Temperature measurement of plasma-assisted flames: comparison between optical emission spectroscopy and 2-color laser induced fluorescence techniques

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Abstract
Accurate thermometry of highly reactive environments, such as plasma-assisted combustion, is challenging. With the help of conical laminar premixed methane-air flames, this study compares two thermometry techniques for the temperature determination in a combustion front enhanced by nanosecond repetitively pulsed (NRP) plasma discharges. Based on emission spectroscopic analysis, the results show that the rotational temperature of CH(A) gives a reasonable estimate for the adiabatic flame temperature, only for lean and stoichiometric conditions. The rotational temperature of $N_2(C)$ is found to significantly underestimate the flame temperature. The 2-color OH-PLIF technique gives correct values of the flame temperature.

Introduction

As the temperature is an essential parameter in combustion science, decades of optical diagnostic improvements have led to several accurate techniques, which offer good spatial and temporal resolution of measurements. On the other hand, from the global need for decreasing pollutant emissions and fuel consumption, new combustion strategies have emerged. Each of these new combustion modes has specific requirements and issues, which often increase the complexity of experimental investigations. For example, plasma-assisted combustion studies have shown promising results for extending the flammability limits [1] or for controlling combustion dynamics [2]. Unfortunately, accurate thermometry of highly reactive environments such as plasma-assisted flames remains challenging.

With the help of well-known laminar flames, this study compares two thermometry techniques for the temperature determination of premixed flames enhanced by nanosecond repetitively pulsed (NRP) plasma discharges. Based on Optical Emission Spectroscopy (OES) analysis of the emitting zone, the first thermometry technique has been validated for the temperature determination of non-equilibrium plasma discharges in air, at atmospheric pressure [3]. Thus, the rotational temperature of the N_2 (C) state gives a good estimate of the temperature of the gas. Similarly, as suggested by different authors [4,5], the rotational temperature of CH (A), naturally present in premixed hydrocarbon flames, could be used to determine the flame temperature. The second technique is based on the 2-color planar laser induced fluorescence (PLIF) of the hydroxyl radical, produced by both the plasma and the flame. Flame thermometry by 2-color OH-PLIF is commonly used in combustion science [6], and has been

recently validated for temperature determination in post-non-equilibrium plasma discharges [7,8].

Experimental setup

The experimental setup comprises a Bunsen burner, able to stabilize conical methane-air premixed flames with equivalence ratios varying from 0.8 to 1.3. The outlet of the burner is a quartz tube of 10-mm inlet diameter and 1-mm thickness. The flame is stabilized over this tube. In order to keep the size of the flame constant, the velocity of the mixture flow is kept fixed at four times the laminar flame speed, taken from [9].

The NRP discharges are generated by a solid state pulse generator (FID Model FPG 10-30NM10) in the bottom part of the flame between a copper-tape, wrapped at the outlet of the tube, and a stainless steel ring of 24-mm inlet diameter, 1-mm thickness, centered on the burner and placed 5 mm above the tube (see Fig. 1). The nanosecond pulses are up to 6 kV in amplitude, 10 ns in duration, and are applied at a repetition rate of 30 kHz.
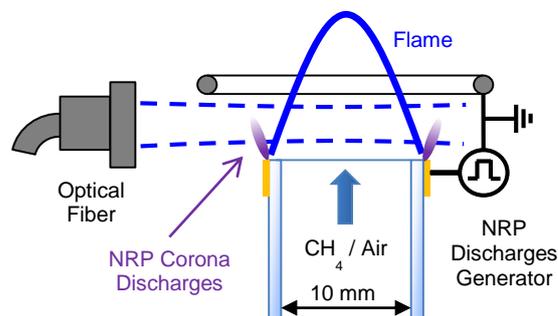

Figure 1: Experimental setup with spatial location of the OES measurements.

In order to ensure that the flame temperature was close to the adiabatic flame temperature, preliminary measurements were performed with R-type thermocouples of 125 µm diameter (Omega P13R). These measurements were made 5 mm above the quartz tube for equivalence ratios of 0.8 and 0.9. The thermal losses were taken into account and the temperatures was corrected accordingly [10]. The difference between the measured flame temperature and the adiabatic temperature was found to be less than 20 K, validating our setup for the evaluation of temperature measurement techniques (see blue triangles on Fig. 6).

Diagnostics

The OES measurements were performed with an Acton SpectraPro 2750i spectrometer (focal length 750 mm, grating 1200 lines/mm) fitted with an ICCD Camera (PIMAX Princeton Instruments). The spectral resolution was 0.11 nm. The light emission from the bottom part of the flame (see Fig. 1) was collected through an optical fiber (Ocean Optics QP600-4-UV-VIS) and the corresponding area of spatial integration was about 7 mm^2. The global integration time was set to 10 s.

The experimental spectr; were then compared with simulations performed with SPECAIR [11] for N_2 (C-B), and LIFBASE [12] for CH (A-X), for various rotational and vibrational temperatures. As this study focuses on the gas temperature, only the rotational temperatures are presented and discussed in this paper.
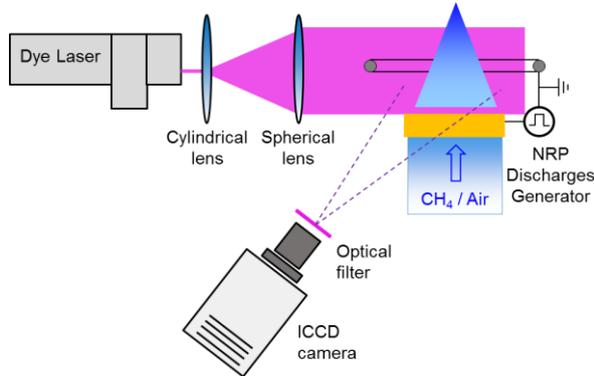

Figure 2: Two-color OH-PLIF setup.

The studied flames were stationary; therefore, the 2-color PLIF systems comprised a single PLIF system (see Fig. 2). The measurements were not performed simultaneously but one after the other. Excitation was provided by a Continuum ND6000 Dye laser, pumped by a Nd:YAG laser. The two excitations lines (P_1(7) at 285.005 nm and Q_2(11) at 285.073 nm) were chosen according to the literature [13] for measurements of temperature around 2000 K:

$$\frac{\Delta E}{k} = 2046 \ K$$

where ΔE is the quantum energy difference between the two rotational levels of excitation and k is the Boltzmann constant. The measurements were realized in the linear regime of fluorescence, with an energy per laser pulse of 6.5 mJ and a laser sheet of 10 mm height.

The fluorescence signal (S_f) was imaged onto a 512 x 512 px^2 ICCD camera equipped with a 10-nm band pass filter (Asahi Spectra ZBPA310). In order to increase the signal-to-noise ratio, and to smooth the shot-to-shot laser fluctuations, each fluorescence image is an accumulation of 150 acquisitions. The temperature field was then obtained from the fluorescence images by:

$$T = \frac{2046}{log \left(C \frac{S_{f1}}{S_{f2}} \right)}$$

where T is the temperature, S_{f1} and S_{f2} are the fluorescence signals related to the excitation lines 1 and 2, and C is a calibration constant. This constant has been determined by fitting the temperature of the stoichiometric flame at 2222 K.

Results and discussion

Examples of measured and simulated spectra of CH (A-X) and N_2 (C-B) are presented in Figs. 3 and 4, respectively. The uncertainty in temperature determination is given by the two best lower and upper fits of each experimental spectrum. The CH (A-X) spectra were recorded without enhancement of the flame by NRP discharges, while the N_2 (C-B) spectrum was obtained for enhancement of the premixed flame by NRP corona discharges, as defined by Pai *et al.* [14], *i.e.* with a negligible heating of the gas by the plasma. The influence of the equivalence ratio on the rotational temperatures of N_2 (C) and CH (A) is summarized in Fig. 6.

The results show that the rotational temperature of CH (A) (red bullets in Fig. 6) gives a good estimate of the adiabatic flame temperature, only for lean and stoichiometric conditions. For rich mixtures, the adiabatic flame temperature decreases while the rotational temperature of the CH (A) state increases with the equivalence ratio. Therefore, even for a flame without plasma discharges, this molecule cannot be used for the temperature determination of the combustion zone.
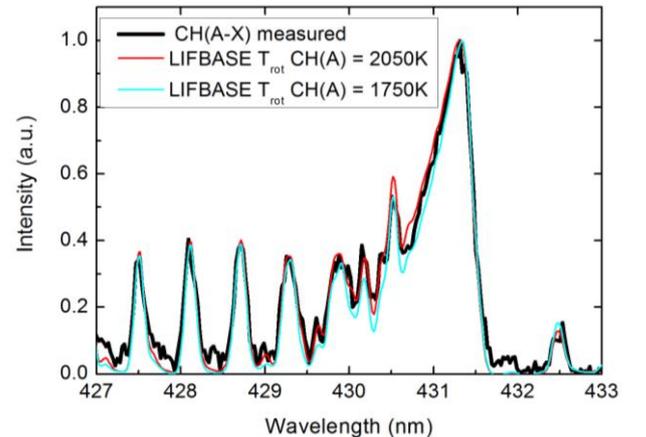

Figure 3: Example of comparison between measured CH (A-X) spectra and simulations performed with LIFBASE [10].

The rotational temperatures of N_2(C) were measured at the onset of light emission from the excited nitrogen.

Thus, it is assumed that no ultra-fast heating of the gas by the plasma has occurred [3]. The rotational temperature of N_2 (C) underestimates the adiabatic flame temperature for the whole range of equivalence ratios investigated (green triangles in Fig. 6). The rotational temperature of N_2 (C) varies from 1500 to 1650 K. However, as shown in Fig. 7, where the normalized adiabatic temperatures and rotational temperatures of N_2 (C) are compared, the relative effect of the equivalence ratio is correctly captured. The light emission from the N_2 (C) state occurs during the first tens of nanoseconds after starting to apply the high voltage pulses. During this time, even if the plasma is highly nonequilibrium, the rotational temperature of this excited state cannot be less than the translational temperature of the gas. On the other hand, there is no known process of NRP discharges cooling the gas. Therefore, a rotational temperature lower than the adiabatic flame temperature could have two possible explanations:

- The emitting N_2 (C) is not located in the hottest part of the flame front, neither in the burnt gases, *i.e.* the NRP discharges are not located in the domain of maximum temperature.
- Due to thermal losses at the outlet of the quartz tube, the flame temperature where the NRP discharges are located is significantly lower than the adiabatic flame temperature.

Further investigations will be necessary to determine which of these two mechanisms is responsible for the observed disagreement. Until then, it is not possible to conclude about the use of N_2 (C-B) spectra for determining the flame temperature.
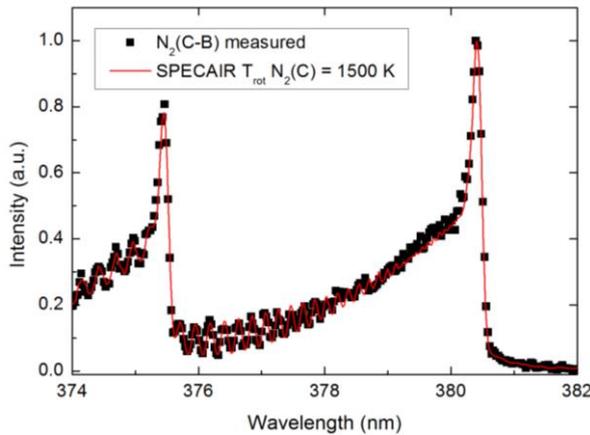

Figure 4: Example of comparison between measured N_2 (C-B) spectra and simulations performed with SPECAIR [9].

An example of the temperature field obtained by 2-color OH-PLIF for a flame, without enhancement by NRP discharges, with an equivalence ratio of 1, is presented in Fig. 5. Only the area inside the cyan rectangle has been taken into account for the temperatures presented as cyan squares in Fig. 6. The error bars correspond to the root mean square of the temperature fluctuations in this area. The 2-color OH-PLIF technique gives a good estimate of the adiabatic

flame temperature. The values as well as the trend as the equivalence ratio is varied are correctly reproduced. Thus this technique is appropriate for temperature measurements of premixed flames.
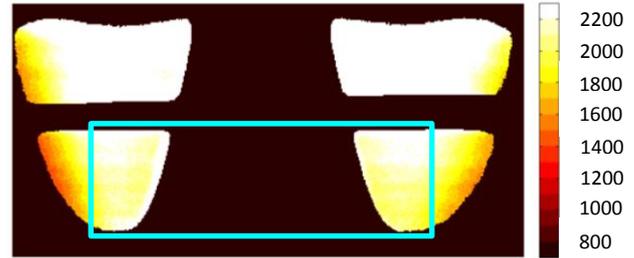

Figure 5: Example of a temperature field (in Kelvin) obtained with the 2-color OH-PLIF technique for a premixed stoichiometric CH_4/air flame.
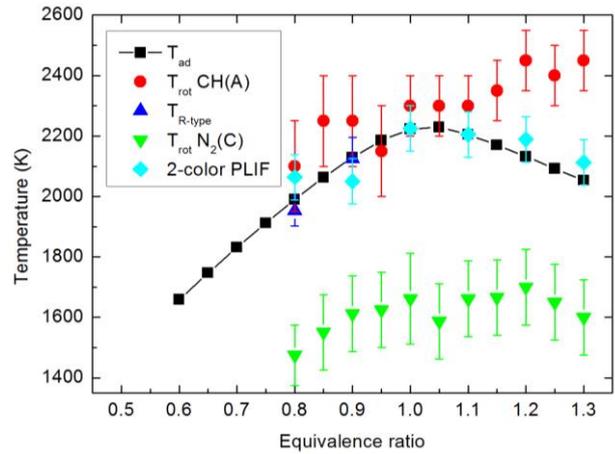

Figure 6: Comparison between different temperature measurement techniques for equivalence ratios varying from 0.8 to 1.3.
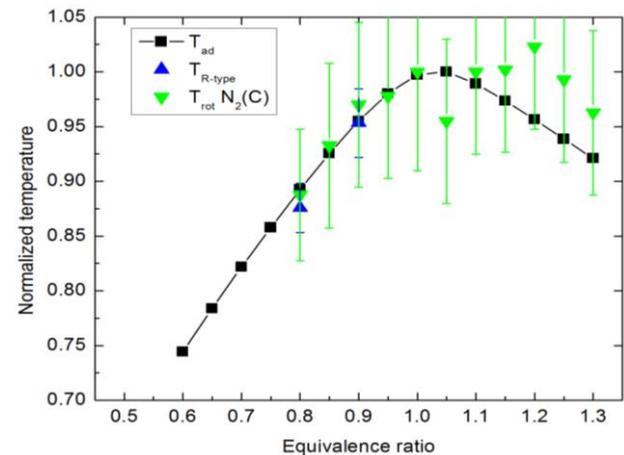

Figure 7: Comparison between normalized adiabatic flame temperatures and rotational temperatures of N_2 (C) for equivalence ratios varying from 0.8 to 1.3.

Figure 8 presents a comparison of the temperatures measured by 2-color OH-PLIF of plasma-assisted

methane air flames, and flames without enhancement by NRP discharges. The PLIF system was synchronized with the NRP discharges and the temperature measurements were made 1 μs after applying NRP corona discharges. The fluorescence images are accumulated over 150 acquisitions. Figure 8 indicates that for lean, stoichiometric and rich mixtures, the heating by the plasma is less than 150 K. However, the uncertainty is too big to quantify accurately the heating induced by the NRP corona discharges.
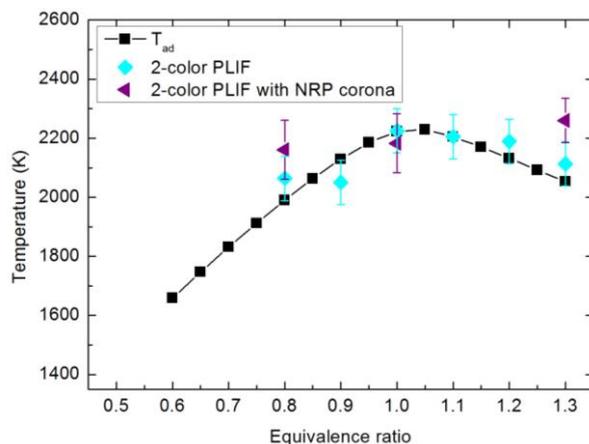

Figure 8: Comparison of temperatures of flames and temperatures of flames enhanced by NRP corona discharges, measured by 2-color OH-PLIF.

Finally, comparisons with other techniques such as Coherent Anti-Stokes Raman Scattering (CARS) [15] or Spontaneous Raman Scattering (SRS) [16] on a similar canonical configuration would be of great interest.

Conclusions

In this study, conical premixed methane-air flames have been used for the evaluation of two thermometry techniques, suitable for plasma-assisted combustion investigations.

The results show that even if the rotational temperature of CH (A) gives a good estimate for the adiabatic flame temperature, for lean and stoichiometric conditions, this molecule cannot be used as a good tracer of the adiabatic flame temperature.

On the other hand, the rotational temperature of $N_2(C)$ is found to underestimate the flame temperature by about 600 K. However, the effect of the equivalence ratio on the temperature evolution has been caught. Further investigations are necessary to conclude about this thermometry technique.

The 2-color OH-PLIF technique gives correct values of the flame temperature. However, with an accuracy of about 100 K, this technique is not sufficiently precise for determining the heating induced by non-thermal plasmas such as NRP corona or glow discharges.

References

[1] Starikovskiy, A., Aleksandrov, N. "Plasma-assisted ignition and combustion", *Progress Energy Combust. Sci.* 39(1), 61-110 (2013)

[2] Lacoste, D.A., Moeck, J.P., Durox, D., Laux, C.O., Schuller, T., "Effect of nanosecond repetitively pulsed discharges on the dynamics of a swirl-stabilized lean premixed flame", *J. Engineering Gas Turbines Power* 135: 101501-1 (2013)

[3] Rusterholtz, D.L., Lacoste, D.A., Stancu, G.D., Pai, D.Z., Laux, C.O., "Ultrafast heating and oxygen dissociation in atmospheric pressure air by nanosecond repetitively pulsed discharges", *J. Phys. D: Appl. Phys.* 46: 464010 (2013)

[4] Brockhinke, A., Krüger, J., Heusing, M., Letzgus, M., "Measurement and simulation of rotationally-resolved chemiluminescence spectra in flames", *Appl. Phys. B* 107, 539-549 (2012)

[5] Bruggeman, P.J., Sadeghi, N. Schram, D.C., Linss, V., "Gas temperature determination from rotational lines in non-equilibrium plasmas: a review", *Plasma Sources Sci. Technol.* 23: 023001 (2014)

[6] Eckbreth, A.C., "Laser diagnostics for combustion temperature and species", *Combust. Sci. Technol. Book Series*, 2nd Edition (1996)

[7] Sainct, F.P., Lacoste, D.A., Kirkpatrick, M.J., Odic, E., Laux, C.O., "Temporal evolution of temperature and OH density produced by nanosecond repetitively pulsed discharges in water vapour at atmospheric pressure", *J. Phys. D: Appl. Phys.* 47, 075204 (2014)

[8] Yin, Z., Adamovich I.V., Lempert, W.R., "OH radical and temperature measurements during ignition of H-2-air mixtures excited by a repetitively pulsed nanosecond discharge", *Proc. Combust. Inst.* 34 (2), 3249-3258 (2013)

[9] Mazas, A.N., Fiorina, B., Lacoste, D.A., Schuller, T., "Effects of water vapor addition on the laminar burning velocity of oxygen-enriched methane flames", *Combust. Flame* 158 (12), 2428-2440 (2011)

[10] Hindasageri, V., Vedula, R.P., Prabhu S.V., "Thermocouple error correction for measuring the flame temperature with determination of emissivity and heat transfer coefficient", *Rev. Sci. Instruments* 84, 024902 (2013)

[11] Laux, C.O., "Radiation and Nonequilibrium Collisional-Radiative Models," von Karman Institute Lecture Series 2002-07, Physico-Chemical Modeling of High Enthalpy and Plasma Flows, eds. D. Fletcher, J.-M. Charbonnier, G.S.R. Sarma, and T. Magin, Rhode-Saint-Genèse, Belgium (2002)

[12] Luque, J., Crosley, D.R.,"LIFBASE: Database and spectral simulation (version 1.5)", *SRI International Report* MP 99-009 (1999)

[13] Saiki, Y., Kurimoto, N., Suzuki, Y., Kasagi, N., "Active control of jet premixed flames in a model combustor with manipulation of large-scale vertical structures and mixing", *Combust. Flame* 158, 1391-1403 (2011)

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[14] Pai, D.Z., Lacoste, D.A., Laux, C.O., "Transitions between corona, glow, and spark regimes of nanosecond repetitively pulsed discharges in air at atmospheric pressure" *J. Appl. Phys.* 107: 093303 (2010)

[15] Lanier, s., Bowman, S., Burnette, D., Adamovich, I.V., Lempert, W.R., "Time-resolved temperature and O atom measurements in nanosecond pulse discharges in combustible mixtures", *J. Phys. D: Appl. Phys.* 47(44): 445204 (2014)

[16] Lo, A., Cessou, A., Boubert, P., Vervisch, P., "Space and time analysis of the nanosecond scale discharges in atmospheric pressure air: Gas temperature and vibrational distribution function of N_2 and O_2", *J. Phys. D: Appl. Phys.* 47: 115201 (2014)