



## Laser induced breakdown spectroscopy for local equivalence ratio measurement of kerosene/air mixture at elevated pressure

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### ABSTRACT

The quantitative local equivalence ratio measurements of premixed kerosene/air gas at elevated pressure by the laser induced breakdown spectroscopy (LIBS) technique were performed in a high temperature test cell, which was designed and manufactured for simulated supersonic combustion state. According to in-situ experimental conditions in supersonic combustion, the correlations between the values of emission intensity ratio of H/O, H/N and the equivalence ratio of premixed kerosene/air mixture in different gas pressures was established. The dependence of the FWHM of H<sub>α</sub> on pressure of the mixture was also measured. Therefore, the laser diagnostic method for quantitative local equivalence ratio measurements of kerosene/air mixture by the LIBS technique in supersonic combustion was developed in laboratory. The fundamental data was prepared for on-line quantitative local equivalence ratio measurements of kerosene/air mixture in supersonic combustion experiments.

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### 1. Introduction

In combustion, fuels must be mixed into air in an appropriate ratio to release more energy. The mixed ratio of fuel and air (the equivalence ratio,  $\Phi$ ) determines the efficiency of combustion and the generation of pollutants when it affects the ignition, transitions and stability of combustion. Therefore, measurement of the local equivalence ratio is essential in maintaining the stability of the combustion process, as well as in minimizing soot emissions. It is also a key factor to keep appropriate equivalence ratio in combustion chamber for supersonic air-breathing engines, in which chemical reactions takes place in the supersonic gas flow. Hitherto, the accurate measurement for the distribution of hydrocarbon fuels is still one of the most critical keynotes in SCRAMjet that is the soul of supersonic vehicle. Therefore, the development of optical diagnostic techniques, which could measure fuel–air ratio accurately owns a great significance for the studying of high efficiency engine system and SCRAMjet.

Laser induced breakdown spectroscopy (LIBS) is a powerful diagnostic method that can be applied to combustion systems for measurements of fuel–air ratio, fuel composition and temperature. In this technique, a sufficiently high energy laser pulse is employed to create local plasma by focusing into a small volume.

The high peak power laser pulse breaks chemical bonds and initializes the fragmentation of molecules into elemental components in the focus. Then, high temperature plasma is produced while the atoms dissociated from target molecules are excited. The emission of these excited atoms in their relaxation process can be recorded and analyzed to determine the elemental compositions of sample qualitatively and quantitatively. In LIBS, if the plasma is optically thin and in local thermodynamic equilibrium (LTE), the intensity of spectral emission line has a linear correlation with the population of the atomic excited state, as the following formula:

$$I = CNh\nu \exp\left[-\frac{E}{kT}\right] \quad (1)$$

where  $I$  is the intensity of the spectral emission line,  $N$  is the population of the excited state of atomic species,  $h\nu$  is the energy of single photon of excited laser,  $E$  is the excitation energy of the atomic species,  $k$  is the Boltzmann constant,  $T$  is the absolute temperature and  $C$  is the constant which relates to instrument parameters, element property and other factors. In Eq. (1),  $C$ ,  $h\nu$ ,  $E$ ,  $k$  and  $T$  are all constants for the specific spectral emission lines and other given experimental conditions. That is to say, the intensity of the spectral emission line varies directly with the population of the atomic excited state in certain conditions. In LIBS experiment, the relative intensities of atomic spectral lines are often employed to get the qualitative information of the atomic species with keeping from effects of instrument parameters and the jitter of laser energy. For instance, the ratio

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between emission intensity of hydrogen and oxygen (H/O) or C/N in LIBS is usually used to get the equivalence ratio of hydrocarbon fuels within the probe volume.

The LIBS technique is one of the best tools for the fuel–air ratio (the equivalence ratio) measurements in combustion. It possesses many advantages, e.g. high temporal and spatial resolution, low requirements for environment, fast response time, non-intrusiveness and so on. It collects the emission of excited atoms, which is unaffected by the molecular state (e.g. burned or unburned gas), differing from other optical diagnostic techniques, which record the emission of excited molecules. Therefore, it can be applied to analysis of any matter regardless of its physical state, providing the possibility for real time multi-element analysis. Furthermore, the LIBS technique is particularly effective in harsh, hostile, toxic or dangerous environments and it has been employed in the planetary environment exploration [1–3]. For example, the LIBS technique was utilized to analyze the components of Mars soil by NASA in MSL project in 2009 [4]. Moreover, there are many studies that have been successfully performed for local equivalence ratio measurements of lower carbon number hydrocarbon fuels (e.g. CH<sub>4</sub> and C<sub>3</sub>H<sub>8</sub>) and air flames on low pressure premixed chamber by LIBS [5–10]. Experiments in which the laser induced plasma ignition and equivalence ratio measurements by LIBS were achieved simultaneously have also been carried out [11–13]. Additionally, some LIBS on-line investigations were accomplished. e.g. Rai et al. [14] have been applied LIBS technique to in-situ component analyze in combustion and in plume of a simulated CH<sub>4</sub>/air rocket. The quantitative local equivalence ratio measurements of methane/air in a standard motor by LIBS under laboratory conditions were also executed by Ferioli et al. [15].

However, most of the previous LIBS investigations in combustion for quantitative equivalence ratio measurements are limited in low pressure premixed combustion or in lower carbon number hydrocarbon fuels. The experimental data obtained from these investigations does not suit to kerosene, a perspective jet fuel in supersonic combustion. Since kerosene is a kind of mixed complex composed of many kinds of higher carbon number hydrocarbon fuels. Its breakdown threshold and spectra are greatly different from that of the lower carbon number hydrocarbon fuels like methane. Furthermore, it is well known that at elevated pressure, the spectral line profile, the emission efficiency and the lifetime of spectral line are all changed remarkably because of the intensified collision. The relative intensities of different spectral lines as well as the results of quantitative equivalence ratio measurements are also greatly affected. For these reasons, the LIBS experiments of kerosene in elevated pressure should be performed in detail before executing LIBS diagnostic for the supersonic combustion of kerosene. In addition, it is significant that kerosene is a complex hydrocarbon fuel mixture which owns strong regionalism. Taking jet fuel for instance, it contains hundreds of components like alkane, alkene, cyclohydrocarbon, aromatics and so on. Its exact components are usually varies with habitats, producers and vintages. Therefore, the LIBS investigation for the specific kerosene (e.g. the RP-3 model) is very necessary.

In this paper, the LIBS investigation of RP-3 kerosene was carried out in a high temperature elevated pressure test cell, which was designed and manufactured for simulated supersonic combustion state. The quantitative local equivalence ratio measurements of premixed kerosene/air gas by the LIBS technique were preformed. The correlations of values of emission intensity ratio (H/O and H/N) in LIBS with the equivalence ratio of kerosene/air mixture in different gas pressures were established. The calibration experiments of quantitative local equivalence ratio measurements of kerosene/air mixture in different gas pressures were also accomplished considering the pressure effect on the FWHM of H<sub>α</sub>.

## 2. Experimental apparatus

The used kerosene in present experiment is RP-3 kerosene. It is conventional national aviation kerosene, which is composed of many kinds of alkane, alkene, cyclohydrocarbon, aromatics, etc. In this kerosene, volume percentages of saturation hydrocarbons, aromatics and unsaturated hydrocarbons are 92.5, 7 and 0.5, respectively. The molecular formula of RP-3 kerosene closes to C<sub>11</sub>H<sub>22</sub>. The characteristic data of the specific kerosene was summarized in Table 1.

The experimental setup of quantitative local equivalence ratio measurements for kerosene/air mixture at elevated pressure by LIBS was illustrated in Fig. 1. As shown in Fig. 1, the LIBS measurements were performed in a stainless steel flow cell designed and machined by ourselves. The cell is a high temperature elevated pressure test cell equipped with optical accessible quartz windows whose transmission region is in the range of 200–3000 nm. The temperature of the cell could be varied from room temperature to 350 °C by hiring a temperature controller while the pressure could be adjusted in 1–4 atm region with a pressure sensor. A premixed gas sample containing a suitable ratio of vaporized kerosene and air supplied from the liquid vaporizer (Model: DLILS Turbo Vaporizer, BROOKS Instrument) through stainless steel pipes and controlled by individually calibrated mass flow controller (Model: Coriolis Mass Flow meter and SLA5851S Thermal Mass Flow meter, BROOKS Instrument) was slowly passed through the cell. The LIBS signals of premixed kerosene/air gas were generated from the excited atoms in the microplasma, which was created by focusing the second harmonic output of a Nd:YAG laser (Model: Quanta-Ray190, Spectra-Physics) at 532 nm. The plasma emission was collected by a lens system, passed through a fiber and then, focused into an echelle spectrometer (Model: Mechelle5000, Spectral range: 200–850 nm, Spectral resolution: 0.125 nm at 500 nm, Andor Technology) equipped with an ICCD camera (Model: iStar DH734, minimum optical gate width < 5 ns, Andor Technology). The output of the ICCD was inputted into a digital board and then analyzed with a computer data acquisition system. Experimental synchronization is controlled through a multi-channel digital delay pulse generator (Model: DG535, Stanford Research System). The grating efficiency of the echelle spectrometer was calibrated by a NIST standard traceable tungsten halogen lamp (Model: 63976, Oriel Instruments) and wavelength calibration of the system was proceeded with a standard Hg–Ar lamp.

In experiment, the temperature of liquid vaporizer was kept at 180 °C (± 1 °C) while that of the cell was set at 200 °C (± 8 °C), in order to vaporize liquid kerosene thoroughly and avoid the condensation in the test region. The flow rate of air was set at 3 L/min while that of kerosene varied in the range of 7.5–28.5 g/h, which was corresponding to the equivalence ratio in the 0.47–1.8 region. The second harmonic output of a Nd:YAG laser was focused via a quartz lens with a focal of 150 mm to form a local plasma. The single pulse energy of excited laser was set as 120 mJ (± 5 mJ) unless otherwise specified. The emission of LIBS was collected by a 250 mm focal length quartz lens at the

**Table 1**  
Physicochemical properties of RP-3 kerosene.

Density (g/cm <sup>3</sup> ) at 20 °C	0.8079
Flash point (°C)	50
Viscosity (mm <sup>2</sup> /g) at 20 °C	1.67
Distillation (°C) at 10%	179
Mass ratio of C/H	6.04
Aromatics content (%) in mass	15.0
Crystallization point (°C)	–59

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