Feature Article

Laser Diagnostics of Soot Formation Processes in Diesel Spray Flame

Tetsuya Aizawa

In order to develop clean next-generation diesel engines with ultra-low exhaust particulation emission, it is important to fully understand the soot formation processes occuring in the spray flame in the engine cylinder. The authors employed laser spectroscopy and imaging techniques to examine when and where the formation and growth of soot precursors occur in the diesel spray flame. The chemical composition and the growth processes of the soot precursors were further investigated by a newly-developed laser-spectroscopic technique called Excitation-Emission Matrix (EEM) using a multi-wavelength excitation laser source. The examination of diesel spray flame using the EEM technique revealed that the formation and growth of soot precursors can be delayed or suppressed by lowering the ambient temperatures and oxygen concentration in the cylinder.

Introduction

Diesel engines are expected to play an important role as environmentally friendly and energy efficient powertrains in the mitigation of global warming. However, the emission regulations for diesel engines are ever tightening. In order to further suppress the particulate emission without sacrificing the superior thermal efficiency, it is becoming necessary to employ the real-time model-based prediction and control of the particulate emission trend, taking account of various combustion modes and transient engine operating conditions. The emission of soot, a major component of the particulate, from diesel engines is known to be governed by the "balance between formation and oxidation" of soot in the cylinder. However, currently available soot models are not capable of reasonably predicting the "balance between formation and oxidation" under various combustion modes and engine operating conditions.

In addition, varieties of "smokeless" diesel spray combustion concepts, based on the control of fuel injection and ambient conditions, are attracting a growing attention. However, the soot suppression mechanisms of these concepts have not been experimentally investigated in detail. A number of numerical simulation studies of the soot suppression mechanisms using detailed chemical kinetics have recently been reported, but the experimental data needed for the validation of the simulated results are lacking.

In order to investigate the soot formation process in a diesel spray flame, the author has recently conducted spectral measurements of laser-induced emission[1] at two different excitation wavelenghes (266 and 355 nm) and simultaneous 2-D imaging of soot precursors and soot particles in a diesel spray flame using laser-induced fluorescence (LIF) and laser-induced incandescence (LII) techniques[2,3]. As shown in Figure 1, the measurement results revealed that the soot precursors are observed in the central region of the spray flame immediately after ignition and the soot particles gradually appear from the periphery of the soot precursor region and extend downstream. The author further investigated the the chemical composition and the growth processes of the soot precursors and also the effect of engine operating conditions (ex. ambient gas, fuel, and injection conditions) on soot formation processes in diesel spray flame by a newly-developed laser-spectroscopic technique called Excitation-Emission Matrix (EEM) using a multi-wavelength excitation laser source[4]. The present paper reports the measurement principles, experimentals, and some examples results of EEM analysis of soot formation processes in diesel spray flames.

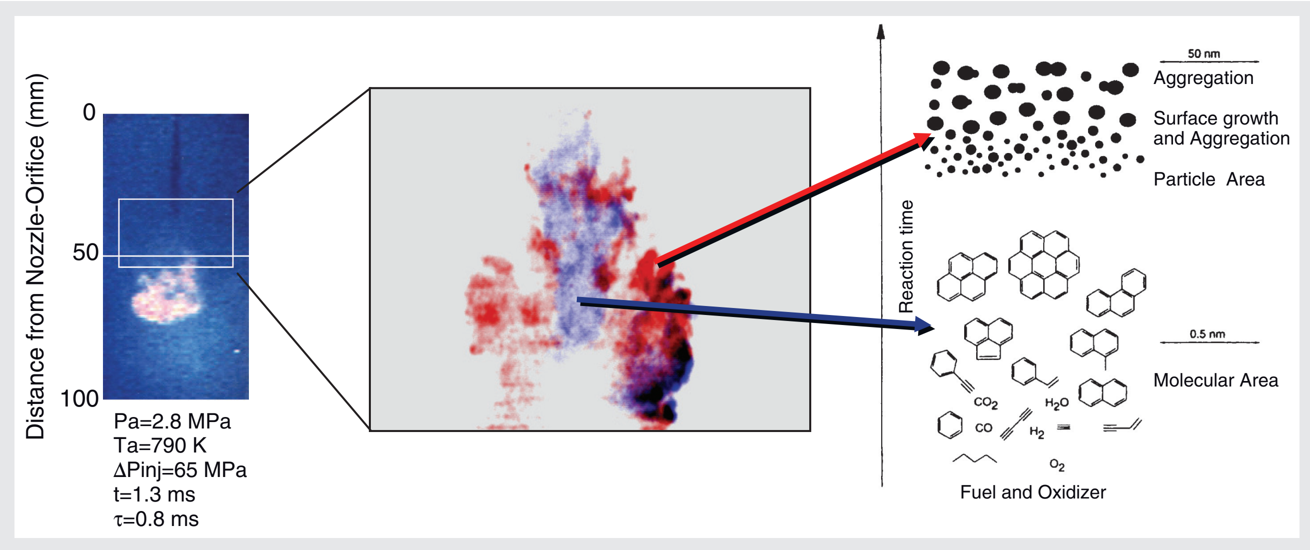

Figure 1　Simultaneous images of LIF from soot precursors (blue) and LII from soot particles (red) in a diesel spray flame

Excitation-Emission Matrix (EEM)

EEM is a version of multi-wavelength excitation fluorometry techniques invented by Christian in 1970's and a detailed explanation of the instrumentation and the measurement theory of EEM is found in Ref.[5] and [6]. The energy states of many chemical components are known to get excited by absorption of light, and then get relaxed by emission of light. The wavelengths of the absorption and the emission of light are unique to each chemical component. Figure 2 shows example 3-D plot of absorption (Excitation) wavelengh on x-axis, Emission wavelength on y-axis and the emission (fluorescence) intensity on z-axis of pyrene, a representative component of soot precursors known as poly-cyclic aromatic hydrocarbons (PAHs). One notices that pyrene has absorption band around 330 nm and emission band around 390 nm, respectively. Primary absorption and

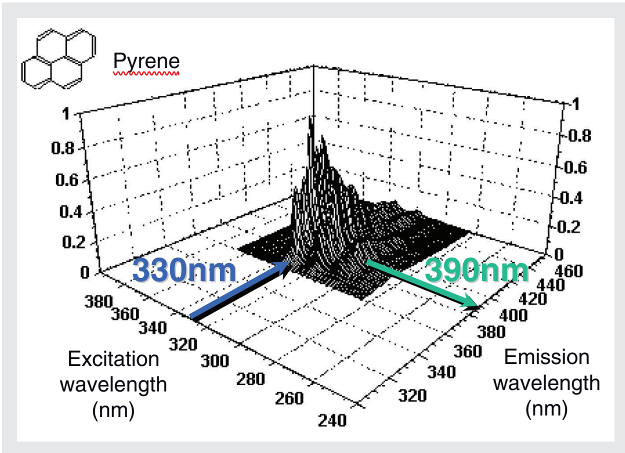

Figure 2　Example EEM of pyrene

emission bands of a number of representative PAHs obtained from literature[7] are schematically presented on a reference EEM map on the right of Figure 3. The EEM

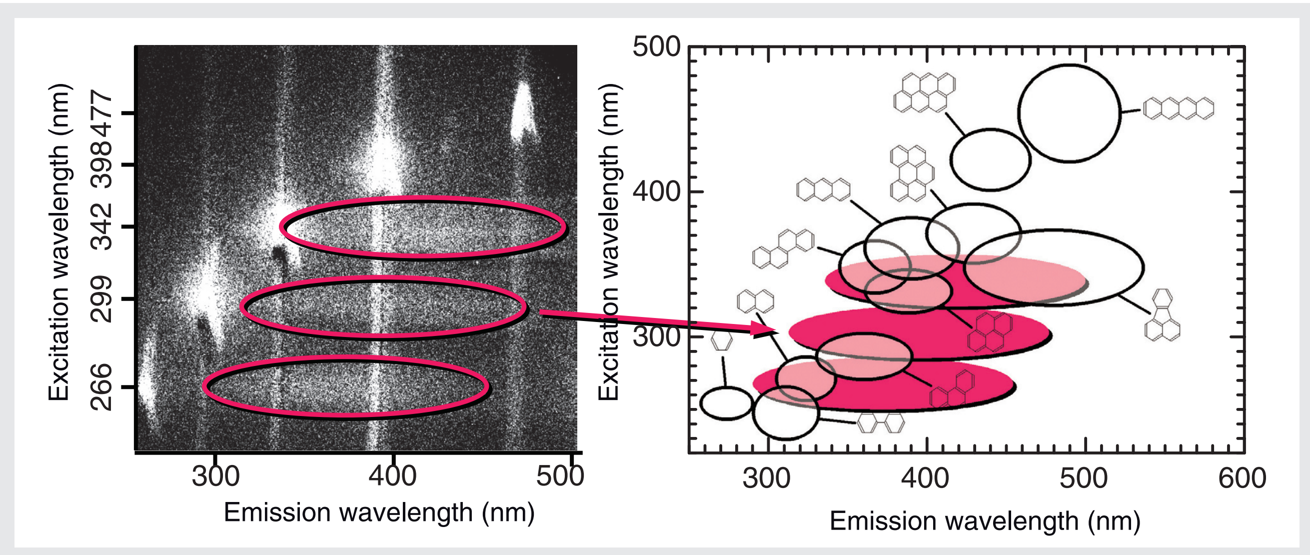

Figure 3　EEM measured in a diesel spray flame (left) and EEM map of representative PAHs (right)

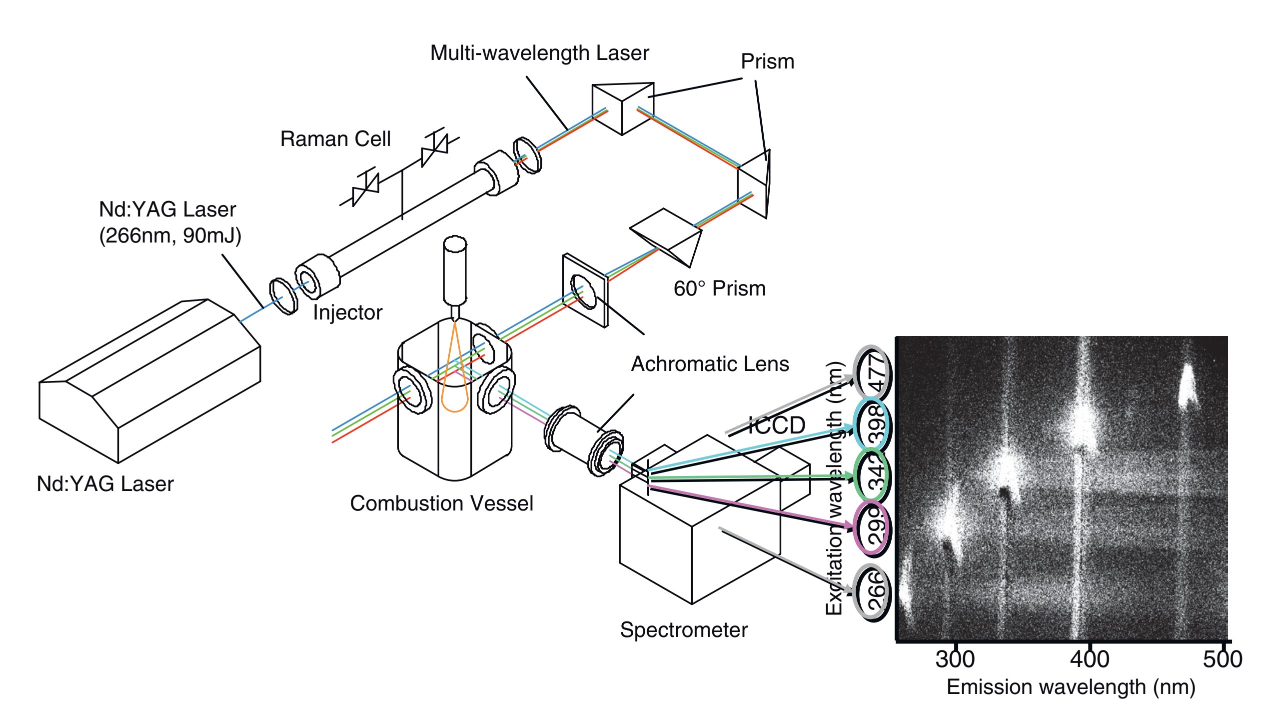

Figure 4 Optical setup for EEM measurement of diesel spray flame

map shows that each PAH component has its own distinctive spectral region and there is also a general tendency that larger PAHs have the absorption and the emission bands in longer wavelength regions. Shown on the left of Figure 3 is an example of measured EEM image which were obtained in a diesel spray flame as described in the following section. By comparing the measured EEM image with the reference EEM map, one may analyse which PAH components exist in a diesel spray flame.

Experimental Setup

Figure 4 shows the experimental setup for EEM measurements in a diesel spray flame used in the present study. The 4th harmonic (266 nm, 60 mJ) of a pulsed Nd:YAG laser is converted to a coherent multi-wavelength "rainbow" laser light by a high-pressure Raman cell described below. A 60-degree triangular diffraction quartz prism vertically disperses the multi-wavelength laser, and a UV achromatic lens focuses the laser lights into a series of focal points aligned along the central axis of a diesel spray flame in an optically accessible constant volume combustion vessel described below.

The top-to-bottom height of the series of the focal points used for the measurements, from the shortest to the longest wavelengths (in the present study 266, 299, 342, 398 nm), was about 5 mm. The LIF of PAHs from each focal point in a diesel spray flame, excited by different wavelengths, is collected by another UV achromatic lens and is separately focused onto a series of focal points on the entrance slit of a spectrometer. An ICCD camera at the exit port of the spectrometer simultaneously captures the LIF spectra excited by different laser wavelengths along a series of horizontal rows as shown in the example image on the left of Figure 3.

Multi-wavelength light source is an essential element required for the measurement of EEM. In a conventional EEM fluorometry setup, a broadband light source, such as high-pressure mercury lamps, has been used. However, the power and the spectral density of such light sources are not sufficient for single-shot fluorescence measurements in a diesel spray flame. The light source used in the present study is a coherent multi-wavelength "rainbow" laser which is composed of a pulsed Nd:YAG pumping laser and a Raman cell frequency converter. The details of the multi-wavelength light source have been explained in a previous publication[8].

The EEM measurements of diesel spray flames were conducted using an optically accessible constant volume combustion chamber. The chamber has a 183 mm-tall rectangular shape with a 56 mm×56 mm square cross section and a volume of 560 cm^3, enabling an investigation of a transient non-impinging diesel spray flame. A common-rail fuel injector with a single-hole nozzle is mounted at the center of square chamber head and the fuel spray is injected along the central axis of the chamber. Three quartz windows with a view field diameter of 35 mm are mounted on side walls of the chamber for optical measurements. Diesel-like high temperature and high pressure experimental conditions with desired ambient oxygen concentrations are simulated by spark-ignited premixed combustion of acetylene, hydrogen, oxygen and nitrogen mixture.

EEM Measurements of Diesel Spray Flame

The EEM technique was applied to the analysis of soot formation processes in diesel spray flames under different ambient conditions. Figure 5 shows an example result of high-speed laser shadowgraphs and EEM images of a diesel spray flame at ambient temperature of 940 K and oxygen concentration of 21%. The black region in the laser shadowgraphs corresponds to sooty region in the spray flame. All the EEM images are ensemble-averaged images of up to 10 different combustion events at the same condition for an improved signal-to-noise ratio. The ignition delay, which is the time duration required for the fuel spray injected into high pressure and high temperature ambient gas to evaporates, mixes with the ambient gas and auto-ignites, was approximately 0.8 ms. The timing of the ignition delay is indicated by a triangle mark on the bottom edge of the shadowgraph. The EEM measurement timings of 0.6 ms, 1.0 ms and 2.0 ms corresponds to before ignition, right after ignition and well after ignition, respectively.

In the EEM images, bright spots aligned from the top right to the bottom left of the image and vertical stripes overlapping with the spots are the laser light scattered by gas or soot particles and inner surfaces of the combustion chamber, which are not important for the measurement and should be ignored. The broad horizontal stripes are the LIF from PAHs excited by the laser light. This LIF was not observed in a non-combusting evaporating spray injected into an ambient gas with no or very low oxygen, which proves that this LIF is not be due to the aromatic impurities originally contained in the fuel but due to combustion generated components. The LIF exhibits very broad and featureless emission regardless of the excitation wavelength, which is typical of fluorescence due to large molecules such as PAHs.

From Figure 5, one notices that the EEM measured at t=0.6 ms, before ignition, already exhibits PAH fluorescence. As the measurement timing progresses, the intensity of LIF increases and the wavelength of LIF broaden towards red. These results suggest that smaller

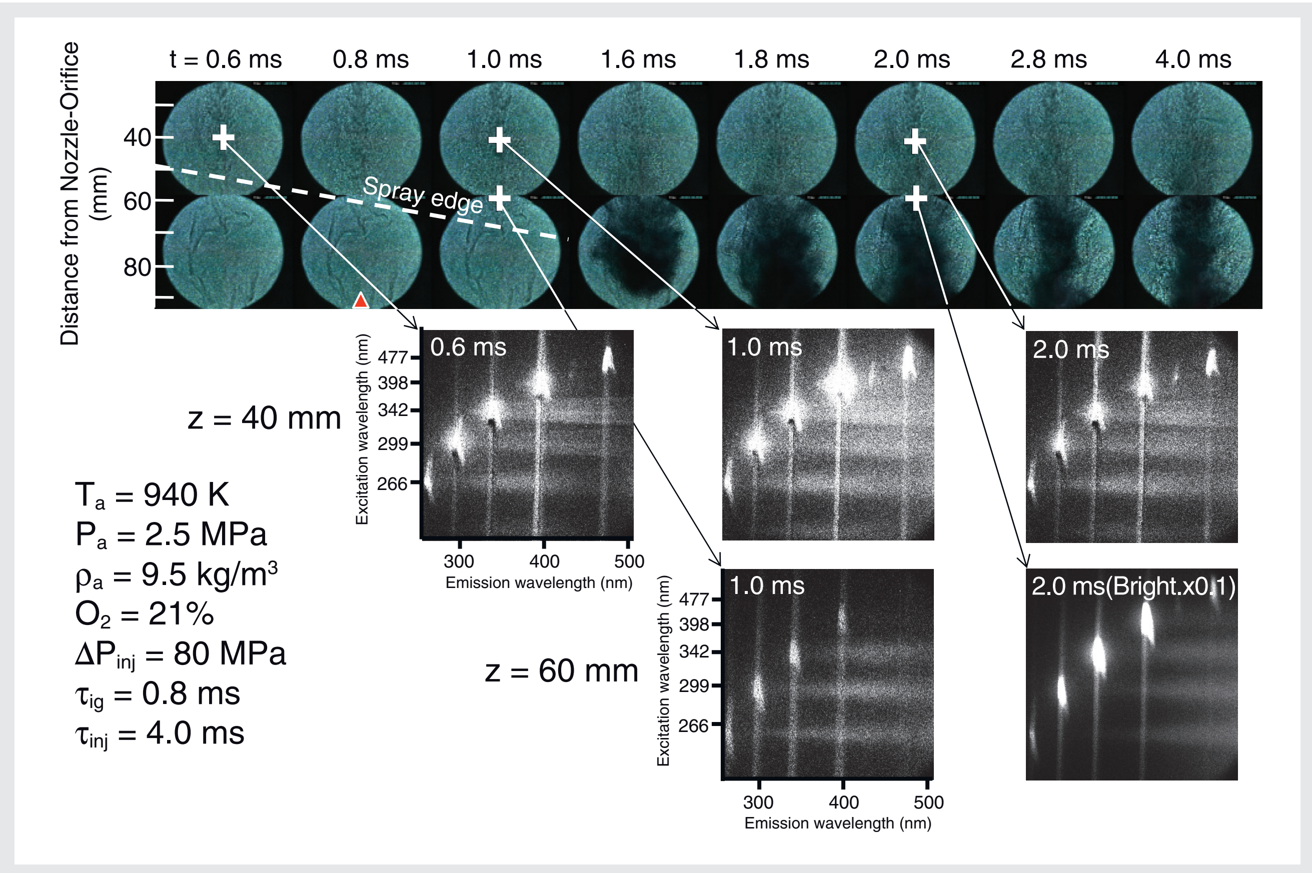

Figure 5 Example EEM images and laser shadowgraphs of diesel spray flame at standard condition

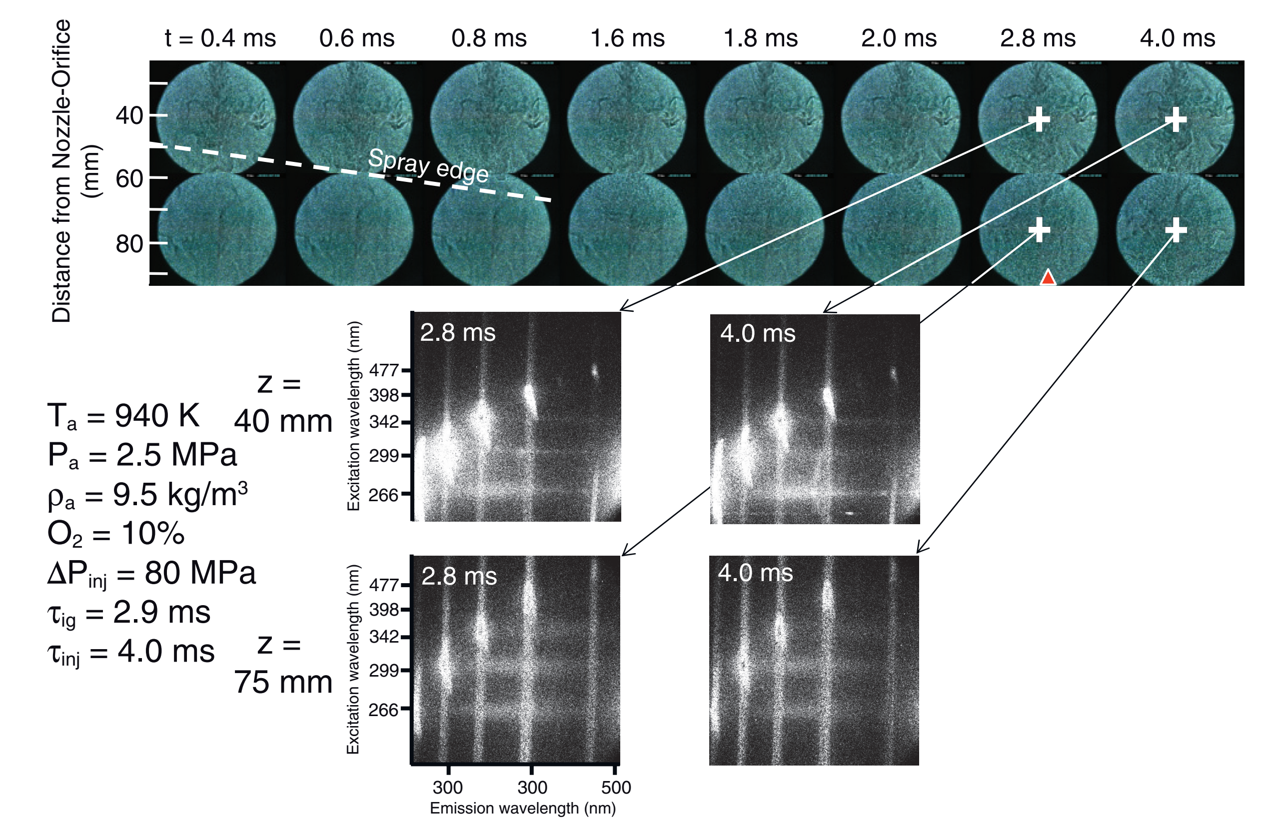

Figure 6 Example EEM images and laser shadowgraphs of diesel spray flame at lowered ambient oxygen concentration of 10%

PAHs formed in the spray grow to larger PAHs. At z=60 mm and t=2.0 ms, where soot is observed in the laser shadowgraph, a significantly brighter emission than the previous broad LIF appears in a longer wavelength region on the right end of the EEM image. Note that the relative brightness of the EEM image of z=60 mm and t=2.0 ms is lowered by 0.1 times than the other images. This bright emission is a laser-induced incandescence (LII) from soot particles in the spray flame. LII is known to exhibit a monotonously brightening identical emission spectrum beyond 400 nm independent of the excitation wavelength, because LII is a thermal radiation from the soot particles and emission wavelength is determined by the temperature of the laser-heated soot particles which is not dependent on excitation wavelengths.

Figure 6 shows high-speed laser shadowgraphs and EEM images of a diesel spray flame at lowered ambient oxygen concentration of 10%, which corresponds to engine operation condition with exhaust gas recirculation (EGR). Lowering the ambient oxygen concentration causes elongated ignition delay of 2.9 ms. The high-speed laser shadowgraphs shows that the soot formation in the spray flame is significantly reduced by lowering the oxygen concentration down to 10%. The EEM images show that the LIF is observed primarily in a wavelength region shorter than 400 nm, which is attributable to relatively smaller PAHs, despite the late measurement timings such as t=4.0 ms. Although not shown in the figure, it was also verified that the smaller PAHs are already detected well before ignition, around t=1.2 ms, by more instantaneous single-shot EEM measurements. Therefore, the emission wavelength of the LIF do not significantly vary with the location in the spray and throughout the combustion duration. These results show that smaller PAHs formed in the spray before ignition do not grow into larger PAHs and soot particles throughout the following combustion duration under the ambient oxygen concentration of 10%.

The EEM analyses of soot formation processes in diesel spray flame were also conducted at different ambient temperatures. The results showed that the formation and growth of soot precursors can be delayed or suppressed by lowering the ambient temperature. Figure 7 summarizes the results of the EEM measurements of diesel spray flames at different ambient temperatures. In order to provide an overview of the effect of ambient temperature on the soot formation process, Figure 7

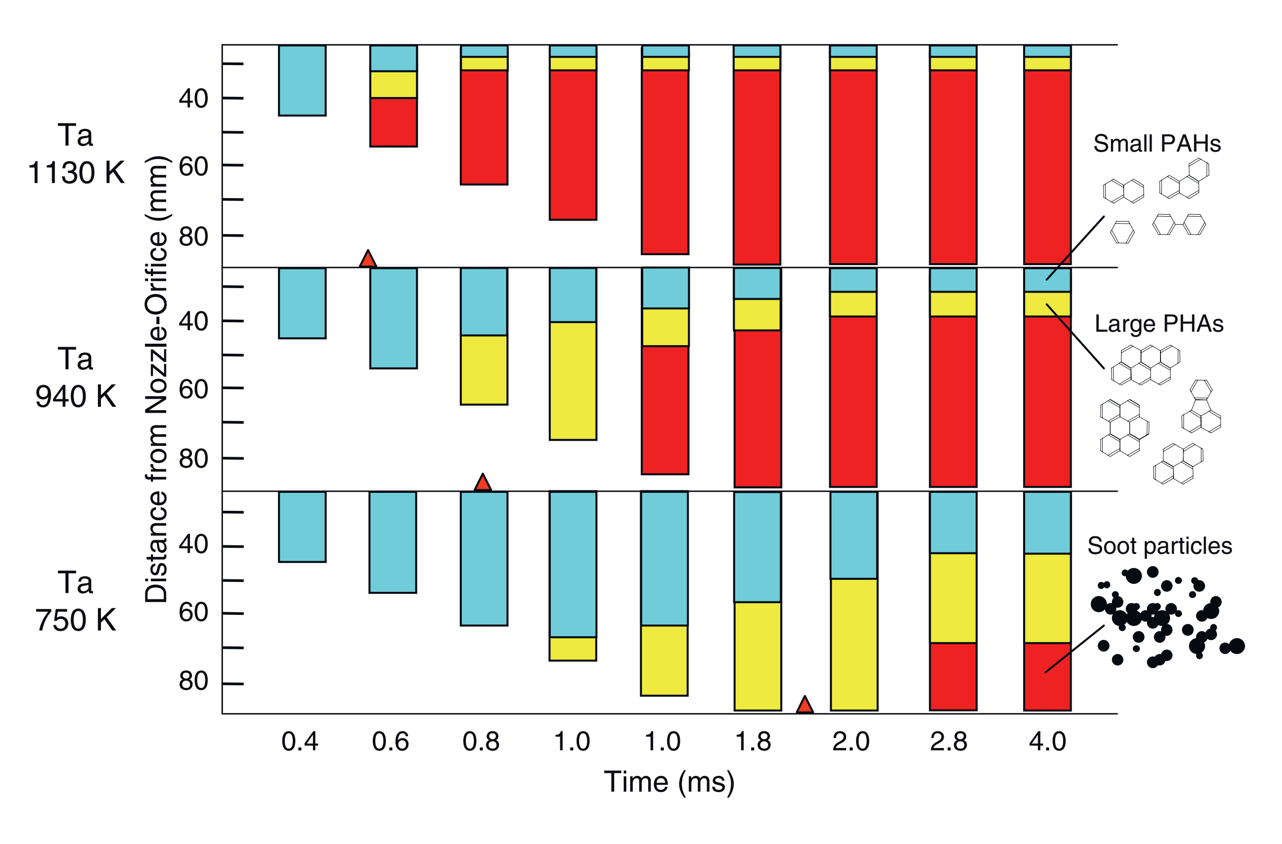

Figure 7 Soot formation process in a diesel spray flame at different ambient temperatures

conceptually illustrates the soot formation process in a diesel spray flame by dividing it into three stages; small PAHs, large PAHs and soot particles. Figure 7 clearly shows that the timing and region, when and where PAH growth and soot particle formation are observed, are delayed and extended downstream by lowering the ambient temperature.

Conclusions

In order to investigate the early soot formation processes in diesel combustion, spectral analysis of polycyclic aromatic hydrocarbons (PAHs) formed in the early soot formation region in a diesel spray flame was conducted via newly-developed excitation-emission matrix (EEM) technique using a multi-wavelength laser source. The EEM measured in diesel spray flames at different ambient temperatures and oxygen concentrations revealed that the formation and growth of soot precursors can be delayed or suppressed by lowering the ambient temperatures and oxygen concentration. The experimental data gained in the present research, including species, relative abundance and spatial distribution of different PAHs in the spray flame, are unique and difficult to be gained by other conventional experimental techniques. The final goal of the present research is, however, not only to investigate the soot formation processes, but also to investigate temperature and equivalence ratio in the soot formation region and soot oxidation processes in a diesel spray flame using the laser-spectroscopic diagnostic techniques, and then to integrate all these information into a fast and simple soot model for practical emission prediction. Application of such model to real-time engine control will enable the development of highly efficient and clean next generation engine system and thus will contribute to the mitigation of global warming and energy problems.

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Tetsuya Aizawa
Lecturer in Residence
Doctor of Mechanical Engineering
Department of Mechanical Engineering Informatics
Meiji University