Two-dimensional measurement of 
hydrocarbon fuel concentration using multiple 
laser-induced plasma-forming regions

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Abstract: A two-dimensional measurement of fuel distribution in a gasoline spray flow was performed using multiple laser-induced plasma-forming regions. Multiple plasma-forming regions were generated by a laser sheet with a low breakdown threshold for a two-phase flow. To observe the formation of multiple laser-induced plasma-forming regions, shadowgraphs were imaged using a high-speed camera. Hydrogen and oxygen atomic emissions from the plasma-forming regions were obtained by attaching bandpass filters to the high-speed camera, and a two-dimensional visualization of the fuel distribution in the wide plasma-forming region was obtained by dividing the hydrogen line-filtered image with the oxygen line-filtered image. The result complements a novel method for two-dimensional measurement of instantaneous fuel concentration in the reacting flow by utilizing laser-induced breakdown spectroscopy (LIBS).

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1. Introduction

To ensure a stable combustion in engines fueled by liquid hydrocarbon, it is important to control properties such as flame structure, temperature, pressure, flow velocity, and fuel distribution. The measurement of these properties in the flow of air-breathing combustion engines including jet engines and scramjet engines, is performed using devices such as pitot tube, hot wire anemometer, thermocouple, and gas chromatograph [1,2]. However, even if the physical probes could endure the extreme conditions inside a combustor, they cannot be inserted into it without interrupting the flow [3]. To solve these problems and overcome the limitations of existing measurement probes, an optical means was suggested to perform such measurement roles inside the combustors [4].

To analyze the flame structure, flow velocity, concentration of species, density, temperature, and equivalence ratio, methods such as planar laser-induced fluorescence (PLIF), particle image velocimetry (PIV), Raman spectroscopy, laser Doppler velocimetry (LDV), and laser-induced breakdown spectroscopy (LIBS) have been applied to combustor diagnostics [5]. By applying LIBS inside a supersonic combustor, simultaneous measurements of the concentration and gas density in harsh environments can be performed [6]. Previously published research has shown that only two peaks of an atomic spectrum are needed to estimate equivalence ratios [7,8]. Keiffer et al. has proven that H/O intensity ratio and H/N intensity ratio obtained from hydrocarbon and air mixtures have linear relations with equivalence ratio and constructed calibration curves [8]. Even though LIBS can provide various data, including the fuel distribution under diverse conditions, it can only provide such data for a single plasma at a time. In conventional LIBS, a laser beam is concentrated at one point to excite molecules and generate plasma. Additionally, a spectrometer and an intensified charge-coupled device (ICCD) are used to collect plasma emissions and provide spectra of atomic lines from a single-point plasma. Also, attempts have been made to analyze the two-dimensional characteristics of a single plasma [9,10], to image a two-dimensional elemental distribution by repeated laser-induced breakdowns [11,12], and to visualize a two-
dimensional gas-to-particle phase transition in flames using laser-induced nanoplasmamas [13]. Moreover, a line LIBS technique has been developed to measure the 1D fuel-to-air ratio in laminar and turbulent flames [14]. However, no studies have been done to observe the formation of multiple laser-induced breakdowns in a sheet region and apply them to the measurement of an elemental composition distribution.

In our previous work, we developed and tested a miniaturized device for fuel concentration and flame diagnostics, which we named ‘LIBS plug’ [15,16]. The device was intended to provide a novel feedback control strategy for flame stabilization with simultaneous in situ combustion flow diagnostics in a scramjet engine combustor. The LIBS plug was constructed using two photodiodes and two bandpass filters which pass either the H (656.3 nm) or the O (777 nm) atomic line. Using the LIBS plug, the construction of a calibration curve between the H/O intensity ratio and the equivalence ratio in both a single gas phase flow and a two-phase flow was performed. We also suggested methods of measuring the equivalence ratio inside a gasoline spray flow and performing flame diagnostics with the LIBS plug. In our previous study, we analyzed the laser-induced breakdown characteristics inside a two-phase flow [17], demonstrating that the two-phase flow experiences a drastically reduced breakdown threshold and multiple breakdowns in the laser path when a single laser beam is used. Since the plasma volume exceeded the size of fuel droplets, the signal of the atomic emissions from the plasma represented chemical compositions of both fuel droplets and air. Subsequently, the present research showed that the equivalence ratio at the focal point could be estimated using a laser-induced plasma even inside a two-phase gasoline flow.

This study proposes a measurement method for the two-dimensional hydrocarbon fuel distribution, using characteristics of a laser-induced breakdown in a two-phase flow, and specific lines of LIBS that are required to perform a fuel distribution analysis. To produce multiple plasma-forming regions in a specific region, a laser sheet was constructed using a high-energy pulsed laser. However, since the laser energy was not strong enough to create a full two-dimensional plasma sheet over the region of interest, and the threshold of the breakdown in liquid is significantly lower than gas, the plasma-forming regions were limited only near where the fuel droplets were present. Hence, 30 shots of images were averaged to compensate plasma-absent regions in each shot. The shadowgraph imaging performed by a high-speed camera confirmed the formation of multiple breakdowns. Two bandpass filters were sequentially located in front of the high-speed camera to collect either the H or the O atomic emissions from multiple plasma-forming regions. The attenuation of the plasma emission intensity was observed, which aligns with the results obtained using the conventional LIBS and the LIBS plug. The two-dimensional H/O intensity ratio was obtained by dividing the H-filtered image with the O-filtered image. To estimate the exact fuel-air ratio from the H/O intensity ratio, a calibration curve between two-dimensional H/O intensity ratio was obtained from multiple plasma regions, and the equivalence ratio was established from a uniform droplet stream. In the gasoline spray flow, the increase of the H atomic line emission and the decrease of the O atomic line emission were both observed two-dimensionally, according to an increasing gasoline flow rate. The direct linear relation between the H/O intensity ratio and the equivalence ratio was then applied to the non-uniform spray nozzle flow, to obtain the final distribution of two-dimensional fuel distribution. To validate our result, we compared it to the H/O intensity ratio obtained using the conventional LIBS system at the same position and flow condition.

2. Methods

2.1 Experimental setup

The main purpose of this work is to visualize the two-dimensional fuel concentration in a two-phase spray flow using multiple laser-induced plasma-forming regions. The multiple
plasma-forming regions were generated by an Nd:YAG laser and a high-speed camera was used to capture the plasma-forming regions.

The experimental setup is illustrated in Fig. 1(a). The Nd:YAG laser (Surelite III, Continuum) used to generate the laser sheet had the wavelength of 1064 nm and a pulse duration of 5 ns. To produce multiple plasma-forming regions using a single laser beam, the energy of the used laser was set to 968 mJ. Laser-induced plasmas were detected using a high-speed camera (HPV-X2, Shimadzu) with a recording speed of 10 million frames per second, which was positioned vertically from the focal point of the laser sheet. The exposure time of each frame was 50 ns and the interval of the frames was 100 ns. The high-speed camera provided a resolution of 400 x 250 pixels, and a field of view was measured to be 65.1 x 40.7 mm. To collect the H (656.28 nm) and the O (triplet at 777.194 nm, 777.417 nm, and 777.539 nm) atomic lines of the plasmas, the bandpass filters (656FS10-50 and 777FS10-50, Andover) with center wavelengths (CWL) of 656 nm and 777 nm and a full width at half maximum (FWHM) of 10 nm were located in front of the high-speed camera in each procedure. The acquired shadowgraph images verified the multiple laser-induced breakdowns. A continuous wave (CW) laser with a wavelength of 532 nm was used as an illumination source, while neutral-density filters and a bandpass filter (532FS03-50, Andover) centered at 532 nm were used to prevent the appearance of a plasma emission. A bandpass-filtered image and the shadowgraph image of the multiple plasma-forming regions are shown in Figs. 1(b) and 1(c), respectively.

To validate the results obtained using multiple laser-induced breakdowns, the results were compared to those obtained from a conventional LIBS experiment, where a LIBS system was used in the same flow field. To generate a single plasma, a laser energy of 100 mJ was used in combination with a plano-convex lens. The plasma emission was collected using an optical collector and fed to the spectrometer (Andover Mechelle 5000) and ICCD (Andor iStar). For measurements, a gate delay time of 400 ns and a gate width of 2 μs were selected, and the H/O intensity ratio was calculated for each point of the spray flow.

![Fig. 1. (a) Experimental setup, (b) H-filtered image of the multiple laser-induced breakdowns (delay time of 300 ns and exposure time of 50 ns), and (c) shadowgraph image of the multiple plasma-forming regions (delay time of 1 μs and exposure time of 50 ns).](image)

### 2.2 Experimental procedure

The atomization of liquid gasoline was done by a siphon nozzle (Delavan 30609-2). An attached dosing pump (Simdos 10, KNF) and a pulsation damper (FPD 10, KNF) were used to control and provide a continuous fuel flow rate. The air flow rate was controlled by a mass flow controller (MFC, TSC-230, MKP) and the inlet flow temperature and pressure were set to 298.15 K and 101.3 kPa, respectively. Shadowgraph imaging and fuel distribution measurement were done in the siphon nozzle flow while a Bunsen burner flow was used to
construct calibration curve. To construct calibration curve between the H/O intensity ratio obtained from multiple plasma regions and the equivalence ratio, a uniform droplet stream was generated with ultrasonic vibrating plate nebulizer and sent to a Bunsen burner with a nozzle diameter of 12 mm. In this case, the laser sheet was irradiated right above the nozzle to avoid intervention of the laboratory environment. The experimental flow conditions are presented in Table 1.

Between the Nd:YAG laser and the flow, three cylindrical lenses were attached to form a planar laser sheet. The height of the laser sheet was 25 mm, and the focus was located 8 mm above the center of the siphon nozzle. To compare the breakdown characteristics of a single plasma-forming region and multiple plasma-forming regions, shadowgraph imaging of a single-point laser-induced plasma was also conducted by replacing the cylindrical lenses with a spherical lens. In the single plasma case, a lower laser energy was used because its plasma continuum emission was so intense that interference occurred during shadowgraph imaging. Five different experimental conditions were set, each having different flow rates of fuel, as presented in Table 1. To see the averaged fuel-to-air ratio distribution over the two-dimensional region, 30 shots of images were ensemble-averaged for each flow condition.

### Table 1. Flow conditions of experiments.

| Experimental condition | Shadowgraph imaging | Construction of calibration curve | Measurement of fuel distribution |
|------------------------|---------------------|-----------------------------------|---------------------------------|
|                        | Single plasma-forming region | Multiple plasma-forming regions | Multiple plasma-forming regions |
| Gasoline flow rate [mL/min] | 5 | 5 | 1.5–3.7 | 1.25–10 |
| Air flow rate [L/min] | 6 | 6 | 15 | 6 |
| Laser energy [mJ] | 200 | 968 | 968 | 968 |

3. Results and discussion

3.1 Multiple laser-induced plasma-forming regions

The shadowgraph images of the formation of multiple plasma-forming regions are presented in Fig. 2, where Fig. 2(a) shows images for a laser focused on a single point in air, Fig. 2(b) a laser focused on a single point in the spray flow, Fig. 2(c) a planar laser sheet focused in air, and Fig. 2(d) a planar laser sheet focused in the spray flow. In the images of Fig. 2, the laser beam is propagated from the right. In air, the laser beam focused on a single point produced a single plasma, forming a strong shock wave, whereas, the one in the gasoline spray flow produced several plasma-forming regions. Our previous research has already described the formation of multiple breakdowns in a two-phase flow [17]. The breakdown threshold for a two-phase flow decreased as the droplet size increased, and multiple plasma-forming regions were created along the laser beam path in front of its focal point. Similar results were obtained using a two-phase spray flow by Kawahara et al., who suggested that the droplet lens effect may explain the displacement of a laser-induced plasma [18]. To produce shadowgraph images for a planar laser sheet (Figs. 2(c) and 2(d)), the focal length of the last lens of the sheet optics was 300 mm and the energy of the laser beam was set to 968 mJ, significantly higher than the energy required for a single breakdown. Under these conditions, the region of the laser sheet, in which the laser energy density exceeds the breakdown threshold, becomes widely distributed. In Fig. 2(d), the formation of hundreds of plasmas can be seen together with their propagation and combination. However, in the case of air without droplets, this sheet laser beam cannot cause air breakdown because the breakdown threshold in air is considerably higher than it is in the gasoline spray flow.
The regions of plasma sheet formation with respect to certain gasoline flow rates are shown in Fig. 3. On average, multiple plasma were formed with a height of 20.0 mm and a width of 17.2 mm. The focal line of the laser sheet was located right above the center of the siphon nozzle, but most of the plasma-forming regions were in front of the focal line. When a laser beam induces a plasma, a large amount of energy is absorbed by the medium, which leads to an immense reduction in energy [19]. For this reason, it is assumed that, after multiple plasma-forming regions were generated, the laser irradiance beyond the breakdown threshold from the front side of the focal line loses its energy. As seen in Fig. 3, an increase in the gasoline flow rate of the spray is followed by the overall plasma region being slightly shifted forward, toward the direction of the incoming laser. This indicates that higher gasoline flow rates cause the breakdown to occur closer to the laser. Our previous study presented an analysis of the relation between the breakdown threshold and the droplet size [17], which demonstrated that the breakdown threshold decreases with an increase in the droplet size. This phenomenon was also observed in this experiment, with the droplet size increasing as the gasoline flow rate increased, resulting in the decrease of the breakdown threshold of the total flow. As the breakdown threshold decreased, the location of the breakdown region shifted in the direction of the incoming laser.
To confirm the formation and attenuation of multiple plasma-forming regions, we need to analyze the same manner, a comparison with the established plasma analysis method was conducted. The characteristics of a single plasma-forming region and multiple plasma-forming regions in a gasoline spray flow are presented in Fig. 4. The emission intensity of the single laser-induced plasma-forming region was analyzed using the conventional LIBS and the LIBS plug. In the conventional LIBS, its spectrum was recorded every 100 ns between 200 ns and 1000 ns of delay, with a gate width of 100 ns. The emission intensity of H (656.28 nm) and O (777.53 nm) atomic lines was then selected. When using the LIBS plug, aided by photodiodes and filters, the atomic signals of H and O were obtained every 4 ns. To counteract the interference of the plasma continuum emission, our previous works utilized a LIBS plug signal, which was selected after 250 ns and averaged to obtain H and O atomic emissions [16]. Similar results were reached for the multiple laser-induced plasma-forming regions by attaching bandpass filters in front of a high-speed camera. Images taken every 100 ns were averaged to obtain the emission intensity of the plasma-forming regions. Here, the intensities were averaged in the range between 310 ns and 2010 ns in the H and O images, thereby minimizing the intervention of the plasma continuum.

Fig. 4. Normalized emission signal intensity of (a) H (656.3 nm) and (b) O (777 nm), obtained using three different measurement techniques.
3.2 Measurement of two-dimensional fuel concentration

Figure 5 shows the calibration curve between the two-dimensional H/O intensity ratio obtained from multiple laser-induced breakdowns and the equivalence ratio of gasoline-air mixture. H and O line-filtered images of multiple laser-induced plasma regions formed in uniform droplet stream were obtained and divided to get a two-dimensional H/O intensity ratio. The laser sheet was focused right above the Bunsen burner and the analysis region had a width of 12.0 mm and a height of 15.2 mm on average. The equivalence ratio was changed from 0.83 to 2.06 by increasing gasoline volume flow rate. At each flow rate, averaged two-dimensional H/O intensity ratio was obtained and the H/O intensity ratio was ensemble-averaged to construct a mean value of H/O intensity ratio. The fitting curve was expressed with the red line in the figure and had coefficient of determination ($R^2$) of 0.9513.

![Graph showing calibration curve between H/O intensity ratio and equivalence ratio](image)

Fig. 5. Calibration curve between the H/O intensity ratio and the equivalence ratio.

The results of H and O line-filtered images of the multiple plasma-forming regions in the spray nozzle flow are shown in Figs. 6(a) and 6(b), respectively. With an increase in the gasoline flow, the H-filtered signal intensity of the multiple plasma-forming regions increases, while the O-filtered signal intensity decreases. The opposite behavior of the hydrogen and oxygen emission intensity at different flow rates has already been shown by other studies [7]. This result confirms that the laser-induced plasma provides localized data in a two-phase flow. The intensity of the central part of the plasma sheet was particularly high, which is naturally due to a Gaussian form of the laser itself. In the LIBS, it is necessary to normalize or divide the two-line signals to obtain a quantitative result because the variance in intensity of the laser energy is highly correlated with the plasma emission [20].

The image obtained by dividing the H-filtered image by the O-filtered image at each flow rate is shown in Fig. 6(c). Once the two images were divided, the effects of the variance in intensity of the laser sheet disappeared and a more precise distribution of the fuel was obtained. The H/O intensity ratio was then replaced to the equivalence ratio using the calibration curve in Fig. 6. As shown in Fig. 3, the center of the siphon nozzle is located 8 mm below the lower left of the region of the multiple plasmas. As this figure shows, the overall equivalence ratio increases with the increase in the flow rate, and the equivalence ratio increases when it is obtained closer to the nozzle exit.
Fig. 6. Averaged (a) H (656.3 nm) and (b) O (777 nm) filtered image of multiple plasmas in different gasoline flow rates and (c) two-dimensional image of the equivalence ratio.

To validate the results, the two-dimensional image of the H/O intensity ratio obtained by dividing each filtered image was compared to the H/O intensity ratio obtained using the conventional LIBS system at certain heights. Data from the conventional LIBS was obtained by shifting the point of measurement by 2 mm at a specified height. The H and O spectra obtained using the conventional LIBS at the height of 15 mm from the siphon nozzle are shown in Fig. 7. As the radial distance increased, the intensity of the H atomic line decreased and the intensity of the O atomic line increased. The H/O intensity ratio decreased in the radial direction.

The data obtained by both the plasma sheet measurement and the conventional LIBS system is summarized in Fig. 8. In both cases, the gasoline flow rate was 10 mL/min and the H/O intensity ratio was obtained at heights of 10 mm, 15 mm, and 20 mm above the nozzle. As each pixel of the high-speed camera acted as a detector for the laser-induced plasma sheet, the H/O intensity ratio was obtained at 210 points between the radial distance of 0 mm and 30 mm at each above-mentioned height. With the conventional LIBS system, the plasma locations were chosen at an interval of 2 mm from the center of nozzle at each height. The results show that the H/O intensity ratio obtained by both the conventional LIBS and the filtered imaging of the laser-induced plasma sheet show a similar scale and characteristics. The overall tendency of the H/O intensity ratio to decay in the radial direction was observed in the results of both methods, and the fuel of the spray flow was seen spreading when the height of the plasma increased.
4. Conclusion

Multiple plasma-forming regions were generated in a two-phase flow and used to measure the two-dimensional fuel distribution in a gasoline spray flow. Multiple plasma-forming regions were obtained by enlarging the area of the laser sheet having the irradiance above the breakdown threshold of the liquid, which is substantially lower than the air. To determine the distribution and characteristics of the multiple plasma-forming regions, a high-speed camera was used for the shadowgraph imaging which confirmed the plasma formation. The collected bandpass-filtered images of the multiple plasma-forming regions led to detection of the H and O atomic line emissions. The distribution of H/O intensity in a spray nozzle flow at each flow
rate was confirmed by dividing the data from the H-filtered image by that of the O-filtered image, and then converted into the equivalence ratio, using the calibration curve acquired from the uniform droplet stream. The results were validated by comparison with those of the conventional LIBS system applied to the same flow condition. The H/O intensity ratio obtained from both the filtered plasma sheet imaging and LIBS at the same location was similar in scale and maintained the observed spreading behavior of the fuel in the spray flow. Thus, the present technique advances the instantaneous measurement of the two-dimensional distribution of fuel in the combustion chamber of liquid-fueled engines.

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**References**

1. B. T. Vu and F. C. Gouldin, “Flow measurements in a model swirl combustor,” AIAA J. 20(5), 642–651 (1982).
2. G. Cox, “Gas velocity measurement in fires by the cross-correlation of random thermal fluctuations-a comparison with conventional techniques,” Combust. Flame 28, 155–163 (1977).
3. S. Tavoularis, *Measurement in fluid mechanics*, (Cambridge University, 2005), p. 193.
4. A. C. Eckbreth, *Laser diagnostics for combustion temperature and species*, vol. 3. (CRC Press, 1996), p.1.
5. K. Kohse-Höinghaus, R. S. Barlow, M. Aldén, and J. Woffram, “Combustion at the focus: laser diagnostics and control,” Proc. Combust. Inst. 30(1), 89–123 (2005).
6. B. McGann, C. D. Carter, T. M. Ombrello, S. Hammack, T. Lee, and H. Do, “Gas property measurements in a supersonic combustor using nanosecond gated laser-induced breakdown spectroscopy with direct spectrum matching,” Proc. Combust. Inst. 36(2), 2857–2864 (2017).
7. P. Stavropoulos, A. Michalakou, G. Skevis, and S. Couris, “Quantitative local equivalence ratio determination in laminar premixed methane-air flames by laser induced breakdown spectroscopy,” Chem. Phys. Lett. 404(4–6), 309–314 (2005).
8. J. Kiefer, J. W. Tröger, Z. S. Li, and M. Aldén, “Laser-induced plasma in methane and dimethyl ether for flame ignition and combustion diagnostics,” Appl. Phys. B 103(1), 229–236 (2011).
9. Y. L. Chen, J. W. L. Lewis, and C. Pariggar, “Spatial and temporal profiles of pulsed laser-induced air plasma emissions,” J. Quant. Spectrosc. Ra. 67(2), 91–103 (2000).
10. P. Gregorčič, J. Diaci, and J. Možina, “Two-dimensional measurements of laser-induced breakdown in air by high-speed two-frame shadowgraphy,” Appl. Phys. A 112(1), 49–55 (2013).
11. 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 B At. Spectrosc. 88, 63–68 (2013).
12. J. H. Yang, S. J. Choi, and J. J. Yoh, “Towards reconstruction of overlapping fingerprints using plasma spectroscopy,” Spectrochim. Acta B At. Spectrosc. 134, 25–32 (2017).
13. Y. Zhang, S. Li, Y. Ren, Q. Yao, and C. K. Law, “Two-dimensional imaging of gas-to-particle transition in flames by laser-induced nanoplasmas,” Appl. Phys. Lett. 104(2), 023115 (2014).