Laser-induced breakdown spectroscopy measurements of mean mixture fraction in turbulent methane flames with a novel calibration scheme

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ABSTRACT

Laser Induced Breakdown Spectroscopy (LIBS) was applied to homogenous methane–air mixtures of a wide range of compositions ranging from only air to only fuel in order to establish a new calibration scheme suitable for LIBS measurement of mixture fraction in turbulent non-premixed flames. Both a portable low resolution spectrometer equipped with a CCD detector with wide temporal detection window and a monochromator with a fast gated ICCD camera were employed to monitor the plasma emission. The results obtained using the two detection systems were fully consistent, suggesting that LIBS can be used successfully with the CCD detector that is more suitable for industrial applications. From the spectroscopic analysis, it was shown that the ratio of the intensities of H_α (656.3 nm) over O (777.3 nm) (H_α/O) and of C_2 ($\Delta \upsilon = 0$, $d^3\Pi_g$ –#x03B1;$^3\Pi_u$) over CN ($\Delta \upsilon = 0$, $B^2\Sigma^+ - X^2\Sigma^+$) ($C_2$/CN) depend monotonically on the mole fraction of methane in the ranges of 0.0–0.3 and 0.3–1.0 respectively, therefore providing a new scheme for measurement of mixture fraction in non-premixed systems spanning the complete range of equivalence ratios. The technique was also applied to a swirling recirculating premixed flame and it was found that the equivalence ratio is successfully measured in both reactants and products. Based on the above, LIBS experiments were then carried out in turbulent axisymmetric non-reacting and reacting jets, and the mean mixture fraction determined by the aforementioned calibration curves was in good agreement with empirical correlations, while the rms measurement showed the expected trends, demonstrating hence the usefulness of the technique.

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1. Introduction

During the past few years, Laser Induced Breakdown Spectroscopy (LIBS), a laser-based spectroscopic or analytical technique, has been developed and successfully applied for combustion diagnostics purposes [1–3]. By strongly focusing the beam of a laser system in the flame region, dissociation of the present molecules and/or ionization is occurring forming the plasma. After the collection and monitoring of the plasma-emitted radiation and its spectroscopic analysis, one can retrieve information about the fuel content (and/or the Air-to-Fuel Ratio) of various combustible mixtures and infer the local chemical composition [4], determine parameters of the flame such as the temperature [5], and/or identify the fuel used [6]. After the first application of LIBS in hydrocarbon–air mixture by Schmieder [7], a large number of studies have been presented in various combustion systems under different conditions. In the majority of the previous LIBS works, the ratio of some spectral lines of atomic origin like the ratio H_α/O (at 777 nm) [4,6,8–15] or O/H_α [16], the ratio H_α/N (at 567 nm or 744 nm) [17–19] or sometimes both [20–22], are employed most of the times for the determination of the equivalence ratio and rarely for the determination of the concentration (in %) of a constituent. Sometimes emission lines attributed to carbon atoms are also used since these emission lines are directly linked to the fuel. Thus, either the ratio of a carbon line to a nitrogen, hydrogen or oxygen line like C/H_α and C/N (at 500.5 nm) [5], the ratio C (at 833 nm)/N (at 744 nm) with C (at 833 nm)/O (at 777 nm) [23], or C (at 711.3 nm)/O (at 776.6 nm) and C (711.3 nm)/ N (at 746.3 and 743.8 nm) [24], or the ratio of the intensity of a carbon line

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to the sum of intensities of a nitrogen and an oxygen line C (at 711 nm)/[N (at 744 nm)+O 777-nm] [24] have been successfully used. However, there are limited calibration curves based on the ratio of a molecular band to atomic lines like CN (integrated from 705 to 733 nm)/N (at 746.3 nm or 743.8 nm), or CN (integrated from 705 to 733 nm)/O (at 776.6 nm) or CN (integrated from 705 to 733 nm)/[N (at 744 nm)+O (at 777 nm)] [25], or based on the intensity of molecular bands like CN ($\Delta \upsilon = 1, 0, -1, B^2\Sigma^+ - X^2\Sigma^+$) [26], CN ($\Delta \upsilon = 0, B^2\Sigma^+ - X^2\Sigma^+$) [27,28].

Most of the work above concerned lean mixtures. To expand LIBS to fully non-premixed systems, Do et al. [29] have employed the ratio H_α/N (at 568 nm) and the N/H_α for the determination of the fuel concentration in methane- and ethylene–air mixtures with fuel mole fractions up to 90%. The analysis and the fuel measurement were retrieved from emission spectra which have been obtained with a detection window of 20 ns duration using an ICCD camera. From a more practical point of view, such a delicate detection system would be a rather limiting factor for the LIBS application under very aggressive industrial conditions. In addition, this work has not explored in detail the application of the technique and the calibration scheme to turbulent non-premixed flames.

Concerning the precision of the single shot results, which is necessary for instantaneous mixture fraction (or equivalence ratio) measurements in turbulent non-premixed systems, some investigations have demonstrated [24,25] that the equivalence ratio calculated from the instantaneous measurements is accurate even if there is some dispersion of the values of the equivalence ratio greater in fuel-rich in comparison to the fuel–lean mixtures. However, there is no extensive study on the precision of LIBS for turbulent systems.

Finally, it is important to note that in turbulent combustion one must be able to measure in both reactants and products. In fully premixed flames, the carbon to oxygen atomic ratio does not change across the flame (once any small-scale differential diffusion effects have been smoothed out), but the species composition and temperature are very different. The decrease in density and the large amounts of CO_2 in the hot products suggest that a higher energy is needed to produce a spark, and the presence or not of a spark has been exploited to actually establish the boundary of the flame brush [10,30]. However, if the laser energy is high enough, one may expect that a spark will always be created in both reactants and products and hence a measurement of the same equivalence ratio across a premixed flame would suggest that LIBS can measure the mixture fraction in combusting systems irrespective of the degree of reaction, which is a necessary requirement for the measurement of mixture fraction in non-premixed flames.

Examination of the aforementioned literature suggests that no molecular line other than the one of cyanogen band has been employed as an indicator of the amount of fuel in the mixture so far, and that up to now, the technique has been applied only to a small range of compositions. However, when non-premixed turbulent combustion systems are under investigation, with the corresponding severe changes of fuel concentrations both locally and temporally, further work is needed (i) to develop calibration strategies spanning a wider range of mixtures than before; (ii) to ensure plasma creation in both the low- and high-temperature regions; and (iii) to investigate the precision of single-shot results.

This paper explores the application of LIBS technique for local and instantaneous measurements of the equivalence ratio in turbulent premixed and non-premixed flames. The key novelty lies in the use of the technique for highly turbulent systems and for a much wider range of equivalence ratio than before using a new calibration strategy. In the remaining of this paper, we describe the LIBS measurements using two different detection systems as applied in uniform mixtures spanning a very wide range of compositions so as to create calibration curves, which are then used

for the study of turbulent premixed and non-premixed flames. The paper closes with a summary of the most important conclusions.

2. Experimental methods

2.1. Flow conditions

For the needs of the present research, initially homogeneous flowing methane/air mixtures, exiting from a $d = 23$ mm inner diameter (ID) tube, were used (burner (I) in Fig. 1). The flow rates of the mixture varied from 10 SLPM to 200 SLPM, which corresponds to velocities of 0.4–8.0 m/s, and therefore Reynolds numbers of 586–11720. Based on the fact that the mixtures were fully mixed and uniformly distributed across the pipe, in the immediate exit of the pipe (i.e. at low height to diameter (h/d) values; $h/d < 0.4$), the acquired results were used as calibration curves for the subsequent investigation. In order to characterize the methane–air mixtures of different compositions, the mole fraction of methane, X_{CH_4}, was in the range from 0 to 1.

Following the calibration, turbulent flames were investigated. For non-premixed flames, the burner (II) (Fig. 1) consisted of two coaxial stainless steel tubes; a mixture of 70% methane and 30% air by volume (equivalence ratio 22.21) was supplied from the inner tube of 5 mm ID and with the length-to-diameter ratio 128, while laminar airflow was coming out from the outer tube of 200 mm ID to surround and protect the mixture and/or the flame from disturbances from the environment [31]. The jet exit velocity of the mixture was 17 m/s and the velocity of the surrounding air was 0.1 m/s. In some experiments, the CH_4 volume fraction was 30%, which is too low for a jet turbulent flame to stabilize and therefore the measurement of the fuel at the spark location corresponds to a measurement of an inert tracer.

For premixed systems, a recirculating methane–air flame, which resulted from the bluff-body burner (III) (Fig. 1) developed by Cavaliere et al. [32] but adjusted to the framework of the present investigation, namely without the enclosure, was studied. In this configuration, the CH_4–air mixture was supplied from a pipe of length 350 mm and ID 37 mm, which in the center contained a conical bluff-body (45° half-angle) with diameter 25 mm. A swirler produced an azimuthal velocity component, which reinforced the recirculation zone in the wake of the bluff body and caused a widening of the flame. Note that the flame is exposed to the ambient air, so it is expected the equivalence ratio to drop to zero at large distances from the flame brush; however, to remain the same in the reactants and the products across the flame. For these experiments, the equivalence ratio was 0.81 and the bulk velocity at the annulus was 10.2 m/s.

In all cases the reactants, namely the methane and the air, were supplied by a high purity bottle (99.99%) and an air compressor respectively, while their flow rates were regulated and measured by calibrated rotameters. The air compressor was equipped with a drier and with moisture and oil filters, which resulted in dry air being delivered. This is important because even small amounts of moisture in the atmospheric air can contribute to the hydrogen line in the LIBS spectra. However such contribution in the intensity of hydrogen line arising from the humidity present in the air was taken into account and subtracted in any case from the rest of the results shown.

2.2. LIBS experimental setup and data acquisition

Two sets of experiments were performed. The first set was with uniform mixtures and was aimed at establishing the suitability of calibration schemes and the importance of the detection system and the laser energy. The second set was with the turbulent