

Measurement of two-phase hydrocarbon spray flows using laser plasma spectroscopy

S.H. Lee¹, J.J. Yoh²

¹Division of Physical Metrology, Korea Research Institute of Standards and Science,

267 Gajeong-ro, Yuseong-Gu, Daejeon 34113, South Korea

²Department of Mechanical and Aerospace Engineering, Seoul National University,

1 Gwanakro, Gwanakgu, Seoul 08826, South Korea

E-mail (corresponding author): seokhwan.lee@kriss.re.kr

Abstract

Performance and emission characteristics of direct injection (DI) engines are greatly influenced by many factors such as the local fuel/air equivalence ratio and condensed-fuel concentration. Picturing the inhomogeneous distributions of the flow properties is essential in understanding the ignition processes in two phase hydrocarbon spray flows. Simultaneous laser ignition and spectroscopy is a scheme that enables rapid determination of the local equivalence ratio and condensed fuel concentration during a reaction in two phase spray flows. In parallel with laser ignition, the equivalence ratio and droplet characteristics such as the concentration, size, and distribution of hydrocarbon spray flows are simultaneously obtained for a feedback control system. The plasma characteristics of fuel droplets are evaluated initially by shadowgraph, and the high-speed imaging of air and spray breakdown provides visualization of the transition from the plasma to a flame kernel. The flow fields of the spray are obtained using the time-resolved PIV (Particle Image Velocimetry) method during the laser ignition. The spectrum in the spray is evaluated according to droplet characteristics such as size and number density. The probability density function is used to analyze the interaction between the fuel droplets and the laser plasma with laser-induced breakdown spectroscopy (LIBS) measurements. In this research, we have conducted quantitative analysis of the LIBS signals according to the equivalence ratio, droplet size, droplet number density and droplet concentration for development of a control strategy for flame ignition and stabilization with simultaneous in situ two-phase hydrocarbon flow diagnostics.

Keywords: two-phase hydrocarbon spray flows, laser induced plasma, droplet, laser induced breakdown spectroscopy (LIBS)

1. Introduction

A variety of optical techniques have been used previously to measure the local equivalence ratio, including infrared (IR) absorption, planar laser-induced fluorescence (PLIF), and Raman scattering. Spontaneous Raman scattering and coherent anti-Stokes Raman spectroscopy can provide species concentration and gas temperature/density. However, the Raman signal is very weak and is thus susceptible to fluorescence/emission interferences. Furthermore, the complexity of pulsed Raman scattering measurement may limit its application in laboratory environments. Alternatively, laser-induced breakdown spectroscopy (LIBS) has high potential in combustion applications due to its high emission intensity and minimal system complexity. Ferioli et al. [1] used LIBS on engine exhaust gas to illustrate the ability of this technique to measure the equivalence ratio of SI engines. Do et al. measured fuel concentration and gas density simultaneously in a supersonic wind tunnel using LIBS [2]. In [3], a two-dimensional LIBS was proposed as a meaningful diagnostic tool for flame analysis.

Measuring fuel properties such as equivalence ratio and liquid phase fuel volume fraction at possible ignition and/or flame residence locations in a SI engine is key for executing a feedback control strategy since the properties can potentially suggest optimal ignition/stabilization locations under harsh combustor conditions. Focused laser energy can also be used as a tool for successful ignition and to aid flame enhancement at preferred locations. In mostly gas phase, the laser ignition and spectroscopic measurement have been conducted in reacting flows. The characteristics of laser-induced breakdown and spectrum in two phase flow are vastly different from those in the gas phase [4]. Understanding of the interaction between laser-induced breakdown and fuel spray is needed for laser ignition and quantitative measurement of fuel concentration. Therefore, the primary objective of this study is to investigate flame ignition processes in two-phase hydrocarbon fuel spray with simultaneous measurements of equivalence ratio and liquid phase fuel concentration. An unprecedented investigation of laser-induced ignition is conducted in the spray flame with high-speed imaging in conjunction with quantitative and simultaneous flow property measurements using LIBS. Three atomic and molecular emission line intensities (H- α (656 nm), O (777 nm) and

C_2 (516 nm)) are monitored, which potentially provide better accuracy in the species concentration measurements. The idea behind the present study is a combination of laser ignition and spectroscopy for feedback control of fuel injection. This is a desirable scheme since such real time information onboard an DI engine for instance can be constantly monitored and fed back to the control loop to improve the mixing process and minimize emission of unwanted species and combustion instability, preventing the degradation of vehicle performance.

2. Experimental method

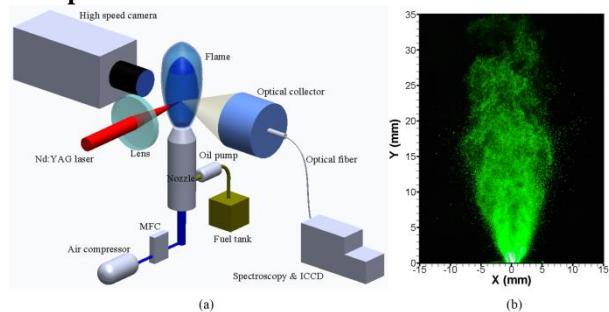


Figure 1: (a) Experimental setup, (b) scattering image of the droplet.

Figure 1(a) shows the experimental setup for the laser ignition and laser-induced plasma spectroscopy while 1(b) shows a scattering image of the spray. A spray of gasoline droplets was mixed with air at the nozzle tip. The spray nozzle was a Delavan SN 30609-2 air-assisted siphon nozzle. The fuel was directed to the nozzle through a pipe inside the flow channel. A second pipe supplied the nozzle with an atomization air flow, which impacted the fuel jet just before the nozzle exit. The droplet diameter distribution was between 20 and 50 μm at the nozzle exit. The fuel was a commercial gasoline obtained from the local gas station. The fuel flow rate was fixed to 10 ml/min by the oil pump. Compressed air was injected into the nozzle through a MFC (Mass Flow Controller). The air flow rate was 10 L/min and the equivalence ratio at the nozzle exit was 8.9.

For laser ignition and diagnostics, a Nd:YAG laser (Continuum, Surelite I) at a wavelength of 1064 nm and 5 ns pulse duration was used. The beam was focused through a 100 mm convex lens with 100 mJ laser energy to generate the plasma. The emission spectra were collected at the 1 μs delay time after laser irradiation. For visualization of the plasma and flame kernel, a high speed camera (Phantom v711) was arranged perpendicular to the laser beam path and used to observe the breakdown from a sideway perspective as shown in Fig. 1 (a). It provided broadband luminosity images at a sustained repetition rate of 390 kHz, an exposure time of 2 μs and a resolution of 128 x 64 pixels. The field of view was determined to be 20.9 x 10.5 mm with a calibration target. The plasma light was collected by a quartz lens with a 100 mm focal length, which was perpendicular to the direction of the laser used for the laser-induced plasma spectroscopy. The collected plasma light was sent to an echelle spectrometer (Andor Mechelle 5000) with 0.1 nm

resolution and an ICCD camera (Andor iStar) to record the signal. The delay time and ICCD exposure time were 1 μs and 50 μs , respectively. Several peaks ($H : 656 \text{ nm}$, $O : 777 \text{ nm}$, $C_2 : 516 \text{ nm}$) were selected for analysis of the spray. The shadowgraph method was used to obtain the droplet size and distribution with high magnification. Nd:YAG laser (minilite, continuum) was diverged through a concave lens having a focal length of 30 mm. The diverging beam was collimated by a convex lens having a 500 mm focal length. Passing through the droplet stream, the collimated beam was focused by a convex lens having 100 mm focal length. The laser beam was captured by a CCD camera (Nikon) through the magnification lens. The measurement size was 300 $\mu\text{m} \times 300 \mu\text{m}$ which had 1470 x 1470 pixel size. If a droplet is present in the test section, the laser beam was strayed from its direction as to allow shadowgraph imaging. The magnification was adjusted by changing the distance between the collecting lens and camera. The real droplet diameter and number density were extracted from image processing such as median filter, Binarization, and edge detection methods. The accuracies of the measured droplet diameter and number density were about 5% and 20%, respectively. The relatively high error of number density was originated from the unsteady feature of the spray. The plasma volume was also evaluated by the same shadowgraph technique.

The flow fields in the gasoline droplet stream were obtained using the PIV (Particle Image Velocimetry) method. Gasoline droplets and a dual pulsed Nd:YAG laser (Minilite, Continuum Inc.) were used as the seed particles and illuminating light source, respectively. The images showing the illumination of the gasoline droplets was captured by a high-speed CCD camera (Phantom v711). The delay time between the dual laser pulses was 5 μs . For vector processing, a cross-correlation algorithm based on the fast Fourier transform is applied, and the interrogation windows with 32 x 32 pixels overlapped as much as 50%.

3. Results

Figure 2 shows the sequences for the temporal development of laser-induced air breakdown and gasoline spray ignition. All images in the sequence are averages of the recordings from five individual breakdowns using in-house matlab code. The raw image was converted to an emission intensity contour for comparison of air and spray. The focal point in air and spray was at the (0, 0) position, and the laser beam was irradiated from the right side to the left. The breakdown was initiated at a location slightly before the focal point of the lens, in the region with the highest emission propagating toward the incoming laser beam. First, the air breakdown gradually adopted a bimodal appearance. The emission of air breakdown disappeared after about 50 μs . The second column in Fig. 2 shows average images of the spray ignition. The third row shows three lobes along the path of the laser beam before the focal point in the spray ignition. The first and second lobes resulted from local breakdowns initiated by fuel droplets. The third lobe resulted from a cluster of small

breakdowns. This effect was also observed by Kawahara et al. [5] and is caused by droplets acting as micro-lenses. The emission intensity of lobes in the spray is higher than in the air, since the laser beam is absorbed by the fuel droplets. The three lobes became blurred and fibrous after 7.8 μ s in spray. The transition from plasma to a flame kernel was observed at this time. The emission intensity was observed at about 100 μ s. This observation is in agreement with ref. [4].

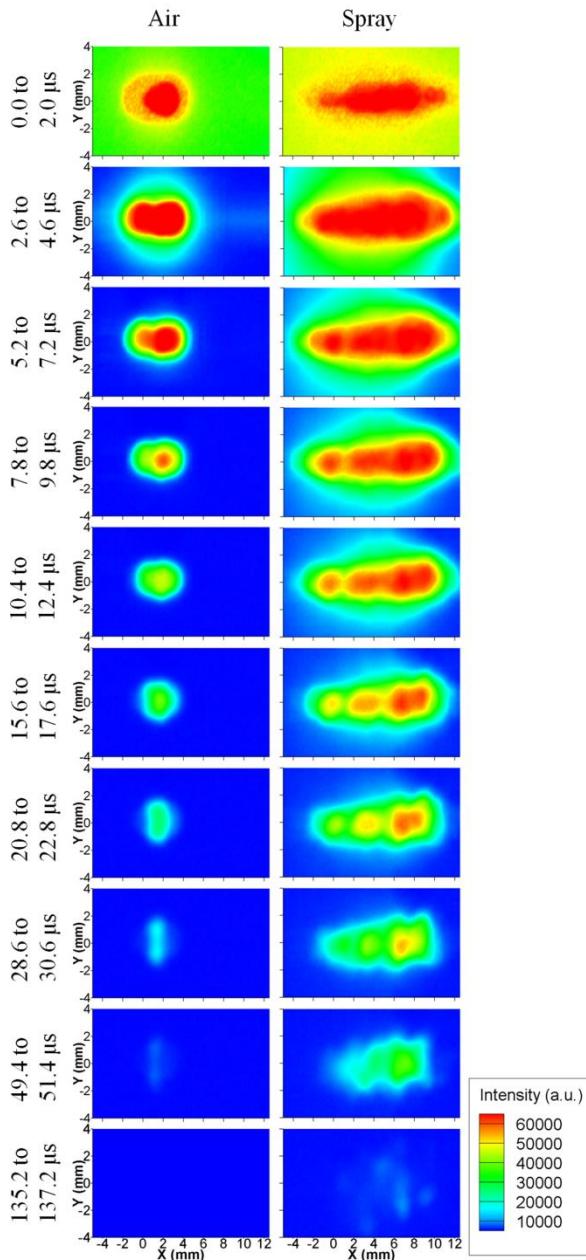


Figure 2: Sequences of the temporal development of laser-induced air breakdown (left column) and gasoline spray ignitions (right columns).

Figure 3 shows the mean intensity of high speed images for air breakdown and spray ignition. The emission intensity was averaged for 5 individual images. The emission intensity of air breakdown decreased rapidly within 10 μ s. Afterwards, the intensity decreased slowly and approached the detection limit after about 30 μ s, which is in agreement with the above mentioned study

[4]. The intensity for spray ignition decays exponentially over time. The intensity of plasma light decreases rapidly and the transition from plasma to a flame kernel occurs. Chemiluminescence is generated in the flame kernel from combustion radicals. So, the emission intensity of the spray ignition lasts for 150 μ s. This tendency is similar to published results [4], but the transition from plasma to flame kernel with gasoline droplets was observed for the first time in this study. The size of the gasoline fuel droplet easily decreases due to the high volatility of the fuel. So, the emission intensity is high in the initial flame kernel and is maintained for a long time due to the high ignitability of gasoline fuel.

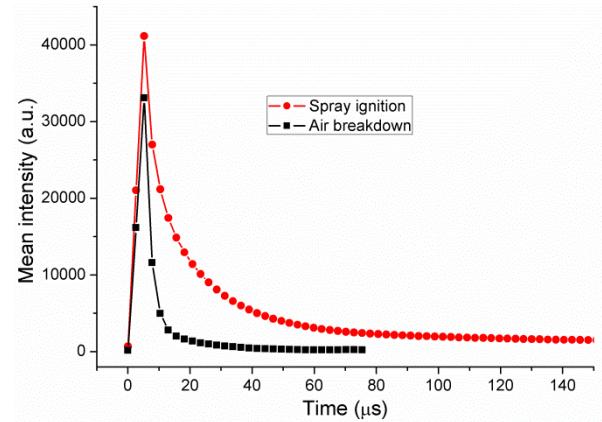


Figure 3: The mean intensity of high speed images for air breakdown and spray ignition.

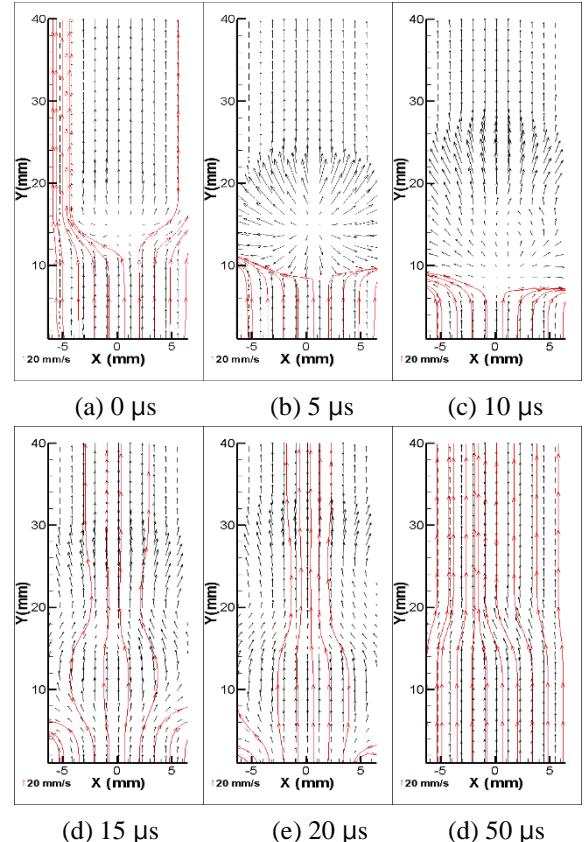


Figure 4: Temporal flow velocity fields according to the delay time after laser-induced air breakdown.

Though the high pressure and temperature of the laser induced a breakdown to achieve the laser ignition, they generate a shock wave that can disturb the flow field. This disturbed flow field has to be refreshed for accurate LIBS measurement. The refresh time scale of the flow fields is determined by the flow velocity and the laser repetition rate. Figure 4 shows the temporal flow velocity fields and streamlines according to the delay time after generating laser-induced breakdown. A stream flows from the lower side to the higher side, and the average flow velocity of the original stream is about 3.35 cm/s. When laser-induced breakdown is generated at the focused spot (0 mm, 15 mm), the flow spreads in all the directions as shown in Fig. 4 (a) and (b). The laser induced breakdown is considered as a source with a short time duration. The stagnation point moves towards the downward direction as the delay time increases (0–10 μ s). After 10 μ s, the original stream starts to recover as the source flow weakens. The original stream is almost recovered at a delay time of 50 μ s, as shown in Fig. 4 (d). This means that the flow fields are ready to obtain the next LIBS signal after a delay time of more than 50 μ s. Therefore, our LIBS measurement has sufficient refresh time (100 μ s) according to current experimental conditions (flow velocity: more than 5 cm/s, laser repetition rate: 10 Hz).

When the focused laser beam is irradiated onto the spray, the probability of a droplet at a specific position depends on the droplet size and the number density. If a droplet exists at the measurement position, the density at the measurement position is much higher than without a droplet. The density is related to the plasma intensity, which is proportional to the LIBS base intensity. So, the probability of droplet existence at the measurement position can be determined using the LIBS base signal. Figure 5 (a) shows the laser induced plasma spectra for the C₂ signal at a height of 10 mm in spray for different base intensities (low, middle and high). Figure 5 (b) shows the laser induced plasma spectra for H and O signals at a height of 10 mm in spray for different base intensities (low, middle and high). The base intensity was averaged from 320 nm to 350 nm. Differences were observed in the spectra even when the laser irradiated the same position. The base intensity increased if a fuel droplet was present at the measurement position. Also, the C₂ signal in the presence of a droplet (high base intensity) was about 5 times higher than in the absence of a droplet (low base intensity) as shown in Fig. 5 (a). When the laser beam irradiated the gasoline droplet, fragmentation of the droplet occurred, resulting in the C₂ signal. On the other hand, the difference in H intensity in the presence or absence of a droplet was much lower than the difference in the C₂ signal intensity as shown in Fig. 5 (b). Also, the difference in O intensity in the presence or absence of a droplet is very low. This means that the H and O signals were barely related to the existence of a droplet.

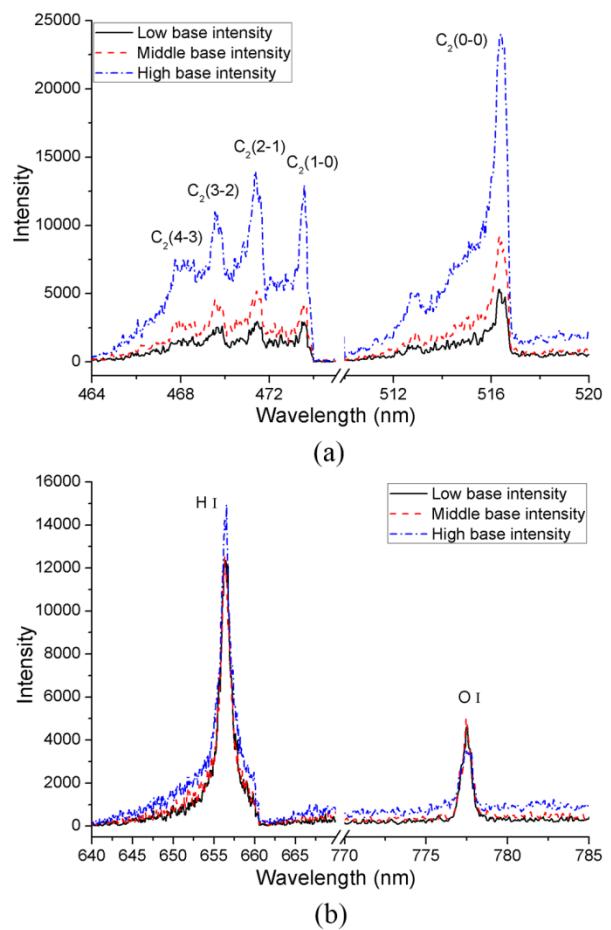


Figure 5: (a) C₂ spectra, (b) H, O spectra at 10 mm height in spray for different base intensities.

We found that the C₂ signal was related to the size of the droplet and the number density in spray. Therefore, the relation between the droplet characteristics (size and number density) and the LIBS signals was investigated using PDF analysis. Figures 6 (a) and (b) show the PDF of the droplet diameter and that of the C₂/base intensity ratio from laser induced plasma at a height of 10 mm, respectively. The C₂/base ratio can provide the liquid phase fuel concentration regardless of droplet existence. Also, normalization to the base signal can reduce measurement errors such as fluctuation of laser energy. Figures 6 (c) and (d) show the probability density function of the droplet diameter and that of the C₂/base intensity ratio from laser induced plasma at a height of 15 mm, respectively. Figure 6 (e) and (f) show the probability density function of the droplet diameter and that of the C₂/base intensity ratio from laser induced plasma at a height of 20 mm, respectively. The number density of the droplets in plasma volume was 275, 267 and 209 at 10 mm, 15 mm and 20 mm, respectively. The droplet diameter decreased with increasing height in Fig. 6 (a), (c) and (e) and the C₂/base ratio also decreased with increasing height in Fig. 6 (b), (d) and (f). The droplet diameter and the C₂/base intensity ratio have a Gaussian distribution, indicating that the C₂/base ratio is related to the droplet diameter. When the laser is focused on a droplet, the C₂/base ratio is proportional to the droplet size. Thus measuring the C₂ signal leads to the information about the droplet size or number density. The

signal is a weighted average of the droplet diameter in the plasma volume. When the laser induced plasma is generated, the droplet in the constant plasma volume evaporates completely due to the high laser energy.

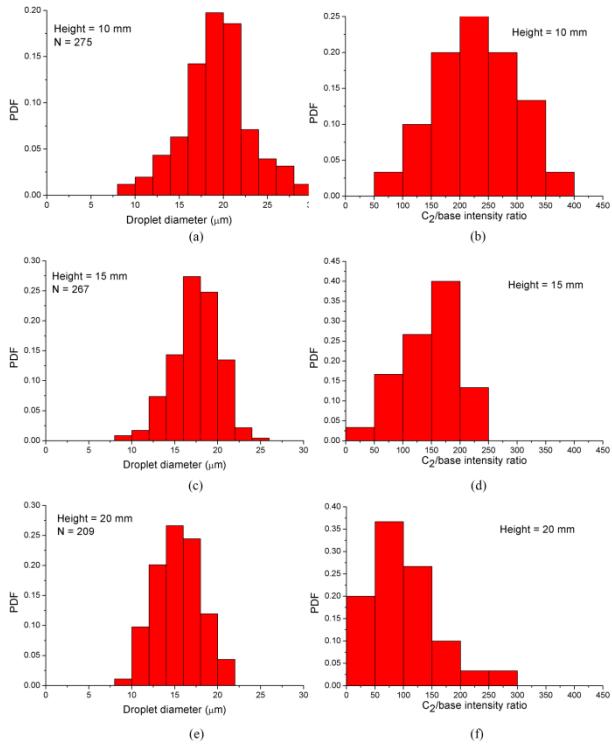


Figure 6: (a) Probability density function of the droplet diameter for different heights (a) 10 mm, (c) 15 mm and (e) 20 mm, probability density function of the C_2/base intensity ratio for different heights (b) 10 mm, (d) 15 mm and (f) 20 mm.

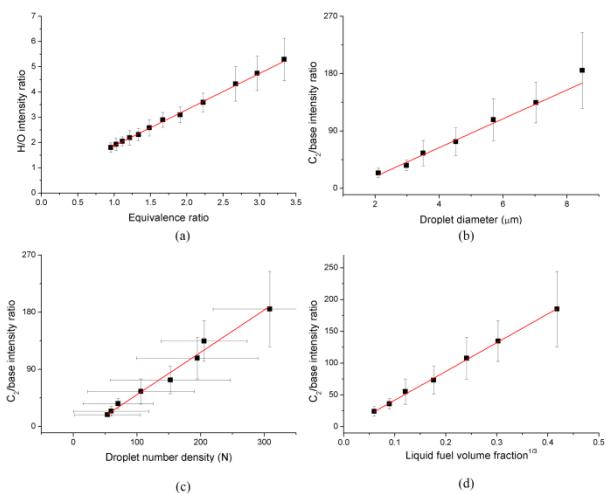


Figure 7: Calibration curves (a) H/O ratio according to the equivalence ratio and (b) C_2/base intensity ratio according to the fuel droplet diameter (c) C_2/base intensity ratio according to the fuel droplet number density and (d) C_2/base intensity ratio according to the cube root of liquid fuel volume fraction.

Quantitative analysis of the spray was conducted in a uniform droplet stream to obtain an equivalence ratio and liquid fuel concentration. Figure 7 (a) shows the H/O LIBS intensity ratio according to the equivalence ratio. The H/O intensity ratio was almost linear in accordance

with the equivalence ratio. The H and O signals originated from the hydrocarbon fuel and air, respectively. Previous works have demonstrated that the H/O ratio is usually linear according to the equivalence ratio in gaseous fuel [1-3]. Though droplets exist in the flow, the H/O ratio remained linear according to the equivalence ratio in this study. Thus, we confirmed that the H/O intensity ratio could be used to obtain the equivalence ratio in the two-phase flow condition.

Figure 7 (b) and (c) show the C_2/base intensity ratio according to the fuel droplet diameter and the C_2/base LIBS intensity ratio according to the droplet number density in a uniform droplet stream, respectively. The C_2/base ratio is almost linear according to the droplet diameter and the droplet number density. The fuel droplet volume was calculated from the droplet diameter obtained with the shadowgraph. Figure 7 (d) shows the C_2/base intensity ratio according to the cube root of the liquid fuel volume fraction. The liquid fuel volume was obtained by multiplying the number density and the droplet volume. The liquid fuel volume was then divided by the constant plasma volume, which was 0.0108 mm^3 . Interestingly, the C_2/base intensity ratio has a linear relation with the cube root of the liquid fuel volume fraction. In other words, the intensity (I) of the molecular band signal from laser induced plasma is proportional to the cube root of the liquid fuel volume (V_{fuel}). This relation is defined as $I \propto (V_{\text{fuel}})^{1/3}$. This confirms that the equivalence ratio and the liquid fuel concentration can be obtained simultaneously from the laser induced plasma during ignition.

Figure 8 (a) and (b) show the equivalence ratio and liquid fuel volume fraction for three different heights along the radial direction for a spray. The calibration curves of Fig. 7 (a) and (d) were used to quantify the equivalence ratio and the fuel volume fraction, respectively. The diffusion of the fuel along the radial direction is shown for different heights in Fig. 8 (a). The atomization and evaporation of the fuel droplet for different heights can be observed in Fig. 8 (b). The equivalence ratio and droplet characteristics affect the ignition, extinction, stability behavior, combustion efficiency, and pollutant emissions such as CO, soot, and NOx. These two parameters can be used to monitor fuel injection systems utilizing laser ignition.

Simultaneous laser ignition and spectroscopy is a scheme that enables rapid determination of the local equivalence ratio and the condensed fuel concentration during a reaction. Using this technique, one can thoroughly investigate the influence of the measured flow properties on the spray flame ignition/stabilization processes and possibly, transient flame propagation into the internal engine. Also, the experimental data sets taken under various operation conditions can provide a practical guidance for the design of actual engine, e.g., optimizing fuel injection rate/location and ignition location. This is a desirable scheme since such real time information onboard an engine for instance can be constantly monitored and fed back to the control loop to improve the

mixing process and minimize emissions of unwanted species and combustion instability, preventing a degradation of engine performance.

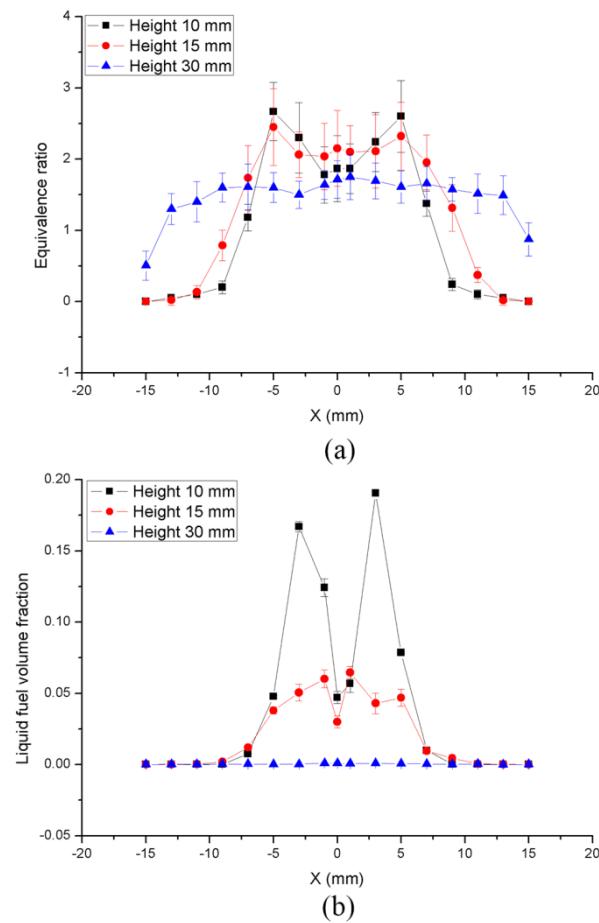


Figure 8: (a) Equivalence ratios and (b) liquid fuel volume fraction along the radial direction for different heights (10 mm, 15 mm and 30 mm).

4. Conclusion

Simultaneous laser ignition and laser-induced plasma spectroscopy on a spray flame were employed for development of a control strategy. The plasma characteristics were evaluated by visualization of the spray. Based on the shadowgraph imaging of the plasma within 1 μ s of the pulse irradiation showed that the gasoline droplets absorbed the laser energy along the path of the beam, and the generated plasma propagated toward the incoming laser beam. The transition from the plasma to the flame kernel in fuel spray was then visualized by high speed imaging. The plasma emissions in the spray were replaced by a much weaker combustion chemiluminescence. Small breakdown along the laser beam path occurred in the spray due to a micro-lens effect from the fuel droplets. The intensity of the air breakdown decreased rapidly whereas the intensity of the spray breakdown decreased steadily, becoming invisible to the camera after 150 μ s, which is 3 times slower than the air breakdown. The flow fields of the spray are obtained using the time-resolved PIV method during the laser ignition. The refresh time scale is obtained in our experimental conditions for accurate LIBS measurement.

For laser-induced breakdown spectroscopy, the probability density function of the LIBS signal was obtained to detect the fuel droplets in sprays, which are highly stochastic. We found that the C₂/base intensity ratio was highly related to the size and the number density of the fuel droplets. The H/O intensity ratio and C₂/base intensity ratio were used to obtain the equivalence ratio and the liquid fuel concentration in the spray, respectively. The quantitative equivalence ratio and liquid fuel volume fraction were obtained from the laser irradiation, which also causes ignition in the spray. The present results can be utilized for a future feedback controller that uses a single laser source for both optical breakdown in spectroscopy and subsequent ignition of the two-phase spray. The quantitative and simultaneous measurements ultimately provide practical guidance of (i) optimizing flow conditions for initiating/sustaining combustion reactions in a spray flame, (ii) placing an ignition source at optimal location, and (iii) determining minimum energy requirement for ignition in a fuel spray. Therefore, a novel feedback control strategy for flame ignition and stabilization simultaneously with in-situ combustion flow diagnostics onboard can be developed via the present study.

References

- [1] F. Ferioli, P. V. Puzinauskas, and S. G. Buckley, “Laser-induced breakdown spectroscopy for on-line engine equivalence ratio measurements”, *Appl. Spectrosc.*, **Vol. 57**, pp. 1183-1189, 2003.
- [2] H. Do, C. D. Carter, Q. Liu, T. M. Ombrello, S. Hammack, T. Lee, and K.-Y. Hsu, “Simultaneous gas density and fuel concentration measurements in a supersonic combustor using laser induced breakdown”, *Proc. Combust. Inst.*, **Vol. 35**, pp. 2155-2162, 2015.
- [3] S. H. Lee, H. T. Hahn, and J. J. Yoh, “Towards a two-dimensional laser induced breakdown spectroscopy mapping of liquefied petroleum gas and electrolytic oxy-hydrogen flames”, *Spectrochim. Acta Part B: At. Spectrosc.*, **Vol. 88**, pp. 63-68, 2013.
- [4] G. C. Gebel, T. Mosbach, W. Meier, and M. Aigner, “Optical and spectroscopic diagnostics of laser-induced air breakdown and kerosene spray ignition”, *Combust. Flame*, **Vol. 162**, pp. 1599-1613, 2014.
- [5] N. Kawahara, K. Tsuboi, and E. Tomita, “Laser-induced plasma generation and evolution in a transient spray”, *Opt. Express*, **Vol. 22**, pp. A44-A52, 2014.