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Jean-Baptiste Perrin-Terrin, Nicolas Vaysse, Daniel Durox, Ronan Vicquelin, Sébastien Candel, et al.. Plasma-assisted combustion of hydrogen swirling flames: Extension of lean blowout limit and NO<sub>x</sub> emissions. Proceedings of the Combustion Institute, 2024, 40 (1-4), pp.105546. 10.1016/j.proci.2024.105546 . hal-04665064

**HAL Id: hal-04665064**

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Submitted on 30 Jul 2024

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# Plasma-assisted combustion of hydrogen swirling flames: extension of lean blowout limit and NO<sub>x</sub> emissions.

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## Abstract

A key challenge in the use of hydrogen in practical combustors is to stabilize the flame at equivalence ratios close and even below the LBO limit. Plasma-Assisted Combustion (PAC) has been shown to improve the ignition, stabilization, and blowout margins of combustion systems for a great variety of fuels. In this work, Nanosecond Repetitively Pulsed (NRP) discharges are applied to extend the lean blowout limit (LBO) of a partially premixed hydrogen-air combustor atmospheric test rig equipped with a special unit injecting gaseous hydrogen in crossflow into a swirling air stream. The plasma is formed between this injector and an electrode placed in the central recirculation zone. The LBO limit of a 2.2-kW, V-attached flame is extended from 0.220 to 0.177 (20% reduction) using NRP discharges with a plasma power of about 1% of the nominal flame power. Spectroscopic measurements indicate that the plasma operates in the non-equilibrium spark regime: a low ionization degree is observed, and the temperature in the interelectrode gap is moderate (3500 K on average). It is found that plasma discharges increase the OH\* concentration at the flame foot, in the vicinity of the plasma location. Burnt gases analyzed at the outlet of the system indicate that low NO<sub>x</sub> concentrations are being produced, even at stoichiometric conditions where the Emission Index reaches its maximum value of 1.1 gNO<sub>x</sub>/kgH<sub>2</sub>. The application of the NRP discharges increases the NO<sub>x</sub> concentration at all equivalence ratios. To minimize the power consumption and the NO<sub>x</sub> increase, the NRP discharges are applied with a duty cycle where the plasma is discharged during about half of the time. It is shown that the NO<sub>x</sub> concentration scales linearly with the plasma power and thus that NO<sub>x</sub> emission induced by the plasma can be mitigated by 60% with this technique while still preserving the LBO extension.

*Keywords:* Plasma-assisted combustion; Hydrogen swirling flames; Lean blowout; NO<sub>x</sub> emissions

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## Information for Colloquium Chairs and Cochairs, Editors, and Reviewers

### 1) Novelty and Significance Statement

The novelty of this research lies in the use of plasma discharges in pure hydrogen swirling flames. Extending the lean blow-out limit as well as measuring NO<sub>x</sub> emissions of such flames when applying NRP discharges is a novel addition to the existing knowledge in the field of plasma-assisted combustion. It is significant because it provides technological proofs of the capacity to operate hydrogen combustion in very lean conditions. It is apparently the first time that plasma discharges are applied to extend the operability limit of pure hydrogen flames.

### 2) Author Contributions

- J.-B. P.-T. performed the experiments, processed the data (plasma part) and wrote the paper.
- N.V. performed the experiments, processed the data (combustion part) and wrote the paper.
- D.D. supervised the research, designed the experiments, analyzed the data and edited the paper.
- R.V. supervised the research.
- S.C. supervised the research and edited the paper.
- C.L. supervised the research, analyzed the data, edited the paper and obtained the funding.
- A.R. supervised the research, analyzed the data, edited the paper and obtained the funding.

### 3) Authors' Preference and Justification for Mode of Presentation at the Symposium

The authors prefer **OPP** presentation at the Symposium, for the following reasons:

- The audience would not need to be familiar with the experiment to comprehend an oral presentation of this standalone work.
- Both hydrogen and plasma-assisted combustion are topical issues, likely to arouse discussion.
- The topic of hydrogen combustion in ultra-lean conditions is key for the future of decarbonized combustion.

## 1. Introduction

Combustion of renewable hydrogen is a potential solution to decarbonize many industrial sectors requiring high-temperature heating. However,  $H_2$  combustion comes with many new challenges for combustion chambers. The hydrogen/air laminar burning velocity is higher than for conventional fuels, causing flashback issues depending on the injector, and the adiabatic flame temperature is also augmented, indicating a risk for higher  $NO_x$  production [1, 2]. The high molecular diffusivity of hydrogen makes it prone to thermo-diffusive instabilities whereas hydrogen enrichment may modify the thermo-acoustic coupling in combustion chambers [3]. Extending the operability domain of hydrogen flames to ultra-lean conditions could be a path to safe combustion. However, the proximity with the extinction limit brings up stability issues.

The flame stability can be improved by the application of plasma in burnt or fresh gases. This technique is called Plasma-Assisted Combustion (PAC). Various methods can be used to generate plasma, and one of the most power-effective is the Nanosecond Repetitively Pulsed (NRP) discharge. These discharges use 1-25 kV amplitude voltage pulses lasting a few nanoseconds and repeated at frequencies of the order of 10-100 kHz. Depending on these parameters, several plasma regimes can be reached with NRP discharges [4, 5]. For PAC applications, a spark regime is preferred due to its hydrodynamic, chemical, and thermal effects that enhance combustion [6].

Many PAC experiments using NRP discharges were conducted during the last two decades showing various combustion enhancements. The ignition delay can be reduced [7], thermo-acoustic instabilities can be controlled and mitigated [8–10] and lean blow-out limits can be extended [11–13]. These effects were observed not only on lab-scale burners but also on semi-industrial scale combustors (70 kW [10], 100 kW [14]). They were also demonstrated on a wide variety of fuels: propane [8], methane [8, 9, 11–13], liquid heptane and dodecane [13], kerosene [15], and ammonia [16].

Regarding hydrogen, plasma-assisted combustion of pure  $H_2$  has focused mostly on the ignition enhancement [17], in supersonic conditions [18], at low pressure (0.2 bar) [19] and at high pressure (1-12 bar) [20]. A recent study also focused on the deflagration to detonation transition using NRP discharges and showed that the plasma accelerates the hydrogen combustion [21]. Despite these promising effects of NRP discharges on  $H_2$ /air combustion, to the authors' knowledge no studies have focused on the extension of the lean blow-out limit.

Lean combustion is interesting to reduce the burnt gases temperature, and thus mitigate  $NO_x$  emissions. This subject is of particular importance for hydrogen combustion since the  $H_2$  adiabatic flame temperature is, as already mentioned, substantially higher than for conventional fossil fuels at the same equivalence ratio [1]. Exploring ultra-lean conditions is the first step to reducing flame temperature. Among the studies of LBO limit extension using NRP discharges, Lacoste et al. compared the  $NO_x$  emissions of a propane/air flame with and without the plasma at different equivalence ratios [22]. When the plasma was applied, they measured an increase in  $NO_x$  emissions with the plasma power, a conclusion also mentioned by [10]. More recently, Choe et al. [16] applied NRP discharges in ammonia/air flame and showed that the plasma can reduce by 30% the  $NO_x$  levels at the exit of the burner. More surprisingly, they observed a decrease in  $NO_x$  when the deposited energy increased. This change in the  $NO_x$  emissions trends suggests that the effect of PAC on  $NO_x$  emissions highly depends on the fuel.

In this work, NRP discharges are applied in a swirl-stabilized  $H_2$ /air burner, the SICCA-H2 configuration. The LBO limit is measured with different NRP parameters and the burnt gases are analyzed at the outlet of the burner. An extension of the LBO limit to ultra-lean conditions is shown with a limited  $NO_x$  penalty.

## 2. Experimental setup

### 2.1. The SICCA-H2 PAC facility

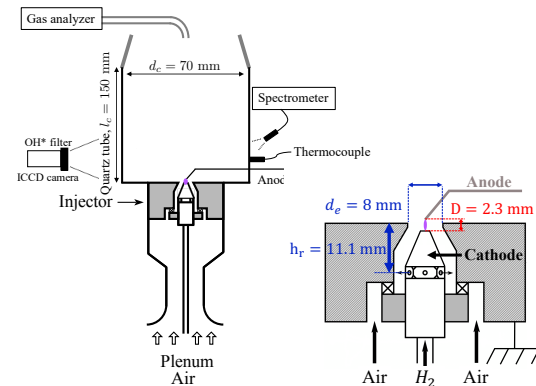


Fig. 1: Left : Schematic view of the SICCA-H2 experimental setup. Right: Schematic view of the CFI-X3 cross-flow injector, equipped with a 1-mm diameter tungsten electrode for NRP discharges.

The experimental configuration is derived from the SICCA-H2 burner presented in [23] which comprises an air plenum, a tangential swirler, a cross-flow injector for gaseous hydrogen (denoted CFI-X) and a cylindrical combustion chamber. In this work, the hydrogen injection head is longer and has a truncated conical shape as presented in Fig. 1. The hydrogen is injected in cross-flow in the swirled air flow through 6 radial holes of 1.2 mm in diameter. In this work, the swirl number is kept constant at a value  $S = 0.60$ , determined from laser Doppler velocimetry axial and tangential velocity profiles. The quartz

1 chamber is 150-mm long and surmounted by a 41-mm 56  
2 high 15° converging nozzle that reduces recirculation 57  
3 effects when measuring exhaust gas composition. Air 58  
4 and hydrogen mass flow rates are controlled by two 59  
5 *Bronkhorst* EL-FLOW meters with full-scale capacities 60  
6 of 210 NL/min for air and 20 NL/min for hydrogen 61  
7 and a typical relative accuracy of 0.8%. The 62  
8 hydrogen mass flow rate is kept constant and thus the 63  
9 flame thermal power is fixed at  $\mathcal{P}_{th} = 2.2$  kW. The 64  
10 equivalence ratio is varied through the air flow rate 65  
11 while keeping a constant fuel flow rate. Thus, the 66  
12 Reynolds number varies between 8,000 and 36,000. 67

13 NRP discharges are applied with a 1-mm diameter 69  
14 tungsten electrode with an end located at  $D = 2.3$  mm 70  
15 above the injector tip, in the central recirculation 71  
16 zone. The plasma is formed in this area, between 72  
17 the electrode (anode) and the injector (grounded cathode). 73  
18 Pulses of a fixed duration of 10 ns are generated 74  
19 by an FPG 30-100MC4K pulse generator (FID 75  
20 Technology). The generator is externally triggered 76  
21 by a Pulse-and-Delay BNC model 574, enabling frequency 77  
22 control and time delay adjustment with other 78  
23 devices. The NRP discharges have a positive polarity 79  
24 and their frequency and incident voltage are varied 80  
25 throughout this study in the range 15-30 kHz and 81  
26 0.5-1.5 mJ. 82

## 27 2.2. Diagnostics

28 The voltage and the current delivered by the 83  
29 pulses are monitored with voltage (Lecroy PPE20kV) 84  
30 and current (Pearson 6585) probes placed in the middle 85  
31 of the 5-m cables connecting the pulser and the 86  
32 electrode. The time-integration of the product of the 87  
33 voltage by the current gives the energy deposited by 88  
34 each pulse in the interelectrode gap. Then, the product 89  
35 of the frequency by the energy gives the plasma 90  
36 power. More details on these electrical measurements 91  
37 can be found in the appendix of [11]. In the rest of 92  
38 this work, each plasma configuration will be designated 93  
39 by its frequency and the energy deposited per 94  
40 pulse. 95

41 The spectra of the flame and the plasma-assisted 96  
42 flame are recorded using an OceanOptics 97  
43 Maya2000Pro deep-UV spectrometer. A Princeton 98  
44 Instruments PI-MAX4 intensified camera equipped 99  
45 with an Asahi Spectra XBPA310 optical bandpass filter 100  
46 centered on 310 nm is used to capture OH\* chemiluminescence 101  
47 images of the flame. The exhaust gas composition 102  
48 is measured using an ECOM J2KN analyzer. The sample 103  
49 probe is placed at the center of the conical exit nozzle, 104  
50 as illustrated in Fig. 1. A non-heated 4-m hose links 105  
51 the sample probe to the gas analyzer. 106

## 53 2.3. Protocol

54 In this part, the different experimental protocols 110  
55 used in this work are described. 111

### *Determination of the equivalence ratio at the LBO limit*

58 A K-type thermocouple placed on the external quartz 59  
60 tube surface measures the wall temperature 30 mm 61  
62 above the backplane. When thermal equilibrium is 63  
64 reached at the combustor wall, the LBO limit is measured 65  
66 by increasing the air flow rate at a rate of 0.1 (NL/min)/s, 67  
68 resulting in an equivalence ratio variation of  $0.01 \text{ min}^{-1}$ . 69  
69 To detect the blowout two methods are tested. The first 70  
71 consists in recording the time evolution of the air flow 72  
73 rate value and the output signal of a microphone placed 74  
75 at the chamber outlet. The abrupt change in the microphone 76  
77 signal indicates the extinction. Then, the equivalence ratio 78  
79 at the LBO limit is determined via postprocessing of the 80  
81 recorded signals. The second method consists in a direct 82  
83 acoustic detection of the extinction by the experimenter and 84  
85 a direct reading of the air flow value on the controller. 86  
87 Both methods are in good agreement and feature a constant 88  
89 uncertainty of 1% on the air flow rate value at extinction. 90  
91 Thus, the typical uncertainty on the gain in LBO is 4%, 92  
93 computed as a combination of the latter reading uncertainty 94  
95 and statistical uncertainties. In the present work, the results 96  
97 are obtained with the second method. 98

### *Measurement of the OH\* chemiluminescence*

82 To measure the OH\* chemiluminescence when the 83  
84 NRP discharges are applied, the camera is externally 85  
86 triggered by the BNC generator to adjust the time delay 87  
88 with the plasma emission. For the non-assisted flame, 89  
90 the camera is internally triggered. 91

### *Application of the NRP discharges*

88 In this work, NRP discharges are applied in two 89  
90 modes: 91

- The continuous mode, which is the most commonly used in the literature.
- The duty-cycle mode, used in [14, 24].

92 The pulsing pattern of the duty-cycle mode is described 93  
94 in Fig. 2. This mode consists in repetitively applying 95  
96 bursts of NRP discharges. At a given frequency, the 97  
98 duty-cycle is described by the number of pulses ON in the 99  
burst ( $N_{ON}$ ) and the number of following pulses OFF ( $N_{OFF}$ ). 100  
101 In Fig. 2,  $N_{ON} = 6$  and  $N_{OFF} = 5$ . The duty-cycle ratio is 102  
103 defined as  $r_{DC} = N_{ON}/(N_{ON} + N_{OFF})$  and corresponds to the 104  
105 fraction of the cycle during which the plasma is on. The 106  
107 continuous mode corresponds to the limit case where 108  
109  $r_{DC} = 1$ . In the duty-cycle mode, the plasma power is 110  
111  $P_{plasma} = f \times E_p \times r_{DC}$ . Thus, the lower the duty-cycle ratio, 112  
the lower the plasma power.

### *Burnt gases analysis*

107 After each variation of the experimental conditions, 108  
109 a few minutes are necessary for the measured dry concentration 110  
111 to stabilize. Once stabilized, their values are noted. The 112  
analysis of the dry gases composition gives, among others, the 113  
dry mole fractions of O<sub>2</sub>,

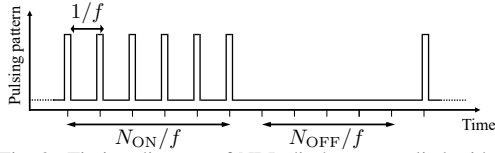
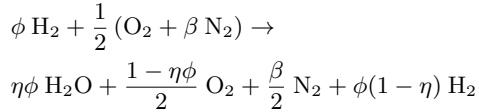


Fig. 2: Timing diagram of NRP discharges applied with a duty-cycle, here with  $N_{ON} = 6$  and  $N_{OFF} = 5$ .

1 NO, NO<sub>2</sub> and H<sub>2</sub>. Because the unburnt H<sub>2</sub> rapidly saturates the ECOM analyzer (the mole fraction limit is 23000 ppm), H<sub>2</sub> cannot be measured when the combustion efficiency is not close to unity. The following global chemical equation is used for the post-processing of the dry mole fractions:



7 where  $\beta = n'_{\text{N}_2}/n'_{\text{O}_2} = 3.76$  is the nitrogen-to-oxygen ratio in fresh air and  $\eta = n''_{\text{H}_2\text{O}}/n'_{\text{H}_2}$  is the combustion efficiency. The number of moles  $n'_i$  and  $n''_i$  respectively stand for the reactants and the products of species  $i$ . The mole fraction of oxygen under dry conditions can be expressed as:

$$X_{\text{O}_2}^m = \frac{n''_{\text{O}_2}}{n''_{\text{O}_2} + n''_{\text{N}_2} + n''_{\text{H}_2}} = \frac{1 - \eta \phi}{1 + \beta + (2 - 3\eta)\phi}$$

13 thus the combustion efficiency can be deduced from 14  $X_{\text{O}_2}^m$  and the global equivalence ratio  $\phi$ :

$$\eta \phi = \frac{(\beta + 1 + 2\phi)X_{\text{O}_2}^m - 1}{3X_{\text{O}_2}^m - 1}$$

15 One retrieves the limit case of no combustion, for which  $X_{\text{O}_2}^m = 1/(1 + 2\phi + \beta)$  leads to  $\eta = 0$ , and the case of complete combustion where  $X_{\text{O}_2}^m = (1 - \phi)/(1 - \phi + \beta)$  leads to  $\eta = 1$ .

19 One can then derive the wet mole fractions by multiplying the measured dry mole fractions by a dry-to-wet correction factor  $C$ , defined as:

$$C = \frac{n''_{\text{O}_2} + n''_{\text{N}_2} + n''_{\text{H}_2}}{n''_{\text{H}_2\text{O}} + n''_{\text{O}_2} + n''_{\text{N}_2} + n''_{\text{H}_2}}$$

22 After some algebra one finds that:

$$C = \frac{\phi + \beta/2 - 1}{\phi + \beta/2 - X_{\text{O}_2}^m(1 + \beta + 2\phi)}$$

23 which gives, in the limit case of no combustion, 24  $C = 1$  which is consistent with the fact that 25 no water is produced. In the complete combustion 26 assumption, one retrieves the classical formula 27  $C = (\beta + 1 - \phi)/(\beta + 1 + \phi)$ .

28 The evolution of  $C$  is shown in Fig. 3.

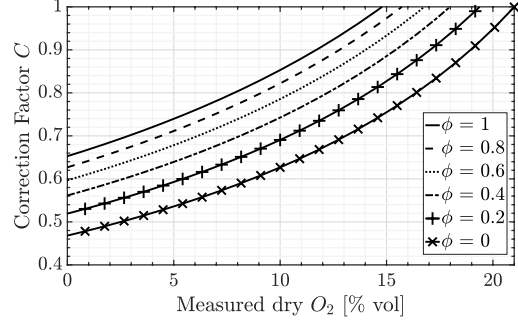


Fig. 3: Abacus giving the correction factor  $C$  linking dry and wet mole fractions, as a function of the measured O<sub>2</sub> dry mole fraction and the equivalence ratio.

### 29 Electrode erosion

30 Electrode erosion was observed both when operating the flame with and without plasma. The erosion speed was about 0.5 mm/h with the flame alone and 0.6 mm/h with the flame and plasma at 40 kHz and 1.5 mJ. For this reason, the electrodes were replaced after each hour of operation to prevent a variation of more than 0.6 mm in the interelectrode gap distance during experiments. Additionally, to minimize this uncertainty for each operating point, several electrodes with different erosion states are used.

## 40 3. Experimental conditions

### 41 3.1. Flame structure

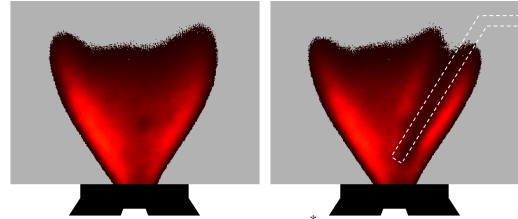


Fig. 4: Line-of-sight integrated OH\* chemiluminescence images of the flame without plasma, at thermal power  $\mathcal{P}_{th} = 2.2$  kW and equivalence ratio  $\phi = 0.231$ , (left) without and (right) with the electrode outlined with white dashes.

42 The observed LBO equivalence ratio of the 43 flame at thermal power  $\mathcal{P}_{th} = 2.2$  kW is  $\phi = 0.220$ . 44 At  $\phi = 0.231$ , corresponding to 105% of the LBO 45 limit, the flame is V-shaped and is attached to the 46 injector flat tip. The OH\* chemiluminescence images of 47 Fig. 4 show that the presence of the electrode brings 48 minimal changes to the flame shape (the electrode can 49 be seen in the right image of Fig. 4).

### 50 3.2. Characterization of the plasma

51 NRP discharges are generated with frequencies 52 in the range of 15 – 30 kHz and with incident voltages

1 in the range of 1.5 – 3 kV. The typical shapes of the  
 2 voltage and current traces are shown in Fig. 5. The  
 3 interelectrode distance is  $D = 2.3 \pm 0.6$  mm. The  
 4 energy is deposited during the initial pulse and the series  
 5 of reflected pulses (as shown in blue in Fig. 5) and  
 6 varies between 0.5 mJ and 1.5 mJ, with an uncertainty  
 7 of  $\pm 100$   $\mu$ J. In these conditions, according to [4], the  
 8 plasma is formed in the spark regime.

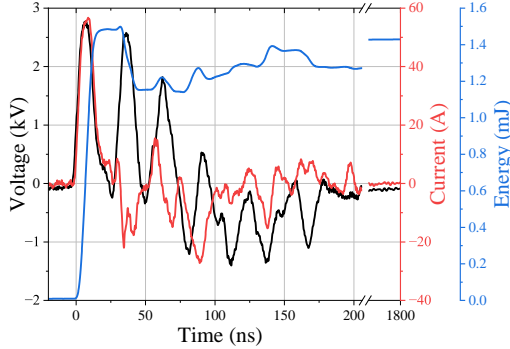


Fig. 5: Typical traces of voltage (black) and current (red) of a pulsed discharge. The energy (blue) is the integration over time of the product of the voltage by the current. In this case, the incident voltage is 2.8 kV, and the deposited energy is 1.42 mJ after 1800 ns.

9 The optical emission spectroscopy measurements shown in Fig. 6 are used to measure the temperature in the plasma region from the rotational temperature of  $N_2(C)$  in the range 360 – 390 nm using Specair [25]. A temperature of  $3500 \pm 300$  K is found for the 15 kHz – 1.5 mJ plasma (applied continuously and with duty-cycle) and  $3800 \pm 400$  K for the 30 kHz – 0.75 mJ plasma. Additionally, an upper bound of the electron number density can be estimated from the broadening of the  $H_\beta$  line at 486 nm [26]. The total broadening observed in Fig. 6 is 1.5 nm. Taking into account the instrumental broadening of about 1 nm, we obtain an upper bound of the ionization degree of less than about 0.2%. Considering the relatively low heating and ionization degree, the plasma is in the non-equilibrium spark regime [5].

25 The spectrum of the flame without plasma is dominated by the black-body emission of the heated electrode. In particular, the broadband blue emission characteristic of the hydrogen flames is not visible. The absorption caused by the water vapor produced by the combustion appears around 950 nm. At 310 nm,  $OH^*$  emission is not intense because the field of view of the spectrometer is centered on the discharge which produces intense emission via the second positive system of nitrogen.

## 35 4. Results

### 36 4.1. Influence of frequency and deposited energy per pulse on LBO extension

38 In a first set of experiments, the frequency is

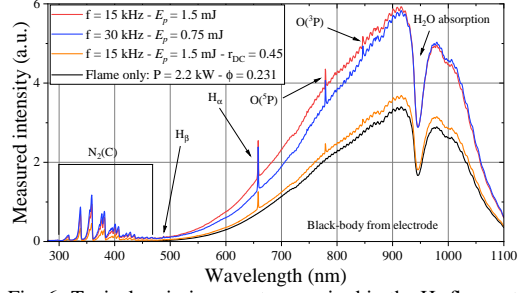


Fig. 6: Typical emission spectra acquired in the  $H_2$  flame at  $P_{th} = 2.2$  kW and  $\phi = 0.231$ . Spectra are acquired during 50 ms and averaged 40 times. The black line is acquired without the plasma. The red and orange lines are acquired with the 15 kHz – 1.5 mJ plasma, applied respectively continuously and with a duty-cycle ratio of 0.45. The blue line is acquired with the 30 kHz - 0.75 mJ plasma applied continuously.

39 kept equal to 30 kHz and the deposited energy per  
 40 pulse varies with values 500  $\mu$ J, 750  $\mu$ J, 1 mJ, and  
 41 1.5 mJ. Fig. 7 shows that the gain in LBO compared  
 42 to the non-assisted case increases linearly with the  
 43 energy, with values of 6%, 11%, 19%, and 23%. Then,  
 44 measurements are performed at a lower frequency,  
 45  $f = 15$  kHz, and show that the gain in LBO also  
 46 increases with the frequency at  $E_p = 0.75$  mJ (from 7%  
 47 to 11%) and  $E_p = 1.5$  mJ (from 20% to 23%). It is  
 48 interesting to compare in Fig. 7 two experiments with  
 49 the same average plasma power. The  $f = 15$  kHz -  
 50  $E_p = 1.5$  mJ and the  $f = 30$  kHz -  $E_p = 0.75$  mJ  
 51 plasmas are on the 22.5-W line (corresponding to 1%  
 52 of the flame thermal power) but it is shown in Table 1  
 53 that the former extends the LBO limit by up  
 54 to 19.5% while the latter only to 10.9%. This indicates  
 55 that in these conditions, increasing the energy  
 56 extends the LBO limit more efficiently than increasing  
 57 the repetition frequency at constant plasma power.  
 58 Because of its larger improvement in LBO limit, the  
 59  $f = 15$  kHz -  $E_p = 1.5$  mJ plasma will be studied in  
 60 more detail in the rest of this work.

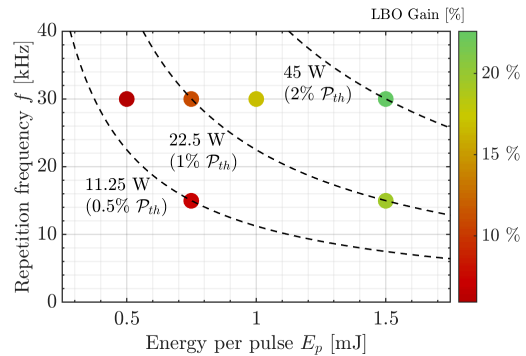


Fig. 7: Mapping of the LBO gain as a function of the repetition frequency and the average energy deposited per pulse. Dashed lines represent isolines of plasma power, expressed as a percentage of the thermal power.

Table 1: LBO measurements for a flame at  $\mathcal{P}_{th} = 2.2$  kW

Frequency [kHz]	-	15	15	30	30
Energy [mJ]	-	0.75	1.5	0.75	1.5
$\phi$ at LBO	0.220	0.204	0.177	0.196	0.170
Gain in LBO [%]	0	7.3	19.5	10.9	22.6
Plasma power [W]	0	11.25	22.5	22.5	45

The effect of the plasma pulses on the chemistry of the flame is analyzed through phase-averaged OH\* chemiluminescence imaging presented in Fig. 8. The images are recorded during the time separating two pulses and show that the plasma locally increases the OH\* radical concentration near the foot of the flame front, where it is applied. Right after the discharge, the OH\* signal is intense in the vicinity of the electrode. Then, the intensity monotonically decreases until the following pulse. This effect is emphasized in the bottom row of Fig. 8, which isolates the contribution of plasma to the OH\* signal by subtracting the OH\* image of the flame without plasma shown in Fig. 4 from the phase-averaged images. It also highlights that the effect of the plasma on the flame is local and does not increase the OH\* chemiluminescence elsewhere in the flame.

#### 4.2. Combustion efficiency

The values of the combustion efficiency presented in Fig. 9 include the flame without plasma and the plasma-assisted flame in the three investigated plasma cases:  $f = 15$  kHz -  $E_p = 1.5$  mJ,  $f = 30$  kHz -  $E_p = 0.75$  mJ and  $f = 15$  kHz -  $E_p = 1.5$  mJ applied with a duty-cycle ratio  $r_{DC} = 0.45$ . At first, the combustion efficiency increases with the equivalence ratio. Then, a plateau value of  $92\% \pm 8\%$  is reached above  $\phi = 0.5$ . However, the measured mole fraction of  $H_2$  is 0 ppm in this domain, indicating that combustion is complete. In stoichiometric conditions, the efficiency is close to 100%. No clear effect of the plasma on the combustion efficiency of the flame is visible in Fig. 9.

#### 4.3. NO<sub>x</sub> emissions

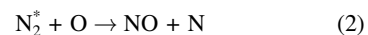
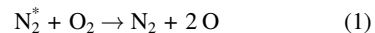
The measured dry NO<sub>x</sub> mole fractions are multiplied by the dry-to-wet correction factor  $C$  introduced in Fig. 3 to obtain their wet values in the flame. This is essential to derive an emission index based on the mass flow rate ratio between NO<sub>x</sub> and injected H<sub>2</sub>. The evolution of this NO<sub>x</sub> Emission Index, plotted in Fig. 10, shows that the NO<sub>x</sub> emissions of the non-assisted flame remain lower than 1.1 g/kgH<sub>2</sub> on the whole investigated operability domain of the flame. This value corresponds to the lower end of the typical Emission Indexes reported in the literature (1-20 g/kgH<sub>2</sub> for jet and coaxial flames [27–29]). The NO<sub>x</sub> emissions increase with the equivalence ratio after a slight decrease in the leanest range. An optimum for NO<sub>x</sub> emissions is found around  $\phi = 0.4$  for all conditions. All plasma pulses investigated in

this work induce an increase of the NO<sub>x</sub> emissions by 0.5 to 1.5 g/kgH<sub>2</sub>. However, applying a duty-cycle (green diamonds in Fig. 10) to the  $f = 15$  kHz -  $E_p = 1.5$  mJ plasma reduces the NO<sub>x</sub> emissions at all equivalence ratios, up to 60% in the leanest domain, while still extending the LBO limit. The one-sigma error bars are fairly large because all sources of error are taken into account. The main uncertainties arise when the oxygen mole fraction is close to zero near stoichiometry or at very low equivalence ratios (especially in Fig. 10). However, the measured mean values still clearly support the reduction of NO<sub>x</sub> emissions in duty cycle mode, which is the intended point of Fig. 10. The effect of this technique on the NO<sub>x</sub> emissions is further investigated in the following section.

#### 4.4. Influence of the pulsing pattern : effect of the duty-cycle on flame stability and NO<sub>x</sub> emissions

To decrease the plasma power without reducing the effect of the plasma on the flame, the NRP discharges are applied in the duty-cycle mode at  $f = 15$  kHz and  $E_p = 1.5$  mJ. This method was already tested by [14, 24] and the energy was reduced without losing the stabilizing properties of the plasma. The power consumption is thus directly multiplied by  $r_{DC}$ . The equivalence ratio is fixed at 105% of the LBO limit obtained with this plasma ( $1.05\phi_{LBO}^p = 0.186$ ). To determine the lowest duty-cycle ratio allowing the stabilization of a flame at this equivalence ratio,  $N_{ON}$  is fixed and  $N_{OFF}$  is progressively increased until the flame is blown off. The gas is sampled in the process. The results are shown in Fig. 11. The extinction limit is drawn as a dotted line and presents the same V-shape as found in [14], with a minimum reached around  $N_{ON} = 100$ . During the ON phase, the pulses generate a stabilizing kernel, enhancing the combustion. During the OFF phase, the enhancement of the flame fades until the next ON phase. For the low  $N_{ON}$ , the aforementioned combustion-stabilizing kernel is not strong enough and the enhancement fades before the next ON phase. When  $N_{ON}$  increases, the kernel is stronger, and thus the maximum  $r_{DC}$  is larger. As  $N_{ON}$  is further increased, for a given  $r_{DC}$ , the OFF phase is also longer and the contribution of the additional pulses to the kernel is not sufficient to sustain the combustion on this longer OFF phase: the flame extinguishes.

Two key plasma-induced reactions must be considered to interpret the results on NO<sub>x</sub> emissions presented in Fig. 11: the beneficial radical production (1) and the detrimental NO formation (2).



where  $N_2^*$  stands for excited electronic states of nitrogen. The flame is stabilized when successive pulses have produced enough radicals through reaction (1).



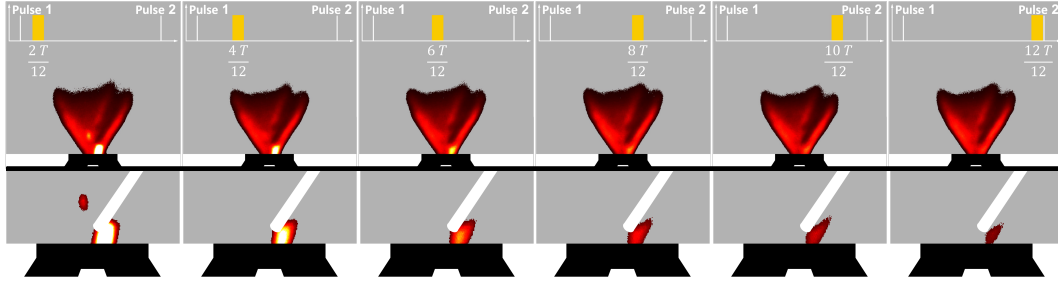


Fig. 8: Phase-averaged  $\text{OH}^*$  chemiluminescence images of the flame between two plasma pulses. The phase number is indicated in the top diagram. The  $f = 15 \text{ kHz} - E_p = 1.5 \text{ mJ}$  plasma is applied. Top: line-of-sight integrated images of the flame. Bottom: Images of the difference between these phase-averaged images and the image of the flame without plasma, presented in Fig. 4. The electrode is added in white on the second row. The exposure time is  $5 \mu\text{s}$ . Twelve phases are recorded, only the even phases are shown.

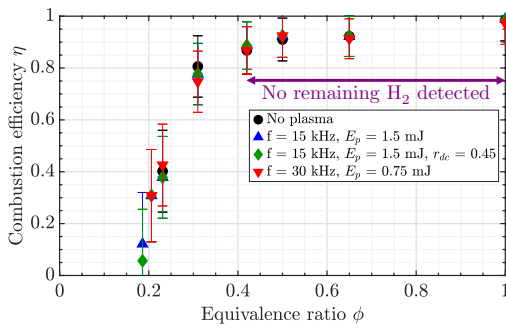


Fig. 9: Evolution of the combustion efficiency as a function of the equivalence ratio, for the flame without plasma, and for the three different discharges applied. The efficiency is computed from the dry  $\text{O}_2$  mole fraction measurement and the equivalence ratio. Uncertainties are propagated starting from the accuracy and resolution of the mass flow meters and the gas analyzer.

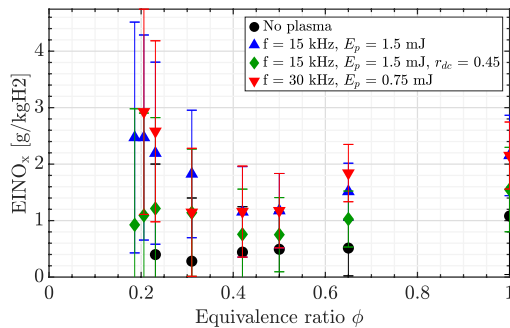


Fig. 10: Evolution of the  $\text{NO}_x$  Emission Index as a function of the equivalence ratio, for the flame without plasma, and for the three different discharges applied. The  $\text{EINO}_x$  is computed from the dry  $\text{NO}_x$  mole fractions, corrected to wet conditions using  $\text{O}_2$  measurements. Uncertainties are propagated starting from the accuracy and resolution of the mass flow meters and the gas analyzer.

1 Above this threshold, additional pulses do not further  
 2 improve stabilization, but they increase NO through  
 3 reaction (2). Thus, the duty cycle mode allows to keep  
 4 the same LBO while minimizing the  $\text{NO}_x$  penalty.

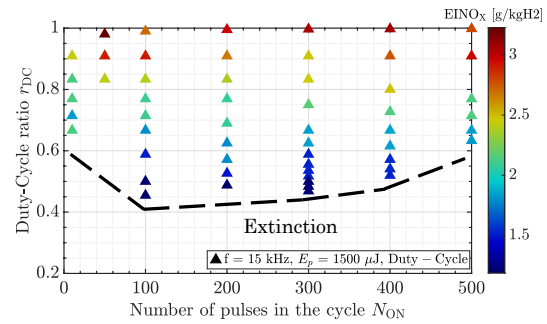


Fig. 11: Map of the  $\text{NO}_x$  Emission Index as a function of the duty-cycle ratio  $r_{\text{DC}}$  and the number of pulses in each cycle, at an equivalence ratio of  $1.05\phi_{\text{LBO}}^p = 0.186$ . The  $f = 15 \text{ kHz} - E_p = 1.5 \text{ mJ}$  plasma is applied. The dashed line shows the minimal duty-cycle ratio required to stabilize the flame.

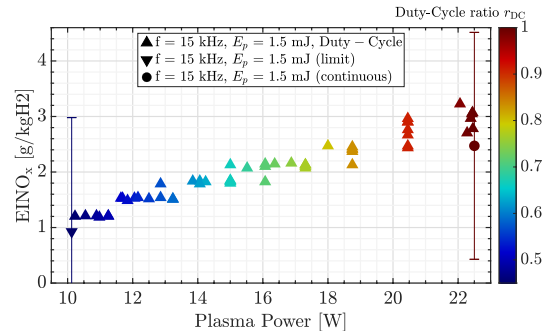


Fig. 12: Evolution of the  $\text{NO}_x$  Emission Index as a function of the plasma power at an equivalence ratio of  $1.05\phi_{\text{LBO}}^p = 0.186$ . The  $f = 15 \text{ kHz} - E_p = 1.5 \text{ mJ}$  plasma is applied. The colors correspond to the duty-cycle ratio. The down triangle corresponds to the measured emissions at the minimal  $r_{\text{DC}}$  value and the circle to the value obtained for continuous plasma operation.  $N_{\text{ON}}$  is varied between 10 and 500.

5 The effect of the pulsing pattern on  $\text{NO}_x$  emis-  
 6 sions is expressed in Fig. 12 as a function of the  
 7 average plasma power, computed as the product  
 8 of the duty-cycle ratio with the continuous plasma  
 9 power. The  $\text{NO}_x$  emissions decrease linearly with

1 the plasma power as the duty-cycle ratio is decreased. 55  
2 This is consistent with the results found in methane 56  
3 flames [10, 14, 22]. 57

## 4 5. Conclusion 58

5 A V-attached hydrogen flame of 2.2 kW is stud- 59  
6 ied. Its equivalence ratio at the lean blow-out limit 60  
7 equals 0.22. NRP discharges are applied to this flame 61  
8 to extend the LBO limit. Various couples of repetition 62  
9 frequencies and deposited energy are tested. In all 63  
10 NRP conditions, the plasma is in the non-equilibrium 64  
11 spark regime, inducing moderate heating (a few thou- 65  
12 sand kelvins) and a low ionization degree (less than 66  
13 0.2%). The effect of the plasma on the flame is obser- 67  
14 ved using  $\text{OH}^*$  chemiluminescence. During the 68  
15 pulse, the plasma locally increases the  $\text{OH}^*$  signal. 69  
16 The increase is confined to the vicinity of the plasma 70  
17 and slowly decays between two pulses. The main 71  
18 findings of this work are: 72

- 19 • The LBO limit can be extended from 0.22 to 73  
20 0.177 (20% gain) with the  $f = 15$  kHz - 74  
21  $E_p = 1.5$  mJ plasma. 75
- 22 • The plasma power required to extend the LBO 76  
23 limit with continuously applied NRP discharges 77  
24 is about 1% of the flame thermal power. 78
- 25 • The plasma power can be reduced to 0.45% of 79  
26 the flame thermal power by applying the NRP 80  
27 discharges with a duty-cycle ratio of 0.45. 81
- 28 • From  $\phi = 0.22$  to 1 (lean operation) the non- 82  
29 assisted flame emits less than 1.1  $\text{gNO}_x/\text{kgH}_2$ . 83
- 30 • The NRP discharges applied continuously in- 84  
31 crease the  $\text{NO}_x$  emission by 0.5 to 1.5  $\text{g}/\text{kgH}_2$ . 85
- 32 • The NRP discharges applied with a duty-cycle 86  
33 ratio of 0.45 increase the  $\text{NO}_x$  emission by 0.2 87  
34 to 0.9  $\text{g}/\text{kgH}_2$ . 88

## 35 Declaration of competing interest 89

36 The authors declare that they have no known 90  
37 competing financial interests or personal relationships 91  
38 that could have appeared to influence this work. 92

## 39 Acknowledgments 93

40 This work was partially supported by the FlyHy 94  
41 project of the Agence Nationale de la Recherche 95  
42 (ANR-21-CE05-0008), the European Research Coun- 96  
43 cil (ERC) under the European Union's Horizon 2020 97  
44 research and innovation program (GreenBlue project 98  
45 under grant agreement No.101021538) and by the 99  
46 Fondation Simone and Cino Del Duca through a sci- 100  
47 entific grant attributed to the project PLATHON. For 101  
48 open access purposes, a CC-BY license has been ap- 102  
49 plied by the authors to this document and will be ap- 103  
50 plied to any subsequent version up to the author's 104  
51 manuscript accepted for publication resulting from 105  
52 this submission. The authors would like to thank Yan- 106  
53 nick Le Teno and the support team of EM2C lab for 107  
54 their technical assistance. 108

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