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Effects of non-steady state discharge plasma on natural gas combustion: Flammability limits, flame behavior and hydrogen production

J.C. Sagás [a,b,*], H.S. Maciel [a,c], P.T. Lacava [b]

[a] Plasmas & Processes Laboratory, Technological Institute of Aeronautics (ITA-DCTA), São José dos Campos, SP 12228-900, Brazil
[b] Combustion, Propulsion and Energy Laboratory, Technological Institute of Aeronautics (ITA-DCTA), São José dos Campos, SP 12228-900, Brazil
[c] Nanotechnology and Plasmas Processes Laboratory, University of Paraiba Valley (Univap), São José dos Campos, SP 12244-000, Brazil

HIGHLIGHTS

- The discharge is the main source of H_2 molecules, which are burned in the flame.
- The flame occurs in cycles in gliding arc assisted combustion.
- Under plasma, the position of the flame is dependent on the equivalence ratio.

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ABSTRACT

The effects of a non-steady state plasma discharge on flammability limits and flame structure of air–natural gas mixtures are investigated. As plasma power increases, flame structure is changed and flammable range is extended. In the absence of a visible flame, a higher hydrogen production is observed, revealing that the discharge is a source of molecular hydrogen. The reduction on hydrogen production inside the flammable range suggests a burning of hydrogen in the flame.

1. Introduction

The use of atmospheric pressure non-thermal plasmas to assist combustion has revealed new chemical and physical effects [1–8]. In great part, these effects are result of the radical pool generated by the plasma [4,5,9,10], although thermal and transport effects must be also considered [4]. Such novel chemical routes are especially attractive in fuel reforming [11–18] and pollutant control [19]. A possible application is in the fuel rich zone of rich-quench-lean gas turbine combustors [20,21]. The injection of radicals in the lean zone leads to a more stable combustion, to an increased combustion velocity and to a reduction of NO_x emission with low levels of CO and unburned hydrocarbons [22].

Non-steady state discharges properties, such as electron density and electron temperature, are useful to the generation of a radical pool [5,23–25]. Besides, they supply the energy necessary to gas mixture ignition due to repetitive breakdowns and glow-to-spark transitions [23–26]. Although this type of discharge has characteristics of a glow discharge (when operating at low current), it is generally called as gliding arc [24,25]. There are several different geometries, such as the so-called planar gliding arc [27], the simple coaxial configurations [28], and the gliding arc in tornado [29]. Although these technologies are growing fast, the complexity of the mechanisms involved in the plasma assisted combustion requires further research to understand plasma effects not only on the combustion chemistry, but also on flame structure.

To evaluate the role of a non-steady state discharge on air–natural gas premixed flames, a plasma reactor with reverse vortex flow (in the so-called gliding arc in tornado geometry) was used [29]. The process was analyzed by optical and mass spectroscopy. Images were recorded by high-speed camera. The measurements were made in fuel rich and lean regimes.

2. Experimental

The discharge tube was made of quartz in order to allow optical access. The gas mixture was injected on a vortex chamber in the

* Corresponding author at: Laboratory of Plasmas, Films and Surfaces, Santa Catarina State University, Joinville, SC 89219-710, Brazil.
E-mail address: julio.sagas@udesc.br (J.C. Sagás).

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front of the reactor, in a typical reverse vortex flow configuration. A helical tungsten wire molded according the flow lines was used as the high voltage electrode. It means that the gas streamlines have the same profile of the helical electrode when the gas flows from the vortex chamber to the end of the reactor. Then, the gas returns by the central axis and exits through the nozzle. The nozzle and the vortex chamber, both made of brass, were grounded. The nozzle acts as an electrode, i.e. the discharge is generated between the helical tungsten wire and the nozzle. The discharge tube has length of 62.0 mm and 22.5 mm of internal diameter and the nozzle has 6.0 mm in diameter. A scheme of the experimental setup is depicted in Fig. 1 and a picture of the plasma reactor can be seen in Fig. 2. More details about the reactor are found in [30].

The electrical power was supplied by an AC transformer (6 kV r.m.s.) in series with a variable resistor (0–30 Ω, 3 kW) in the primary circuit. A homemade high voltage probe 1000:1 was used to measure the r.m.s. voltage, and the temporal behavior of discharge current was monitored using a Hall sensor. Due to the large uncertainties in the current measurement, the applied r.m.s. power was determined by measuring the input current and voltage in the transformer.

In this experimental setup, the discharge is characterized by repetitive cycles of ignition–evolution–extinction. The early stage of the discharge (the gas breakdown) occurs at the nozzle, where the electrodes (nozzle and helical electrode) are closest to each other (4 mm). From this point, the arc elongates under the pushing effect of the gas flow, gliding along the electrodes surfaces until discharge extinction. The extinction occurs due to the voltage limitation imposed by the external circuit. After the extinction, the discharge is restarted, repeating the cycle. In some cases, the new breakdown occurs before the discharge extinction [28].

The reagents were premixed before entering the vortex chamber. The mass flow rates were controlled by needle valves and measured using thermal mass flow meters. The air mass flow rate was maintained constant at 0.80 g/s, which corresponds to a velocity of about 22 m/s at the nozzle in standard temperature and pressure conditions. The natural gas mass flow rate varied from 0.02 to 0.14 g/s. The typical composition of the natural gas used in the present work is (% mol): 88.683% CH_4 (methane), 5.844% C_2H_6 (ethane), 2.339% C_3H_8 (propane), 0.328% i-C_4H_{10} (i-butane), 0.443% n-C_4H_{10} (n-butane), 0.076% i-C_5H_{12} (i-pentane), 0.052% n-C_5H_{12} (n-pentane), 0.025% hexane and higher hydrocarbons, 0.594% N_2 (nitrogen) and 1.616% CO_2 (carbon dioxide).

To characterize the process, equivalence ratio (ϕ) as defined in [30] is used:

$$\phi = \frac{R_{op}}{R_{st}} \quad (1)$$

where R_{op} is the fuel-to-oxidant mass ratio in experimental conditions and R_{st} is the fuel-to-oxidant mass ratio in stoichiometric condition. Thus, for $\phi = 1$ the combustion is stoichiometric and for $\phi > 1$ the combustion is fuel rich. In this work, the equivalence ratio varied between 0.4 and 3.0.

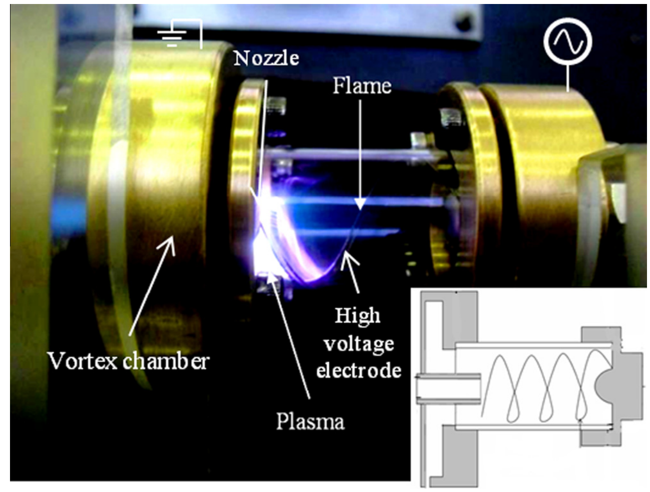

Fig. 2. Image of the plasma reactor with plasma assisted combustion: equivalence ratio of 1.4 and r.m.s. power of 395 W. Electrical-to-thermal power ratio of 12.3%. On bottom at right, a scheme of the reactor. The discharge (purple) is limited to the region between the nozzle and the first turn of the helical electrode. The flame (blue) is stabilized in the central axis. (For interpretation of the references to color in this figure legend, the reader is referred to the web version of this article.)

The discharge was also analyzed by using a set of seven optical emission spectrometers. Each spectrometer covers a range of approximately 100 nm. Therefore, the whole set covers the spectrum in the range 198–876 nm with a resolution of 0.1 nm. The light is captured by an optical fiber that splits itself in other seven fibers (one for each spectrometer). Owing to the different sensitivity of each spectrometer, a calibration was made by using a halogen lamp with tungsten wire. Thus, to compare results from different spectrometers, the intensity measured was converted to irradiance according to the calibration curve. The optical fiber was oriented to the nozzle region, where the discharge was confined (Fig. 2). No lens were used to increase the signal. Also due to the different sensitivities, the integration times were different for each spectrometer and also varied according to the experimental conditions. However, the lowest integration time was 100 ms, which is about 12 times greater than a cycle of the discharge (8.33 ms).

The chemical analysis of the exhaust gases was made with a quadrupole mass spectrometer. For this procedure, the plasma reactor was connected to a post-chamber with 800 mm of length and 160 mm of internal diameter (Fig. 1). The probe of the spectrometer was placed 505 mm from the reactor exit.

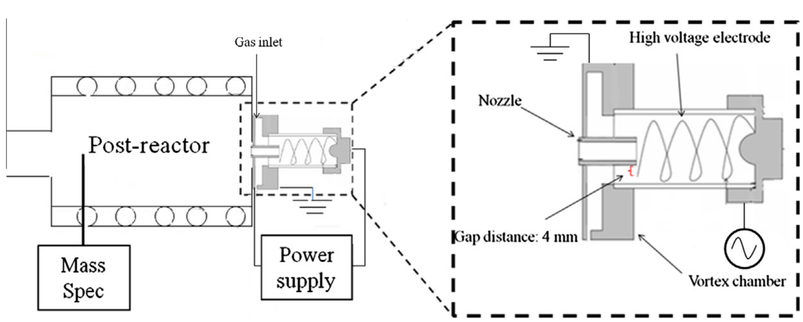

Fig. 1. Experimental setup. In detail, the plasma reactor. The helical electrode reproduces the flow lines.