Fuel 161 (2015) 312–322

Contents lists available at ScienceDirect

Fuel

journal homepage: www.elsevier.com/locate/fuel

Immaturity of soot particles in exhaust gas for low temperature diesel combustion in a direct injection compression ignition engine

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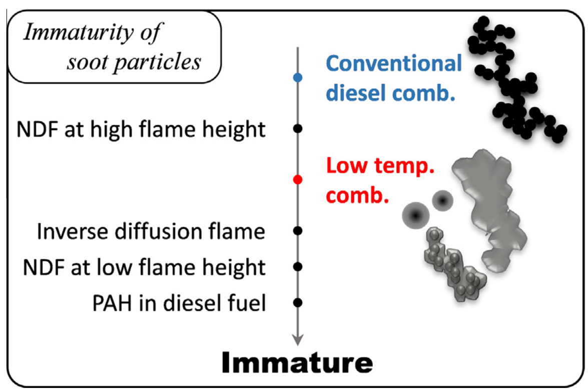
HIGHLIGHTS

- Soot particles for low temperature combustion are oxidized at lower temperature.
- A larger amount of organic fractions was found in particulates of low temperature.
- Soot particles from low temperature diesel combustion are immature.
- Low temperature particulates exhibits clumps of translucent amorphous materials.

GRAPHICAL ABSTRACT



ARTICLE INFO

Article history:
Received 31 May 2015
Received in revised form 5 August 2015
Accepted 27 August 2015
Available online 8 September 2015

Keywords:
Soot
Particulate matter (PM)
Thermogravimetric analysis (TGA)
Low temperature diesel combustion (LTC)
Exhaust gas recirculation (EGR)

ABSTRACT

The particulate matter sampled in exhaust gas from low temperature diesel combustion (LTC) with an exhaust gas recirculation rate of 60% in a direct injection compression ignition engine was comprehensively assessed and compared to conventional diesel combustion using thermogravimetric analysis and elemental analysis. With LTC, relatively significant mass reduction occurred due to the oxidation and desorption of hydrocarbons. 95% of mass was reduced at around 520 °C in LTC, which is lower than for conventional combustion (595 °C). The weight fractions of carbon and hydrogen species found by the elemental analysis after thermogravimetry with inert gas up to 400 °C revealed that the change of weight fractions into higher carbon and lower hydrogen contents were larger for soot particles from LTC. The atomic ratios of carbon to hydrogen (C/H) as a measure of soot maturity were 6.74 and 3.84 for conventional combustion and LTC, respectively. The C/H ratio for LTC is close to that of the precursor particles (1.80) from a normal diffusion flame at low height or young soot particles (2.2) from an inverse diffusion flame, which demonstrates the immaturity of the soot particles from LTC, while the range of the C/H ratio in the conventional combustion was suggestive of highly matured soot particles. Transmission electron microscopy revealed the primary particle and agglomerates from LTC appeared small without distinguishable boundaries. The LTC mode also exhibited clumps of amorphous materials and translucent precursor particles, which were attributed to intermediate substances from the low combustion temperature and abundant hydrocarbon species.

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http://dx.doi.org/10.1016/j.fuel.2015.08.068
0016-2361/

1. Introduction

Low temperature diesel combustion (LTC), as investigated in this study, consists of the partially premixed combustion fueled with commercial diesel in a compression ignition engine using early injection timing and an exhaust gas recirculation [1,2]. Exhaust gas recirculation (EGR) is a promising technology to achieve LTC without significant modification since the recirculated exhaust gas, including carbon dioxide (CO_2) and water vapor (H_2O), replaces a portion of the fresh air in the intake, which reduces the amount of oxygen and its concentration. Along with the early injection timing, this reduced oxygen concentration prolongs the ignition delay, which gives the mixture of fuel and oxidizer sufficient time to form a partially premixed charge in the cylinder. After ignition, the low oxygen concentration and high heat capacity substances such as CO_2 and H_2O can support a relatively inactive combustion with gradual heat release [3]. LTC is characterized by a two-staged heat release rate, low-temperature reaction (cool flame), and high-temperature reaction (hot flame), relative to the conventional diesel combustion of four combustion phases [4–6].

With the increasing stringency of emission regulations, the most beneficial aspect of LTC is the significant reduction of nitrogen oxides (NOx) and particulate matter (PM) [2]. The low combustion temperature can allow the formation rate of NOx to be low. Likewise, the inception of soot particles is not readily permitted because the overall combustion temperature can be lower than the inception temperature, although the temperature in particular areas may be high enough to form incipient soot particles. On the other hand, the oxidation processes of hydrocarbon (HC) and carbon monoxide (CO) are deteriorated by a low combustion temperature, leading to high emissions of these gases in the LTC mode. These unoxidized substances can be related to the formation process of PM particularly.

Many researchers have researched PM in relation to engine operating conditions such as idle, low, and high load. The Argonne group has vigorously studied the structural characteristics of particulate matter such as the diameter of the primary particle, the radius of gyration, and fractal dimensions by means of transmission electron microscopy (TEM) in single-cylinder heavy duty engines [7,8] and four-cylinder light duty engines [9–11]. Typical particulate matter images showed chain-like agglomerates with a highly crystalline structure at high load. At light load, however, their images gave the appearance of a nebulous and amorphous structure, which is regarded as the first observation of precursor particles [12]. The particulate matter was suspected to contain a significant amount of soluble organic fractions, or other liquid phase chemicals [8]. Soot particles using TEM under various engine operations were investigated [13,14]. It was found that the dependence of the categorization on the engine load was not evident [15]. The size of the primary particle was measured using microscopy, and the size of agglomerate evaluated by radius of gyration based on the microscopy was compared to the mobility measurement [16–20]. Soot structures have been studied with respect to the oxidation process, usually occurring in the after-treatment system, and they reported the internal burning mode of soot at an EGR rate of 20%, which is a far faster oxidation mode than the external shrinking mode [21,22].

Particulate matter under LTC with a large amount of EGR has been investigated comprehensively, and it was found that the size distribution of particles in LTC mode was shifted to a smaller range based on the results from a scanning mobility particle sizer [23]. To explain this shift, three possible agglomerate models were suggested: clusters with small primary particles, fewer primary particles with short chains, and very large particles. TEM-based morphology analysis supported the first model in which smaller particles form a fractal aggregate [24].

In a burner flame, highly matured soot aggregates can also be seen in the region beyond the flame front while incipient soot or precursor particles (young soot particles) can be found at low flame heights [25,26]. However, particulate matter from internal combustion engines contains chemical compounds such as soluble organic fractions originating from the engine lubricant and fuel. These compounds, which are absent in the burner flame, can be a challenge, leading to efforts directed at removing them through solvents, evaporation, or gasification. The comparison of soot particles from burner flames with PM from internal combustion engines was performed with elemental analysis in terms of black carbon fraction along with size and morphological investigation by TEM and a mobility particle sizer [27].

It is recognized that the inhalation of the soot particles makes an impact on the potential human health [28]. Several studies have demonstrated the increased toxicity or interstitialization of ultrafine particles compared to fine particles of the same material [29]. Since these particles showed liquid-like properties, the nanometer-sized particles draw our attentions to investigate the adverse effect on human health epidemiologically. In terms of the optical constant, the absorption coefficient of these particles is larger than that of the matured soot [30]. Because the nanometer-particles show liquid-like properties and are found to be semi-transparent in TEM from diesel ignition engines, the similarity of combustion environment to form the soot particles is required to be investigated further.

In terms of soot evolution and maturity, many studies have not been performed to compare soot from the internal combustion engine, especially compression ignition engine, with soot from the burner flame. In this study, the comparison was carried out by the chemical composition as a measure of the soot maturity after separating the soot particles from the organic fraction. Two combustion modes, the conventional combustion and LTC were applied to assume the similarity of the environment for matured aggregates, and nascent or young soot particles, respectively. A temperature-based similarity is plausible, given that temperature plays a significant role in soot particle formation. In addition, thermogravimetric analysis was conducted to examine difference in the soot particle from these two combustion modes during oxidation process of volatile organic fraction and carbonaceous soot particles. TEM was also employed to investigate and compare the morphological characteristics of the particulate matter between these two combustion modes.

2. Experimental setup

2.1. Experimental apparatus and conditions

A single-cylinder, direct injection compression ignition engine was used to investigate the characteristics of soot particles under conventional and low temperature diesel combustion. A schematic of the experimental setup is shown in Fig. 1, and the engine specification is listed in Table 1.

Commercial diesel fuel was pressurized through the filter with a high-pressure pump in a common-rail system, where the pressure was adjusted by a pressure controller (Zenobalti Co., ZB-1200). The timing and duration of the fuel injection were controlled by a programmable signal driver (Zenobalti Co., ZB-9013), which was synchronized by a cyclic encoder for 1800 pulses (Autonics, E50S8). The signal activated the solenoid unit in the injector with a peak and hold driver (Zenobalti Co., ZB-5000). In this paper, the injection