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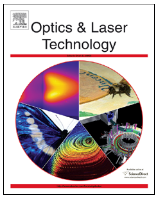

Quantitative mixture fraction measurements in combustion system via laser induced breakdown spectroscopy

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ABSTRACT

Laser induced breakdown spectroscopy (LIBS) technique has been applied to quantitative mixture fraction measurements in flames. The measured spectra of different mixtures of natural gas and air are used to obtain the calibration parameters for local elemental mass fraction measurements and hence calculate the mixture fraction. The results are compared with the mixture fraction calculations based on the ratios of the spectral lines of H/N elements, H/O elements and C/(N+O) and they show good agreement within the reaction zone of the flames. Some deviations are observed outside the reaction zone. The ability of LIBS technique as a tool for quantitative mixture fraction as well as elemental fraction measurements in reacting and non-reacting of turbulent flames is feasible.

1. Introduction

Methods of quickly and rapidly measuring gas composition in combustion systems are of great practical interest. Optical methods such as Raman spectroscopy are quite useful in understanding fluid mixing, optimizing combustion, and minimizing emissions. However, many existing optical methods are limited by the need for some knowledge of the reaction progress, as they measure mole fractions of molecular reactant or product species. Other methods measure condensed-phase (spray) concentrations before combustion, or flame emission directly, to infer composition. Recently, laser induced breakdown spectroscopy (LIBS) became one of the rapidly growing laser based analytical techniques since it allows for the simultaneous sampling, atomization, excitation and ionization of a sample. Spectrochemical analysis using LIBS has shown considerable progress during the last few years and has proven to be extremely versatile, since laser produced plasmas can be created in gaseous [1,2], liquid [3,4] and solid samples [5–7], providing the possibility for real time multi-element analysis not only under laboratory conditions but also in harsh, dangerous and toxic environments. Applications of LIBS typically employ focused pulsed lasers, delivering high peak power pulses capable of inducing a microplasma. The collection and spectral analysis of the light emitted by the plasma permit the qualitative identification of the atomic species being present. Under certain conditions quantitative information can be deduced by the measurement of the intensity of the plasma spectral lines emission. Furthermore, the high temporal and spatial resolution of the LIBS technique makes it particularly well suited for the study of complex phenomena such as flames.

Measurement of mixture fraction is a diagnostics challenge that remains unresolved for combustion applications that involves other than clean non-sooting gaseous flows. Measurement of mixture fraction requires knowing the mass fractions of many species simultaneously to obtain a measure of an elemental conserved scalar. The community has been attempting for years to identify a good tracer that can be used as a conserved scalar and hence replace the need for multi-species diagnostics. The mixture fraction, ξ, as a conserved scalar is basically defined as the ratio of fuel elements' masses to the total mass of the mixture [8]. It can also be defined as the sum of the mass fractions of the fuel elements:

$$\xi = \sum y_{\mathrm{Fi}} \tag{1}$$

where y_{Fi} are the fuel elements' mass fractions. The sum of the fuel and oxidant elements' mass fractions is equal to unity everywhere. Thus the mixture fraction can also be obtained from the following

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equation:

$$\xi = 1 - \sum y_{Oi} \tag{2}$$

where y_{Oi} are the oxidant elements' mass fractions. Thus ξ varies from 0 in the oxidant stream to 1 in the fuel stream. In a dual stream system, fuel and air, the mixture fraction may also be defined as

$$\xi = \frac{y_i - y_{i,O}}{y_{i,F} - y_{i,O}}, \tag{3}$$

where y_i is the mass fraction of element i, e.g. H, and O and F represent the Oxidant and Fuel streams, respectively. More equations can be found in the literature. However, all equations lead to the same definition of mixture fraction as a conserved scalar of the fuel elements within the reacting medium.

Several methods have been proposed to obtain the mixture fraction either from the ratio of the intensities of the spectral lines of C, N, H, and O. Phuoc and White successfully measured fuel-to-air ratios [9] based on simultaneous measurements of the emissions of the H_α line (656.3 nm) and the OI triplet near 777 nm, (777.2; 777.4, and 777.5 nm). Since the H_α line is emitted by electronically excited hydrogen dissociated from the fuel molecules (CH_4, H_2), its intensity is used to represent the fuel concentration. The emission intensity of the OI triplet, which is emitted by electronically excited oxygen dissociated from the air molecules, is used for quantifying the air fraction of the mixture. The ratio of the intensity of these two lines is used to generate a calibration curve of LIBS equivalence ratio measurements. Sturm and Noll [10] used the technique to simultaneously measure C, H, O, and N for gas mixtures containing CO_2, N_2, and C_3H_8 and reported various calibration curves of the LIBS signal versus the atomic abundance ratios. Ferioli et al. [11] used LIBS to measure the equivalence ratio of a spark-ignited engine based on the emission lines of C, N, O and broadband CN (707–734 nm) molecular emission. Measurements were conducted within a range of equivalence ratio from 0.8 to 1.2 using CN/air, C/N, C/O atomic peaks ratios interchangeably. However, all the previous methods do not yet relate the lines intensities to elemental mass fraction and hence obtain the mixture fraction from single shot. The objective of the present work is to demonstrate and evaluate the ability of LIBS technique as a tool for quantitative mixture fraction as well as elemental fraction measurements in reacting and non-reacting environments. Calibration curves for C, N, H, and O elements based on peak intensity and the integration under curves have been introduced. The mixture fraction was calculated using the line intensities ratios of H/O, H/N, and C/(N+O) [12–14]. The effect of the laser energy is also considered for more comprehensive coverage of the several parameters that affect the spectral intensities of the lines.

2. Experimental technique

The LIBS set-up employed in this work is illustrated in Fig. 1. The laser beam of a 6 ns Q-switched Nd:YAG laser (Quanta Ray-Spectra Physics) operating at single shot, 1064 nm was focused with a 50 mm focal length quartz lens in order to create the plasma. The laser beam diameter was 10 mm. The light emitted from the plasma was collected using collection optics in the form of two quartz lenses with equal focal length of 100 mm. This provides a 1:1 image at one end of a one-meter length wide band fused-silica optical fiber with an aperture of 200 μm. This is the limit of the system spatial resolution which is small enough for turbulent flames. The other end of the fiber is connected to a spectral analysis system. This system consisted of an Echelle spectrometer (PI—Echelle, Princeton Instruments, USA) with a gated, Intensified Charge Coupled Device, ICCD, (Princeton Instruments) attached to it. The emission signal was corrected by subtracting the dark signal of the detector through the LIBS software. A gate width of 10,000 ns and a delay of 1000 ns after the laser pulse were chosen for maximizing the intensities of the spectral lines of CI (247.8 nm), H_α (656.3 nm), OI (777.58 nm), and NI (746.8 nm) while maintaining a good temporal resolution. The gate width and delay time have been chosen based on an optimization procedure that maximizes the spectral line intensity. The choice of the gate width and delay time for the spectroscopic data acquisition is accomplished by a computer-controlled system. This allows the observation of the produced plasma at Local Thermal Equilibrium (LTE) since only in such case the densities of the emitting atomic/ionic species can be deduced from measurements of the intensity of spectral lines, and subsequently, the ratio of the number densities derived from the ratio of the measured intensities of the emission lines can be obtained. The trigger signal of the laser is connected to a pulse delay generator (Stanford delay generator DG535) to control the synchronization between the laser and the camera. To ensure timing reliability the delay was set by the delay unit and measured with a fast photodiode (rise time 1 ns) and a fast oscilloscope (500 MHz). The analysis of the emission spectra was accomplished using the commercial software (GRAMS/AI v.8.0, Thermo-electron co.).

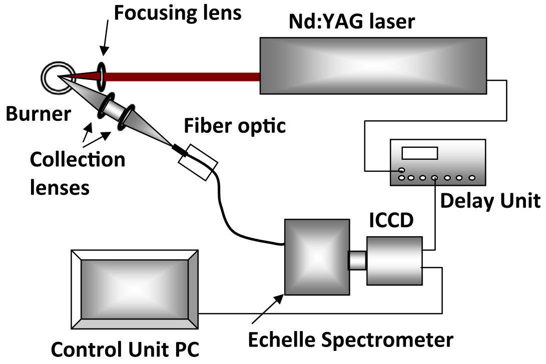

Fig. 1. A schematic diagram of the LIBS setup.

The measurements were conducted in mixtures of air and natural gas (95% methane) at different equivalence ratios. The air and fuel flow rates were controlled using mass flow controllers and mixed at a distance of more than 200 pipe diameters to ensure well-premixed conditions. Then the technique was tested along the reaction zone in a turbulent partially premixed flame. See Mansour [15] for more information about the burner. The calibration curves and the mixture fraction measurements using LIBS technique are presented in the following sections.

3. Results and discussion

3.1. LIBS spectra

Typical emission spectra detected from the laser induced plasma spark in air and combustible gaseous mixtures of air and fuel at different equivalence ratios of 0, 0.467, 0.794, 1.111, 1.587 and 3.175 are shown in Figs. 2 and 3. The spark location was just above the burner rim and in the burner cone centerline. The intensities of the emission spectral lines of CI, H_α, NI, and OI at the same equivalence ratios are shown in Fig. 2. In the single pulse measurements the experimental parameters were optimized for high signal-to-noise ratio. The spectra from single shot measurements show a very small background effect since the baseline due to continuum effect is very small, $< 0.5\%$. The spectral emission