Investigations of carbon monoxide emission in methane flames stabilized by nanosecond pulsed discharges using Mid-IR QCLAS

G.D. Stancu(*)[1,2], D.A. Lacoste[1,2], C.O. Laux[1,2]

[1] *CNRS, UPR 288, Laboratoire EM2C, Grande Voie des Vignes, 92290 Châtenay-Malabry, France*
[2] *Ecole Centrale Paris, Grande Voie des Vignes, 92290 Châtenay-Malabry, France*
(*)gabi-daniel.stancu@ecp.fr

A current application of Nanosecond Repetitively Pulsed (NRP) discharges is the stabilization of lean flames, which are known to produce less NOx. Understanding the plasma-flame stabilization mechanism, reduction of the unburned hydrocarbons and pollutants is of important scientific and technological interest. Carbon monoxide (CO) is a key intermediate species in hydrocarbon combustions. Here, Quantum Cascade Laser Absorption Spectroscopy (QCLAS) at 2055.4 cm^{-1} was used to characterize CO densities emitted by plasma assisted methane-air flames. Open air or confined combustions with and without NRP discharges were studied. The NRP discharges were found to efficiently stabilize lean flames, significantly change the CO density and heat release.

The interest for using lean combustion is the reduction of pollutants, in particular the NOx emission. The disadvantage of the lean combustion is the initiation of flame instabilities, which can lead to flame extinction, hence to unburned hydrocarbons (loss of energy) and pollutants release. One possible stabilization technique is the enrichment of hydrocarbon combustion by hydrogen. However the practical aspects such producing and supply are critical drawbacks.

The nanosecond repetitively pulsed (NRP) discharges have been successfully applied for lean flame ignition and stabilization [1]. The use of NRP discharges is a very promising approach, the discharge power (< 10 W) being less than one percent of the flame power (a few KW). Here, lean CH_4-air premixed flames were stabilized by the NRP discharges at flow rates of 3-10 m/s. For efficient stabilization the discharge was placed in the recirculation zone, behind a bluff body. The NRP discharges were generated using a pin-to-pin electrode configuration (gap distance 4 mm), by 10-ns short high voltage pulses (5-10 kV) at pulse repetition frequencies of 30 kHz. These discharges are strongly non-thermal plasmas (gas temperatures ~ 1000 K, electron temperatures ~ 30000 K) characterized by low energy requirements for high electron and radicals densities [2,3].

Carbon monoxide is one major combustion intermediate species. Emitted in large amounts is a very toxic pollutant and its presence in the exhaust gases is an indication of combustion incompleteness. Its role in multi-component combustion mixture is not entirely understood. For instance new investigations [4] have shown that enrichment of methane with CO can increase the combustion velocity thus pushing the flammability limits towards fuel-lean conditions. On plasma stabilization mechanisms recent studies [3,5] have shown the key role of atomic oxygen, which is largely produced by NRP discharges (dissociation up to 50 % of O_2). However, the CO emission in plasma-assisted combustion was not investigated.

Here, measurements by Mid-IR QCLAS of CO absolute densities in post-flame with and without NRP discharge were compared. The presence of large amounts of water and carbon dioxide in the flame and the exhaust gases (main combustion products) makes the CO detection by Mid-IR absorption techniques very challenging. In addition, the high temperature environment (500 K - 2000K) results in exciting a larger number of transitions from higher rotational and vibrational quantum numbers. The presence of many absorption lines and the high-pressure broadening effect (atmospheric pressure conditions), induce a complex spectral overlapping of plasma-flame species. The absorption spectral simulations have shown a reduced spectral interference for the CO rovibronic transition at 2055.4003 cm^{-1} and thus, this transition was chosen for detection.

A first set of experiments was performed with the flame confined in a metallic tube of 50 cm length and 8 cm diameter. The measurements were performed at the tube output. The temperature of the burned gases at tube output was measured by thermocouples and was found to be 1100 (70) K with negligible variations for the experimental conditions. Abel inversion was applied to the lateral absorbance measurements to obtain local densities. A constant CO molar fraction was found along the tube diameter. Line strength temperature correction was applied to obtain the absolute densities (at 1100 K the line strength increases by a factor 5.5 compared to 300 K).

In figure 1 the CO densities measured in burned gases at 1100 K with and without plasma are shown. Single-pass absorption was used for rich and stoichiometric flames and a White multi-pass cell (20 passes) was used to improve the sensitivity for detection in lean conditions. It can be seen that the amount of CO decreases about three orders of magnitude compared to rich flames (equivalence ratio (ER) >1). Using NRP discharges in lean condition no significant changes of CO density were observed. The burned gases flow in the confinement tube at temperatures in the range of 1100 K – 2000 K. Because the combustion of CO is very efficient ($CO+1/2O_2 \rightarrow CO_2$) at high temperature, its density strongly decreases to $5x10^{13}$ cm^{-3} (8 ppm). In rich flame regime large amounts of CO (< 1%) are measured because the methane combustion in the confinement tube is not finished. When NRP discharge was applied at ER ≥ 1 an increase of about 20-30 % of CO density was measured.

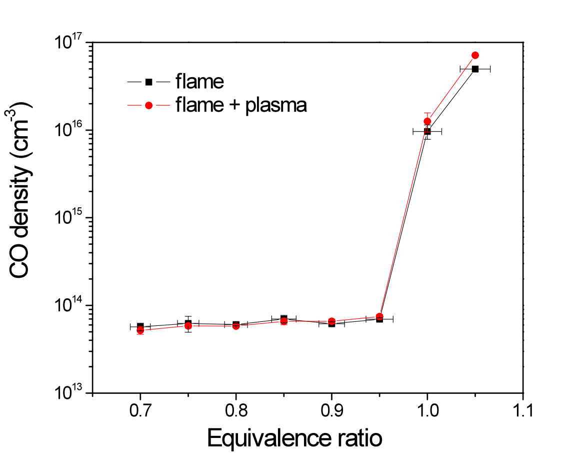

Fig. 1: CO density mesurements in confined system function of ER with or without plasma.

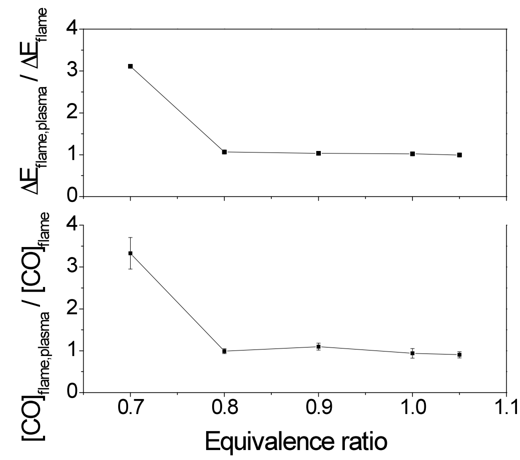

Fig. 2: CO density ratio (bottom) and gas kinetic energy ratio (top) with and without plasma function of ER.

The second set of experiments was performed for the same plasma and flame conditions without the confinement tube. The CO density and temperature measurements were carried out at the same distance from the flame burner. Larger temperature variations (320 K – 570 K) were measured by thermocouples when changing the CH_4-air mixtures and when using NRP discharges. In figure 2 (bottom), the ratio of CO density with and without plasma function of ER is shown. The CO density ratio is about 1 except for ER = 0.7, where the CO density increases about 3 times in the presence of plasma. From the temperature measurements the ratio of the changes of the gas kinetic energy with and without plasma is plotted function of ER (figure 2 top). A similar dependency like for CO ratio is obtained. At ER = 0.7 there is also 3 times increase of the gas kinetic energy in the presence of plasma. This demonstrates, that the use of NRP discharges at the lean flammability limit enhances substantially the combustion degree. Thus, CO emission increases because when using NRP discharges a much larger methane quantity is burned.

NRP discharges are shown to be a very promising technique for the extension of the flammability limits towards lean condition. Mid-IR QCLAS has proven to be a suitable technique highly selective and sensitive, capable of *in situ*, absolute CO density measurements in harsh environments such as plasma-assisted combustion.

References

[1] G. Pilla, D. Galley, D.A. Lacoste, F. Lacas, D. Veynante, C.O. Laux, IEEE Trans. on Plasma Sci., 34, (6) (2006) 2471
[2] C.O. Laux, T.G. Spence, C.H. Kruger, R.N. Zare, Plasma Sources Sci. Technol. 12 (2003)125
[3] G.D. Stancu, F. Kaddouri, D.A. Lacoste, C.O. Laux, J. Phys. D: Appl. Phys., 43 (2010) 124002
[4] A.A. Konnov, G.P. Alvarez, I.V. Rybitskaya, J. De Ruyck, Combust. Sci. Tech. 181 (2009) 117
[5] G.D. Stancu, F. Kaddouri, D.A. Lacoste, C.O. Laux, 40th Plasmadynamics and Laser Conferences San Antonio TX, 3593 (2009) USA