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Applied Thermal Engineering 24 (2004) 1619–1632

APPLIED THERMAL ENGINEERING

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Chemiluminescence sensor for local equivalence ratio of reacting mixtures of fuel and air (FLAMESEEK)

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Received 13 July 2003; accepted 31 October 2003
Available online 1 January 2004

Abstract

This paper describes a Cassegrain optics-based chemiluminescence sensor (CS) for measurements in gas turbine combustors. The chemiluminescence sensor measures the equivalence ratio of reacting fuel and air mixtures, and can identify the flame location, in partially premixed flames. It has the potential for monitoring the degree of premixedness of reacting fuel and air in industrial gas turbine combustors, where operation with lean premixed mixtures is important for reduction of NO_x emissions. The spatial resolution of the sensor is evaluated by comparing OH^* chemiluminescence measurement from the CS with laser induced OH fluorescence, in the cone-shaped premixed flame of a Bunsen burner. The ability of the sensor to measure in a modified micro-gas turbine environment burning a methane/air, as well as, a methane/water/air flame (humidified flame) is also demonstrated.

Keywords: Premixed combustion; Chemiluminescence sensor; OH^*, CH^* and C_2^* excited radicals; OH PLIF; Humidified flame

1. Introduction

Lean premixed combustion is an important method for the reduction of NO_x emissions from industrial gas turbine combustors. Operating in a lean-premixed mode has the advantage of

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doi:10.1016/j.applthermaleng.2003.10.028

avoiding high-temperature, stoichiometric combustion that increases thermal NO_x. Lean fuelling, however, currently has the overriding disadvantage that combustion induced oscillations arise which can destroy the machine. One theory is that these oscillations arise during fuel lean operation because of a feedback mechanism involving appropriately-phased time-dependent fluctuations of equivalence ratio of the reacting mixture. An instrument which could measure spatial and temporal variations of the equivalence ratio of air with fuel could be useful both in the context of verifying the origin of the combustion induced oscillations and in terms of having the simplicity to measure spatial and temporal inhomogeneity of the premixed mixture in industrial gas turbines. The chemiluminescent sensor described in this paper is potentially able to provide these measurements.

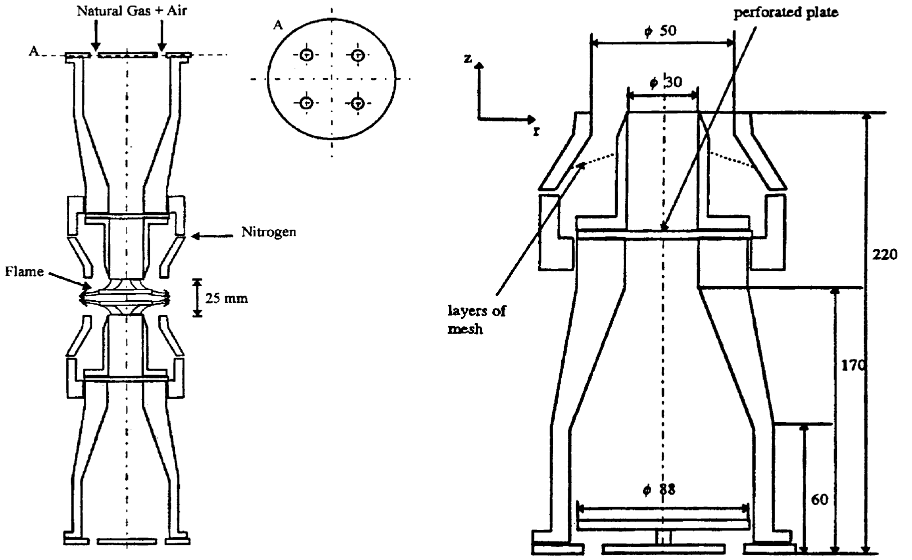
An optical sensor with the ability to measure the fuel/air premixedness and resolve the local flame front, was developed, evaluated and will be tested in a real gas turbine environment, in the context of a project entitled "FLAMESEEK" (FLAME sensors for efficient engine cycles), funded by the European Commission through the fifth framework programme. The partners in FLAMESEEK project include Rolls–Royce Power Engineering plc (UK), Imperial College London (UK), University of Twente (The Netherlands), Lund University (Sweden), Foundation for Research and Technology Hellas/Institute of Chemical Engineering and High Temperature Chemical Processes (Hellas) and Alstom Power SE AB (Sweden).

It has been known for many years that the chemiluminescence from flame excited radicals such as OH^*, CH^*, C_2^*, and CO_2^* can be used to measure aspects of combustion. Recently, for example, Higgins et al. [1,2], and Docquier et al. [3], have used the excited chemiluminescent intensities from OH^* and CH^* to measure the equivalence ratio of premixed, fuel-lean methane/air flames, although their measurements were not spatially resolved because they measured the global chemiluminescence of the flame by using a fiber and a quartz lens. Kojima et al. [4], have reported spatially-resolved measurements of OH^*, CH^* and C_2^* of laminar premixed methane/air flames, but they studied fuel-rich flames.

In this paper, a Cassegrain optics-based chemiluminescence sensor will be presented and its ability to resolve flame fronts will be shown by its comparison with OH PLIF. Then the applicability of the CS at an atmospheric modified micro-gas turbine environment will be assessed. The ability of the sensor to measure equivalence ratio at a CH_4/H_2O/air humidified flame will be demonstrated, and the important effect of the humidity to the measured equivalence ratio by the CS will be studied.

2. The chemiluminescence sensor

The variation of chemiluminescence intensity with flame equivalence ratio and flame strain rate was evaluated in a flow allowing independent variations of flow conditions and combustion parameters, such as bulk flow strain rate and flame equivalence ratio. The so-called "counterflow" burner geometry is suitable for that purpose, because it allowed us to operate the burner within a wide range of flame equivalence ratios and bulk flow strain rates: the spatially stabilized flat flame front was close to the stagnation plane, which had constant stretch rate along the reaction zone. Finally, it offers excellent optical access for measurements. The counterflow geometry used for this work follows the work of Mastorakos [5], Fig. 1. It comprised two opposed brass pipes with inner



(a) (b)

Fig. 1. (a) Counterflow burner geometry and (b) detailed sketch of the lower duct (dimensions in mm).

diameter of $D = 30$ mm, separated by a distance 2H, which can be adjusted to any value up to 200 mm via a vertically moveable lower pipe. The counterflow burner has been described in detail in [6]. The bulk exit velocity of each jet from the pipe, V_0, was varied between 1 and 5 m/s. This corresponds to a bulk flow strain rate, S, ranging from 80 to 400 s^{-1}. The bulk flow strain rate was evaluated as follows:

$$S = \frac{2V_0}{2H}. \tag{1}$$

The equivalence ratio Φ of the premixed mixture was varied from 0.7 to 1.3 for most of the cases studied.

2.1. Instrumentation

The chemiluminescence sensor comprised a Cassegrain optical system, shown in Fig. 2, similar to that of Akamatsu et al. [7]. The use of mirrors, rather than lenses, avoided the generation of chromatic aberrations for different wavelengths, in contrast to lenses (refractive elements), which cannot be corrected for chromatic aberrations over a wide range of wavelengths. The collected light from the CS was focused onto a pinhole placed in front of an optical fiber, which was connected to a purpose-built *photo detector unit* (Fig. 2). The light was split into three parts using two dichroic mirrors for the three wavelengths considered. Each part was directed onto appropriate interference filters specific to the radical considered, at 310.4 nm for the OH^*, 430.5 nm for the CH^*, and 516 nm for the C_2^*. The collected light intensities were transformed into electrical signals by three photomultipliers. The temporal signals of OH^*, CH^* and C_2^* were filtered by a low-pass filters, and digitized simultaneously using a 12-bit A/D card. The MICRO system had a

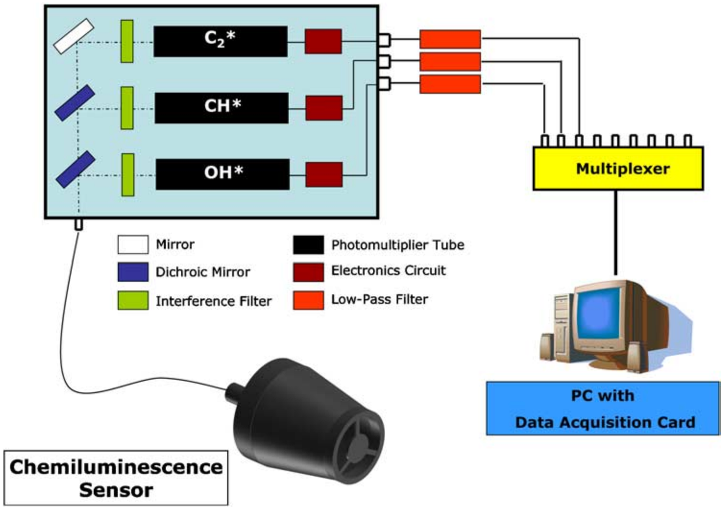


Fig. 2. The chemiluminescence sensor and the photo detector unit.

300 mm focal length, with diameter of primary and secondary mirrors of 150 and 50 mm respectively. This produced a probe volume with nominal diameter and length of 200 microns and 1.6 mm, respectively, according to Akamatsu et al. [7].

2.2. Data processing

Raw intensity signals from the photomultipliers were recorded and then processed according to the following steps:

- A threshold was selected for the raw intensity CH^* signal. This allows detection of signals from the reaction sheet above the background noise.
- Samples which were above the threshold for the CH^* signal were identified. These samples were considered to arise from chemical reaction in the optical probe volume. The corresponding points in time in the OH^* intensity signal were identified to evaluate the instantaneous OH^*/CH^* ratio. Within the interval of the signal record being above the threshold, we identified the maximum values of the CH^* and OH^* signals. If the maxima of the intensities coincided (i.e. they occurred within a temporal tolerance of $2/f_s$, where f_s was the sampling frequency), we calculated the instantaneous value of the OH^*/CH^* ratio from these two maxima. The idea underlying this processing was that only the maxima of the intensities were representative of the local instantaneous reaction rate and equivalence ratio and that intensities before or after the maximum (in a given interval) are typical of chemical reaction initiation or termination.
- The mean and rms of the fluctuation of the samples of the OH^*/CH^* ratio, determined as above, were calculated from the instantaneous OH^*/CH^* ratio taking into account the different efficiencies of the interference filters, dichroic mirrors and the response of the photocathodes at the wavelengths considered. The conversion factors of the current optical system that were used to convert measured intensity ratio into real intensity ratio are:

$$\frac{I_{\mathrm{OH}}^{\mathrm{real}}}{I_{\mathrm{CH}}^{\mathrm{real}}} = 2.720\frac{I_{\mathrm{OH}}^{\mathrm{meas}}}{I_{\mathrm{CH}}^{\mathrm{meas}}}, \tag{2}$$

where I^{meas} and I^{real} represent, respectively, the measured and the real chemiluminescence intensity of the radical considered.

2.3. Calibration of CS for natural gas

Calibration experiments were performed with the counter-flow burner using UK natural gas (94% methane, CH_4) over an equivalence ratio range of 0.7–1.3 and a bulk strain rate range of 160–320 s^{-1}. The measured OH^*/CH^* chemiluminescent intensity ratio was found to be independent of the strain rate, and monotonically dependent on the equivalence ratio. The dependence of OH^*/CH^* ratio on the equivalence ratio, Φ, was approximated by the following curve fit ([8]):

$$\frac{\mathrm{OH}^*}{\mathrm{CH}^*} = 0.497 + 2.107\exp(-(\Phi - 0.7)/0.260). \tag{3}$$

For all flow conditions and for a given value of Φ, the OH^*/CH^* ratio was within 5% of the value given by Eq. (3), which leads to an uncertainty of approximately 0.05 for determination of flame equivalence ratio for $\Phi \leqslant 1.0$. For $\Phi > 1.0$, the chemiluminescent technique could not measure flame equivalence ratio accurately with uncertainties being larger, of the order of 0.1–0.2. Therefore, the curve fit, Eq. (3), may be used only for equivalence ratios less than 1.0. The monotonic dependency of the OH^*/CH^* intensity ratio on equivalence ratio makes the instrument a useful instrument for measuring local flame equivalence ratio in a real burner operating with natural gas at atmospheric pressure. The calibration of the CS for natural-gas-fuelled flames has been described in more detail in Refs. [6,8].

3. Evaluation of the CS with OH PLIF

The spatial resolution of the CS has been evaluated using spatial and temporal resolved planar laser-induced fluorescence, PLIF, of the OH flame radical. The laser technique used in order to evaluate the spatial resolution of the CS, was spatially and temporally resolved planar laser-induced fluorescence, PLIF, of the OH flame radical. The laser source was a cluster consisting of four standard flash-lamp pumped Nd:YAG lasers (BMI, France). The fundamental laser wavelength is 1064 nm. Each laser in the cluster consists of a Q-switched oscillator and a single amplifier, emitting laser pulses with duration of 7 ns. The four laser beams are aligned and frequency doubled according to the scheme shown in Fig. 3.

With this laser cluster a sequence of laser pulses with very short time separation can be obtained, enabling temporally resolved studies of turbulent combustion phenomena. However, the high-temporal resolution capability of the Nd:YAG laser cluster was not used during the present experiments, since the flame studied was a stable laminar Bunsen-burner flame.

The 532 nm output from the Nd:YAG laser cluster was converted to other wavelengths using a dye laser, pumped by the Nd:YAG laser cluster. The dye laser (ND60, Continuum) is narrowband

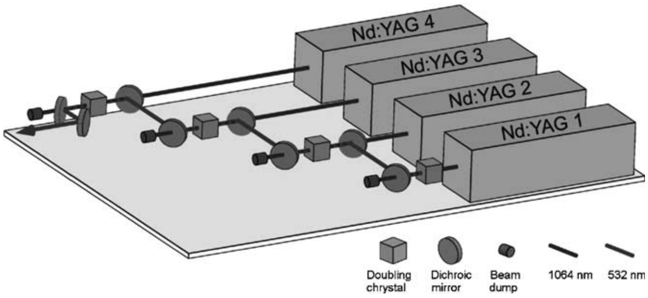


Fig. 3. Schematic overview of the Nd:YAG laser cluster, consisting of four Nd:YAG lasers of LTH.

(0.2 cm^{-1}) and has tunable wavelength with a harmonic generation unit attached to it. For excitation of the OH radical laser light with a wavelength close to 283 nm is used. This wavelength is generated using a Rhodamine 590 dye solution in methanol, with subsequent frequency doubling of the dye laser output using a KDP crystal.

The laser-induced fluorescence images were recorded using an ICCD camera (LaVision Flamestar II), with an image area of 576×384 pixels, and a dynamic range of 14 bit.

A Bunsen burner was used for these experiments, which established an axially symmetric cone-shaped stable flame, when operated with premixed reactants. The radius of the inner nozzle of the burner was $R = 5$ mm. The burner was fuelled by natural gas and air, and the mixture was fully premixed before exiting the nozzle with equivalence ratio, $\phi = 0.85$. The experimental set-up is shown in Fig. 4.

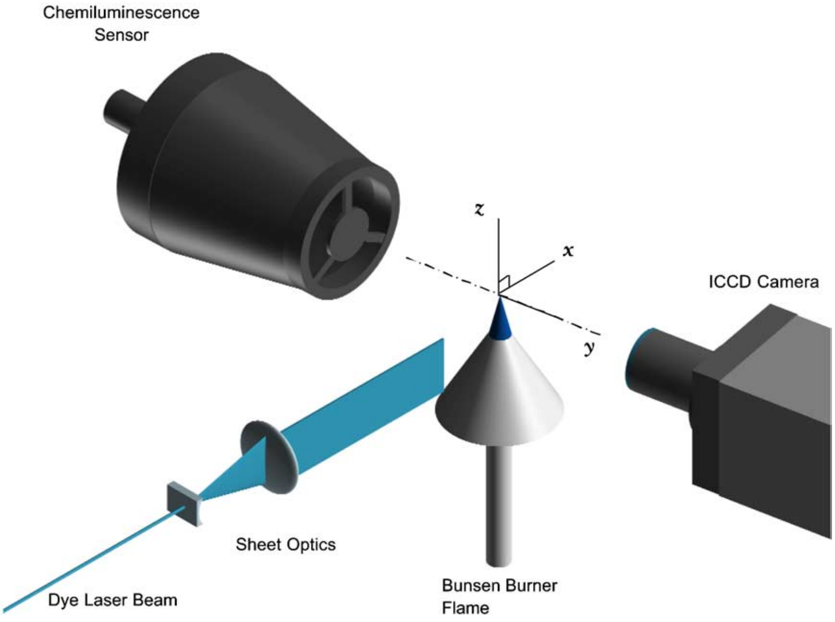


Fig. 4. The experimental setup for OH PLIF and chemiluminescence measurements in the Bunsen-burner flame.

The measurements of the excited species OH^*, CH^* and C_2^* were performed at a distance of $z = 5$ mm downstream of the exit of the burner. Radial profiles of chemiluminescencent intensity were measured in two orthogonal directions at this axial location. For the first direction, the probe volume of the sensor was traversed along the x-axis (Fig. 4), reached the reaction zone and collected emitted light ‘‘tangentially’’ to the flame sheet and moved across until it reached the reaction zone on the other side of the burner axis. The chemiluminescence intensity measurements when the probe volume was inside the reaction zone were very little influenced by background emissions and, therefore, the measurements were mainly due to the reaction in the probe volume and this radial profile is associated with ‘*point measurements*’.

A typical x-direction profile (‘‘*point measurement*’’) of the CS is shown in Fig. 5, where the chemiluminescence intensity from OH^* is compared with the results from the OH PLIF measurements, at the same height above the burner tip ($z = 5$ mm). The figure shows that, for both measurement techniques, two intensity maxima are located at the same ‘‘radial’’ (i.e. x location) positions (symmetrical about the burner axis). However, the PLIF measurements show a wider OH intensity profile outside the reaction zone. However, this is expected since the PLIF technique excites the ground state OH radicals, which are abundant downstream of the reaction zone, in the post-flame gases. The CS, on the other hand, detects only the natural chemiluminescence from the excited radicals formed in the reaction zone. Therefore, since there is no ‘‘combustion’’ in the post-flame gases, the chemiluminescence intensity from OH^* radicals is very low outside the reaction zone. The good agreement for the radial location of the intensity maxima from the LIF and the CS measurements indicates that this is the location of the reaction zone. Nevertheless, the CS measurements indicate a larger intensity around the centerline of the burner where a reaction zone is certainly not present. This signal is due to the collection of light from the flame present outside the probe volume, in front and/or behind the probe volume.

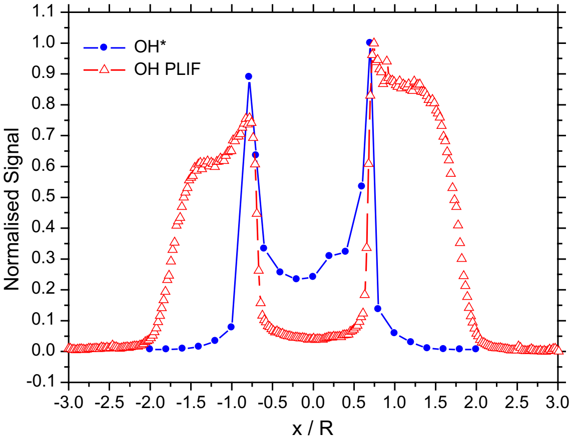


Fig. 5. Comparison between the OH and OH^* intensities measured by OH PLIF and the chemiluminescence sensor respectively. The intensities are normalised with the maximum intensity measured with each instrument and the radial distance is normalised with the burner radius.

4. Measurements with the CS in an atmospheric pressure micro-gas turbine combustor

In order to test the ability of the CS to measure in an environment closer to that of a practical gas turbine, we evaluated the sensor's ability to measure the degree of premixedness of the fuel/air mixture, in a modified micro-gas turbine combustor, running at atmospheric pressure. Unfortunately, the limited optical access to the combustor chamber did not allow us to resolve the flame front, since we could not measure tangentially to the flame, but the ability of the sensor to measure the degree of premixedness under different operating conditions—including a humidified flame—was shown.

The combustion chamber originates from a Volvo micro-gas turbine (VT 40), with operating characteristics for the engine at full load being 4 bar (combustor pressure), 147 kW (thermal power), 600 °C (inlet air temperature) and 355 g/s (air mass flow). The combustor was modified to operate at atmospheric pressure with the corresponding values being 45 kW, 372 °C and 88 g/s, respectively. The combustor operated with natural gas. The inlet air stream could be humidified up to 20% at full load before entering the combustor chamber.

Natural gas was supplied to the pilot burner (diffusion flame) and to the main burner (premixed flame). Consequently, the combustion chamber had the capability for both premixed and non-premixed operating modes. The pilot flame can be characterized as a slightly lean diffusion flame. A part of the air entered through the primary swirler and was mixed with the fuel before it entered the combustion chamber, and this is shown schematically in Fig. 6. The pilot flame was attached to the outer ring of the pilot burner. Before the main flame was swirl-stabilized in the combustor chamber, the fuel/air mixture was premixed. The mixing process started in the secondary swirler, in which the natural gas was perpendicularly injected to the air stream; additional air was added

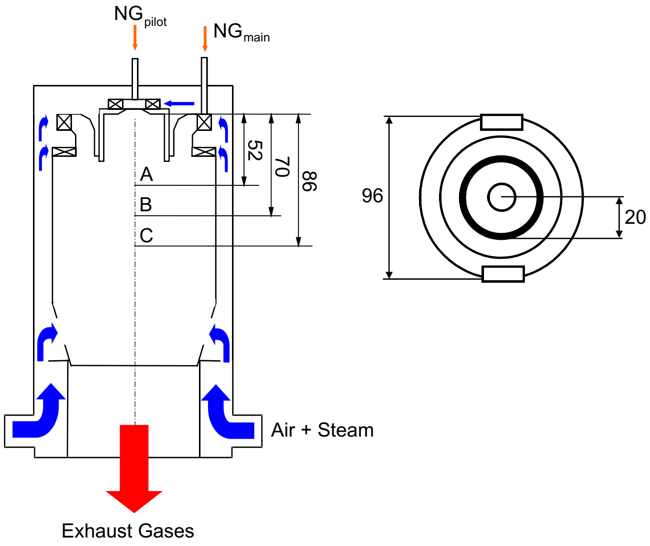


Fig. 6. Left: Flow schematic of the atmospheric combustion rig. Right: The combustion chamber seen from the exhaust gases end.

to the stream in the tertiary swirler. The secondary and tertiary swirlers are counter flowing, which is likely to improve mixing.

Moreover, the combustor has been equipped with two side windows and one bottom window, all in fused silica, providing optical access to the combustion zone. The optical access made it possible to perform measurements with laser/optical-based techniques. In Fig. 7 one of the side windows of the combustor is depicted. The side windows of the combustion chamber, were situated on either side of the diameter of the chamber and were double walled. These windows made it possible to achieve optical access from the fuel/air inlet in the primary zone down to the dilution zone. The window at the bottom of the flue gas channel allowed additional optical imaging of both the flame structure and the flow field. The measurements with the CS were performed through the side window shown in Fig. 7.

During the experiments the atmospheric combustor was operated at three conditions, referred to as GT1, GT2 and GT3, the characteristics of which are presented in Table 1. The two first cases, GT1 and GT2, were without humidified air, whereas in GT3, 8.1 kg H_2O/kg air is used. For all cases, the pilot flame was used *only* during the ignition of the burner. Measurements were performed only under *nominally* premixed fuel/air operation of the combustor, although partially premixed mixture was expected to occur during combustion in the chamber and the measurements presented below confirmed this expectation.

The measurements of the chemiluminescent intensity of the excited radicals of OH^*, CH^* required for measurement of the equivalence ratio, ϕ, were performed at three different planes, A, B and C, and for three different operating conditions of the burner. Plane A was closer to the

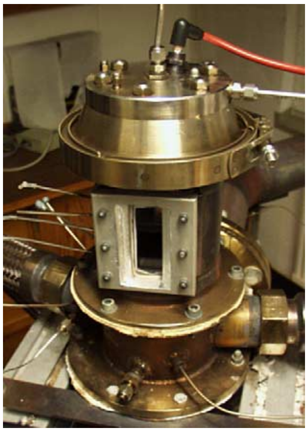


Fig. 7. The side window of the atmospheric combustion chamber.

Table 1
The characteristics for each of the three operating conditions

Case	m_{air} [g/s]	m_{fuel} [g/s]	T_{in} [°C]	Ω [kg H_2O/kg air]	$T_{exhaust}$ [°C]
GT1	83	0.64	340	0	691
GT2	83	0.615	341	0	671
GT3	83	0.637	330	8.1	667

swirlers of the burner at a distance of 52 mm from the exit of the swirlers, and planes B and C were at 70 and 86 mm below this exit, respectively. The position of the different planes where measurements were performed can be seen in Fig. 6.

The optical access of the side windows was nominally 30 mm, but was reduced to approximately 27 mm because of the insulation material that was used for the placement of the windows. This arrangement made impossible chemiluminescence sensor measurements tangentially to the flame, which would have given the best spatial resolution. So, during the measurements the CS probe volume was placed as close to the flame front as possible, but signal was collected from the background before and after the probe volume.

The CS was traversed along the *x*-axis (Fig. 8), and its probe volume was focused inside the burner at a distance of 40 mm from the outer window, which is equal to $r/R_B = 0.24$, where $R_B = 50$ mm is the radius of the inner flame tube of the burner. The different stations where the measurements were performed can be seen visualized by the median dotted line (red dotted line in the web version) shown in Fig. 8.

The measured mean values of the equivalence ratio, for the operating condition GT1, and for the different planes A, B and C, are shown in Fig. 9. The corresponding distributions of the probability functions (PF) of the equivalence ratio of the reacting mixture, ϕ, can be seen in Fig. 10. The values of equivalence ratio of the fuel/air mixture are richer at planes A and C, and slightly leaner at plane B. The same trend was observed for the operating condition GT2, shown in Fig. 11. The operating conditions GT1 and GT2 are quite similar in composition, so no large variations in the time-averaged value of the measured equivalence ratio would be expected. The difference in the measured mean value between operating conditions GT1 and GT2 was of the order of 0.05. The distributions of ϕ are wider at plane A, presumably because the measurements were closer to the mixing region of the fuel/air streams, confirming that the mixture was only partially premixed. The observed discrepancies in the PF of ϕ at the edge of the profiles, i.e. for $r/R = -0.6$ of Fig. 10, plane A, are caused by the limited optical access of the CS at the edges of the combustion chamber.

A humid CH_4/H_2O/air flame ("wet" flame) was also studied during the experiments at the atmospheric combustor. The percentage of humidity was 8.1% (kg H_2O/kg air). The air flow rate

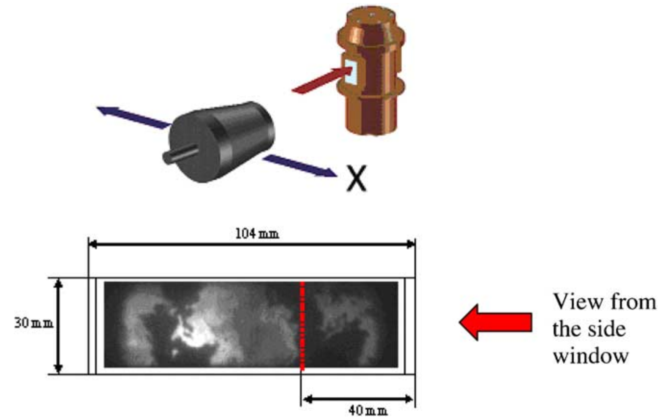


Fig. 8. Measurement stations of the chemiluminescence sensor.

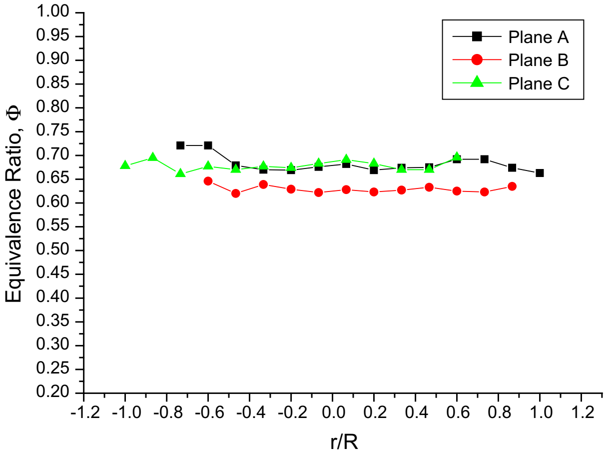
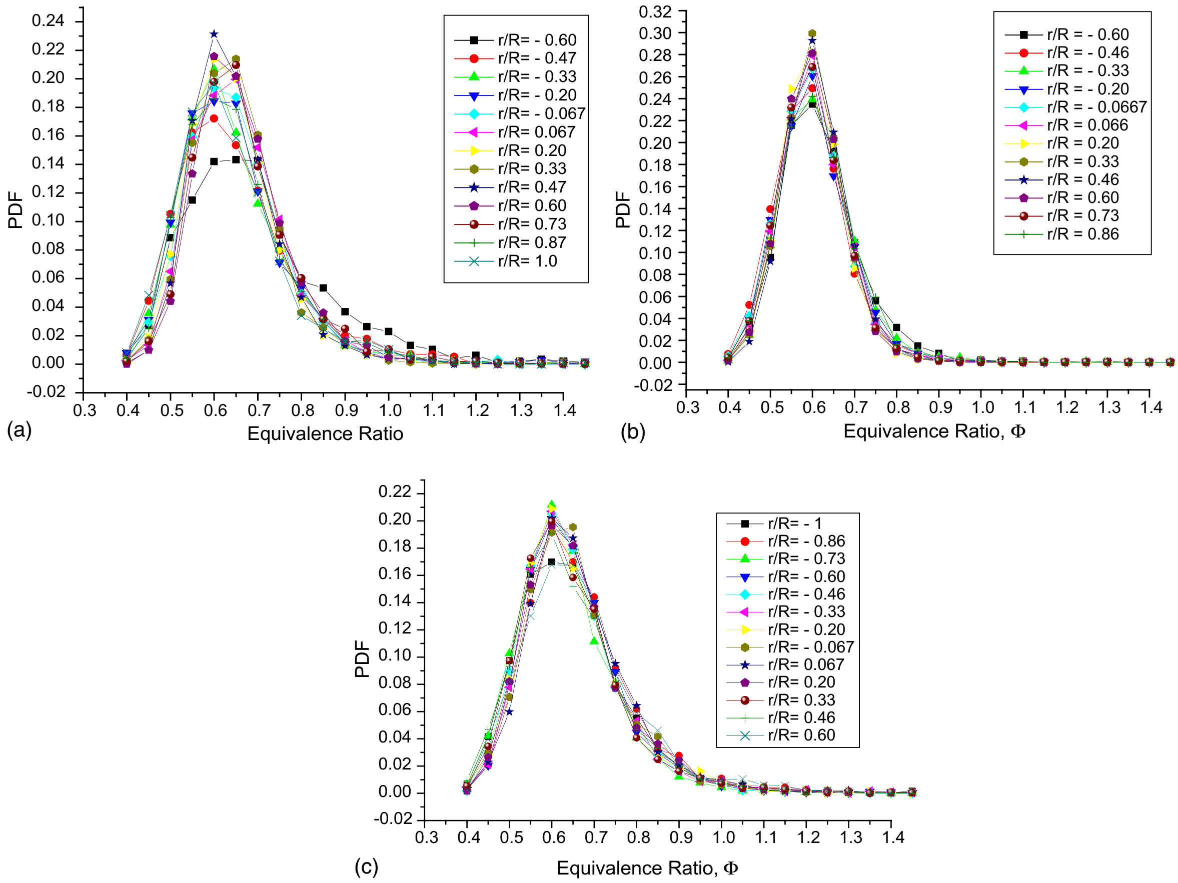


Fig. 9. Mean values of equivalence ratio for the operating condition GT1, at planes A, B and C.



(a)

(b)

(c)

Fig. 10. PF distributions of the equivalence ratio for operating condition GT1, at planes A, B and C.

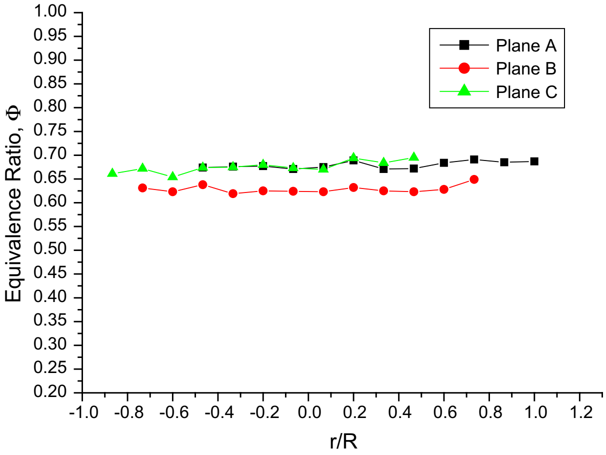


Fig. 11. Mean values of equivalence ratio for operating condition GT2, at planes A, B and C.

and the fuel flow rate were exactly the same as for operating condition GT1, so the results from the humid flame are plotted together with the results for operating condition GT1 at plane C in Fig. 12, and in Fig. 13 the PF of the equivalence ratio are presented. It can be observed that there is a good agreement between the mean values of equivalence ratio of the humidified flame and the CH_4/air flame, suggesting that the addition of water at the combustor did not influence the ratio of the OH^*/CH^* excited radicals, and therefore the equivalence ratio. Another encouraging indication from these measurements is that the CS has the ability to measure in a humidified flame environment.

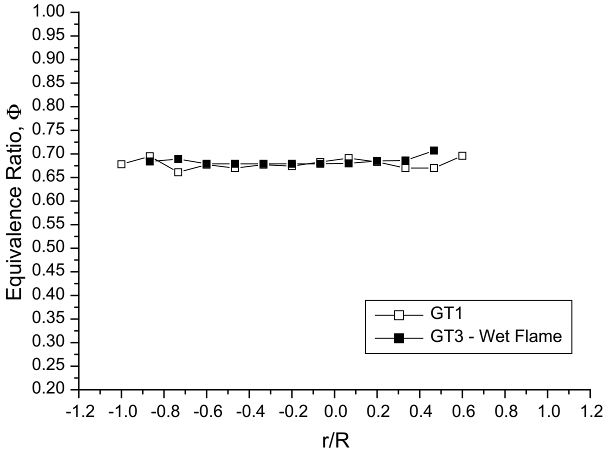


Fig. 12. Distribution of the mean values of the equivalence ratio for the humidified flame and the operating condition GT1, measured at plane C.

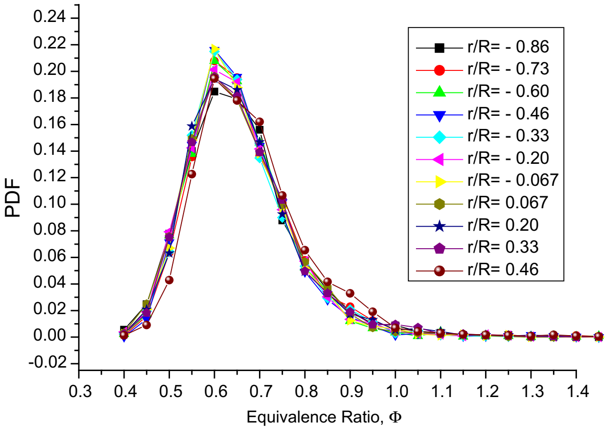


Fig. 13. PF curves for the values of equivalence ratio for the operating condition GT3—humidified flame, measured at plane C.

5. Conclusions—Future work

A chemiluminescence-based technique that has the potential of measuring flame equivalence ratio in a real gas turbine environment was presented in this paper. A Cassegrain telescope optical arrangement was used to provide spatially and temporally resolved intensity measurements, which detected the local flame structure. The calibration of the technique for natural-gas-fuelled flames under atmospheric pressure and variable strain rate has been described. The OH^*/CH^* intensity ratio has the potential for measuring flame equivalence ratio of the reacting mixture in flames at atmospheric pressure burning natural gas. The technique can measure equivalence ratio in lean and stoichiometric mixtures, but higher uncertainties appear for rich mixtures.

1. The evaluation of the CS with OH PLIF has shown that both techniques detected the reaction zone at the same radial position in a Bunsen burner.
2. However, the spatial resolution of the Chemiluminescence sensor is good when the probe is traversed such that the flame sheet is tangential, rather than normal to, the flame. This is due to the fact that when the probe volume of the sensor is located at the center of the flame, the spatial resolution of the sensor is reduced, because the sensor collects light from regions in front and/or behind the probe volume. This may not be a problem if generous optical access can be made available allowing measurement tangentially to the flame front, but can become a serious drawback when such optical access is not available.
3. The ability of the sensor to measure the equivalence ratio in a micro-gas turbine combustor working at atmospheric pressure but otherwise under conditions similar to that of a real gas turbine (air and fuel flow rates, preheating and humidified flame) show that the sensor has the potential for monitoring the degree of premixedness of reacting fuel and air in stationary gas turbine combustors, where operation with lean premixed mixtures is important for reduction of NO_x emissions.

The important effect of pressure to the existing natural gas calibration of the CS is going to be studied in the future, in order to measure the fuel/air degree of premixedness at an industrial gas turbine environment.

Acknowledgement

The authors are grateful to the European Community for sponsoring this work, under contract No ENK5-CT2000-00115.

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