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High-pressure 1D fuel/air-ratio measurements with LIBS

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ABSTRACT

Quantitative, one-dimensional (1D), single-laser-shot, fuel–air ratio (FAR) measurements in both laminar and turbulent methane–air flames were conducted using time-gated nanosecond-laser-induced breakdown spectroscopy (ns-LIBS) line imaging. In the laminar methane–air flames at a pressure of 1–11 bar, hydrogen (H_α) and nitrogen (N_{II}) atomic emission lines at 568 and 656 nm, respectively, were selected to establish a correlation between the line intensities and the local FAR. The spatial calibration profiles of the N/H ratios in the flames at various pressures were obtained in one dimension. The effects of the laser energy and pressure on the stability and precision of the 1D FAR measurements were investigated. It was observed that the N/H correlation is significantly reduced at ~11 bar, which sets the limits of the 1D LIBS-based FAR measurements. Single-laser-shot 1D FAR measurements were conducted in a turbulent flame at atmospheric pressure, and multiline LIBS was performed to extend the measurement area of interest. Spatially and spectrally resolved line LIBS can provide the local FAR with a spatial resolution of ~0.1 mm. These results hold promise for the utilization of ns-LIBS for spatially resolved 1D FAR measurements in turbulent flames at elevated pressures.

1. Introduction

Nanosecond-laser-induced breakdown spectroscopy (ns-LIBS) has been widely applied in scientific and engineering fields, ranging from basic science laboratories to outer space [1]. The optical breakdown due to a focused high-intensity laser beam in solids, liquids, and gases reveals the elemental information of the target with minimal or no requirements for sample preparation. The breakdown process is initiated with the creation of seed electrons near the laser focal position by the front end of the high-intensity pulse [2,3]. The seed electrons, which are generated by a multiphoton ionization process, absorbs the remainder of the same laser pulse through an inverse Bremsstrahlung process and causes avalanche ionization with a rapid growth of the electron density. Hence, the laser-induced breakdown (LIB) process is considered as a probabilistic process [4,5].

In recent decades, the LIBS technique has been utilized for combustion diagnostics based on direct measurements of individual emission lines, delivering a significant amount of quantitative information including the elemental composition, species concentration [6–11], flame density [7,10], temperature [12,13], and fuel–air ratio (FAR) [14–25]. The atomic emissions from a laser-induced plasma are utilized to infer the quantitative composition and species concentrations in the flames [18]. In general, the dependence of the emissions of the laser-induced gas breakdown on the laser energy (LE), the composition of the target gas, and the ambient environment causes significant fluctuations in the spectral line intensity. These factors hinder the reliability of the emission signal for direct correlation to the actual mole fraction or species concentration [26,27]. Do et al. [7] utilized LIBS for the measurement of the gas density based on the direct correlation between the LE scattered/absorbed by a plasma and the density. However, there are significant challenges regarding the measurement accuracy with high laser scattering/absorption by the plasma in real combustion environments due to laser-beam steering, collisional quenching, and the generation of soot particles. Lee and Hedge [12] derived the temperature from LIBS spectra, assuming that the nitrogen signal intensity is proportional to the density or inversely proportional to the temperature. Kiefer et al. [13] took advantage of the breakdown threshold being a strong function of the density, which provides a means for thermometry. Typically, large shot-to-shot signal (i.e., spectral intensity) fluctuations are expected for LIBS-based combustion diagnostics using longer laser pulses. The high signal fluctuations originating from the stochas-

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tic behavior of the gas breakdown process hinder the development of LIBS applications for the quantitative measurement of the species concentration, density, and temperature in combustion environments.

Several studies have demonstrated that the ratios of the peaks of emission lines can be used to correlate and quantify the FAR (or equivalence ratio) in premixed combustion systems [6,14,16–18,28,29] and practical engines [15,19,20,30–32]. Their results also demonstrated that by taking the ratio of signals from various emission lines, the measurement fluctuations can be significantly reduced [14,28,33]. Various emission lines have been used for FAR measurements, including H_α (656 nm)/N_{II} (568 nm) [6,34], H_α (656 nm)/N_I (500 nm) [11,24,33], $H_ß$ (486 nm)/N_I (746 nm) [31], H_α (656 nm)/O_I (777 nm) [8,14,16,17,20,22–25,28,29,32,33,35,36], H_α (656 nm)/N_I (742–746 nm) [20,22,23], C_I (711 nm)/N_I (740–750 nm) [15], C_I (711 nm)/O_I (777 nm) [15], C_I (711 nm)/(N_I (746 nm) + O_I (777 nm)) [18], CN (707–734 nm)/N_I (740–750 nm) [15], and CN (707–734 nm)/O_I (777 nm) [15]. By weighing the advantages and disadvantages for different applications and scenarios, most of the experiments employed time-gated detection to suppress sampling of the plasma continuum emission and flame chemiluminescence, thereby improving the signal-to-background ratio [6,14–20,28–32]. Moreover, other studies have shown that ungated detection with appropriate blocking of the scattered beam can be used for measurements of the FAR [13,25,33].

Although the ratios of various spectral lines provide FAR measurements, all previous occurrences of the detection of the LIBS-based FAR are single-point measurements; that is, the light emitted from the plasma was conventionally treated as a single point source in those experiments. In fact, the plasma and subsequent emission are far from spherical in shape [4,37,38]. Generally, the nanosecond-laser-induced plasma volume has an ellipsoidal shape with a width of tens to hundreds of micrometers and a length of a few millimeters. Hence, the relative spatial position of the plasma recorded by the spectrally resolved detection system (i.e., a spectrometer equipped with a camera) would directly influence the experimental FAR measurements. Moreover, *line LIBS* has potential applications in fluid shear layers with a very fine structure (i.e., a thickness of approximately a micrometer), where both the velocity and shear stress dramatically vary near the boundary.

In this study, spatially and spectrally resolved ns-LIBS was explored for FAR measurements in methane–air flames at an elevated pressure. The primary focus of this study is to expand the dimensionality of the measurement from a point to a line and to present a spatial analysis of the LIBS spectral line emissions for one-dimensional (1D) FAR measurements in turbulent flames. The three major conclusions obtained from the results presented in this paper are as follows. (1) A laminar Hencken flame in a high-pressure combustion chamber is used to comprehensively study the spatial and spectral characteristics of LIBS emission lines with respect to the LE and pressure conditions. (2) Single-laser-shot 1D FAR measurements in a turbulent methane diffusion flame using ns-line-LIBS are demonstrated. (3) We extend single-line LIBS to multiline LIBS using a homemade linear lens array to demonstrate multiline LIBS for FAR measurements in reacting flows.

This paper is organized as follows. The experimental arrangements for high-pressure FAR measurements are described in Section 2. Section 3 presents the spectral and spatial analyses of atomic line emissions in laminar Hencken flames under various pressure conditions. Section 4 presents quantitative 1D FAR measurements for atmospheric-pressure turbulent flames. Section 5 presents a feasibility study of multiline LIBS for large-area FAR measurements. The paper concludes with a summary and discussion for further applications/improvements in Section 6.

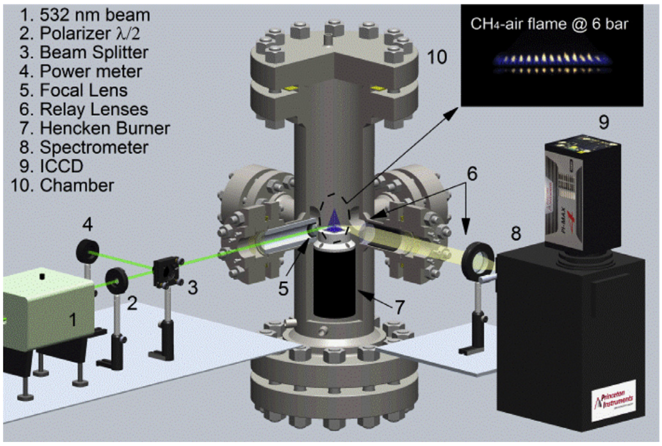

Fig. 1. Experimental setup for ns-LIBS in laminar flames at an elevated pressure. The various components are listed on the left side of the figure. The inset is a snapshot of a laminar methane–air flame at 6 bar.

2. Experimental setup

Figure 1 shows the experimental setup for ns-LIBS measurement in a stainless-steel high-pressure combustion chamber. The dimensions of the inner chamber are 5.85 in. in diameter and 25 in. in height. The chamber has four 2-in. diameter windows for optical diagnostics. The high-pressure chamber can accommodate a 2-in.-diameter burner for the required LIBS-based 1D FAR calibration tests. The exhaust for the gases was operated via a nozzle on the top of the chamber. Air was used as a buffer gas to help push the exhaust away from the flame while also maintaining the overall pressure in the chamber. The flow rates of methane, nitrogen, and air were all controlled by digital flow meters. A cold and dry buffer gas (air or nitrogen) was used to purge the windows continuously during measurement to prevent the condensation of water on the chamber windows.

A laminar flame is generated by a well-calibrated Hencken burner, which is ideal for FAR calibration experiments. A 5.08-cm-diameter Hencken burner (flame area: 2.54 cm. × 2.54 cm) was installed in the high-pressure chamber, as depicted in Fig. 1. The Hencken flame is a diffusion-limited, flow-stabilized, weakly stretched premixed flame. At elevated pressures, the flame is more stretched and flame reaction zone is thinner. When the Hencken burner was operated in the chamber, the gas was exhausted from the top of the chamber through a tube to a fume hood. The chamber temperature and pressure were monitored to ensure that these conditions are maintained during measurement. A Hencken methane–air flame at 6 bar (6×10^5 Pascal) is shown in the inset in Fig. 1.

For LIBS experiments, we employed the second harmonics of a Nd:YAG laser (Powerlite DLS, Continuum, Inc.) with 10-ns-duration laser pulses and a maximum energy of 200 mJ/pulse at a 10-Hz repetition rate. The shot-to-shot laser pulse fluctuation is ~3 %. The laser pulse energy was adjusted by a half-wave plate and polarizer. The incident laser beam was focused 20 mm above the burner surface downstream of the flame front (at the center of the vessel) using a spherical lens with a focal length of + 50 mm. The probe areas were located entirely in the post combustion region for all pressure conditions. The size of the plasma is much smaller than the overall scale of the composite flame (i.e, reacting and post-reacting) structure. The length of laser-induced plasma is limited to a few millimeters while the composite flame width is ~20 mm at the point of measurement. The LIBS emissions from the probe volume were first collected and collimated inside the vessel by a visible-light spherical achromatic lens with focal length