

# Hydrogen addition in a rich premixed ethylene-air flame

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## ABSTRACT

Improving fuel economy and reduction of exhaust emissions have become the major concerns in the combustion community. Due to low pollutant emission and superior combustion characteristics hydrogen/hydrocarbon blends have received increased attention as alternative fuels for numerous power generation applications. The combustion chemistry of hydrocarbon fuels is significantly different from that of hydrogen and the flame structure of composite fuels largely depends on the interaction between the two mechanisms. Studies are needed in order to understand the fundamental combustion chemistry of composite fuels.

In this work we have investigated the effect of hydrogen addition on soot formation in a rich premixed ethylene-air flame produced by a McKenna burner and with an equivalence ratio  $\Phi = 2.34$ . Hydrogen was added up to 50% in volume. Temperature axial profiles of different flames were measured by a radiation corrected thermocouple. A cw Nd:YAG laser was used to measure soot axial profiles. The results indicate that hydrogen has a great influence in reducing the amount of soot in the investigated region of the flame. In order to distinguish between chemical and pure dilution effects, similar flames were also produced by helium addition. Soot reduction has been also observed in this case but to a minor extent. In conclusion hydrogen addition in a premixed ethylene-air flame decreases the soot volume fraction both by a dilution effect and by influencing the chemistry of soot formation.

## INTRODUCTION

Recent research and development in the area of combustion have been motivated by the commitment of preserving a clean environment and to reduce energy consumption. In this direction, hydrogen-hydrocarbon hybrid fuels have received increasing attention. Blends of hydrogen and hydrocarbons show an improved ignitability and flame stability, especially in the lean burning regimes where stable combustion of hydrocarbons is extremely difficult [1]. The use of these fuels has been found to result in improved combustion performance, both in term of combustion ignitability and stability and in term of pollutants emission reduction ( $\text{NO}_x$  and soot). The feature of these fuels are difficult to predict due to the complex and nonlinear nature of chemical kinetics and multi-component diffusion processes. Few of these issues are clearly understood and a primary goal of researches in this field is to investigate fundamental issues surrounding hydrogen-enriched hydrocarbon fuel. After some pioneering work [2] most of the studies performed up to now using hybrid fuels refer to laminar or turbulent jet flames in order to study the stability limits, visible flame appearance, soot and pollutant emission [1,3-7]. The influence of hydrogen addition to fuel on temperature and soot formation in co-flow laminar diffusion flames of ethylene, propane, butane and methane has been experimentally studied [8]. It was found that while methane exhibits a peculiar behaviour, in the case of propane and butane flames hydrogen and helium addition reduces soot by the same amount showing that hydrogen has mainly a dilution effect. On the contrary, in the ethylene flame hydrogen addition reduces soot more than helium, indicating also the presence of a chemical route for soot reduction. More recently, Guo et al. [9], using a computational approach, suggested that the chemically inhibiting effect of hydrogen addition on soot formation can be ascribed both to the H radicals concentration reduction in the surface growth region and to the higher  $\text{H}_2$  concentration in the lower region of the diffusion flame.

In this work, the influence of hydrogen addition to fuel on soot formation was investigated in a premixed ethylene/air flat flame, by using laser diagnostic techniques.

## EXPERIMENTAL APPARATUS

Porous plug McKenna burners are widely used for producing "flat" premixed flame characterized by a wide regions of homogeneous composition and temperature across the flame. The flames are assumed to be mono-dimensional and considered as a standard at least in the lean and close-to-stoichiometric conditions. Nevertheless, McKenna burners are also employed to produce rich premixed flames. It is important to point out that different version of this burners are commercially available, having a stainless steel or bronze porous disk. In a recent paper of Migliorini et al. [10] it was observed that, although fed by the same reactants rates, the two burners made of stainless steel or bronze porous plug produce rich premixed flames very different both in structure and in the amount of soot. In particular the stainless steel burner produces a flame with an annular structure that can not be considered "flat". Therefore studies about soot formation mechanism should be performed in a bronze burner where the assumption of a mono-dimensional flame is better verified.

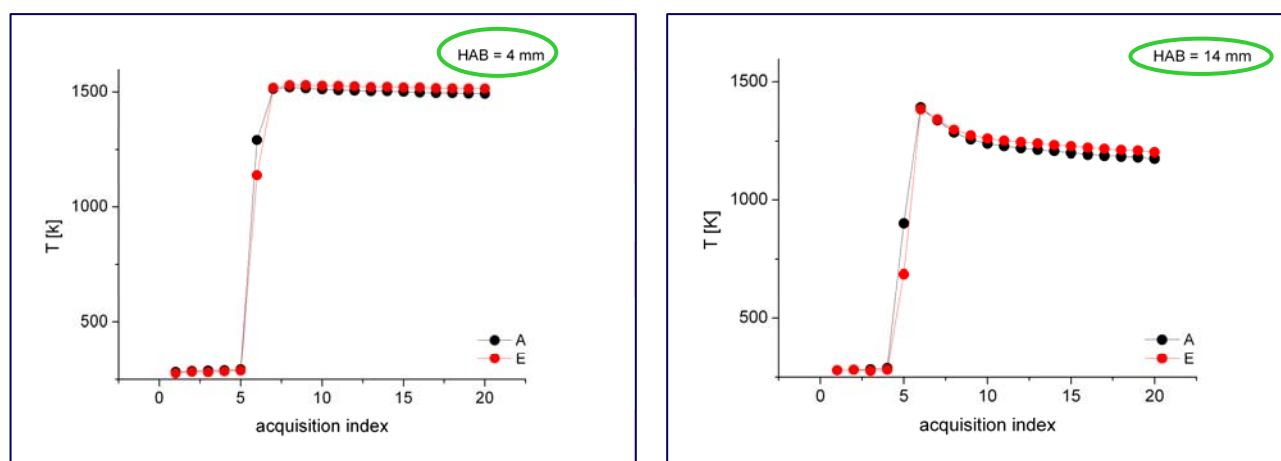
In this work, measurements were carried out on flames produced by a bronze McKenna burner ( Holthius & Associated, Sebastopol, CA). The main body of the burner consist of a water-cooled bronze porous plug of 60 mm diameter. In order to shield the flame from the surrounding air an external shroud of nitrogen, flowing at 15 l/min, was used. Moreover, for flame stabilization, a stainless steel plate of 60 mm diameter was placed at 20 mm height above the burner. The burner was placed on a motorized XYZ table in order to investigate the flames both in the axial and in the radial direction

Investigations were performed on laminar premixed ethylene/air flame with the addition of different percentages of H<sub>2</sub>. For a deeper understanding of the process involved, the same flames were also studied by replacing H<sub>2</sub> with He. The basic flame was an ethylene/air flame of  $\Phi = 2.34$  (flame A). The hydrogen doped flames were obtained by replacing a little amount of flame A ethylene/air mixture with a corresponding amount of hydrogen, leading to different hydrogen percentages, keeping constant several parameters, such as the total flow rate, the cold gas flow velocities (5.84 cm/s) and the C/O ratio (0.775). In this way it was possible to directly compare temperature and concentration profiles for the main species involved in the combustion process. Mass flow-meters (Bronkhorst. AK Ruurlo, The Netherlands) were used for controlling the flow of the gases. The investigated flames are reported in Table 1.

flame	C <sub>2</sub> H <sub>4</sub> [l/min]	H <sub>2</sub> [l/min]	H <sub>2</sub> /C <sub>2</sub> H <sub>4</sub> *100	Air [l/min]
A	1.400	0	0	8.60
B	1.380	0.138	10	8.46
C	1.360	0.272	20	8.34
D	1.346	0.404	30	8.25
E	1.328	0.531	40	8.14
F	1.310	0.655	50	8.03

**Table1.** List of the investigated premixed flames.

Temperature measurements were also performed along the flame axis of the premixed flames, using a 50  $\mu$ m Pt/Pt-Rh (10%) uncoated thermocouple. The thermocouple was inserted in the flame at different heights above the burner and the corresponding voltage output was detected by a voltmeter and acquired by a specific software. The acquisition starts outside the flame and continues after the thermocouple reached the flame axis.



**Fig. 1** Uncorrected temperature history at two HAB of flame A and E.

This kind of procedure, already presented by McEnally et al. [11], has been chosen because it allows to monitor the temperature history of the junction, in particular in the sooting zone. Fig. 1 shows two examples of measurements taken at two different heights while the thermocouple enters the flame and reaches the flame axis at the value of the acquisition index of 9. After this value the thermocouple stays on the flame axis. Low in the flame where soot is not present, the temperature rises and reaches a constant value of approximately 1500 K (uncorrected). High in the flame, where soot is present, the temperature, after an initial increase, shows a constant decrease. This is due to two factors: an initial soot deposit on the junction that changes the emissivity followed by an increase of the diameter. Defining a criterion to choose the exact junction temperature and applying the procedure at different heights above the burner, the gas temperature was evaluated taking into account radiation losses [11-12].

Different optical techniques such as spontaneous emission analysis, laser extinction and laser-induced incandescence (LII) were used during this study. As for the spontaneous emission, the radiation emitted by the flames was collected by

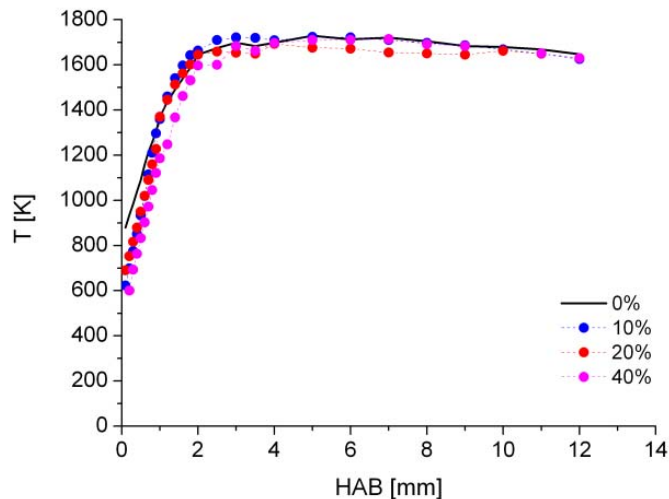
using a Cassegrain objective (Nikon,  $f = 500$  mm, resulting in a 5 m measuring distance), utilized in order to avoid chromatic effects, and a PC driven CCD camera (PCO Pixelfly qe, 1392 x 1024 pixels, 12 bit). An interference filter (647 nm,  $\Delta\lambda = 10$  nm) was placed in front of the CCD camera.

For extinction measurements two different lasers were used, an He-Ne at 632.8 nm and a cw Nd:YAG at 1064 nm. The laser beam was mildly focused on the flame and the transmitted intensity, collected through a two lens system on the entrance of an integrating sphere, was measured with a photosensor (Hamamatsu H5783-01). The signal was processed with a digital lock-in amplifier (Stanford, model SR850 DSP) triggered by a mechanical chopper used to discriminate the transmitted signal against flame emission. The acquisition was driven by a personal computer and each signal resulted from an average over 300 samples.

Laser-induced incandescence technique was implemented by using the fundamental beam (1064 nm, 7 nm FWHM) of a pulsed Nd:YAG laser (Quanta System, SYL 202) operating at 6 Hz. A portion of the beam was selected and imaged on the burner axis by using a lens ( $f = 160$  mm,  $\Phi = 70$  mm). The soot incandescence signal was collected and focused on the entrance slit (1.5 x 3 mm) of a grating monochromator (Hilger and Watts, D 330). The collecting optics consisted of a concave mirror, placed at a certain angle with respect to the incident laser light direction, in order to increase the probe volume, and, consequently, the radiation sensitivity. The LII signal was measured with a photomultiplier (Hamamatsu R955, 2.2 ns rise time) and a fast digital oscilloscope (Tektronix, 1GHz, 5Gs/s), triggered by the laser. The signal, that is the result of an averaging process over 300 individual curves, was stored in a PC and processed with a MATHCAD program. For this kind of measurements no absolute calibration procedure was performed. Concerning the theoretical background of the optical techniques utilized in this work, details are reported in the literature [10] and are not described here.

## RESULTS AND DISCUSSION

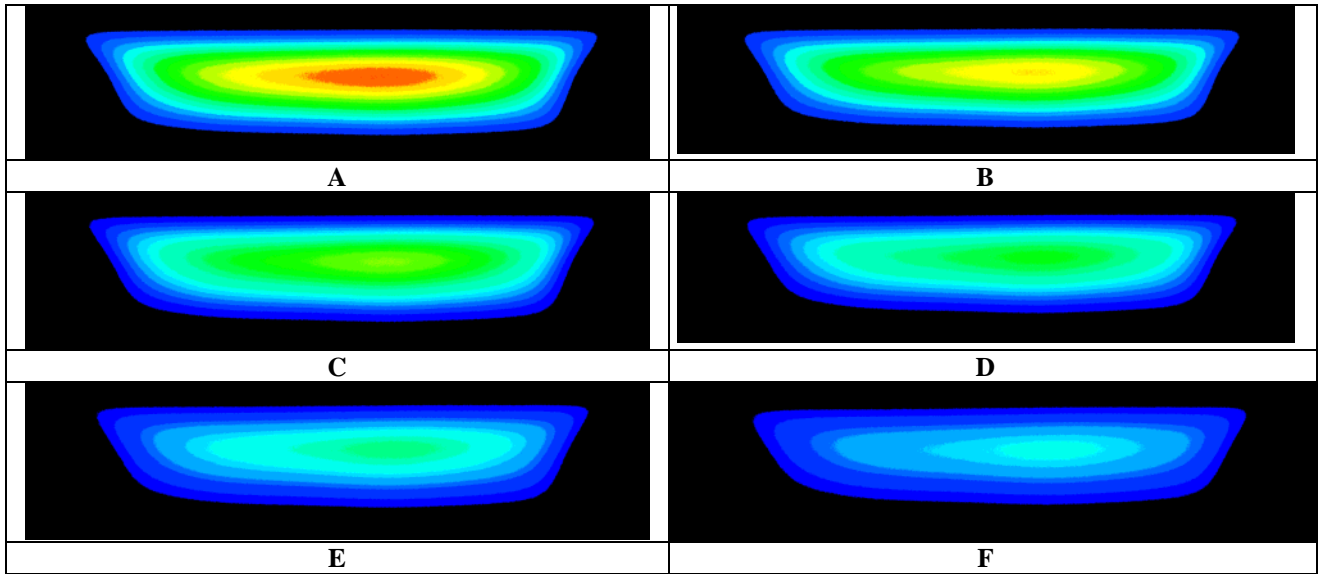
For a first approach in understanding the effects of hydrogen as an additive in this kind of flame, temperature measurements were performed along the flame axis using a 50  $\mu\text{m}$  Pt/Pt-Rh(10%) uncoated thermocouple. The temperature axial profile for flame A, B, C and E are shown in Fig. 2.



**Fig. 2** Comparison between temperature axial profile for flames A, B, C and E.

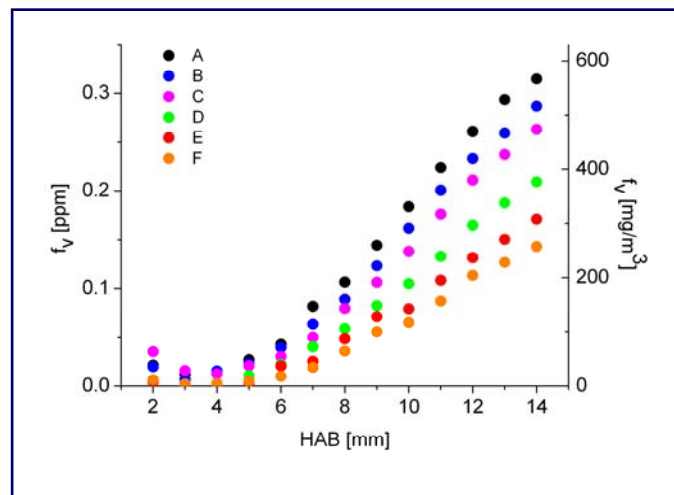
As it can be observed, they are quite overlapped, with only a small reduction of the temperature very low in the doped flames. Therefore, the hydrogen presence in the ethylene flames seems not to affect the temperature field.

Images of the six investigated flame, as detected at 647 nm, are reported in fig. 3. Increasing the hydrogen addition the flame luminosity decreases significantly indicating a strong influence of hydrogen addition on soot formation in spite of a minor influence on the temperature profiles. The intensity of soot radiation is given by  $I_s = \tau \varepsilon(\lambda, fv) I_{BB}(\lambda, T)$ , where  $\varepsilon(\lambda, fv)$  is the soot emissivity depending on the wavelength,  $\lambda$ , and on soot volume fraction,  $fv$ , in a linear way in the low absorption regime.  $I_{BB}(\lambda, T)$  is the black body radiation described by the Planck law and depending on wavelength and temperature,  $T$ . If the temperature profile is identical for all flames the radiation intensity is proportional to the soot volume fraction. The image intensity is then correlated to the total amount of soot present in the flames. Figure 3 shows that the decrease in soot volume fraction is almost proportional to the addition of hydrogen.



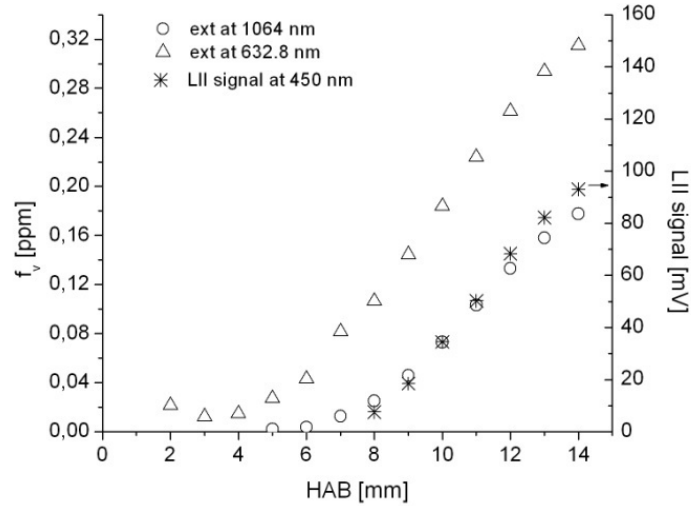
**Fig. 3** Images of the six investigated flames ( $A = 0\% H_2$ ,  $B-F = 10-50\% H_2$ ).

In order to obtain a quantitative measurement of the soot volume fraction distribution in the flames, extinction measurements were performed. For this purpose it is necessary to know the absorption pathlength and the refractive index of soot. The flame images, acquired by the CCD camera as described previously, have been used to measure the soot absorption length,  $L$ , at various heights above the burner (HAB). As for the refractive index of soot, the value of Chang and Charalampopoulos [14] has been adopted. Usually for this kind of measurements a visible laser beam is used. Initially the measurements were performed using an He-Ne laser at 632.8 nm. Figure 4 shows the results of the axial profile of soot volume fraction in ppm and  $mg/m^3$ . The figure shows that absorption starts at about 5 mm HAB and increases linearly with the height. The hydrogen addition decreases the amount of soot in an almost linear way, in good accord with the images of fig. 3.



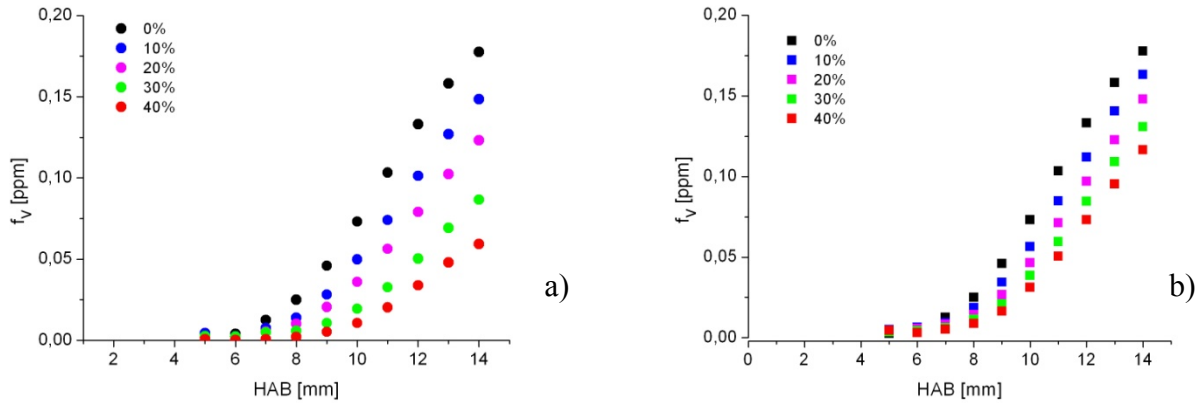
**Figure 4.** Soot volume fraction axial profile for the investigated flames as measured at 632.8 nm.

As described in Ref [18], the values of the soot volume fraction obtained in the red could be overestimated with respect to the values obtained with an IR source such as the fundamental output of a cw Nd:YAG laser at 1064 nm. In order to investigate this possibility we have applied the laser-induced incandescence (LII) technique. As widely reported in the literature [21-24] the peak of the time decay incandescence signal, usually called “prompt” LII, is proportional to the soot volume fraction, while the value of the decay rate after the laser pulse is related to the soot particle size. In Fig. 5 the axial profiles of soot volume fraction measurements for flame A obtained by LII (crosses), by extinction at 1064 nm (open circle) and at 632.8 nm (open triangle) are reported.



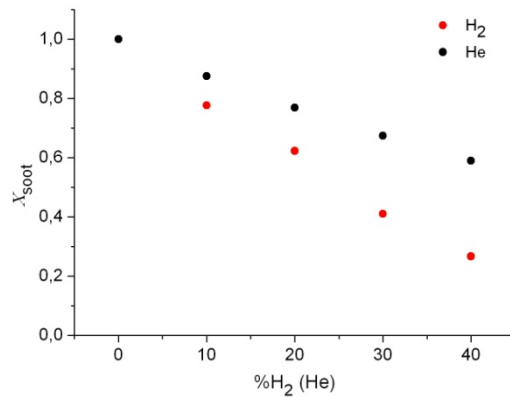
**Fig. 5** Axial profiles of LII signal and  $f_v$  obtained from extinction measurements.

The two sets of extinction measurements present a very different behavior. While with the red laser an extinction and consequently an estimation of  $f_v$ , is measured beginning from  $HAB = 5$  mm, with the IR laser soot particle formation is detected starting from 7 mm. Moreover the slope of the two sets is also different. In the same figure, the value of the incandescence vs the height above the burner is reported and properly scaled on the right axis. The values are almost overlapped with the IR measurements of  $f_v$ . To understand the physical meaning of this different behavior obtained it is important to consider that while extinction measurements are related to absorption coefficient and consequently to the different species absorbing at the wavelength used, LII is strictly dependent on the carbonaceous particle matter. Then, it can be inferred that other gas-phase species are responsible for the absorption in the red. Therefore, in order to investigate the effect of  $H_2$  or He addition on soot volume fraction extinction measurements were performed at 1064 nm.



**Fig. 6** Axial profiles of soot volume fraction at different flames percentages of  $H_2$  (a) and He (b) addition.

In Fig. 6 the axial profile of soot volume fraction are shown for all flames reported in Tab. 2 in the case of  $H_2$  (a) and He (b) addition. As it can be observed, increasing the percentages of  $H_2$  or He in the flame, an important reduction of the soot volume fraction is obtained. Anyway, this reduction results to be more significant in the case of  $H_2$  than the He addition. For example, considering the 14 mm height above the burner,  $f_v$  decreases about three times by adding 40%  $H_2$ , but only 1.5 times the same percentages of He. Also in this case, in order to compare more directly the different flames in term of their tendency to produce soot, a proper criterion was considered. Starting from these axial profiles, and taking into account the flat structure of the soot distribution across each horizontal flame section, the total soot content can be derived. The results are reported in Fig. 7, where the soot conversion,  $\chi_{soot}$ , defined as the ratio between the produced soot [ $mg/m^3$ ] and the fed  $C_2H_4$  [ $mg/m^3$ ], is traced (normalized with respect to flame A) as a function of the hydrogen (helium) percentage.



**Fig. 6** Total soot load and ethylene fractions with reference to the flame A versus the percentages of H<sub>2</sub>(He).

The reduction due to helium addition is less than the one obtained with hydrogen addition. While with He addition the decrease of soot load is only due to a dilution effect, with H<sub>2</sub> both dilution and chemical effects have to be considered.

## CONCLUSIONS

The effect of hydrogen addition to fuel on temperature and soot formation has been investigated in a premixed laminar ethylene/air flame. No significant effects are observed on axial temperature profiles adding an increasing percentages of hydrogen to the fuel. On the other hand, it was found that an increase in hydrogen or helium quantity results in an overall reduction of the soot amount. Nevertheless, different behaviors were observed: both diluents lead to a reduction of soot formation; meanwhile helium shows just a dilution effect, hydrogen induces a stronger reduction of soot, linked with a chemical effect. Moreover comparison between extinction measurements performed in the red and in the IR spectral regions shows that a substantial amount of the extinction must be attributed to non-soot gaseous species especially present in the lower part of the premixed flames. Future work will be devoted to modeling the effect of hydrogen addition by focusing on the processes of nanoparticle formation.

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