Towards simplified monitoring of instantaneous fuel concentration in both liquid and gas fueled flames using a combustor injectable LIBS plug

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ABSTRACT

In this study, instantaneous measurement of fuel concentration and flame diagnostics in both liquid and gas hydrocarbon fueled flame is presented using a simplified laser-induced breakdown spectroscopy (LIBS) approach. The newly developed device or hereinafter the "plug" receives specified lines from the plasma emission without resorting to the conventional LIBS system. Bandpass filters and photodiodes were mounted in the plug to capture hydrogen (656 nm) and oxygen (777 nm) atomic lines. Since H/O intensity ratio has a linear relationship with fuel concentration, the plug's calibration curve between equivalence ratio and H/O intensity ratio is constructed. Single phase and two-phase hydrocarbon fuel fields were measured with the device using gasoline (liquid) and LPG (gas). With the advantage of simplicity of the plug, multiple plasma points and plugs were simultaneously applied in the fuel field and showed possibility of using a single laser device to construct multi-points concentration mapping. Also, flame diagnostics scheme was proposed based on the observed difference of intensity duration between reactive and non-reactive fuel fields.

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1. Introduction

Fuel concentration and flame distribution have a close relationship with stable combustion in an operating engine. Especially in high-speed combustors, because of their unpredictability and susceptibility, measuring instantaneous flow characteristics without interrupting the flow has been one of the biggest issues of interest in the current combustion studies [1]. To overcome disadvantages of existing measurement methods, such as physical interruption of instruments and slow response time, optical measurement techniques have been developed and adopted as combuster diagnostics [2-4]. Especially for high-speed engines such as scramjet engines, studies related to fuel injection and mixing are major concerns since the flow residence time in combustor is of the order of a few milliseconds [5]. In order to promote and verify fuel mixing in scramjet engines, both experimental and computational studies including designing of cavity flame holders have been conducted and identified their flow behaviors [6]. However, as these methods are aimed at predicting the flow inside a combustor, a real-time diagnosis of fuel distribution inside an operating combustor was not possible. Furthermore, in a highly turbulent flow, experimental measurements or computational predictions were not reliable due to unaccountable flow dynamics variables. This limitation calls for the need for instantaneous measurement inside the actual engines.

This study proposes a method of instantaneous detection of fuel concentration in either liquid or gas phase hydrocarbon fuel for flame diagnostics using a simplified laser-induced breakdown spectroscopy (LIBS) device. LIBS has been widely used to instantaneously detect various elements in severe environments utilizing a simple system configuration. A concentrated laser excites molecules and generates plasma at one point. Excited ions and electrons return to their ground states as the plasma cools down, and emit light with respective atomic frequencies. In the conventional LIBS system, plasma emission is collected with spectrometer and ICCD camera and analyzed to give whole spectra of atomic lines for measuring properties such as density and the equivalence ratio of hydrocarbon fueled flames [7]. Also, it is known that H/O intensity ratio obtained from LIBS has a linear relationship with equivalence.
ratio for given temperature and calibration curves were made [8–10].

Even though these earlier studies were aimed at understanding the hydrocarbon reacting flow, the conventional LIBS system had limitations in determining the fuel properties of the environment inside the real operating engine. As those aforementioned studies are mainly focused on constructing and validating relationships between hydrocarbon fuel distribution and LIBS signals, the present study is aimed at developing a simplified instantaneous measurement device that is intended to be installed in an operating combustor. To reduce the excessive volume of the configured spectrometer and ICCD camera, only a set of specific spectral signals was extracted from plasma. As H/O intensity ratio showed linear relationship with equivalence ratio, only H and O signal intensities were selected. The LIBS plug consisted of lens, two bandpass filters for hydrogen as well as oxygen, and two photodiodes. Plasma emission obtained from the plug is immediately changed to electric signal from photodiodes and was transmitted to an oscilloscope. This direct transmission of signal intensity made the delay time designation procedure simple. The main purpose of the study is the instantaneous detection of hydrocarbon-air mixture in both reacting and inert environment using the LIBS plug. The instantaneous detection is especially attractive in high-speed air-breathing engines while providing a simultaneous feedback control for performance enhancement. Such concept is aimed at advancing the field of combustion where the fuel mixing and flame stabilizing at a high-speed condition are still considered challenging. In this study, calibration curves between the plug’s signal and equivalence ratio of gasoline-air mixture and LPG-air mixture were newly established. Analysis of two-phase spray fuel field and single phase fuel field was done with our LIBS plug with gasoline and LPG as the target fuel, respectively. The simple construction of our LIBS plug enabled simultaneous measurements at multiple points. The simultaneous four-point measurements of the fuel distribution were done with an extended experimental setup using 4 plugs, which was not possible with the conventional LIBS setup. This brought a new possibility in application of LIBS in the two-dimensional flame diagnostics. Finally, using a continuous signal of the LIBS plug, identifying how long the plasma continuum emission lasts became possible. Collecting data immediately after plasma continuum emission in just a single shot was possible with a new LIBS plug. This is, not possible by using conventional LIBS setup, since it collects the accumulated intensity along ICCD exposure time. Also, since the decay time of signal intensity changed under two distinct flow conditions, it was possible to identify the difference of the plug’s signal between reacting and non-reacting flow.

2. Experimental setup

2.1. Conventional LIBS setup

Fig. 1 shows conventional LIBS system setup. Nd:YAG laser (Surelite III, Continuum) with wavelength of 1064 nm and pulse duration of 5 ns was used to generate plasma. Plasma emission was collected with optical collector and optical fiber and then sent to spectrometer (Andor Mechelle 5000). ICCD camera (Andor iStar) collects separated lights with respect to its wavelength. In reacting flow, the conventional LIBS setup was used to validate LIBS plug’s signal. Liquid gasoline was sprayed with air by a siphon nozzle (Delavan 30609–2). Spray angle of the siphon nozzle is 40° at 15 cm above the nozzle. The fuel flow rate was controlled with dosing pump (Simdos 10, KNF) with an accuracy within ±2% and is fixed at 0.17 cm³/s. A pulsation damper (FPD 10, KNF) was attached to remove pulsation and provide continuous transfer of the flow. Air flow rate was controlled with Mass Flow Controller (MFC, TSC-230, MKP) which has accuracy of ±1% and is fixed at 166.67 cm³/s.

2.2. LIBS plug implementation and setup

The simplified LIBS plug was designed and produced in this study, a device to acquire data on hydrocarbon fuel distribution and flame without using the bulky spectrometer and ICCD configuration, necessary for a conventional LIBS experiment. In our previous study, the concept of LIBS plug was validated with photodetectors and the initial test device was developed [11]. Though a possibility of the LIBS plug was demonstrated through a rough setup of the previous study, the enhanced LIBS plug’s design was performed to acquire more accurate signal. In this follow-up research, the LIBS plug is presented and all data shown here are obtained with these small sized LIBS plugs. Two lens tubes (SM05V05, Thorlabs) were installed in the plug and its components were sealed in the tubes. Single lens tube had a diameter of 17.8 mm and a length of 26.2 mm. As other components were fixed inside, these two lens tubes determined the total size of the plug. Since H and O atomic lines from LIBS show a strong correlation with hydrocarbon fuel properties, the LIBS plug includes two bandpass filters which pass H atomic line (656.28 nm) and O atomic line (triplet at 777.194 nm, 777.417 nm, 777.539 nm) respectively. Fig. 2 shows the schematic of the plug setup. Bandpass filter (656FS10–12.5, Andover) with center wavelength (CWL) of 656 nm and full width at half

![Fig. 1. Conventional LIBS setup.](image1)

![Fig. 2. A small-sized LIBS plug setup.](image2)
maximum (FWHM) of 10 nm was used to obtain H signal while a filter (780FS10–12.5, Andover) with CWL of 780 nm and FWHM of 10 nm was used to obtain O atomic signal. Both filters have a diameter of 12.5 mm. Two Si photodiodes (FDS010, Thorlabs) are installed in the plug. Their rise time is 1 ns and peak responsivity is 0.44 A/W at 730 nm. Each photodiode was connected to an oscilloscope (WAVESURFER 54MXS-B, Teledyne LeCroy) with BNC cable to get signal voltage in real time. Two N–BK7 plano-convex lenses (LA1074, Thorlabs) with a 12.7 mm diameter were installed in the plug to collect plasma emission for each photodiode while one UV silica plano-convex lens that has a wavelength range of 185 nm–2.1 μm and a focal length of 75.3 mm with a diameter of 50.8 mm (LA4078, Thorlabs) was used to concentrate incident laser beam and produce plasma.

2.3. Experimental procedure

Fig. 3 describes the overall experimental procedure. To construct an accurate calibration curve between the plug’s H/O intensity ratio and equivalence ratio of liquid gasoline, a uniform droplet stream of liquid gasoline was generated with ultrasonic vibrating ceramic plate nebulizer with 1.65 MHz frequency. The LIBS plug’s signal is analyzed in this setup. Then the fuel stream was mixed with air and sent to Bunsen burner with a diameter of 12 mm. The droplet diameter and plasma shape of our setup were measured in our previous research [12]. The air flow rate was fixed at 333.33 cm³/s while the fuel flow rate ranged from 0.02 cm³/s to 0.15 cm³/s to control equivalence ratio. For the calibration curve of gas LPG, the fuel flow rate was fixed at 3.33 cm³/s while air flow rate varied from 31.33 cm³/s to 313.33 cm³/s. LPG flow rate was controlled with MFC (TSC-220, MKP) which has accuracy of ±1%. Mixture conditions including equivalence ratio are shown in Table 1. In both cases, plasma was generated directly above the Bunsen burner to avoid influence of the laboratory environment. Since gasoline and LPG are both mixtures of hydrocarbons, equivalence ratio was calculated based on each fuel’s representative species. Properties of octane and propane were used since the representative species of gasoline is octane and LPG’s major component is propane.

Fuel concentration in cold flow is analyzed with the plug in both two-phase flow and single phase flow. The same siphon nozzle from the conventional LIBS setup were used to generate a two-phase gasoline stream. The flow rate of gasoline fuel and air was also identical with the conventional LIBS setup. A single phase gas flow was generated with LPG gas and air. Premixed LPG gas and air mixture was sent to the Bunsen burner with different flow rates. In this case, laser beams with wavelength of 532 nm was used instead of 1064 nm. The laser beam was generated by the second harmonic generator and the aforementioned Nd:YAG laser. All these experiments were done using laser energy of 100 mJ. The plug’s advantage of simplicity and cost-effectiveness enabled the plug setup to perform a multi-point measurement. Beam splitters and mirrors were used to generate four plasmas at the same time from one incident laser beam. Two plasmas were made 50 mm above the siphon nozzle and the other two plasmas were placed vertically above the two plasmas below. The distance between the plasmas was 40 mm each. Incident laser energy was 430 mJ while the final four laser energies were 124, 107, 110 and 90 mJ respectively. Four plugs were prepared to collect each plasma emission. The difference between energy of each laser beam resulted in each plug signal’s intensity difference. However, since an H/O intensity ratio was obtained from the data, those energy differences were negligible.

Reacting flow was generated by igniting gasoline spray field. Gasoline and air flow rate was also fixed in this setup and the flow was ignited. Since the density of the reacting flow was significantly lower than that of the cold flow, higher laser energy was needed because of the increasing breakdown threshold energy [13]. 300 mJ of laser energy was used to form plasma continuously. The plasma was made 50 mm directly above the siphon nozzle. Table 2 present conditions of targeted flows, and Fig. 4 shows measurement regions. Temperature and pressure of inlet flow were 298.15 K and 101.3 kPa respectively.

3. Results and discussion

3.1. Analysis of the LIBS plug’s signal

The plug’s signals of the plasma emission in different equivalence ratios according to H and O are shown in Fig. 5 (a) and (b) respectively. Each line indicates the average of 40 individual signals and the incident laser pulse energy was fixed at 100 mJ. From the beginning of the plasma generation, instantaneously rising emission of light from both H and O signals was identified. These projecting signals indicate strong plasma background continuum. Atomic signals are seriously interfered by plasma background continuum since the continuum is a broadband emission. Since plasma background continuum lasts more than 100 ns, a signal after 250 ns was used to avoid interference of plasma continuum emission even though an atomic signal appears after 10 ns of plasma generation [14]. Liquid gasoline flow rate changed from 0.08 cm³/s to 0.15 cm³/s to form equivalence ratio from 2 to 4 while air flow rate was fixed at 333.33 cm³/s. Since the LIBS signal of
specific atomic spectra is proportional to the number density of the atom [15], increasing intensity of H signal with the increasing fuel flow rate from the plug’s signal was identified. In Fig. 5 (a), change in the signal of H was observed with different gasoline fuel flow rate. However, as shown in Fig. 5 (b), O signal shape showed uniformity because the same amount of air was injected in each experiment.

Fig. 6 shows the calibration curve between H/O intensity ratio and the equivalence ratio of the gasoline-air mixture using the plug. H/O intensity ratio was calculated in two steps. First, the plug’s signal of H and O averaged between 250 ns and 1000 ns. Then the averaged H signal intensity was divided by the average of O. The equivalence ratio was changed from 0.5 to 4 by increasing gasoline volume flow rate. Inlet flow temperature and pressure were used to calculate gasoline and air densities since no interruption was made to the flow. The fitting curve based on the experimental values was expressed with the red line in the figure and it had coefficient of determination (R²) of 0.9959. The analysis for the conventional LIBS signals in the gasoline spray using a spectrometer and ICCD camera was also done in our previous study [12]. Although uniform droplet stream was formed with ultrasonic nebulizer, fluctuations of two-phase flow appeared between each shot. The fluctuation of H/O intensity ratio in each point increased further in rich condition. The standard deviation increased to 2.97% of the average value when equivalence ratio was 3 whereas it was 1.49% when the equivalence ratio was 0.5.

### 3.2. Fuel concentration in cold flow

#### 3.2.1. Liquid phase gasoline spray flow

Once our calibration curve was made, it was shown that the plug has a linear relationship with the equivalence ratio of two-phase hydrocarbon fuel flow. Fig. 7 shows equivalence ratio for different heights from the nozzle in each radial location for gasoline spray obtained with the single plug. H/O intensity ratio obtained with the plug was converted into equivalence ratio by applying data to the calibration curve. The height of the plasma from the spray nozzle changed from 8 mm to 40 mm and radial distance changed.
from –20 mm to 20 mm with the interval of 2 mm from the center of the nozzle. From Fig. 7, spreading fuel was captured with the plug. Equivalence ratio from the center radial distance dropped significantly when the height of the plasma changed from 8 mm to 16 mm. However, as the height of the plasma increased further, the difference of equivalence ratios at the center radial distance was not clearly observed. Around the center of the nozzle at radial distance from ±2 mm to ±4 mm, signals gradually decreased as the plasma height increased.

With the advantages of the simplicity and small volume of the plug, multiple-plug setup was applied to the distribution of gasoline spray field. Fig. 8 shows the four-plug H/O intensity ratio simultaneously obtained. Single laser pulse was divided into four laser pulses by beam splitters and mirrors. Each laser beam became plasma with four plano-convex lenses. Plasmas 1 and 2 were located at the height of 90 mm from the siphon nozzle while plasmas 3 and 4 were at the height of 50 mm. The distance between plasmas 1 and 2 was 40 mm and the distance between 3 and 4 was the same. A set of plasma moved along radial distance with 2 mm and moved until the total distance reached 40 mm. Each plug’s signal was normalized by its averaged H/O intensity ratio obtained from air. Although each plug’s H/O intensity ratio had a linear relationship with equivalence ratio, differences were observed among signals of plugs. They appeared because of the minute difference of distance and focusing point of each plug. However, after normalizing each plug’s signal, similar appearance and interpretation of spray distribution characteristics were obtained.

3.2.2. Gas phase hydrocarbon-fueled flow

One of the advantages of LIBS is that its outcome is independent from the phase of the target fuel. Although aircraft engines use a liquid fuel inside the combustor, because of their high temperature, most of the fuel evaporate in a short time. Since our LIBS plug is based on the laser-induced breakdown, the plug can detect fuel and air concentrations without being interfered by the phase of the flow. With the following experiments, the LIBS plug shows that it is also applicable to a single gas phase fuel flow. The LPG–air mixture and Bunsen burner were used in this experiment to construct gas phase flow. Fig. 9 shows the calibration curve between H/O intensity ratio and the equivalence ratio of the LPG–air mixture using the plug. H/O intensity ratio was calculated with the same procedure above. The equivalence ratio was changed from 0.25 to 2.5 by increasing the air flow rate. The fitting curve based on the experimental values is expressed with the red line in the figure and it had a coefficient of determination ($R^2$) of 0.9971.
Fig. 10 shows equivalence ratio obtained from LPG-air mixture stream along the various radial distances at different heights with different flow rates of (a) 3.33 cm³/s of LPG with 83.33 cm³/s of air and (b) 10 cm³/s of LPG and 250 cm³/s of air. To obtain the difference of flow characteristics that only occurred by the difference of the flow rate, the same fuel and air ratio were used. While our gasoline-based experiments were done in a two-phase condition, this experiment was done in a single gas phase condition. Fig. 10 (a) and (b) show that fuel distribution follows the vertical exit radius of the Bunsen burner when the flow rate is low, while fuel expands further when the flow rate is high. Results from these experiments show that the plug can measure the equivalence ratio regardless of the phase of the target fuel flow.

3.3. LIBS plug detection in reacting flows

The plug was also implemented in the reacting flow, which was created simply by igniting the same gasoline spray used in the above experiments. The plasma was located 50 mm directly above the siphon nozzle and the laser energy was 300 mJ, which was three times higher than that in the above experiments. In the reacting flow, because of the lower density, higher laser pulse energy is needed to make the stable plasma [13]. To compare reacting and non-reacting flows, the same energy level was used here. The plug’s signal showed different characteristics in the reacting flow. A slower decay of intensity in the H signal of the plug was observed. In air or cold flow, decaying signal intensity was considered as plasma continuum and excluded from data processing procedure. Fig. 11 (a) and (b) show H and O signals from the LIBS plug in gasoline spray droplet stream and flame. Their peak values were normalized since the difference of density and plasma shape brought disparity to their signal intensity. As shown in Fig. 11 (a), H signal inside flame stabilized after about 600 ns while, in droplet stream, H signal stabilized after about 250 ns. However, in case of O signal, both signals are stabilized approximately after 250 ns. It is investigated that temperature affects in broadening of atomic emission lines and stark broadening causes hydrogen line to broaden more than other atomic lines [16]. This establishes the stabilization time of the H/O intensity ratio. Fig. 12 (a) and (b) show plug’s H/O intensity ratio stabilization time in non-reacting flow and reacting flow. In non-reacting flow, signal intensity ratio after 250 ns was averaged since it was stabilized after the moment. However, in the reacting flow, signal intensity ratio before 600 ns was excluded from the calculation since its fluctuation had been continued. This phenomenon was validated by constructing ensemble image of LIBS spectra with the conventional LIBS system. The conventional LIBS spectrum was obtained in every 100 ns delay time until 1200 ns and the gate width in the experiment was fixed at 100 ns. Fig. 13 (a) and (b) show ensemble of the H and O LIBS spectra according to delay time in spray flame. Each spectrum shows an average of ten shots of laser pulse. In the figure, H signal is

![Fig. 10. Equivalence ratio for different heights (10 mm, 40 mm, 70 mm) from the burner in radial location for LPG-air mixture stream at different flow rates. (a) 3.33 cm³/s of LPG and 83.33 cm³/s of air (b) 10 cm³/s of LPG and 250 cm³/s of air.](image1)

![Fig. 11. LIBS plug signals of (a) H and (b) O in droplet stream and gasoline spray flame.](image2)
relaxed after 600 ns and shows distinct peak value while O signal is relaxed in a relatively short delay time. In H signal, before 600 ns of delay time, plasma continuum interrupts H signal to appear clearly. Such an interruption was expressed as continuum extension in the plug’s signal since bandpass filters were used in the plug. In conventional LIBS experiments, to avoid influence of fluctuating plasma continuum, delay time had to be regulated to get relaxed atomic signal. However, the plug made it possible to observe the accurate time of plasma continuum emission disappearance. The signal after inflection point indicates non-interrupted peak signal of H. Since the plug’s signal expresses intensity in real-time, the immediate and the fastest collection of non-interrupted peak signal was possible. Also, since plasma continuum in the flame hinders H atomic signal for much longer than plasma continuum outside the flame does, it is possible to know whether the flow is reacting or not just by comparing the inflection points of the signal. This is an indication that such instantaneous flame diagnostics is suitable for an operating engine.

4. Experimental uncertainty

In this research it was possible to receive the desired atomic signal by installing bandpass filters in front of photodiodes. However, since the bandpass filter passes a certain range of wavelength according to its bandwidth, the plug’s signal presents total intensity of the light corresponding to the range. This also causes plasma continuum interruption problem since the filter’s FWHM is 10 nm. In the follow-up research, a narrow band filter is utilized in the device for obtaining the exact peak value.

Another uncertainty can occur in the experiment because of the fuel condition. Since our experiments were done with commercial grade gasoline and LPG, impurities might have been mixed with the fuel in the process of distribution. Also, fuels were assumed as monotypic substance of main ingredient when calculating the equivalence ratio even though these fuels are combination of diverse hydrocarbons. Using refined fuel in due course of the research can further bring the accurate results including calibration curve of the target fuels.

5. Conclusion

In this paper, a miniaturized LIBS device was designed and
tested. The device was composed of lens, bandpass filters and photodiodes. This LIBS plug has the purpose of being installed in multiple locations inside operating engines. The bandpass filters with 656.3 nm and 780 nm of CWL were implemented in the plug to obtain the H and O atomic signals. The plug can detect the hydrocarbon fuel distribution in either single phase (gas) or two phase (spray) flow fields. The calibration curve between H/O intensity ratio and equivalence ratio on gasoline was made, and the plug was applied to the gasoline spray as well as the LPG-air premixed flow. By using a combination of beam splitters, mirrors, and four plugs simultaneously, a four-point H/O intensity ratio measurement was conducted. Although the conventional LIBS system was a single-point measurement technique, the present approach provides a multiple-point measurement. Also, since the plug is free from designating particular delay time and gate width, the analysis on a single shot result is possible. This is particularly attractive in terms of reducing any effect of plasma continuum emission or atomic line broadening as the inflection point of the signal clearly indicates where the atomic signals appear. By comparing the decay time of the plug’s signal, the region of reactive flow can be identified from the non-reactive region. Therefore, the present study suggests the real-time fuel property analysis and flame diagnostics within operating air-breathing engines by utilizing the multiple LIBS plugs.

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