ATLANTIS PRESS
Advances in Engineering Research, volume 112
4th International Conference on Renewable Energy and Environmental Technology (ICREET 2016)

Soot Morphology and Size Distribution in Diesel Spray Flame via TEM Observation

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Keywords: Diesel; Spray flame; Primary particle; TEM.
Abstract. For better understanding of soot formation and oxidation processes in diesel spray flame, multi-point synchronous thermophoretic soot sampling was taken to obtain more information about soot morphology and size distribution in diesel spray flame. Soot samples were collected by thermophoretic sampling method in a constant volume combustion chamber. 9 different fixed sampling points were adopted for soot sampling and transmission electron microscope observation. Soot morphology and size distribution regularity in cone-shaped diesel spray flame were concluded with combustion status diagnoses. Detailed soot distribution and evolution regularity in both spray centerline horizontal direction and radial direction were gained by experimental results. Soot correlation reaction ratio in different region and combustion stage, reaction affecting factors were deeply discussed.

Introduction

Diesel engine is always accompanied with soot emission issues. In order to reduce emission of diesel particulate matter, detailed understanding of soot formation in diesel combustion is necessary. Traditional soot particle measurement measure soot number, mass and size concentration in combustion aerosols. For measurement convenience, soot particle was usually treated as regular spheres and illustrated by its aerodynamical diameter in measurement. Actual soot particle structure is more complicated and able to reveal further information about soot evolution procedures. New particle sampling methods are extended for detail soot morphological investigations. Transmission electron microscope (TEM) is an advanced technique for soot morphology observation [1,2]. Most research focused on soot evolution procedures and conducted experiment mainly in flame transmission direction. In-flame soot sampling in radial direction was rarely inferred. With different equivalent ratio, temperature and combustion stage, soot morphology, size, distribution and evolution procedure are different in different flame region. Investigation about soot morphology and size distribution through whole spray flame, especially radial direction, is not sufficient. Those information can provide a new aspect to study soot formation processes.

The present study aims to investigate the spray flame soot detail distribution with in-flame multi-point synchronous thermophoretic sampling and TEM observation. Combustion diagnosis were taken to find connection between soot evolution procedures and combustion process.

Experimental method

Diesel combustion for soot sampling was achieved in a premixed-combustion-preheated constant volume combustion chamber. Premixed acetylene and oxygen combustion was used to preheat the combustion chamber and simulate the engine in-cylinder environment. A high-speed digital schlieren optical diagnostic systems equipped at a round quartz window of the chamber was used for combustion flame high-speed observation. Fuel injection, fuel-air mixing, ignition, combustion and extinction processes could be transient recorded. Two operation condition were adopted for soot sampling to represent heavy duty high speed diesel engine operation (case1, 3.7MPa) and light load low speed operation (case2, 1.7MPa).

As seen in Fig.1，a multi-point synchronous soot sampling rig was designed and used in the present study. Considering spray flame profile, spray penetration length and combustion chamber structure, 3 injector nozzle distance Z, 60 mm, 80 mm and 100 mm were chosen. And 3 radial distance r from the centerline, 0 mm, 8 mm and 16 mm were chosen. The 9 sampling grid position are expressed as Z60r0 (Z = 60 mm r = 0 mm), Z60r8, Z60r16, Z80r0, Z80r8, Z80r16, Z100r0, Z100r8, Z100r16 in text. Soot particles were sampled onto the grid surface mainly by the thermophoretic force due to the temperature gradient between the grid and skimming burning jet.

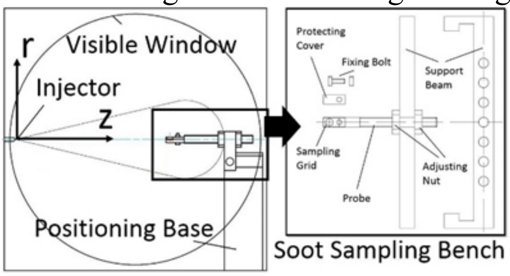

Fig.1.Constant volume combustion chamber soot sampling bench

A high resolution TEM (J FEI Tecnai 20) with a point resolution of 0.19 Å, line resolution of 1.44 Å, magnification of $25\text{-}10^6$ was adopted in this research. The time interval between soot sampling and TEM observation was controlled in 24 hours to maintain soot original form. Before the experiment, the influence of preheating combustion and sampling apparatus on soot sampling were discussed and proved to be insignificant.

Compromising complication and accuracy, soot sampling was conducted 3 times at each sampling position in this research. TEM observation images were digitally recorded with a CCD camera with resolution of 1024×1024. Observation locations were uniform distributed on the TEM grid to suppress statistic fluctuation[3]. TEM images were post processed using commercial software Image-Pro-Plus. Solid soot particle projection profile were identified, average diameter and soot projection area were statistical recorded. Apparently identifiable separately existing single primaries and the ones within the aggregates particles were recognized to measure primary diameter artificially.

Combustion Analysis

Soot sampling result and distribution characteristic were closely associated with flame shape and combustion process. Spray flame photographs recorded by high-speed photography system and combustion pressure curve could reveal many combustion information of the two cases.

Before injection, combustible mixture of acetylene, oxygen and nitrogen were inflated into the combustion chamber and ignited by a spark ignition. High temperature and pressure condition achieved by acetylene combustion was very similar to diesel engine in cylinder environment. After preheating combustion, pressure and temperature in the chamber started to decline by wall heat transfer. At the setting temperature or pressure, injector was triggered to start diesel fuel injection at the specific environment condition (900K). Self-ignition occurred in the middle of the liquid-vapor jet immediately (less than 1 ms after injection). Then the flame propagated towards both upstream and downstream. In about 2-4 ms, a bright cone–shape flame was formed and achieve its maximum area. Fire extinguishment first occurred nearby the upstream injector area after fuel injection termination. Because of the inertia acceleration, large quantity of unburnt fuel impinged to the combustion chamber bottom wall and detained nearby. The flame completely extinguished after bottom wall area combustion finished at about 10~15 ms after injection.

Spray flame evolution process of case1 and case 2 were prone to be analyzed and compared. Set injection timing as time zero, self-ignition happened at 0.4ms in case1, and 0.8ms in case 2. In case1, core flame shaped and reached its max region area at 2.9 ms, and flame radial radius R at Z=60 mm,80 mm,100 mm were 15.4 mm, 21.7 mm and 27.2 mm. In case 2, maximum flame area occurred at 2.7 ms, and flame radial radius R at Z=60 mm,80 mm,100 mm were 12.9 mm, 17.2 mm and 20.3 mm. In case 1 upstream flame extinguishment and downstream wall deposited area combustion all finished earlier than that in case 2. In case 1 cylinder pressure peaks at 7.5ms, and maintain at 3.84 MPa about 3.3 ms, then began to decline. In case 2 pressure rise rate is significantly smaller, cylinder pressure peaks at 7.1 ms, 1.75 MPa, maintain relatively longer (7.0 ms) before declining.

Soot Morphology Analysis

On the purpose of gain brief soot distribution characteristic and soot morphology information, soot TEM observation image of different sampling location was analyzed and compared in this chapter. Fig.2 shows the soot TEM observation images for Z80r0, Z80r8, Z80r16; Z100r0, Z100r8 and Z100r16 at magnification of 6600 and 26000. Dark grey textures appeared in some TEM images were carbon film thermal deformation, which commonly happened during TEM thermophoretic sampling. Many small liquid-like particle could be detected on TEM grid. Those small liquid-like particle were smaller than 20 nm, and soak on the background carbon film. The profile and number of them can’t observed precisely by TEM. According to previous researches [4,5], those small liquid-like particles considered as polycyclic aromatic hydrocarbons (PAH)-containing droplet. Reilly [5] mentioned PAH-containing liquid-like particles as important soot formation intermediates combustion. On this basis, extended multi-point soot sampling location in this study offered plentiful information in whole diesel spray flame soot morphology and size distribution.

Grids located in Z60r0 and Z60r8 of case 1 were burned, being affected by liquid spray area and high temperature. In-flame soot particles where main exist as independent primaries and aggregates. As dense solid soot sampled at r = 0 mm and r = 8 mm, solid soot concentration were dramatically small at Z80r16 and Z100r16. Solid soot distribution were quite similar between Z80r0 and Z100r0. Indicating small soot mass difference from 80mm nozzle tip distance to 100mm at the centerline horizontal direction. Small liquid-like particles, which were tiny and hard to discern, were prevalence existed in those 6 samples, especially at Z = 100 mm locations. The concentration of small liquid-liked particles on r = 16 mm samples were proximate to that on r = 0mm and r =8 mm. Small liquid-liked particles concentration were more uniform at radial direction than solid soot.

Fig.2 shows the 9 different sampling location soot TEM images at the magnification of 6600 in case 2. The environment temperature was relatively lower in case 2 than that of case 1. Thus the Z = 60 mm grids were preserved well. Both solid soot and small liquid- like particles in case 2 were significantly less than case 1. The particle size seems to be smaller, and smaller liquid-like particle concentration was also lower than that in case 1. Solid soot concentration gradually decreased from r0 to r16 at Z = 60 mm, 80 mm and 100 mm. At spray horizontal direction, solid soot mass increased from 60 mm sampling location to 100 mm.

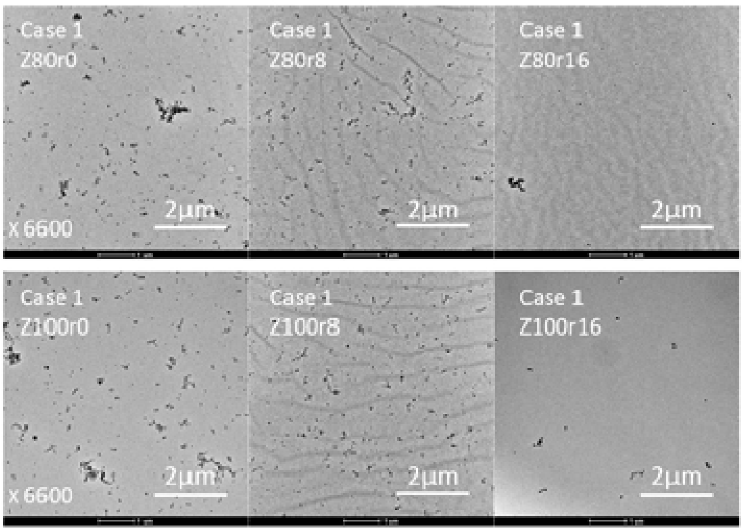

Fig.2.TEM images of soot particles sampled in case 1

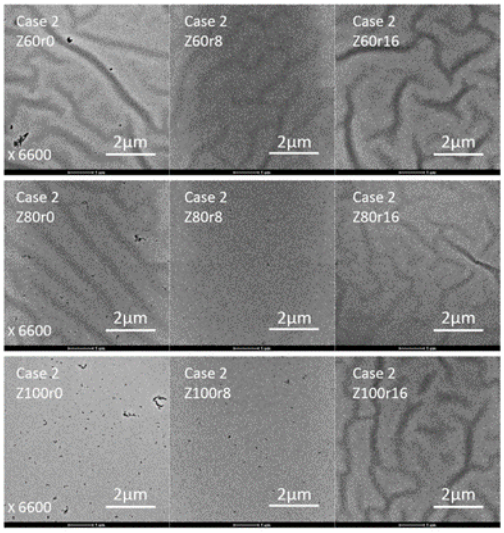

Fig.3.TEM images of soot particles sampled in case 2

Brief soot distribution feature were concluded with different sampling location TEM images. As radial center distance increased, fuel-air mixing were more sufficient and the local equivalence ratio were decreased. Solid soot mass concentration declined from cone spray flame centerline to the edge. Solid soot concentration and spray centerline horizontal change were different in case1 and case2. As combustion reaction rate and fuel injection mass was larger, soot concentration was significantly higher in case 1. Solid soot mass difference at radial and horizontal direction were all implicit in case 1, indicating a widespread high soot concentration area. In low ambient pressure condition of case 2, soot concentration gradually increased from 60mm to 100mm at horizontal direction. It is inferred that soot concentration variation on spray flame centerline were not constant and affected by combustion condition. Small liquid-like particle emerged in case 1 were more than case 2, and not sensitive to center distance r. Generation of small liquid particle was speculate to be affected by fuel mass, rather than equivalent ration.

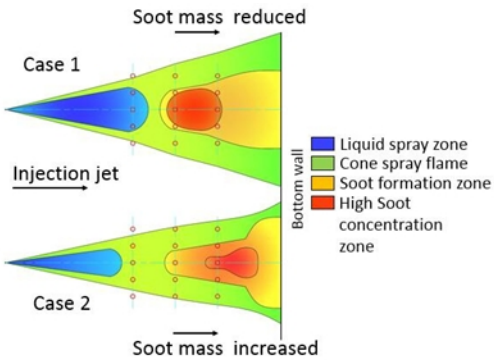

Fig.4.Soot distribution sketch of case 1 and case 2

Soot parameter variation from Z80r0 to Z100r0 shown opposite trend in case1 and case 2. Combustion reaction rate was faster, environment pressure and temperature was higher, soot consumption was higher in case 1. Surrounding PAH precursors was concentrated in this condition. Abundant solid soot were generated in the middle stage of combustion at Z = 80 mm. Then oxidation reaction overwhelmed in soot formation, many PAH precursors and soot were burned. Primary particle diameter and total soot mass declined because of surface oxidation reaction in late combustion stage at Z= 100 mm. Particle diameter and projection area were determined both by primary diameter and primary particle number, so average particle size increased from Z80r0 to Z100r0 against total soot mass. In case 2 combustion reaction was more moderate, and late combustion stage in the bottom wall deposition area was apparently longer. Solid soot generated in middle stage were relatively less. Afterwards soot nucleation and surface growth reaction continued happed frequently, soot number, primary diameter, particle size sustained increased from Z80r0 to Z100r0. The influence of soot oxidation reaction were relatively insignificant. Thus primary particle diameter and total soot projection area At at Z100r0 were all larger than Z80r0. Fig.3 shows the brief in flame soot distribution regulation.

Conclusions

Soot multi-point synchronous sampling were conducted in 9 different location in two diesel spray flame. Solid soot and liquid-like precursor distribution were investigated and discussed with combustion process diagnostic. Common soot distribution regularity and affecting factor were concluded with observation result and soot formation mechanism. The result shows that in-flame soot mainly exists in form of independent primary particle and small aggregation particle. Average soot size were less than 80 nm, large aggregate particle number were very small. Aggregation reaction was restrained by high temperature environment, and affected by soot number density. Soot PAH precursors commonly existed in spay flame, affected mainly by fuel mass. While solid soot concentration were high near the cone-shaped flame centerline and decreased as radial center distance r increased. Solid soot distribution were mainly decided by local equivalent ratio and affected by the downstream wall impinging zone near the combustion chamber bottom. Soot distribution from middle stage combustion at Z = 80 mm to late stage at Z =100 mm vary in different combustion condition. In high environment pressure and high fuel injection pressure of case1, combustion reaction ratio was faster, large amount soot were generated in the middle stage combustion, then oxidized in the late stage. In case 2 combustion were more moderate, soot concentration was significantly smaller. Soot nucleation and surface growth reaction were overwhelmed on oxidation reaction. Soot primary diameter, particle size and mass density keep increasing in the late stage combustion.

Acknowledgement

This research was financially supported by the National Science Foundation (51576083).

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