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Morphology and nano-structure of soot in diesel spray and in engine exhaust

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HIGHLIGHTS

- Soot was sampled in two locations in diesel spray flame and also at engine exhaust.
- Morphological and nano-structural analysis was done by TEM image processing.
- Soot particles become larger and more fractal along the spray axis.
- Engine-out particulates become smaller and less fractal compared to spray soot.
- Engine-out particulates become nano-structurally more ordered compared to spray soot.

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ABSTRACT

This work investigates how morphology and nano-structure of the soot particles produced in a diesel spray flame evolve due to the rise in the temperature/pressure caused by the piston motion. The soot particles were sampled from the exhaust line of a diesel engine and were compared to the soot particles directly sampled from the spray flame of the same injector, in a constant volume chamber. Analysis of the high resolution transmission electron microscope (HRTEM) images acquired from the soot samples at two axial locations in the spray flame shows that the soot aggregates grow in size, become longer, more fractal and attain lower surface to volume ratio along the spray axis. However, the soot particles produced in the spray flame become smaller, shorter, less fractal, more compact and attain higher surface to volume ratio when emitted from the engine. This is discussed to be due to the increase in the oxidation rate by temperature rise caused by piston motion, and the consequent oxidation induced fragmentation of the soot aggregates. The effect of oxidation is also evident on the size of the soot primary particles, as the engine-out soot is found to have smaller primary particles compared to the soot initially produced in the spray flame. Nano-structural analysis shows that the crystallite size in the engine-out soot is increased compared to the soot originally produced in the spray flame. This suggests that among two possible effects of temperature on diesel soot crystallite size, being crystallite size increase due to enhanced graphitization from one hand, and crystallite size reduction due to oxidation enhancement from the other hand, the former effect is dominant and the soot particles become structurally more ordered under the effect of temperature rise caused by piston motion.

1. Introduction

In the diesel engine, the fuel is sprayed into the high-pressure high-temperature environment inside the cylinder. In the rich parts of the non-premixed flame polycyclic aromatic hydrocarbons (PAHs) are formed as a result of fuel pyrolysis in high ambient temperature. Such compounds form small sheets of graphene which then stack on each other and form soot nuclei. These precursors of soot, grow in size by merging with each other due to collision or by surface growths, and form the soot primary particles. Finally the primary particles aggregate and form chain-like agglomerates that are emitted to the environment [1]. Such soot particles, also known as particulate matters (PM), have adverse effects on human health and environment and their emission is strictly regulated.

Nano-structural characteristics of soot primary particles, such as the size of crystallite size, tortuosity of graphene layers and their inter-layer distance have recently gained the attention of researchers due to their correlation with oxidative reactivity and for the insights they can provide for fundamental understanding of the

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soot formation process [2–7]. The size and morphology of PM is also of importance as the size is a major regulatory parameter and together with particle morphology affects the way the PM interacts with human body and impacts the environment [8–11].

Due to the practical significance of soot particles emitted from diesel engines, in recent years many researchers have tried to establish the correlation between the engine operating conditions and the physiochemical characteristics of soot particles [12–24]. However, contradictions are frequent in the reported results. For example, Lapuerta et al. [14] reported a decrease in the size of the primary particles by engine speed while it was not confirmed by Zhu et al. [13]. Yehliu et al. [15] reported a decrease in crystallite size due to the injection timing retardation while Xu et al. [17] reported the opposite. Al-Qurashi et al. [16] reported a reduction in crystallite size due to exhaust gas recirculation (EGR) while Li et al. [25] noticed an increase. The soot reactivity was found to be reduced by EGR in a previous study by the author [26] in contrary to the report of Al-Qurashi et al. [16] on the same issue.

The contradictive findings can be largely due to the fact that in diesel engines (unlike laboratory flames) the combustion parameters such as temperature, pressure, residence time and oxygen availability that directly influence the soot formation cannot be independently controlled. For example, advancing the combustion phasing changes the residence time but is also likely to increase the bulk in-cylinder temperature. Another category of interconnections that further complicates the analysis of the soot formation process in diesel engines is the effect of the in-cylinder global condition on the spray configuration and the consequent effects of the spray configuration on the soot particle formation. For example, variation in injection timing beside changing the residence time and the global in-cylinder temperature, changes the spray lift-off length due to the different pressure and temperature at the time of injection.

Therefore, in order to fully understand the effect of engine operating parameters on soot characteristics, the effect of spray configuration on soot particles should be understood. For this reason, direct sampling of the soot particles from the diesel spray flames inside constant volume chambers (CVCs) have been attempted by some researchers [27–30]. However, all of such studies reported in literature have been performed with single hole injectors, and no direct comparison is made with the soot emitted from the engine at the corresponding conditions. If the results of the spray sampling are to be used for analyzing the characteristics of the diesel engine soot, first a clear understanding of the differences and similarities between the soot sampled from the spray flame and the soot released from the engine should be established.

It is of interest to know to what extend the size, morphology and nano-structure of the soot particles emitted from a diesel engine are product of the spray combustion, and to what extend are affected by the global in-cylinder conditions, namely the pressure and temperature rise due to the piston motion.

The necessity of such investigation has been also noticed by other researchers. Aizawa et al. [28] recently reported the effect of fuel chemical composition on the soot particles characteristics, through direct sampling from the spray flame inside a CVC. Contrary to the existing literature on the engine-out soot, they found that the effect of fuel composition on morphology and nanostructure is insignificant, questioning the relevance between the soot directly sampled from the spray and the actual engine-out soot. They suggested that the engine effect (rise of temperature and pressure due to piston motion) may be a reason for the discrepancy observed. However their data was not directly comparable to the engine-out soot data in literature because of different operating conditions.

Due to the above mentioned motivations, the current study was planned to take the soot sample from the exhaust line of a diesel engine, and compare it to the soot particles directly sampled from the spray flame of the same injector, injecting in an ambient condition identical to the engine in-cylinder condition, but inside a CVC with no temperature and pressure rise due to the piston motion. This comparison sheds light on the extend of the engine effect (temperature and pressure rise by piston motion) on soot characteristics after formation inside the spray.

The analysis is based on the image processing of high resolution transmission electron microscopy (HRTEM) images, which has been proved to be a credible research tool in the past few years [5,28,31–35]. Beside nano-structural analysis, morphological evolution of the particles inside the spray and in the engine exhaust are studied through defining and quantifying various morphological parameters which are of importance with regard to regulations, health impact or oxidative reactivity of the soot particles.

2. Experiments

2.1. Experimental apparatus

2.1.1. Engine

A single-cylinder heavy-duty naturally aspirated diesel engine was used for the engine experiment. The engine configuration including cylinder head, valve-train, injector and piston are based on the commercial multi-cylinder Doosan DL06 engine. The in-cylinder pressure was measured by a piezoelectric pressure transducer (Kistler; Type-6056A) and the intake air pressure was measured using a piezo-resistive pressure transducer (Kistler; Type-4045A5). The fuel (commercial diesel) was pressurized by a high pressure fuel pump driven by an electro-motor. The pressure inside the common rail was regulated by a pressure controller (Zenobalti Co.; ZB-1200) while fuel injection was controlled by an engine controller (Zenobalti Co.; ZB-9013). The engine and the injector specifications are listed in Table 1.

As for the operating point, the engine speed of 1200 rpm and the injection quantity of 25 mg was chosen as a typical representative operating point for a heavy duty diesel engine. The injection timing was swept in order to find the maximum indicated mean effective pressure (IMEP). Detailed operating conditions and the combustion data are shown in Table 2 and Fig. 1 respectively.

An in-house designed soot sampler was used for soot sampling. The sampler is composed of an inner rod on which TEM grid is mounted, inside an outer tube with an opening at one side. The inner rod can rotate by means of a fast servomotor (Hitec “Ultra Speed” HS-7940TH) and switch the sampler status between close and open positions as shown in Fig. 2. The servo-motor is controlled by a programmable throttle controller (Zenobalti Co.; ZB-2200) which can open and close the sampler for a certain number of combustion cycles. The sampler was installed on the exhaust line in a way that the lower part was extruded into the exhaust flow. Once the TEM grid is exposed to the hot exhaust flow, the soot particles stick to the grid due to the thermophoretic effect. The number of cycles should be decided in a way that enough number of soot particles are deposited on the grid but at the same time over-accumulation of particles should be avoided, so that a

Table 1
The specifications of the engine and the injector.

Engine specifications		Injector specifications	
Number of Cylinders	1	Injector type	Solenoid-actuated
Displaced volume (cc)	981	Tip configuration	SAC
Stroke (mm)	125	Number of orifice	8
Bore (mm)	100	Spray cone angle (deg)	146
Compression ratio	17.4	Orifice diameter (mm)	0.146
Number of Valves	4	HFR(cc/30 s)@100 bar	460