burner tests, was very challenging due to sealing around the fast-moving sampling probe. The same issue applies to the soot sampling system in a diesel engine with more harsh ambient conditions due to jet-wall interaction, in-cylinder flow and a

risk of clashing with moving engine components such as valves and a piston.

The difficulties and constraints associated with the modification on the engine and the functional requirements of the soot sampling system are demonstrated in Fig. 4. The figure shows isometric and cross-sectional view of three-dimensional CAD drawings. The used TEM grid is a carbon-coated copper layers with 400 meshes and has a circular shape with 3-mm diameter and thickness less than 0.1 mm. The grid needs to be intruded into the piston area so that it can be placed within the sooting diesel flame. To avoid interference with the moving piston, a portion of the piston bowl-rim was removed as explained previously. One of the optical liner windows was replaced with a dummy metal window to hold the sampling probe inside the combustion chamber as shown at the left of Fig. 4. A copper washer and back screw was used for sealing. The back screw also was used to fix the position of the soot sampling probe.

Previous researchers used a thin plate-type grid holder to minimise flow disturbance, which was found to vibrate during the sampling experiments [37]. Therefore, a

cylindrical sampling probe was used in this study as shown in Fig. 4 (top-right). The sampling probe has a shallow circular pocket where the grid is placed and a cover plate to fix the grid position. A setscrew was used to fasten the plate cover onto the sampling probe body. The opening for soot collection is machined on the probe body, which provides a restrictive passage to retain a cold grid surface for effective quenching of any further reactions.

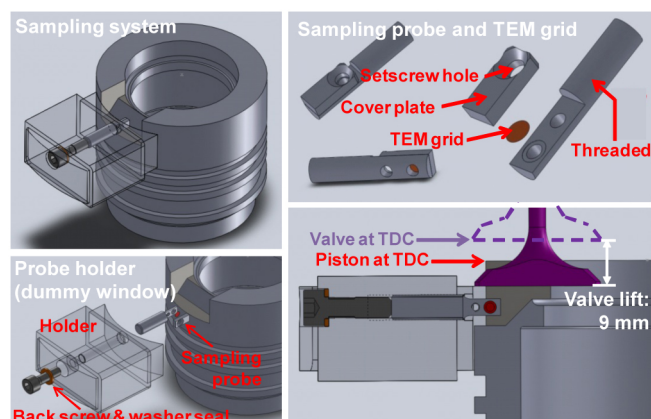


Figure 4. Isometric and cross-sectional view of three-dimensional drawings of the cut-out piston, sampling probe, and probe holder. The valve position relative to the sampling probe is also shown.

It was noted that the position of the sampling probe was constrained by the fast-moving engine parts including a valve above and piston below with very small clearance of about 2 ~ 3 mm and therefore was fixed throughout the experiments. The TEM grid was located about 5 mm from the liner wall and 37.8 mm from the nozzle. Since the sampling probe was stationary, the grid was exposed to the fresh air during the motoring cycles, sooting diesel flame during the firing cycles and then ambient gas with residuals during another motoring cycles before the grid was removed and stored for TEM imaging. This sampling approach might raise questions about potential contamination of the grid from combustion products or engine oil other than soot particles. To resolve this concern, experiments were conducted at no firing condition, which showed no objects in the TEM images. In our previous experiment conducted in a preburn-type constant-volume combustion chamber, the grid was placed outside of the sooting regions of the jet, which also showed no soot particle deposition [26].

Transmission Electron Microscope Imaging and Post-Processing

A transmission electron microscope (JEOL 1400) with a point resolution of 0.38 nm and an accelerating voltage of 120 kV was used to analyse sampled soot particles. Figure 5 shows a procedure of TEM imaging and image post-processing. The TEM grid exposed directly to sooting diesel flame is placed inside the vacuum chamber where the

electrons form a magnified image on a fluorescent screen. A CCD camera with a resolution of 11 mega pixels was used to digitise soot particle images. The TEM magnification was set at 50,000 $\times$ to obtain a good balance between the number of particles per image and image quality for post-processing. An example image is shown at the top-right of Fig. 5 with a scale bar depicted at the bottom-left corner of the image. Similar to refs. [24, 25, 26, 27, 28], soot particles seen in the TEM images are aggregates of many primary particles. Some of the particles appear to be single primaries while larger aggregates with various shapes and structures are also observed.

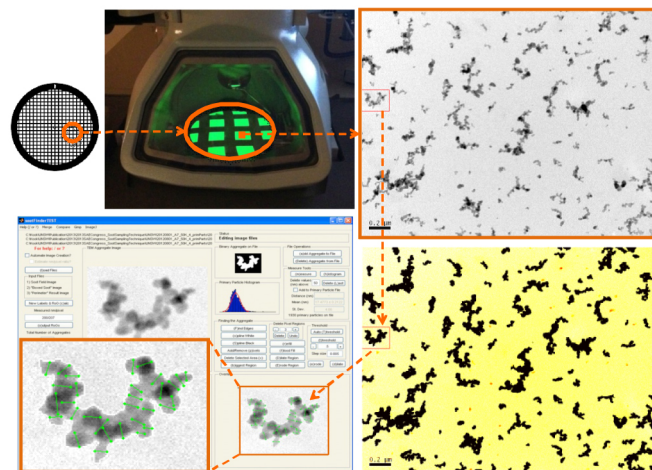


Figure 5. Transmission electron microscope (TEM) imaging and image post-processing.

Digitised TEM images were then analysed using an in-house developed Matlab code [26]. Firstly, boundary detection was performed to obtain binary images, which were used to label each soot aggregates as shown at the bottom-right of Fig. 5. The projected area and radius of gyration for individual aggregates were also calculated using a Matlab code as shown at the bottom-left of Fig. 5. In the example image, more than 100 soot aggregates were detected from the automated boundary-detection process. For primary particles, the current image processing technology cannot differentiate overlapped primary particles within the aggregates of complex structures. Therefore, a manual selection of primary particles was performed for each aggregate. An example for a selected aggregate shows that many spherical primary particles stack up together to form a long stretched aggregate. The green lines overlaid on the aggregate image connect one end to the other of primary particles and illustrate how we selected them. An uncertainty of this manual selection process is discussed in great details in a concurrently submitted paper [38]. The total number of these primaries exceeds 1900 for the selected TEM image.