Spectroscopic analysis on a plasma assisted CH₄-air turbulent swirling flame

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Abstract: The present study reports a preliminary spectroscopic analysis of the Gliding Arc Plasma effects on the CH₄-air combustion by Optical Emission Spectroscopy. A gliding arc system have been applied to a turbulent flame in a swirl burner in order to improve the flame stabilization and reduce the pollutant emissions An analysis of the plasma/flame UV visible radiations have been collected and analyzed according to the plasma setting with the aim to optimize the plasma effects on combustion assistance.

Keywords: gliding arc, plasma, combustion, spectroscopic analysis.

1. Introduction

The development of human activities and technologies requires more and more energy consumption which still remains meanly provided by fossils energy. This growing consumption induces huge pollutant emissions accentuating the sticky problem of global warming. In this context, the development of new combustion technologies involves more fuel efficiency, environmentally friendly processes and needs working in lean mixture. However, the use of leans conditions induces flame disturbances especially in terms of stability leading up to flame extinction in several cases. Among the possibilities of flame stabilization in lean mixture, the use of non-thermal plasma sources to control combustion processes represents a promising technologies. Indeed, non-thermal plasma sources can be a low energy consumption system, and have robust and flexible designs suitable to combustion system requiring only weak modifications of the combustion device [1-3].

Many previous studies have already been carried out on the use of plasma sources for combustion assistance by application of a dielectric barrier discharge (DBD) [4] or more usually by pulsed discharges in plasma torches [1, 5]. However only few publications reported results on the gliding arc discharge (Glidarc) effects on combustion devices. Among the rare analysis based on this plasma reactor type, some studies have been devoted to the plasma/combustion interactions in premixed flame [6], the plasma effects on the ignition enhancement in a diffusion flame [7], methane oxidation processes by using rotating Glidarc [8] or flammability limits and hydrogen production [9]. These studies have clearly demonstrated the relative benefits (or disadvantages) of plasma assistance for combustion processes [10-11], hugely depending on the plasma reactor parameters. In this context, based on the opportunity of using gliding arc discharge in combustion control, a project has been developed to implement a double gliding arc reactor in a coaxial swirl burner in order to investigate the possibilities of flame stability enhancement and gas pollutant reduction in a turbulent flame.

In the field of plasma assisted combustion, three major pathways have been identified [12]: Heating and chemical active species generation leading to the ignition and combustion chemistry enhancement, intensification of the air-fuel mixing in the flow, and flow structure management for the flame front stabilization. These effects are induced by the inherent characteristics of the plasma phase namely: a fast local ohmic heating at the plasma/flame interface, non-equilibrium excitation and dissociation of particles (air, fuel molecules) both by electronic impacts and UV radiation, momentum transfer within the applied electromagnetic fields and the generation of shockwaves/instabilities. Nevertheless, although the general plasma effect mechanisms are known, the plasma/flame interactions remain relatively unclear to allow a comprehensive adequacy between the plasma and flame parameters. Face to this complex medium at the interface between plasma physic and combustion chemistry, this works presents some preliminary results of the gliding arc plasma (GAP) effects on a turbulent CH₄/air flame as a function of plasma power supplies conditions (intensity and frequency). The analysis have been performed on the UV/visible radiations by Optical Emission Spectroscopy (OES) within the plasma, plasma/flame interface and flame front to evaluate the effect of the plasma in the point of view of chemical activities.

2. Experimental setup

The used burner and flame configurations are described in details in previous works [13, 14]. Briefly, the combustion device used to generate the turbulent CH₄/air flames is a coaxial burner equipped with a swirler in the annular tube (cf. Fig. 1). The fuel (CH₄) is delivered in the central part of the coaxial tube and injected in the combustion chamber through 8 circular pinholes. The air passes through the eight vanes of the swirler within the annular tube inducing a helical movement of the air flow. The burner is installed inside a rectangular combustion chamber $(0.48 \times 0.48 \times 1 \text{ m}^3)$ operating at atmospheric pressure. Each face of this chamber is equipped with 6 rectangular optical access allowing visualization and spectroscopic experiments along the entire length of the plasma and the flame. The experiments have been carried out on this turbulent flames (Re > 4500) for different equivalence ratios between 0.55 and 1.2, depending of the experiments.

The two gliding arcs for plasmas generation have been directly installed on the burner presented in the previous section as presented on the Fig. 1. The central tube for fuel injection has been lengthened to serve as central electrode for the plasma system. Two surrounding stainless steel electrodes have been placed symmetrically on the burner above the annular tube to threat the maximum volume of gases.



Fig. 1. Description of the Gliding arc plasma system implementation on the burner

The head of the burner is composed by a piece of alumina both to set the peripheral electrodes and ensure a good electrical insulation from the rest of the burner device. These two electrodes have been specifically shaped to follow the helical movement imposed by the swirler in the annular tube. Two identical pulsed power supplies have been developed to generate the gliding arcs, each one controlling one discharge between the central tube and the surrounding electrode. By this way it is possible to work with one or two gliding arcs in the flame. These power supplies allow to change the plasma characteristic by adjusting the frequency and the power. The pulse duration is fixed at 500 ns with a pulse frequency variable from 1 to 30 kHz. Primary voltage is adjustable between 0 and 350 VDC generating secondary voltage between 0 and 20 kV. Under this system, gliding arcs are produced with an instantaneous power up to 150 W, intentionally selected to be negligible in comparison with the burner power. The choice of flame and plasma parameters on the flame stabilization have been qualitatively checked before each spectroscopic measurement by direct visualization, an example is presented on the Fig. 2. As expected, the effect of plasma on flame stabilization is clearly observed: change of the flame color due to chemistry modification and species radiations, change of the flame shape and behavior related to a partial hanging of the flame on the surrounding electrode edges. In addition, two intense whitish zones appear close to the baseplate of the electrodes which corresponds to plasma stretching areas.



Fig. 2. Photo of the turbulent flame with swirler (Sn=1.4): a°) without GAP and b°) with GAP. $Q_{AIR} = 150 \text{ L/min}$, $Q_{CH4} = 19 \text{ L/min}$ and $P_{FLAME} = 11.3 \text{ kW}$.

For the purpose of the identification of the chemical pathways leading to a better flame stabilization, an optical arrangement for measuring the plasma and flame UV/visible radiations by Optical Emission Spectroscopy (OES) have been implemented. The optical setup used for OES experiments is shown on Fig. 3. This system collects the plasma/flame radiations by a set of 2 inches UV fused silica plano-convex lenses with focal length of 200 mm and 540 mm respectively leading to a linear magnification of 0.37. The choice of the first focal length has been imposed by the dimension of the combustion chamber. These radiations pass through an optical UV fused silica optical fiber bundle to be collected in the entrance slits $(d = 10 \,\mu m)$ of two separated Optical Multichannel Analyzer (OMA). All the optical devices are placed on a three axis motorized translation plate, allowing a thin adjustment of the spatial and the focal lens. The first OMA (Avantes ULS2048Cl-EVO) is used for a quick overview of the radiations over the 300-550 nm broad spectral range where the key reactive species ae located. A second OMA system (ACTON Sp2750i) equipped with an ICCD camera (Princeton PIMAX gen II) have been implemented in parallel to complete the preliminary results. This OMA works with an extensive spectral range between 200 and 800 nm to collect particle emission from the UV to the near IR. Three different grating are mounted on a turret inside the spectrometer to finely adjust the spectral range and spectral resolution $(\Delta\lambda < 0.01 \text{ nm})$. This high resolved spectrometer allows to measure the rovibrational structure of molecular radiative species and to determine plasma temperatures (rotational, vibrational, electronic,...) based on comparison between experimental and theorical spectra using the Specair[®] software [15]. Wavelength calibrations (position and apparatus function broadening) of the spectrometer have been operated with rare gas low pressure spectral lamps (Ar, He and Hg). Due to the strong variation of the collection efficiency over the large

spectral range, the intensity calibration curve have been made by using a calibrated deuterium-tungsten lamp.



Fig. 3. Schematic view of the optical setup for OES experiments

3. Results and discussion

Prior to the experiments, an inventory of the radiative atomic and molecular species formed in the plasma have been realized without fuel (in absence of flame) in order to evaluate the possibilities of key species formation for flame stabilization. The Fig. 4 represents the plasma emission spectra as a function of the pulse frequency in the center of the GAP. An increase of plasma radiation is observed with the rise of frequency, especially for f = 20 kHz, highlighting the more intense energy deposition within the gas with the frequency.



Fig. 4. Effect of the pulse frequency on the plasma radiation: $Q_{AIR} = 30 \text{ L/min}$, $U_{ARC} = 230 \text{ V}$

For all conditions, the emission spectrum between 300 and 400 nm is over dominated by the molecular structure of the second positive system of N_2 . In the UV, only molecular bands form NO are clearly observed. As a consequence, no molecular bands of O_2 are distinguished in these spectral ranges. Only atomic lines of oxygen and nitrogen are noticed in the near IR close to 745 and 777 nm respectively. These atomic emission clearly

demonstrate that the energy deposit is sufficient to form N* and O* reactive radicals. These results widely observed in air plasma at atmospheric pressure [16], highlight the effective production of reactive species like O*, key specie involved in combustion chemistry. However, the formation of reactive species such as NO and N*, intermediate reactants in nitrous oxide formation, can also have a non-negligible effect on the NO_x pollutants production as combustion products. Following these first results demonstrating the possibility of GAP implementation on the burner with only low mechanical modifications, we have tested the interest of plasma generation for flame stabilization and measured the flame emission spectra with and without plasma. An example of obtained results is presented in the Fig. 5.



Fig. 5. Effect of the plasma within the flame at the edge of the electrode. $Q_{TOTAL} = 169 \text{ L/min}, \phi = 1.2$, Swirl 1.4

We observed a huge influence of the plasma in this area at contrary to the flame alone characterized by only weak emission of OH and CH radicals. The identification of the spectral lines [17] highlights the formation of several radicals such as OH (306 nm), CN (388 nm), CH (431.5 nm), NH (337 nm), NO or C₂ (512 nm). Only H atomic lines (656.3 nm) are clearly distinguished except some metallic lines due to electrodes erosion. As a consequence, the plasma bring some benefits on radical production such as CN or NO greatly increasing with the frequency. Nevertheless, the plasma seems do not affect significantly the quantity of key species involved in combustion processes as CH, H, O or OH according to the weak increase of their emission lines. Although the observation of N₂ radiation is not directly interesting (because it can be considered as a non-reactive for combustion chemistry), the vibrational bands structure are sufficiently resolved to be used for temperature measurements within the plasma or the plasma/flame interface. Therefore, the plasma temperatures have been determined by a fitting procedure between the experimental and theorical spectra using Specair® Software [15]. An example of results obtained in the flame front (at the border with the plasma phase) with plasma assistance (f = 20 kHz) is shown on the Fig. 6.



Fig. 6. Example of fit on the 320 -340 nm emission spectrum in plasma assisted combustion. $Q_{TOTAL} = 169$ L/min, $\phi = 1.2$, Swirl 1.4, f_{PLASMA} = 20 kHz

The vibrational and rotational temperatures obtained by the fitting procedure are respectively close to 3601 and 3443 K in this condition against 2812 and 2757 K for a plasma at f = 12 kHz (not represented). Consequently, the plasma seems to generate a slight thermal effect throughout the formation of radicals at the boundary between the flame front and the plasma phase. This phenomenon is clearly enhanced with the increase of the energy deposit by frequency adjustment leading to a higher local production of radicals (mainly NH and CN) with higher temperatures.

4. Conclusion

A preliminary spectroscopic study of the plasma effects on CH₄-air swirling flame at atmospheric pressure has been performed in order to have a better understanding of the chemical mechanisms involved in plasma assisted combustion. A double gliding arc system have been implemented in an existing burner only with low mechanical modifications and operating with a relative low consumption (P = 150W). Analysis of the flame and the plasma radiations by optical emission spectroscopy have been carried out on this turbulent flame in absence or presence of the plasma. The results show the interest of plasma assistance for flame stabilization through the local production of radicals and weak thermal effect, this phenomenon enhanced with the energy deposit (increase of the frequency). Nevertheless, the major species formed (such as N, NO, CN) are typical intermediate reactants in NO_x mechanism in combustion processes and seems to be responsible for the weak NO_x emission increase observed in plasma/combustion. These preliminary results need further investigations in spectroscopic analysis to develop these first observations on the gliding arc plasmacombustion interactions.

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6. References

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