Laser induced fluorescence investigation of the chemical impact of nanosecond repetitively pulsed glow discharges on a laminar methane-air flame

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Abstract

This paper reports on an experimental investigation of the chemical impact of nanosecond repetitively pulsed (NRP) glow discharges on a laminar methane-air flame. The chosen configuration was a lean wall stabilized flame where NRP discharges were generated across the flame front. After careful selection of the excitation lines, planar laser induced fluorescence of OH and CH was conducted. Comparisons between the OH and CH fluorescence of a base flame (without plasma actuation), and those obtained during the steady state and the transient regimes of plasma actuation, were performed. First it is shown that during the steady state regime, the intensity of OH and CH fluorescence in the flame could be increased by up to 40% and 10%, respectively. In addition, the life time of OH fluorescence in the discharge channel was estimated to be between 3 and 4.5µs. The transient regime at the beginning of plasma actuation showed that the flame began to be affected by the discharges long before OH fluorescence could be detected in the discharge channel, upstream of the flame. After 40ms of plasma actuation, OH intensity began to increase simultaneously in both the flame and the discharge area. Based on current knowledge of nanosecond discharge chemistry, explanations for these results are proposed.

Keywords: Plasma-assisted combustion, NRP discharges, OH PLIF, CH PLIF, Non-equilibrium plasma

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1. Introduction

In the last two decades, non-thermal plasmas produced by nanosecond repetitively pulsed (NRP) discharges have shown promising results for plasma-assisted combustion applications [1–3]. For example, they have been successfully used to improve the ignition of flammable mixtures [4–8], enhance the lean blow-off limit [9–11], or control combustion instabilities [12–14]. The results obtained were usually attributed to thermal, chemical, and/or transport effects of the discharges [3].

Under conditions relevant to combustion applications, two regimes of NRP discharges can be distinguished, the NRP glows and the NRP sparks [15]. NRP sparks have a strong thermal and hydrodynamic effect [16, 17], coupled with a chemical actuation of the gas [17, 18]. On the other hand, the NRP glows have a minimal thermal and hydrodynamic effect [19], and the chemical impact is dominant. This weak regime of the NRP discharges has recently attracted more attention because it can effectively influence combustion [14], with a small penalty in terms of nitric oxide (NO) emission [8, 20, 21].

Chemical characterization of NRP discharges for plasma-assisted combustion applications is ongoing [22]. Targeted species are produced by discharges generated in fuel-air mixtures, with a potential impact on combustion chemistry. For example, atomic oxygen (O) [23–25] and atomic hydrogen (H) [26], have been measured in different plasma-assisted combustion configurations. Similarly, several studies have reported on the measurements of methyldyne radicals (CH) [27] and hydroxy radicals (OH) [5, 24, 28–30]. However, these studies focused mainly on ignition or on enhancement of combustion by non-equilibrium discharges at sub-atmospheric pressures. To the best of our knowledge, the effect of NRP glows on the OH and/or CH distributions of a fully developed premixed flame, at atmospheric pressure, has not yet been investigated. These results would be of interest to clarify the plasma action on flame dynamics, as well as for challenging the modeling efforts on the coupling between non-equilibrium plasma mechanisms and combustion chemistry.

The main objective of this study is to characterize the impact of NRP glow discharges on the OH and CH distributions of a premixed flame at atmospheric pressure. To facilitate future comparisons with numerical simulations, an axi-symmetric two-dimensional configuration was chosen, with NRP glows generated across the flame front. With this configuration, each experiment provides information about the chemical effect of NRP glows on the fuel-air mixture, the flame front, and the burnt gases.

2. Experimental Setup and Procedure

The experimental setup included a plasma-assisted combustion (PAC) burner equipped with electrical diagnostics, and a planar laser-induced fluorescence (PLIF) system. Both systems are detailed in this section, along with the experimental procedure followed.

2.1. PAC Burner

A schematic of the PAC burner used is presented in Fig. 1. It consisted of a stagnation plate burner where a lean methane (CH₄)-air laminar flame could be stabilized. The diameter of the burner outlet was 10 mm and the distance between the burner outlet and the quartz plate was 10 mm. The equivalence ratio was 0.70, and the bulk velocity at the outlet of the burner was 1.2 m/s. The thermal power released by the flame was then about 230 W. A co-flow of nitrogen (N₂) prevented the flame from attaching to the burner lip.

Figure 1: Schematic of the PAC burner with a photograph of the methane-air flame with NRP glows (not to scale).

Two pin electrodes were positioned 9 mm apart along the symmetry axis of the burner. The grounded electrode was a tungsten pin with a radius of curvature of about 50 µm, embedded in the quartz plate. The anode, connected to the high-voltage output of a nanosecond pulse generator (FID GmbH FPG Series), was a 100-µm diameter tungsten wire. When high-voltage pulses were applied to the electrodes, NRP glows formed along the centerline, perpendicularly to the local flame front (see Fig. 1). Note that the presence of the anode affected the flow field out of the nozzle, and in turn, the flame...
shape. The curvature of the base flame (without NRP glows) originated from a profile of the flow that was no longer top-hat.

In this experiment, NRP glows (as defined in [15]), were obtained by applying high-voltage pulses of 8 kV amplitude and 10-ns duration. They were applied at a pulse repetition frequency (PRF) of 10 kHz. Typical electrical measurements, obtained with a high voltage probe (Tektronix P6015A) and a current transformer (Pearson Model 6585) are shown in Fig. 2. The corresponding deposited energy, calculated as the integral of the product of voltage and total current, was around 90 ± 40 µJ. As the total current was used, due to the charge and discharge of the stray capacitance of the electrodes assembly, the instantaneous energy fluctuates. However, over the entire duration of a pulse, these fluctuations compensate and the value after about 100 ns corresponds to the energy deposited in the discharge. For a PRF of 10 kHz, this energy per pulse corresponded to an average electrical power deposited by the discharge of about 0.9 W, i.e., less than 0.5% of the flame thermal power.

![Figure 2: Typical voltage and current measurements of a single glow discharge and corresponding energy (PRF = 10 kHz).](image)

2.2. PLIF System

A conventional nanosecond PLIF system was used to investigate OH and CH distributions in the PAC burner. The second harmonic of a Nd:YAG laser (Continuum PL DLS 9010) was used to pump a dye laser (Continuum ND6000), coupled with a doubling system (Continuum UVT-3). The laser beam was first sampled for shot-to-shot energy monitoring and then shaped by means of cylindrical and spherical lenses, as a sheet of 10-mm height and about 200-µm thickness, passing by the central axis of the PAC burner. To obtain a homogeneous excitation, only the central region of the laser sheet was used.

Fluorescence was collected using an intensified CCD camera (Princeton PI-MAX 4 1024i) equipped with a f/2.8 100 mm UV lens. A LaVision bandpass filter centered at 320 nm, with 40-nm bandwidth, associated with a custom Semrock AFRL-0002 filter (described in [31]) was used to collect only the fluorescence signal, rejecting background emissions and laser elastic scattering and reflections. The exposure time was 200 ns for all conditions.

Synchronization between NRP glows and PLIF measurements was achieved with two Berkeley Nucleonics BNC 575 delay generators.

2.3. Procedure

Two different aspects of the actuation by NRP glows were investigated: (i) the transient response of the flame at the beginning of plasma actuation; (ii) the steady state regime for a continuous actuation by NRP glows.

For the transient response of the flame, the PLIF images of the base flame were recorded first. Then, assuming reproducible experimental conditions, phase-locked PLIF measurements were performed 300 ns after the second discharge, the 10th, the 100th, etc. After the 10,000th NRP glow discharge (i.e., after 1 s of plasma actuation), the steady state regime was reached. This delay of 300 ns after the discharges allowed for strong PLIF signal in the discharge area and absence of chemiluminescence from the plasma.

For the steady state condition, PLIF images of the base flame were also recorded. Then, NRP glows were applied continuously and, after a few seconds, phase-locked PLIF measurements were performed. Different delays between the discharge and the camera trigger were selected to explore and characterize the entire inter-discharge period.

To increase the signal to noise ratio, each PLIF image was obtained by averaging up to 250 single shots. For each experimental condition, five average PLIF images were recorded. The post-processing of these images consisted of background subtractions and color coding, only.

3. PLIF Excitation

Different excitation and detection schemes have been used for OH and CH detection in plasma-assisted combustion [22]. For example, Winters et al. [30], chose the OH $\Lambda^2\Sigma^+ - \chi^2\Pi (0,0)$ excitation (308 nm), for its high excitation and fluorescence rates, while Li et
al. [29], preferred the OH $A^2\Sigma^+ - X^2\Pi (1,0)$ excitation (281 nm), with a collection of the fluorescence signal from the (0,0) band, near 310 nm.

To facilitate OH and CH measurements with the same PLIF system, the present study measured CH through excitation and detection within the R-branch transitions in the CH $C^2\Sigma^+ - X^2\Pi (0,0)$ band [31]. Transitions in the C-X band are spectrally close to OH lines from the A-X (0,0) and (1,1) bands [32]. Accordingly, transitions in the A-X band were considered for the OH PLIF measurements.

To finalize the selection of the excitation lines, a wavelength scan between 311.28 and 311.55 nm was performed. The fluorescence of the flame enhanced by NRP glows in the steady state regime, 300 ns after a discharge, was collected.

Figure 3a shows the average fluorescence response in the flame area as a function of the excitation wavelength (red line), as well as Lifbase simulations of OH [33] (black dashed line), in the partially saturated regime of fluorescence, for a temperature of 1850 K. Simulations and experiments were in relatively good agreement.

In order to reach a relatively good agreement with the experimental spectrum, it was necessary to overpopulate the level $v' = 1$, corresponding to the fluorescence peak at 311.43 nm (gray dashed line).

Figures 3a and 3b allow the comparison of the relative fluorescence intensities in the flame and in the discharge. For the OH S21(2) line of the (1,1) band, the fluorescence was similarly intense in the discharge and in the flame. Therefore, this excitation wavelength was selected to allow measurements of OH in both areas for a same setting of the PLIF system. A second excitation line was also selected, the O12(4) of the (0,0) band. At low temperatures, this line should give the strongest fluorescence signal for the range of wavelength considered. Examples of PLIF images obtained for these two lines are shown in Figs. 4b and 4c.

Note that the fluorescence of CH (rotational lines in green) could not be appreciated in the analysis of Fig. 3. However, on the images, CH fluorescence could be observed in the flame front for an excitation at 311.438 nm, i.e., corresponding to the CH R2(10) and R1(10) lines (see Fig. 4a). Considering the laser line width (about 5 pm), it was not possible to separate the excitation of these two lines. In the following, the excitation line for CH will be referred as the CH R1(10).

In order to disregard the shot-to-shot variation of laser intensity, the saturated regime of fluorescence was preferred. For CH, the saturation was achieved with a laser energy larger than about 3 mJ per pulse, while for the

Figure 3: Average fluorescence signals in flame (a) and discharge filament upstream of flame (b), as a function of the excitation wavelength. Comparisons with simulations with Lifbase [33] for A-X band of OH (black and gray dashed line). Rotational lines of C-X band of CH are also indicated (green).

Figure 4: Examples of CH and OH PLIF images for different excitation lines, without plasma discharges (left column) and with NRP glow discharges (right column), at $t = 300$ ns. Color scale is arbitrary, but intensities are consistent.
two selected OH lines, a laser energy larger than about 6 mJ per pulse was necessary. Note that under the specified experimental procedure, the comparison of the relative intensity of PLIF images obtained with two different excitation lines has no meaning.

4. Results and Discussion

In this section, the steady state regime of plasma actuation is reported first. A second section details measurements conducted at the beginning of the plasma actuation, referred to as the transient response of the flame to plasma actuation. Results are provided in terms of spatially- and temporally-resolved intensity of fluorescence signal.

4.1. Steady State Actuation

For a continuous NRP glow actuation, the temporal evolution of OH and CH fluorescence was studied between two pulses. PLIF images were collected at different times after a discharge, starting from 300 ns to 98 µs. The jitter in these phase locked measurements was about ± 10 ns.

Figure 4 compares the PLIF images obtained for the base flame (left column) with those obtained in the steady state regime of plasma actuation, 300 ns after an NRP glow (right column). Even if the shape of the flame front was affected by the plasma actuation, for CH (Fig. 4a) the fluorescence intensity at the flame front was almost unchanged. Also, in the discharge filament upstream of the flame as well as in the burnt gases, no CH fluorescence was detected. In the OH PLIF images (Figs. 4b and 4c), the impact of the NRP glows on the fluorescence intensity in the flame was found to depend on the excitation line. In addition (and as expected from their selection), the intensity of fluorescence in the discharge channel was negligible for the O12(4) excitation line, compared to the fluorescence in the flame area, while for the S21(2) line, a similar fluorescence signal could be observed in both areas of the image.

By design, plasma actuation mainly affected the axis of the burner, therefore, the fluorescence intensities along the centerline of the burner are compared in Fig. 5, for different delays between discharges. During plasma actuation (red, blue and grey profiles), the OH and CH fluorescence intensity and distribution in the flame did not change during the forcing period. For this reason the flame is said to be in a steady state regime.

Figure 5: Fluorescence distribution along the centerline for different excitation wavelengths.

Compared to the base flame (black), the plasma actuation induced a small increase of about 10% of the CH fluorescence in the flame front (Fig. 5a). In addition, no fluorescence of CH could be detected in the NRP discharge area for any delay, upstream or downstream of the flame front. In Grisch et al. [27], for rich CH₄-air mixtures, two very thin zones of CH fluorescence, located at the periphery of the plasma channel, could be observed for nanosecond spark discharges inducing a gas heating of about 2500 K. However, no results associated with the fluorescence of CH were reported for lean mixtures, known to induce less CH fluorescence in flames [34]. Further investigation will be necessary to determine whether the lack of CH fluorescence in the NRP glow discharge area compared to nanosecond sparks originated mainly from a difference in plasma chemistry or from a difference in gas composition.

Effects of NRP glows on the OH fluorescence in the flame front and in the burnt gases were similar for both excitation lines (see Figs. 5b and 5c). Compared to the base flame, the OH fluorescence in the flame front increased by about 40%. In addition, a small increase of the OH fluorescence in the burnt gases, close to the cathode (height above the burner of 8–10 mm), could be noticed. However, in the discharge area upstream of the flame front (height above the burner of 0.5–2 mm), a relatively strong OH fluorescence signal was obtained with the S21(2) excitation line (Fig. 5c), while with the
O12(4) line (Fig. 5b), the strong fluorescence from the flame prevented proper rendering of the discharge.

Figure 6: Temporal evolution of average OH fluorescence intensity in discharge channel (height above the burner of 0.5–2 mm). Error bars represent extreme values obtained for five independent measurements.

Figure 6 shows the time evolution of the OH fluorescence in the discharge channel in the fresh gases, for the S21(2) and O12(4) excitation lines. In both cases the OH intensity followed an exponential decay with time constants of 3 \( \mu \text{s} \) and 4.5 \( \mu \text{s} \), respectively. These results are aligned with those in [35], for a CH\(_4\)-Air mixture at low gas temperature.

According to [35], OH lifetime has a strong dependence on the temperature and it can feature multiple peaks. The first peak is due to OH generation by plasma produced species, such as atomic oxygen, e.g. \( \text{O}(^1\text{D}) + \text{CH}_4 \rightarrow \text{CH}_3 + \text{OH} \). At higher gas temperature, a second peak can be observed. It is related to chain propagation/branching, occurring below the self-ignition threshold and limited by accumulation of intermediate species [35]. During ignition, a third peak occurs. The two latter peaks are not observed here – a single exponential-like decay is recorded instead.

Fig. 6 also shows that there was no significant OH accumulation in between pulses in the fresh gases ahead of the flame. The maximal fluorescence intensity was obtained within the first 0.5 \( \mu \text{s} \) after the discharge, regardless of the excitation line, and the signal fell below the detection level in less than 30 \( \mu \text{s} \). This result contrasts with other works, such as [27], where for a stronger discharge, the OH fluorescence was shown to peak later and exhibited a long plateau and slow decay. Finally, Fig. 6 shows that the discharges in the fresh gases did not ignite directly the gases. The effect of the discharges might consist in a very partial oxidation of the fuel, where small amounts of methanol and formaldehyde could be formed, leading to a temperature increase as well [36].

4.2. Transient Response of the Flame

Compared to the base flame, the OH and CH fluorescence distribution in the flame during plasma actuation differed significantly. The previous section showed that in the steady state regime of plasma actuation, the OH and CH fluorescence in the flame did not respond to individual discharges. However, it could be expected that flame changes would not occur instantly after the first discharge, making flame conformity to plasma actuation by NRP glows and interesting issue.

Figure 7 displays 1-mm slices of OH PLIF images around the centerline of the burner for the S21(2) excitation line. Each slice was imaged 300 ns after a different numbers of applied discharges (increasing from left to right). Thus, this figure presents the transient regime of OH fluorescence as a function of the number of applied discharges (or time after the beginning of plasma actuation).

After about 100 pulses (10 ms), the flame began to move upstream, in agreement with previous results obtained for laminar flame subjected to forcing by NRP glow discharges [14]. The flame reached its nearly final location after about 350 discharges (35 ms). Only after the flame began to stabilize (400 discharges or 40 ms), OH fluorescence appeared in the discharge channel, upstream of the flame. Between the 400th and 500th discharge, the intensity of OH fluorescence–both in the flame and in the discharge–increased. This increase continued for about 600 ms; afterward fluorescence intensity remained relatively constant.

This temporal analysis of OH fluorescence at the beginning of plasma actuation by NRP glow discharges shows that: (i) the flame began to be affected by NRP
glow discharges before OH fluorescence upstream of the flame front reached a detectable level, and (ii) even if OH can be rapidly produced by NRP glow discharges in the fuel-air mixture (less than 300 ns according to Figs. 6 and 5), the cumulative effect of multiple discharges was necessary to modify the flame and the conditions in the discharge-gap so that OH could be generated and detected. After 400 discharges, OH appeared in the discharge channel, and its intensity in the flame increased, suggesting that the chemistry changed, with a possible increase in the temperature of the NRP glow discharges. Further investigation of the discharge by means of measurements of electric field, electron density and temperature, could shed more light on this dynamic process.

5. Conclusions

Investigation of the chemical impact of NRP glow discharges on a lean laminar methane-air flame was conducted by OH and CH PLIF. Due to a strong non-equilibrium distribution of the OH population in NRP glows, the selection of the excitation line for OH fluorescence required measurement of an experimental fluorescence spectrum. The lines selected for OH were the S21(2) of the (1,1) band and the O12(4) of the (0,0) band. The main findings were:

- Compared to the base flame, OH and CH fluorescence intensities in the flame increased by up to 40% and 10%, respectively, during plasma actuation. At steady state, these intensities did not vary between pulses: OH and CH distributions in the flame did not respond to individual discharges.
- Fluorescence of OH in the NRP discharge channel upstream of the flame reached a detectable level after 400 discharges; at steady state, its decay time after a discharge was 3 to 4.5 μs, depending on the excitation line.
- At the beginning of plasma actuation, the flame moved upstream before the appearance of OH fluorescence in the discharge. Once it reached its stable location, OH fluorescence increased in both the discharge and the flame, suggesting that different mechanisms were at play at the beginning and in the steady state regime of plasma actuation by NRP discharges.

Acknowledgments

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References

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