

Investigation of Soot Formation in Laminar Diesel Diffusion Flame by Two-color Laser Induced Incandescence

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ABSTRACT

Soot emissions in the combustion process of diesel engines are greatly harmful to the environment and human health. Consequently, there is large interest and great efforts in decreasing soot emission from diesel engines to meet the increasingly stringent emission standards. The mechanisms of soot formation and oxidation so far have not been well understood. Laser induced incandescence (LII) is particularly suited to measure the instantaneous spatial distribution of the soot volume concentration, which can offer much needed detailed information of soot distribution for better understanding of soot formation and oxidation. In this paper, a two-color laser induced incandescence (2C-LII) technique was implemented for measuring absolute soot volume fraction in a laminar diesel fuel diffusion flame. Based on LII signal of the same point in the flame recorded at two wavelengths, the temperature of the laser-heated soot particles was first derived and then the soot concentration of the point could be acquired by calibration at the same time. The 2D soot concentration distribution in the flame was obtained by mapping. Measurements were performed in a laminar diesel and biodiesel/air diffusion flame by 2C-LII. It was found that with the biodiesel blended in the diesel fuel, the maximum soot concentration and the soot distribution area decrease, which are consistent with the test results of diesel engines.

INTRODUCTION

The compression ignition of the stratified mixture in diesel engines is the main reason for soot emission. Due to the complex combustion and soot formation chemistry and interaction between turbulence and chemistry in a real engine, soot formation and oxidation are still not well understood. Although soot emission can be greatly reduced by particulate traps aftertreatment, it is still desired to achieve an efficient and clean in-cylinder combustion with low or even zero soot formation by adjusting injection strategies, changing combustion chamber shape, enhancing air flow and so on. Therefore, it is necessary to obtain a detailed knowledge of the spatial and temporal distributions of the soot concentration in the cylinder.

The study of soot formation mechanism and control is mainly dependent on the real time measurement of soot

in the flame. Recent investigations and applications indicated that as a useful laser diagnostic technology, laser induced incandescent (LII) could measure soot volume fraction in the flame without disturbing the combustion field. Its advantages over other measurements are high spatial and temporal resolutions with a relatively strong signal, which have drawn increased attention [1]. The 2D LII image has to be calibrated to get quantitative soot volume fraction. The quantitative gravimetric sampling and light extinction method were commonly used to calibrate soot concentration, which are very useful to the development of LII technology. However a restriction of the light extinction method is that the test results are path integrated thus the nonuniform concentration fields can not be easily resolved. The quantitative gravimetric sampling has disadvantages of disturbing the combustion and some impurity is collected [2,3]. A new calibration method was proposed by Smallwood et al. [4] in 2002. Based on the two-color pyrometry theory, two light filters were used to detect LII signals at two wavelengths. The temperature of laser heated soot particulate can be obtained from the ratio of the two LII signals [4] and consequently the absolute soot volume fraction can be inferred from the absolute LII intensities. Because of its high precision, simple equipment and on-line calibration, this new calibration method has gained popularity [5].

In this paper a LII measurement system was set up. Based on the two-color LII theory proposed by Smallwood et al [4], the 2D LII image was calibrated and then the quantitative soot volume fraction in the flame was acquired. The LII system was used to measure the laminar diffusion flame of ordinary diesel fuel and biodiesel.

MEASUREMENT THEORY

LII was first noticed by Eckbreth when he did research work of Raman spectroscopy in flame in 1977 [6]. He found that the Raman spectroscopy was overwhelmed by the presence of soot particles that emitted laser modulated incandescence. He was able to relate the time dependence of this interference to laser particulate heating, heat transfer to the medium, particle vaporization, and indirectly to the particle size. Melton performed numerical calculations to investigate the possibility of developing a soot diagnostics based on this laser heating of particles. He concluded that the radiant

emission from heated soot particles is proportional to soot concentration [7].

The principle of the LII method is based on heating of soot particles in the flame by a pulsed laser sheet with the duration of less than 20 ns. The temperature of the soot particles within the laser pulse increases rapidly and reaches about 4000 K. As the temperature of the laser heated soot particles increases, the blackbody radiation intensities from these particles increase dramatically. Through filter, the radiation signal, which is proportional to the soot volume fraction in the laser sheet, is recorded by Intensified Charge Coupled Device (ICCD). After the calibration, soot 2D concentration distribution can be acquired based on the recorded radiation signal. The biggest advantage of LII is that the radiation from the heated soot particles is much stronger than that of flame background, and then the flame luminosity can be suppressed.

In the calibration process, the LII signal emitting from soot particle is recorded by a detection system. After the calibration of LII signal, the quantitative soot volume fraction can be acquired. Some research teams investigated the quantitative soot concentration measurement by multiplying the LII intensity by a calibration factor C :

$$f_v = C \times I_p. \quad (1)$$

Here I_p is the LII intensity recorded by the detection system and f_v is the soot volume fraction. The calibration factor C was obtained by correlating the LII intensity I_p with a known soot volume fraction f_v , which was either measured independently such as by light attenuation or gravimetric sampling, or known in a well-controlled carbon black generator. As long as there are no significant changes between the conditions of the calibration and the LII measurements, the deduced volume concentration should be accurate enough. However, it is quite difficult to keep the same condition of the calibration as the actual measurement. Some changes such as local gas temperature, the chemical compositions of the particulate matter, and the ambient pressure etc. will result in errors.

Snelling et al. [8] proposed a new method to calibrate LII signal, which correlates LII signal intensity with soot particle temperature and soot volume fraction. The soot temperature heated by the laser was calculated by two-color method, and then the absolute local soot volume fraction was acquired based on the methodology summarized as follows [8]:

The spectral radiance per unit solid angle of a known source such as tungsten ribbon lamp or black body furnace is

$$R_{CAL}(\lambda, T) = \frac{2c_0^2 h \varepsilon(\lambda, T)}{\lambda^5} [\exp(\frac{hc}{k\lambda T}) - 1]^{-1}, \quad (2)$$

where $\varepsilon(\lambda, T)$ is the emissivity as a function of wavelength λ and temperature T . c_0 is the speed of light, h is Planck constant and k is Boltzmann constant. When the gain of the detection system is G_{CAL} , the observed LII signal is V_{CAL} . A calibration factor can be defined as the following form

$$\eta(\lambda) = \frac{V_{CAL}}{R_{CAL}(\lambda, T) * G_{CAL}}. \quad (3)$$

It means that the sensitivity of the detection system in a wavelength is a constant when the system is set up. For primary soot particles whose diameters d_p are in general quite small relative to the wavelength, the particles are in the Rayleigh limit, which can be approximately defined as $\pi d_p / \lambda < 0.3$. At temperature T_p , the total radiated power per unit volume of particle at wavelength λ is

$$\phi_p(\lambda, T_p) = \frac{48\pi^2 c^2 h}{\lambda^6} [\exp(\frac{hc}{k\lambda T_p}) - 1]^{-1} E(m_\lambda). \quad (4)$$

Here $E(m_\lambda)$ is a function of the soot refractive index and is called the soot absorption function. In the laser sheet of thickness w_b , the soot cloud with the concentration of f_v will produce an experimental voltage signal expressed as

$$V_{EXP} = \eta(\lambda) G_{EXP} f_v w_b \frac{1}{4\pi} \phi_p(\lambda, T_p). \quad (5)$$

From equations 4 and 5, the soot volume fraction is obtained as

$$f_v = \frac{V_{EXP}}{\eta w_b G_{EXP} \frac{12\pi^2 c^2 h}{\lambda^6} E(m_\lambda) [\exp(\frac{hc}{k\lambda T_p}) - 1]^{-1}}. \quad (6)$$

In order to get f_v from equation 6, the soot particle temperature must be known, which can be measured by the two-color method. Because $\exp(hc/k\lambda T_p) \gg 1$, equation 4 could be expressed using Wien approximation. From equations 4 and 5, the ratio of LII signals at λ_1 and λ_2 can be obtained as follows

$$\frac{\lambda_2^6 E(m_{\lambda_1})}{\lambda_1^6 E(m_{\lambda_2})} \exp[-\frac{hc}{kT_p} (\frac{1}{\lambda_1} - \frac{1}{\lambda_2})] = \frac{V_{EXP1} \eta_2 G_{EXP2}}{V_{EXP2} \eta_1 G_{EXP1}}. \quad (7)$$

The value of the $E(m_\lambda)$ is 0.3 at λ_1 and λ_2 [5], and then the soot particle temperature and the soot volume fraction can be acquired.

EXPERIMENT SETUP

The LII measurement system was set up as shown in Fig. 1. The Nd:YAG laser is Brilliant B produced by Quantel company, which can give out 10 Hz 532nm wavelength light with the FWHM of the temporal pulse

around 7 ns. If the laser energy is too high, the vaporization will occur on the particle surface, which results in the decrease of soot volume fraction with the attenuation of LII signal. If the laser energy is too low, the soot particle could not be heated to a sufficiently high temperature to emit significantly higher incandescence intensity above the flame emission. The laser energy should be tuned

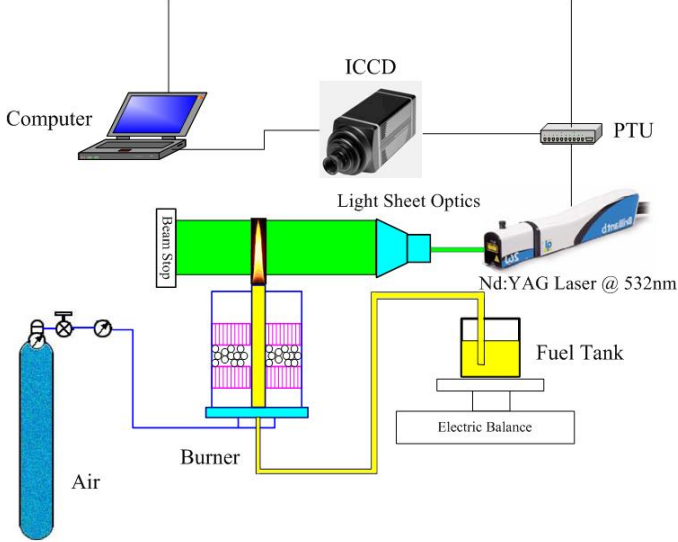


Fig.1. Schematic diagram of the LII experiment setup

appropriately so that the LII signal does not or weakly depend on the laser energy and is proportional to the local soot concentration [9]. As shown in Fig.2, the normalized LII signal is a function of the laser pulse energy. There is a 'plateau regime' of LII signal when the laser energy reaches 90 mJ. The experiment should be implemented in the 'plateau regime' and then the LII signal is somewhat insensitive to laser energy. Therefore in the experiment the energy of the laser pulse is 100 mJ. The beam with the diameter of around 9 mm was expanded by several cylinder lenses to form a uniform laser sheet (1 mm × 50 mm) across the measurement volume. The elastic scattering signal of soot particle in the flame and burner wall would affect the detection of LII as well as the laser induced fluorescence (LIF) of C₂ and PAHs. Since the scattering occurs in the laser pulse duration and the fluorescence induced by the laser decays rapidly about 20 ns whereas the LII signal lasts about several hundred ns, thus the record time of ICCD can be delayed. With appropriate filters and short gate time, the background luminosity such as natural flame luminosity can be rejected with the improvement of signal-to-noise ratio. A Princeton ICCD PIMAX2 was used to record LII 2D images, the gate width of which was set to 20 ns with a delay of 40 ns after the laser pulse. The center wavelength of the filter is 590 nm with FWHM 30 ns.

Many previous LII studies focused on the diffusion flames with gaseous fuels, but there are not enough data available concerning the liquid fuel flames. The atmospheric pressure laminar diffusion flame in this study was produced by the combustion of diesel fuel in air. The schematic diagram of the liquid burner is shown

in Fig.3, which was designed based on the oil lamp principle. The center of the burner is a steel tube with the diameter of 8 mm, which is connected with fuel tank by a pipe. The liquid surface in the tank keeps level. The fuel vapor for combustion is supplied by a sintered metal immersed in the fuel. Through glass beads and honeycomb plate, the co-flowing air (co-flowing diameter was 100 mm, co-flowing flux was 6 m³/h) was provided to keep a stable flame. The flame height was controlled by adjustment of fuel depth or the outer part of the sintered metal. An electronic balance was used to weigh the fuel and the fuel flow rate was derived from the weight loss of the fuel. In the experiment the fuel flow rate was fixed at 3.57 mg/s and the height of flame is about 45mm. Fig. 4 shows a photograph of the test flame.

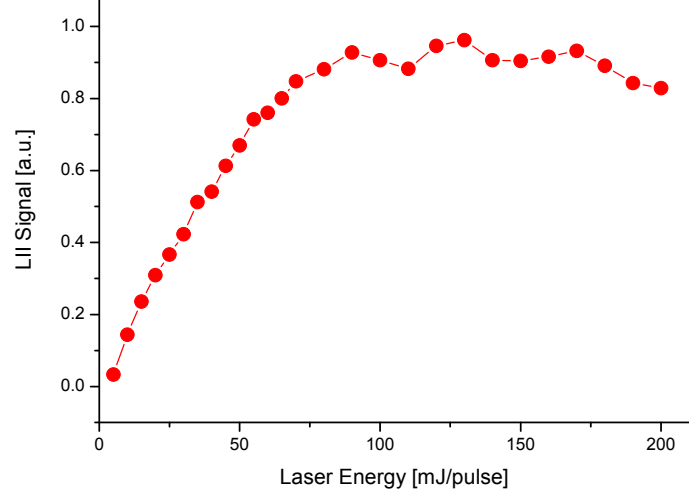


Fig.2. Laser energy curves

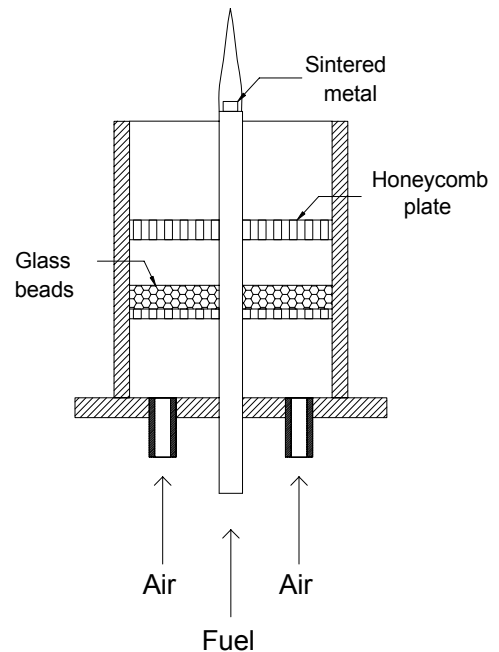


Fig.3. Laminar diffusion flame apparatus

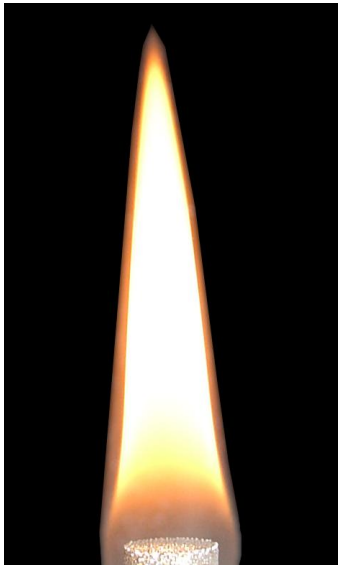


Fig.4. A photograph of the test flame

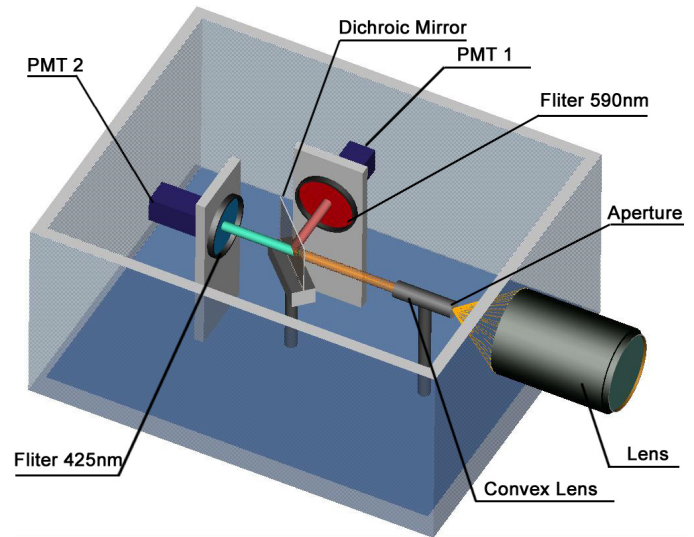


Fig.5. LII calibration equipment

As shown in Fig.5, the calibration equipment is composed of two photo multipliers tubes—PMT (Hamamatsu 6780-20), whose rise time is less than 1ns. The LII signal from measurement volume in the flame was imaged to an aperture by a lens. The aperture limits the detection volume as a cylinder, whose diameter is 1 mm and the height equals to the laser sheet thickness. The aperture is put on the focus of a convex, through which the LII signal forms a beam with the diameter of 6 mm. The beam is split by a dichroic mirror and reaches the PMT cathode respectively. A filter is applied to each PMT. The center wavelengths of the filters are 425 nm and 590 nm with FWHM 50 nm and 30 nm. The system was assembled in a metal box which was put on a translation stage. The soot concentration of the measurement volume can be obtained by scanning the flame.

Before performing measurements, a calibration of the LII calibration equipment was performed with a black body furnace of known spectral radiance by recording the light emission. In the calibration process, the temperature of the black body furnace was controlled from 1900 K to 2500 K. From equations 2 and 3, the calibration factor $\eta(\lambda)$ can be acquired.

The laser beam with 532 nm was expanded to a sheet by cylinder lenses and passed through the center line of the flame vertically. At the same time a synchronization signal was sent to programmable timing unit (PTU). After receiving the synchronization signal, PTU controlled the gate time and delay of ICCD, the 2D LII image was recorded and the relative soot volume fraction distribution was acquired. Then a detection area with the diameter of 1 mm in the flame was measured by the calibration equipment, getting the soot volume fraction in the measurement volume. By mapping the calibration points to the 2D LII image, the quantitative soot volume fraction in the laser sheet was acquired.

RESULTS AND DISCUSSION

A biodiesel (B100) laminar diffusion flame was used to calibrate the LII system. The biodiesel was extracted from palm oil. The signal-to-noise ratio was not very good in the bottom of the flame where polycyclic aromatic hydrocarbons (PAHs) were believed to be in relatively high concentration. PAHs are common soot precursors in flames, and their fluorescence extends over a large spectral range in the visible. It is therefore difficult to separate the PAHs fluorescence from the LII signal with detection filters. With the increase of the laser fluence the contribution of PAHs interference increases and affects the signal recorded by PMT. So the calibration locations were at 25 mm height about burner (HAB) where the PAHs were almost zero. The interval of horizontal scan was 0.4 mm. In Fig. 6, the typical incandescence time decay curves of a test volume (HAB= 25 mm, radial position= 0 mm) were obtained for both investigated wavelengths. Each curve was built by averaging 200 individual curves on the oscilloscope (LeCroy WaveRunner 104Xi). By the ratio of the two LII decay curves of Fig. 6, the time behavior of the soot temperature can be calculated as shown in Fig. 7. The synchronization signal from Nd:YAG laser was sent out at 0 ns. At 30 ns the signal was disturbed because of Q-switch opening. As shown in Fig.6, at 75 ns the laser pulse was sent, whose temporal profile was measured by a high speed silicon detector (Thorlabs DET10A). Within the laser pulse, the LII signals increase rapidly because the soot particles continue to absorb laser energy. The peak temperature of soot particle was about 3880 K, from which the soot volume fraction was inferred. After reaching the maximum temperature, the soot particle was cooled down for hundreds of nanoseconds after the laser pulse toward that of the surrounding gases mainly due to heat conduction. The cooling rate of the soot particle was dependent on the particle size, so the decay curve of the particle temperature was used to evaluate soot primary particle diameter [10,11]. This is another important

application of the LII technology in the recent years. Although 200 individual LII signal curves were used to build the LII signal decay curves for calculating the temperature, the resultant temperature curve in Fig.7 was still somewhat noisy, especially at longer times where the signal intensity was much lower. Due to the exponential dependence of soot volume fraction on temperature, Eq.(6), it is obvious that a small error on the laser heated soot temperature might strongly affect the estimation of the soot concentration. So it is necessary to improve the signal-to-noise ratio to obtain smoother curves of the temperature decay, which is the key point to LII measurement.

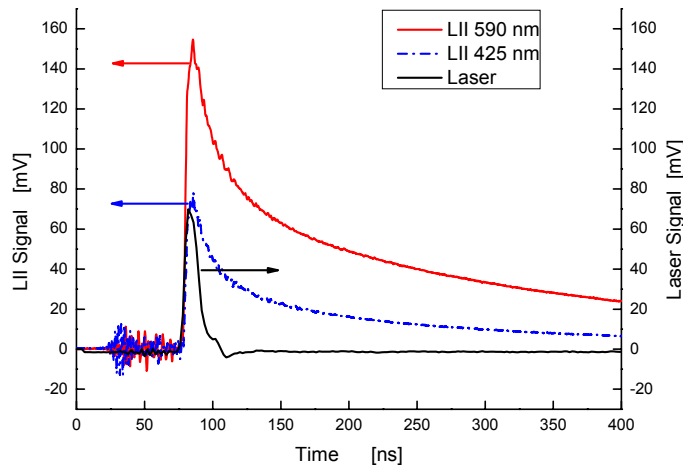


Fig.6. Two-color LII signal and laser signal

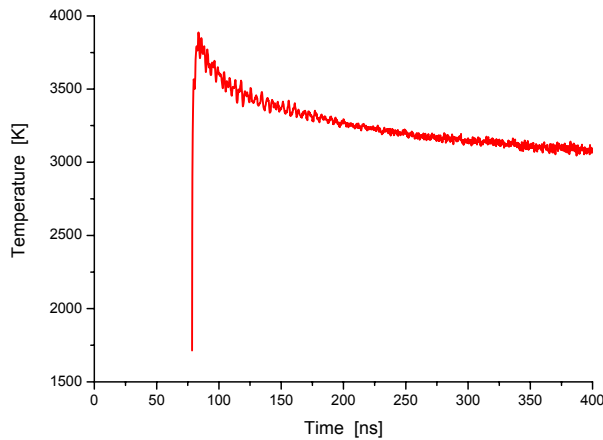


Fig.7. Soot temperature

The qualitative soot concentration distribution of biodiesel flame was acquired by averaging 50 LII images recorded by ICCD. By mapping calibration points to the 2D LII image, the 2D quantitative soot concentration image was obtained. The result of the flame at 25mm HAB is shown in Fig.8, the blue solid curve is the soot profile from the 2D image and the red points are the calibration data. There is a relative agreement between the calibration points and the 2D image except a point in the right of flame.

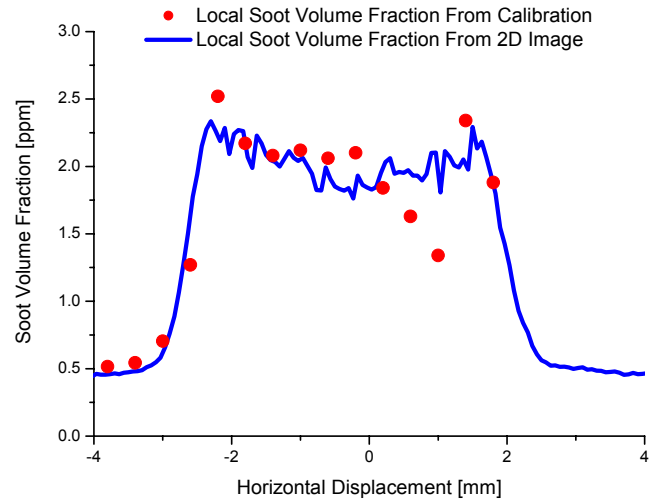


Fig.8. Two-color LII calibration

It is easy to find two little peaks in the same height. The signal-to-noise ratio near the flame edge is not good either due to very low soot concentration. The difference at the extremities of profiles will cause errors when estimation temperature at the edge of the flame. So it is important to improve signal-to-noise ratio by optimizing the detection system. Furthermore the horizontal scanning interval should be reduced to 0.1 mm and more calibration data are required, which can improve the measurement precision.

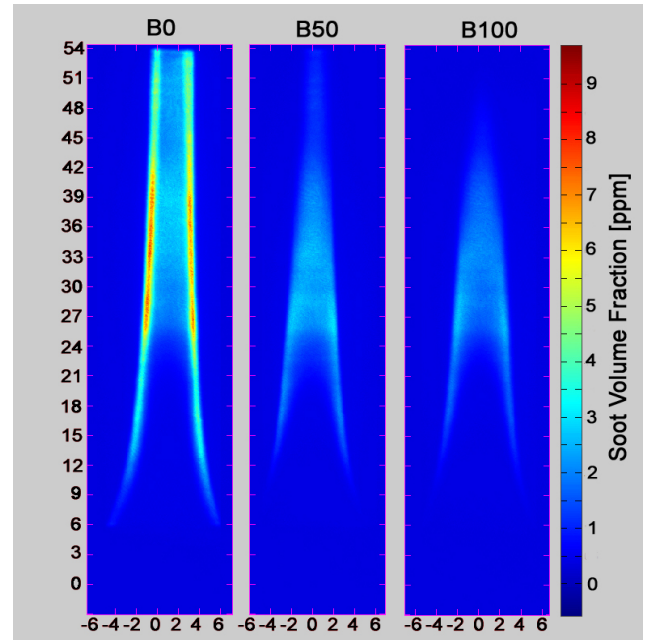


Fig.9. Calibrated LII images in the flames

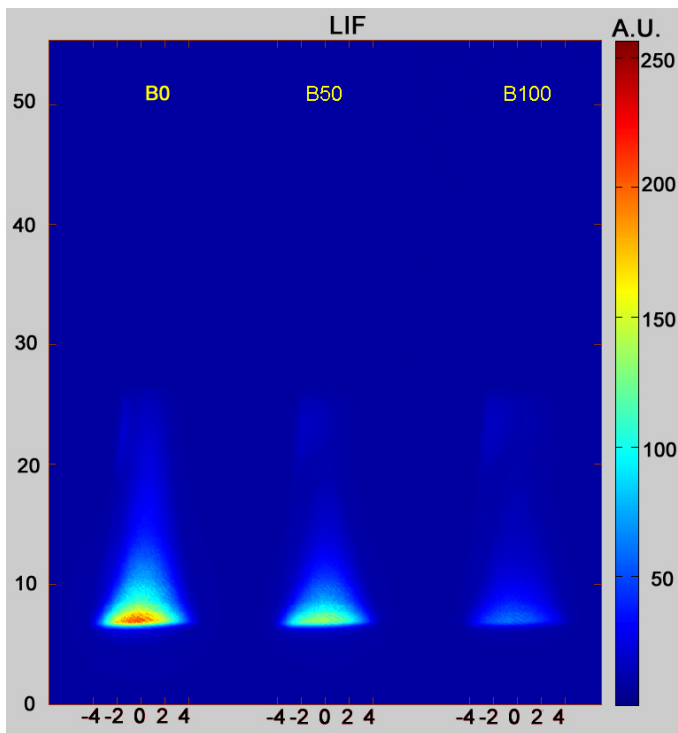


Fig.10. FIL images in the flames

According to the amount of biodiesel blended in the conventional diesel fuel, three fuels are named as B0 (100% diesel fuel), B50 (50% diesel fuel and 50% biodiesel, volume basis) and B100 (100% biodiesel). The quantitative soot volume fractions of these three flames are shown as Fig.9. The soot rich concentration was distributed in the wings of the B0 flame while in the top of B50 and B100 flames. The two lines in the case of diesel flame indicates the fact that the flame is smoking, i.e., soot escapes from the flame tip or soot particles are not fully oxidized and can survive. The maximum soot volume fraction in the B0 flame is about 8.5 ppm. With the increase of the amount of biodiesel in the diesel fuel, the maximum soot concentration decreases. In the B100 flame the maximum soot volume fraction is about 3.7 ppm and the soot distribution area is smaller than those in the flames of B0 and B50. The results can be explained from two points. First, compared with the diesel fuel, the biodiesel has less PAHs, which is the soot precursor. With the more biodiesel blended in, the amount of PAHs will be reduced, so the soot in the flame decreased. Figure 8 demonstrate the PAHs distribution in the flames. The PAHs fluorescence was excited by 266 nm laser. It was shown that the intensity of PAHs fluorescence reduced as the biodiesel amount increased. Second, biodiesel is one kind of oxygenated fuels. The oxygen atom can accelerate combustion process and the absence of oxygen in the middle of diffusion flame can be improved. Then the soot particle oxidation is speeded up and the fuel can be combusted more completely, which leads to less soot emission.

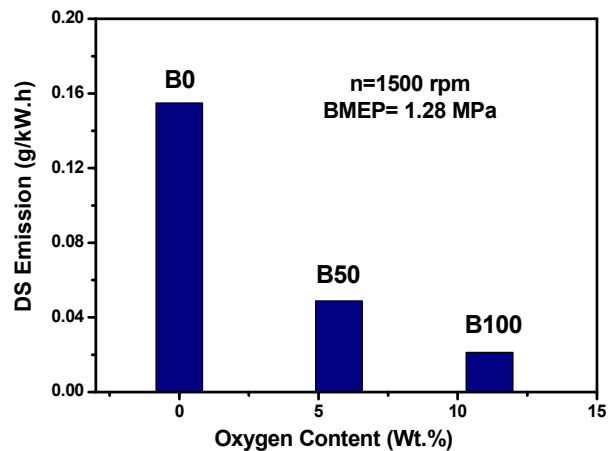


Fig.11. Dry soot Emission of B0, B50 and B100

In order to measure the soot emission of B0, B50 and B100, a diesel engine was used to combust the three fuels. The test engine was Cummins ISBe6 direct injection diesel engine, with a turbo-charger and an intercooler, meeting Euro III standard. The engine specification is shown in Table 1.

Table 1 Engine specifications

Engine Model	Cummins ISBe6
Type	4-stroke, in-line, turbo-charging, intercooling, direct injection
Cylinder number	6
Displacement (L)	5.9
Compress ratio	17.5
Rated power/speed	136 kW / 2500 r·min ⁻¹
Max toque/speed	670 N·m /1500 r·min ⁻¹
Fuel supply system	High pressure common rail

PM was sampled on filters (TX40HI20-WW of PALL Corporation) by AVL SPC-472 partial flow diluter smart sampler. The filters were extracted in dichloromethane (CH₂Cl₂) and deionized water, separating the PM into two parts: dry soot (DS) and none soot fraction (NSF). The filters were weighed by an electronic balance with 10µg sensitivity. In figure 11, it can be observed that DS emission decrease consistently as more biodiesel is blended. B50 resulted in a 69% DS reduction and B100 resulted in 86% DS reduction compared with B0. Lots of researches also show that in diesel engines fueled with biodiesel blend fuels, the soot emission was greatly reduced [12-14].

CONCLUSION

In the paper a two-color laser induced incandescence measurement system was set up. Several test volumes

were selected as the calibration points in the flames. The LII signal time decay curves of the test volumes were obtained for both investigated wavelengths, from which the soot volume fraction can be calculated. Mapping the calibration points to the 2D image, the quantitative soot concentration distribution can be acquired, which is very useful to the soot mechanism research in an optical engine. Three kinds of fuel flames were used for LII measurement, some conclusions can be drawn as follows:

1. The LII measurement system was set up. Based on the new calibration method proposed by Snelling et al., the 2D LII image was calibrated and the quantitative soot concentration distribution was obtained.

2. The LII measurement system was used to measure soot concentration in the flame produced by biodiesel blend fuel. There is a relative agreement between the calibration points and 2D image, which proves the LII system works well.

3. The soot rich concentration occurs in the wings of the B0 flames while in the top of B50 and B100 flames. With the biodiesel blended in the diesel fuel, the maximum soot concentration and the soot distribution area decrease, which are consistent with the test results of diesel engines.

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