Combustion and Flame 162 (2015) 4479–4485

Contents lists available at ScienceDirect

Combustion and Flame

journal homepage: www.elsevier.com/locate/combustflame

Direct spectrum matching of laser-induced breakdown for concentration and gas density measurements in turbulent reacting flows

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ARTICLE INFO

Article history:
Received 12 June 2015
Revised 28 August 2015
Accepted 28 August 2015
Available online 29 September 2015

Keywords:
LIBS
High-speed turublent combustion
Laser breakdown
Concentration
Density measurement

ABSTRACT

A direct spectrum matching method for laser-induced breakdown spectroscopy is proposed to simultaneously measure gas density and concentration in turbulent reacting environments with improved measurement accuracy. The breakdown spectrum recorded in the target flow is directly matched with a spectrum out of a database consisting of various emission spectra recorded under well-defined conditions in a range of gas density and composition. It is shown that the wavelength, intensity and line width of the atom/ion emission lines in the spectrum indicate atom composition and gas density that are independent of parent molecular species in the target flow. Once a matching spectrum (within 550–830 nm containing O, H, N, and C lines) in the database of a known gas condition is found, the concentration and gas density at the location of the breakdown can be accurately derived. A 532-nm Nd:YAG laser with 10-Hz pulse repetition rate is used to induce breakdown in fuel/air mixtures in a variable pressure combustion chamber to build the spectrum database.In addition, it is used in a cavity flameholder of a model supersonic combustor to measure the gas density and concentration fields in a turbulent reacting environment. All the measurements are completed within 100 ns after laser firing, before breakdown affects the flow and the fast evolving environment alters the breakdown spectrum.

1. Introduction

Laser-induced breakdown spectroscopy (LIBS) is a technique that allows for quantitative measurement of gas properties in challenging environments [1–3]. A laser beam focused with sufficient energy to ionize, dissociate, and excite molecules in a small volume about the focal point creates a small volume of plasma, typically less than 1 mm^3 [1–8]. Electrons of the gas constituents near the focus absorb the photon energy through the inverse Bremsstrahlung process to cause excitation, dissociation and ionization [9]. The prominent mechanism of the laser-induced plasma development is assumed to be through cascade ionization as opposed to multiphoton ionization due to the relationship between gas density and laser energy required to induce breakdown, [10]. Then, the excited species in the laser-induced plasma (e.g., atoms and ions) emit photons containing information on atom composition and gas density. Atom composition in the breakdown plasma determines the wavelength and intensity of the photon emission with individual emission line strength being a function of the atom/ion concentration. In addition, the unperturbed temperature and pressure, i.e., the gas density, at the focal point where the plasma is produced before breakdown, has a great effect on the line width and overall emission strength, since the gas density determines the number of molecules absorbing the laser photons and the number of atoms that emit photons. (Note that throughout this manuscript gas density refers to the ratio of pressure to temperature prior to the formation of the plasma.) The quantitative LIBS measurement is based on the monotonic correlations of the quantities that are extracted from the emission spectra (e.g., emission line widths, strengths and their ratios) and the physical gas properties (e.g., gas density and concentration) of target media.

Implementing the measurements in rapidly evolving turbulent reacting environments, however, is challenging because the breakdown plasma, surrounding high-speed turbulent flows, and combustion chemistry interact to increase measurement uncertainty [11–15]. In addition, large and rapid variations of gas properties such as density, species concentration, pressure, and temperature in the harsh environment make the measurement difficult. Therefore, a method for deriving gas properties within the shortest time is essential such that the measurement is not affected by the interaction of the breakdown with the surroundings. In previous work [2, 3], a nanosecond-gated LIBS (n-LIBS) method was developed to instantaneously measure species concentration in a reacting flow. This newly developed

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http://dx.doi.org/10.1016/j.combustflame.2015.08.021
0010-2180/

n-LIBS technique minimizes the total measurement time below 100 ns (versus $> 1\ \mu s$ in conventional LIBS [4–8]) enabling the application of the technique to turbulent supersonic flows [2, 3]. Density dependence of the n-LIBS is also investigated and is critical for measurements in compressible and/or reacting environments. Previously, several atomic emission line intensities and their ratios were used as the species/atom concentration indicators [1–8]. However, it was found that the ratio of emission line intensities is also a function of gas density, which limits the application of the method. Furthermore, not only do the emission line intensities (and their ratios) change with gas density, but also the line widths and spectrum baseline profiles are sensitive to the density variation. Therefore, simply extracting the line intensity from the spectra is not the best approach vis-a-vis measurement accuracy.

In this study, we are proposing a Direct Spectrum Matching (DSM) method that accounts for all the detailed spectrum features including the multiple emission line intensities/widths (more than 15 atomic/ionic emission lines), overlapping line structures, baseline profiles, etc. With the DSM method, the goal is to find the best match of the spectrum from a database containing spectra collected at well-defined gas conditions. The DSM process is just like the fingerprint matching process, thus requiring an intensive spectral database. Therefore, the contribution from each atom to the spectrum profile (e.g., sensitivity of line strength to atom concentration) is quantitatively evaluated in this study. It is used to explore the possibility of accurate and empirical spectrum profile prediction that is potentially filling the gaps between the data points in the database and extending the property measurement range. In order to provide the proof of the concept, several sample spectra recorded within a cavity flameholder (in a Mach-2 crossflow) are used to demonstrate the feasibility of the technique in practice.

2. Experimental setup

2.1. Overview

There were two separate experimental setups used in the course of the experiment: one with a continuous supersonic wind tunnel with a model combustor (Research Cell 19, RC-19, at Wright-Patterson Air Force Base) for the collection of test spectra containing gas property information in the compressible turbulent reacting flow, and the other with a variable pressure combustion chamber (VPCC) to create a reference database, or spectrum database, covering the gas property ranges of the RC-19 experiments. In both cases, a 532-nm, Q-switched Nd:YAG laser (Spectra Physics GRC-170) operating at 10 Hz was used to induce breakdown. To maintain a constant spatial and temporal beam profile independent of beam energy, a half wave plate mounted on a rotation stage in conjunction with a thin-film polarizer was used to vary the beam energy. The volume of the plasma, which was approximately 1 mm^3 for our conditions, depends on the local gas density, and this entire volume was used for emission collection.

The beam energy required to induce breakdown is dependent on the density of the target media. A beam energy of 300 mJ per pulse exiting the laser was used for testing in both the VPCC and RC-19, which was found to be sufficient to induce breakdown in the range of gas densities found in the RC-19 tunnel. Further increases in energy (beyond the threshold value) elevate the overall signal strength of plasma emissions but do not alter the relative intensities and emission line widths, i.e., the baseline-subtracted and normalized spectra (post-processed spectra) are independent of laser energy.

2.2. Variable pressure combustion chamber setup

The VPCC is a cylindrical chamber with four optical access windows. A vacuum pump controls the pressure inside the chamber between 760 and 50 torr. Within the VPCC a modified premixed Hencken burner was mounted on automated linear actuators allowing for the position of the burner to be adjusted. The burner size is $25\times25\ mm^2$ and is surrounded by a 10 mm wide inert gas shroud. Unlike a typical Hencken burner, the burner in the VPCC is modified such that the fuel and oxidizer, or other combinations of gasses, are premixed before reaching the burner exit. Species concentrations in the gas mixture were modified by changing the volumetric flow rates of C_2H_4, CH_4, CO_2, O_2, N_2, and compressed air with electromechanical flow controllers, calibrated with a piston type flow meter (MesaLabs Dry-Cal Definer 220). A lens (17.5 cm focal length) located inside of the VPCC focused the laser beam at the center of the burner, approximately 2 mm above the exit plane, which is the most upstream location possible for calibration/measurement; here, the gas conditions will be well known. Figure 1 shows the layout of the setup. Burner exit velocity was set to be higher than the flame speed so that the measurement was always within the reactant gases. An intensive database was constructed in the VPCC that covers fuel concentrations from 0–70% C_2H_4 in air at pressures from 500–50 torr; this fully covers the species concentrations and gas densities found in the supersonic combustor. A fast spectrometer (Kaiser HoloSpec f/1.8) and camera (Andor iStar DH320T-18U-73) located at the spectrometer exit plane captured the plasma emissions. A long-wave-pass filter (Semrock BLP01-532R-50) blocked scattering from the laser beam, and a Nikon camera lens (58 mm, f/1.2) collected the emission and focused it on the spectrometer entrance slit. The intensified camera recorded the emission 75 ns after the laser beam arrived at the focal point with a 10 ns gate width. Due to the short gate time, the flame chemilunenescence had no detectable effect on the LIBS spectra.

2.3. RC-19 setup

The model supersonic combustor used for the testing is part of a continuous supersonic wind tunnel operating at Mach 2 with an inlet temperature of 590 K. The combustor contains a cavity flameholder as shown in Fig. 2. Measurements were taken in and above the cavity flameholder. Fuel was injected in the upstream direction from the ramp at the downstream side of the cavity. The same laser setup was used as during the calibration experiment, though the focusing lens was located outside of the wind tunnel, as shown in Fig. 2. The focal point in the supersonic combustor was positioned with an automated 3-axis translation optical table on which the laser system and all the optical components were installed.

The converging laser beam for generating the breakdown plasma in the tunnel was transmitted through an access window made of fused-silica. Strong converging and diverging reflections from the windows on both sides of the tunnel were blocked to protect optics using a heat sink made of copper. The laser path was adjusted to be slightly off normal to the window, approximately 2°. The converging and diverging reflections returned at the angle of incidence, resulting in 4° between the incoming and reflected beams, allowing the reflected beam to be captured by the copper heat sink. Spectrum collection was accomplished with the same camera and spectrometer setup as in the calibration experiments.

3. Results

3.1. Characteristics of the plasma emission spectrum

Figure 3 shows typical spectra obtained in the VPCC at various pressures with a constant fuel concentration ($X = 12.5\%$ by volume of C_2H_4 in air); the spectra are normalized by the peak intensity of the 568-nm N emission line. The wide spectral window used here, 550 nm – 830 nm, provides multiple emission lines of N, O, C, and H. Of particular interest for the DSM method are the emission lines that have strong signal and are not overlapped with other emission