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STUDY OF SOOT FORMATION IN LAMINAR NON-PREMIXED FLAMES DILUTED WITH INERT GAS

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Abstract. *The natural gas from Brazilian pre-salt layer contains high concentrations of inert gases in its composition. Thus, it is of fundamental interest of oil industry to characterize the flames produced from this fuel. Therefore, the main objective of this work is the qualitative evaluation of the soot production in laminar non-premixed flames for different dilutions. The flames were stabilized on a Santoro burner in which the flame power was held constant while the proportion of inert gas added to the fuel stream was increased. It was used either carbon dioxide or nitrogen as inert gas in dilution process. The soot measurements in each case were performed applying the laser-induced incandescence technique. Images of the incandescence intensity, which are proportional to the amount of soot produced, were taken and the amount of soot produced was compared in each case. The dilution leads to a suppression in the soot production for both inert gases, especially in flames diluted with carbon dioxide.*

Keywords: *soot formation, inert gas dilution, laser-induced incandescence, natural gas.*

1. INTRODUCTION

Soot is of great interest in engineering, since its formation increases the heat transfer by radiation, which can be a positive or negative characteristic for the considered system. In industrial heating equipment, such as a boiler, the presence of soot increases the heat exchange by radiation from the flame to the system walls. Thus, where a large and uniform heat transfer is required, the presence of soot in combustion may be positive. On the other hand, the excessive presence of particulate matter in diesel engines is an example where soot is harmful. These particles resulted from an inadequate combustion process can become logged in the seat of valves, preventing a proper seal in the combustion chamber and lead to an inadequate compression in the cylinder, as shown by Caetano, *et al.*, 2015. Another example is in gas turbines, where the presence of soot affects the life of blades due to abrasion. The emission of this type of particulate into the atmosphere causes harm to the environment, since there is a significant deterioration in air quality. Also, it induces respiratory problems in humans and it is associated with the occurrence of cancer.

There are several techniques able to measure the properties of soot in flames, each one with their own limitations. Optical techniques using laser are often the method of choice due to accuracy, spatial and temporal resolution. In addition, they are non-intrusive and have simple application.

According to Hadef *et al.*, 2010, over the past 20 years, laser-induced incandescence has proven in numerous studies to be a useful tool in soot volume fraction measurements, resolved in time and space for a wide range of applications, such as laminar and turbulent flames, combustion within the engine cylinders, and characterization of the engine exhaust gases. This technique provides quick results of the soot volume fraction with high spatial and temporal resolution, and performs real-time measurements, according to Vander Wall, *et al.*, 1994.

Soot formation in pure hydrocarbon flames or fuel mixtures is the subject of several studies, as described by Machado, 2015. However, the effect of diluting fuel with inert gases is not complete explored. The increase in use of alternative energy sources such as biogas evidences the importance to study the effects of fuel dilution, considering this gas is commonly found mixed with methane from syngas.

In addition to the use of fuels such as biogas, carbon dioxide is present in the natural gas from new sources of oil, such as the Brazilian pre-salt layer, which, according to Ferro and Teixeira, 2009, has carbon dioxide concentrations up to 12% in volume. In this scenario, the importance of soot production study in hydrocarbon flames diluted with inert gases is evident.

In this study, the formation of soot in laminar diffusion flames diluted with inert gas was evaluated employing the laser-induced incandescence technique (LII). Different levels of dilution and two diluent species, namely carbon dioxide and nitrogen, were explored.

2. BACKGROUND

2.1 Soot formation in flames

According to Turns (2000), soot is a product generated by a partial combustion of hydrocarbons. In a non-premixed flame, soot formation occurs at the fuel rich region and in temperatures between 1300 K and 1600 K. This process occurs in four steps: (i) formation of precursor species, (ii) formation of incipient particles (iii) growth and agglomeration of particles, and (iv) oxidation of particle, as described by Glassman (1988).

The first step comprises the acetyl radical reactions and polycyclic aromatic hydrocarbon (PAH) formation that appears to be an important bridge between the fuel molecules and the primary particles of soot. This step has great influence of chemical kinetics, although the mechanisms and chemical identification of the precursor species are still subject of study.

The second stage comprises the conversion of macromolecules of hydrocarbon in particles with critical sizes greater than 3000 atomic mass units. This process occurs by nucleation from the PAH, forming little soot particles with variable molecular weight.

In the third step, the small primary soot particles continue their trajectory in the flame. In this step occurs the growth of the primary particle by surface growth and the agglomeration with other particles.

The final step occurs when the particle reaches a region with oxidant species. During the passage of the particles through this region, they may be totally oxidized. If all particles are oxidized, there is no soot emission. Otherwise, a sooty flame is observed.

The formation of soot is a characteristic of the fuel associated with the conditions under which combustion occurs, and there is not a parameter that defines the amount of soot formed by the amount of fuel burned, according to Glassman (1988). The type of fuel allows estimating only the tendency to the formation of soot. Furthermore, the soot formed by different fuels does not present significant difference after the formation of the aromatic rings.

2.2 Effects of dilution with inert gas

The insertion of diluents in the fuel stream with total flow rate kept constant reduces the visible flame length. This reduction occurs due to the decrease of the amount of fuel as the dilution increases.

According to Turns *et al.*, 1993, dilution effects analyzed in constant power flames, due the constant fuel flow rate, have shown that laminar flames diluted with nitrogen up to 60% in mass, i.e., maximum of 35% in volume, do not present significant visible flame length variation. It shows that flames suffer greater influence from buoyancy than the momentum from fuel jet. In this condition, the nitrogen addition does not significantly affect the density of the gases produced by the reaction. They also observed that nitrogen dilution in methane flames reduces the radiant fraction in turbulent flames, and also to propane flames, ethylene and mixtures of carbon monoxide with hydrogen. This reduction in radiant fraction with increasing dilution is more intense for sooty fuels.

Mishra and Kumar, 2010, shown that laminar flames of liquefied petroleum gas (LGP) diluted with nitrogen present greater free soot formation length in comparison to pure LGP flames. Also, was observed a decrease of yellowing light from the flames, indicating a decrease in soot formation.

3. METHOD

3.1 Overview of studied flames

The results were obtained from laminar non-premixed flames of natural gas (NG). Either carbon dioxide (CO2) or nitrogen (N2) were used to obtain different dilution levels. The flow of natural gas was constant in all flames, thus the flame power was constant. Table 1 presents the studied cases. The flames were stabilized in a Santoro burner, proposed by Santoro *et al.*, 1983, in which the annular flow was set to 100 ls/min.

Table 1. Flames configurations.

Nomenclature	Flow at 300 K and 101,325 KPa [ls/min]				Dilution
	NG	CO2	N2	Total	[% Volume]
NG	0.5	0	0	0.5	0
C10	0.5	0.056	0	0.56	10
C20	0.5	0.125	0	0.63	20
C30	0.5	0.214	0	0.71	30
N10	0.5	0	0.056	0.56	10
N20	0.5	0	0.125	0.63	20
N30	0.5	0	0.214	0.71	30
N40	0.5	0	0.333	0.83	40

3.2 Evaluation of soot production

According to Hadef, *et al*., 2010, the LII technique involves heating the soot particles to approximately 4000 K using a high power laser pulse. Figure 1 presents the laser apparatus employed:

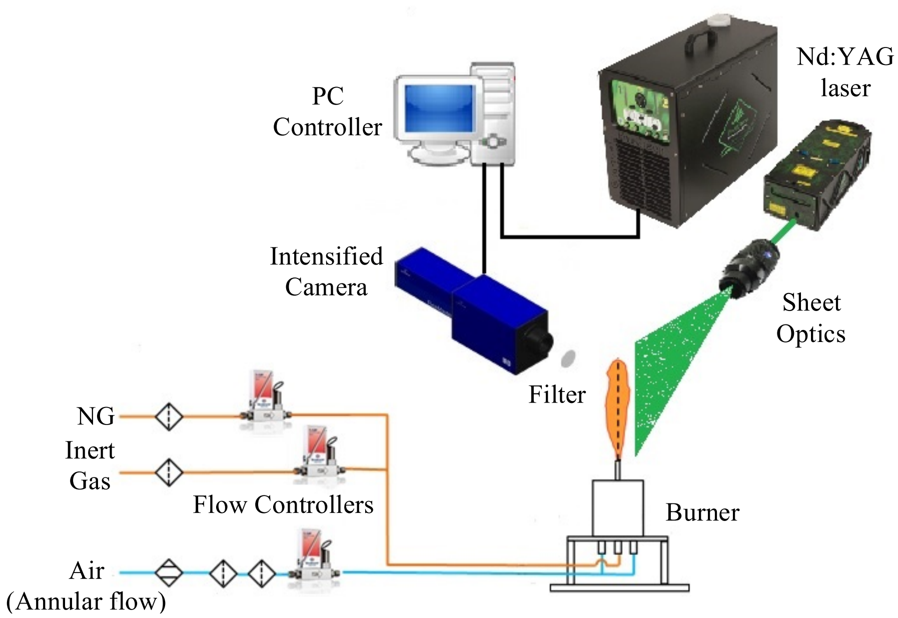

Figure 1. Laser apparatus representation.

Soot is a broadband absorber, and although the absorption cross-section increases with decreasing wavelength, in principle any wavelength in the visible or IR regions can be used to heat the particles for LII detection. Ultraviolet excitation wavelengths of 266 and 355 nm have been shown to generate LII, but shorter UV wavelengths preferentially promote photo dissociation instead of particle heating, as showed by Schulz *et al*., 2006. Nd:YAG lasers are frequently used for LII excitation because these lasers are generally reliable, readily available and provide high-power pulses with typical pulse durations of 8-15 ns.

The system used to perform LII technique was the Soot Master from LaVision GmbH. The laser Nd:YAG (Quantel, Evergreen) used has 200 mJ per pulse, with a 10 Hz repetition rate. Each pulse has 7.4 ns of duration and was performed at 532 nm of wavelength producing a laser light beam of 5 mm of diameter. A light sheet is formed with a height of approximately 100 mm at the center of the burner, using a cylindrical negative lens (f = -10 mm) in tandem with a plan-convex lens of f =30 mm. The waist thickness obtained has about 500 µm, which was placed in the flame center.

The image results considered to each dilution ratio is an average taken from 600 measurements. Hadef *et al*., 2010, acquired one hundred of LII images for each case studied, thus the number of measurements used in this work seems to be sufficient to achieve good accuracy. The exposure time of the CCD sensor was set to 3.100 µs, which capture the phosphorescent signal of an image intensifier with delay set to 5 ηs, gate of 40 ηs and 50% of gain. The distance between the center of the burner and the camera lens was adjusted to 0.547 m and the focal length to 24.7 mm. The optical band-pass filter employed (placed in front of the camera) is centered at 360 nm, to prevent detection of laser light scattering from soot particles and to reject most background luminosity.

The laser spatial and temporal profiles have a marked impact upon the LII signals that are generated. The laser temporal profile has been received little attention since most researchers are currently employing similar Nd:YAG lasers with approximately 7 – 10 ns pulse durations. However, spatial profile of the laser beam has received considerable attention, since the particles in the probe volume experience different fluence and should be heated to different peak temperature, as showed by Schulz, *et al*., 2006. The Nd:YAG laser apparatus used in this work has a top-hat profile, as informed by manufacturer, wich result in an approximately constant fluence for the volume probe.

The laser fluence has a large impact on LII-signal intensities. Temporally-integrated and time-resolved LII signals depend strongly on fluence since different heat-loss mechanisms dominate at different laser fluences. According to Michelsen, *et al*., 2003, at fluences above 0.2 J/cm^2 for 532 nm excitation in a flame, the peak signal is nearly fluence independent for an 8 ns long laser pulse with a homogenous top-hat spatial profile.

When the signal is gated, i.e. it is temporally integrated over the gate width, the signal also increases with increasing fluence at low fluences. At fluences just above 0.2 J/cm^2 for 532 nm excitation and above 0.3 J/cm^2 for 1064 nm excitation, the gated LII signal from soot in flames is frequently observed to be nearly independent of fluence, as showed by Axelsson, *et al.*, 2000. Considering the information above, a fluence of about 0.21 J/cm^2 was used in this work, due to excitation laser fluences within this plateau region have been preferred in soot volume fraction measurements in practical applications with strong laser attenuation or laser power fluctuations.

4. RESULTS

Figure 2 presents the flame photographs at the top and the image obtained from LII signal for flames with carbon dioxide dilution at the bottom. Figure 3 presents the same results, for nitrogen dilution.

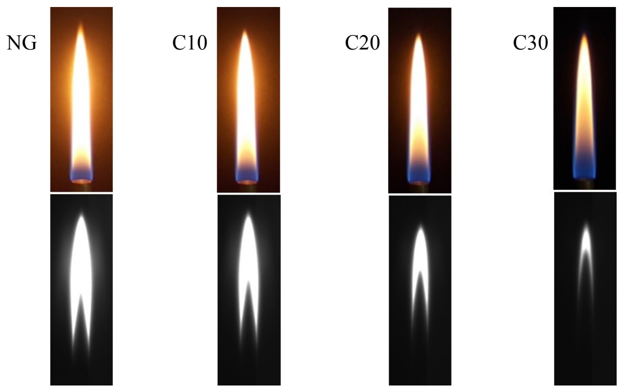

Figure 2. Flame photographs and, below, LII images for each studied flame diluted with CO_2.

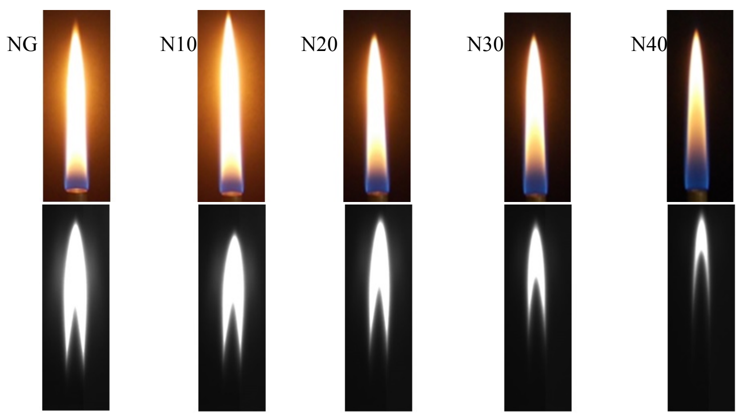

Figure 3. Flame photographs and, below, LII images for each studied flame diluted with N_2.

The direct flame images show a reduction of the flame luminosity as the inert gas is added. This evidence indicates a reduction of soot production for higher dilution levels. The images of LII signal present an increase in the free soot formation length. This behavior is the same for dilutions with both carbon dioxide and nitrogen. The higher luminous intensity region occurs for higher heights above burner, as the dilution is increased.

The LII technique detected the presence of soot for dilutions of up to 30% of carbon dioxide, although yellowing flame was still observed in direct image, indicating the presence of soot. Dilutions with nitrogen were performed detecting presence of soot in conditions up to 40% of dilution.

In order to map the soot presence in different portions of the flame, the LII signal intensity is analyzed in several cross sections at different highs above the burner (HAB). The profiles were obtained in the height range from 40 to 90 mm above the burner, with 10 mm pitch. Figure 4 and Fig. 5 shows these profiles for all configurations presented in Tab. 1. The results of incandescence intensity (vertical axis) were normalized with the maximum signal obtained for the pure flame case and are shown on the same scale for all cases. The horizontal axis represents the flame thickness normalized by the diameter of the fuel injector tube (*x/D*).

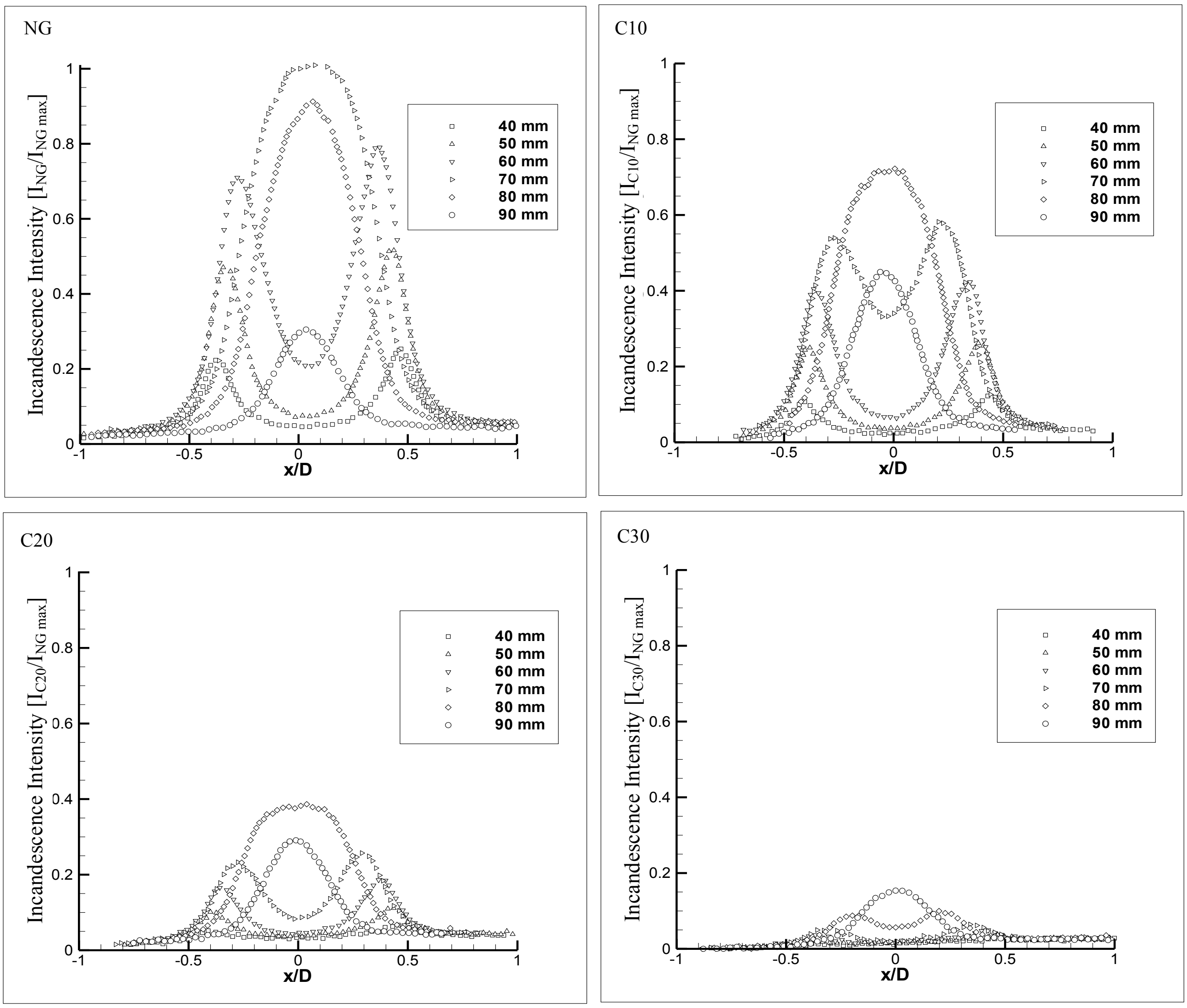

Figure 4. Profiles of LII normalized signal for NG flame configuration and flames diluted with CO2, according to Tab. 1, for different heights above burner.

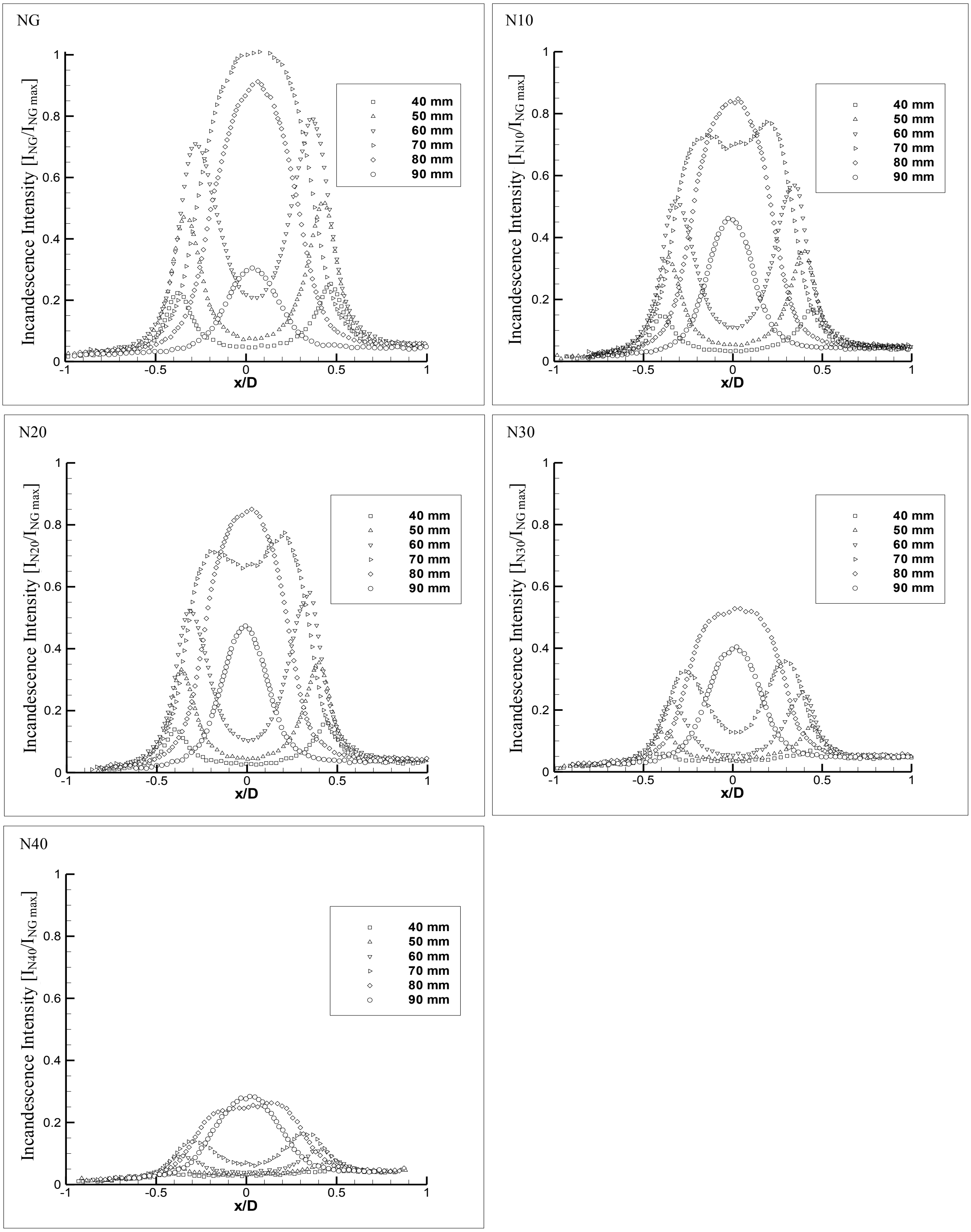

Figure 5. Profiles of LII normalized signal for NG flame configuration and flames diluted with N2, according to Tab. 1, for different heights above burner.

The results show a decrease of the soot signal with the dilution level for both carbon dioxide and nitrogen. However, flames diluted with carbon dioxide presented a higher reduction in soot formation when compared with flames diluted with nitrogen.

The reduction in soot formation with inert dilution showed in the present results can be explained by a chemical effect, as showed by Axelbaum *et al*., 1988, Axelbaum and Law, 1990 and Glassman, 1998. This chemical effect can be interpreted as the fuel concentration reduction in the reaction zone, which provides a lower concentration of soot precursors and, consequently, soot formation itself.

There are also changes in adiabatic temperature of the flame, in each dilution configuration. According to Glassman (1988), the soot formation is determined by the competition between fuel pyrolysis reactions, oxidation of soot precursors and oxidation of soot itself. Increasing temperature enhances the two processes, but the oxidation reactions are more enhanced than pyrolysis reactions. In non-premixed flames, the pyrolysis and oxidation reactions do not occur in the same region. Thus, reducing the temperature decreases the precursors formation rate due to the decrease in the pyrolysis reactions intensity. Since flames diluted with carbon dioxide results in lower adiabatic temperatures than flames diluted with nitrogen, the greater suppression of soot production in flames diluted with this inert gas is an expected result.

However, some considerations regarding the obtained results must to be done. According to Fig. 4, for the same cross section, the peak of the signal intensity is greater at $x/D > 0$, even though the laser incidence occurs at the opposite side. It can be explained by the inhomogeneity on the image intensifier sensor, that present growing sensibility from left to right hand side. New measurements are under work to correct such effects.

Attenuation of the LII signal due to extinction effects in the path from the volume probe to the ICCD camera are believed to be negligible for low soot volume fraction as in the present study.

Moreover, the flame temperature for each dilution case is not equal. Then a direct comparison between different dilution ratio of same inert gas and comparison of different inert gas is not possible. The effect of the flame temperature reduction due to dilution on the LII signal is under investigation in our group through a modelling approach. The next efforts will be directed to include the considerations above in order to quantify the total soot production and to identify the region of maximum soot production for each dilution case.

5. CONCLUSION

The evaluation of soot production of laminar non-premixed flames of natural gas was performed applying the laser induced incandescence technique. The flames studied were diluted with carbon dioxide and nitrogen.

The results for soot production of several regions of the flame shown that the addition of nitrogen causes a reduction in the soot formation. For the same dilution ratio, the carbon dioxide caused a higher reduction, even considering the limitations of the technique employed.

The addition of both inert gases also increased the free soot formation length.

Dilution of natural gas with inert gases changed the flame characteristics, like the color observed in the flame, as well as the soot production. The effect of inert dilution on soot formation has two sources. The first one is the reduction of precursor concentration. The second is the reduction of the flame temperature, which explains the more intense soot suppression in CO2 diluted flames.

6. ACKNOWLEDGEMENTS

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7. REFERENCES

Axelbaum, R. L., Flower, W. L. and Law, C. K., 1988. "Dilution and temperature effects of inert addition on soot formation in counterflow diffusion flames". *Combustion Science and Technology*, V. 61, p. 51-73.

Axelbaum, R. L. and Law, C. K., 1990. "Soot formation and inert addition in diffusion flames". *Symposium (International) on Combustion*, V. 23, n. 1, p. 1517-1523.

Axelsson, B., Collin, R. and Bengtsson, P. –E., 2000. "Laser-induced incandescence for soot particle size measurements in premixed flat flames". *Appl. Opt.*, V. 39, p. 3683-3690.

Caetano, N. R., Soares, D., Nunes, R. P., Pereira, F. M., Schneider, P. S., Vielmo, H. A. and van der Laan, F. T., 2015. "A comparison of experimental results of soot production in laminar premixed flames". *Open Eng.*, V. 5, p. 213-219.

Eckbreth A. C., 1977. "Effects of laser-modulated particulate incandescence on Raman scattering diagnostics". *J. Appl. Phys*. V. 48, p. 4473-4483.

Ferro, F. and Teixeira, P., 2009. "Os desafios do Pré-Sal". *Câmara dos Deputados*. Brasília, p. 79.

Glassman, I., 1988. "Soot formation in combustion processes". Symposium (International) on Combustion, V. 22, n. 1, p. 295–311.

Glassman, I., 1998. "Sooting laminar diffusion flames: effect of dilution, additives, pressure and microgravity". *Symposium (International) on Combustion*, V. 27, n. 1, p. 589–1596.

Hadef, R., Krüger, V., Geigle, K. P., Tsurikov, M. S., Schneider-Kühnle, Y. and Aigner, M., 2006. “Measurements of soot size and concentration in the laminar premixed flame”. *Oil Gas Sci. Technol.,* V. 26, p. 691-703.

Hadef, R., Geigle K. P., Meier W. and Aigner M., 2010. “Soot characterization with laser induced incandescence applied to a laminar premixed ethylene-air flame”. *International Journal of Thermal Sciences*, V. 49, p. 1457-1467.

Machado, I. M., 2015. *Caracterização experimental da radiação térmica emitida por chamas não pré-misturadas de metano diluído com* CO_2. Master thesis, Federal University of Rio Grande do Sul, Brazil.

Melton, L. A., 1984. “Soot diagnostics based on laser heating”. *App. Opt*., V. 23, p. 2201.

Michelsen, H. A., Witze, P.O., Kayes, D. and Hochgreb, S., 2003. “Time-resolved laser-induced incandescence of soot: the influence of experimental factors and microphysical mechanisms”. *Appl. Opt.*, V. 42, p. 5577-5590.

Mishra, D. P. and Kumar, P., 2010. “Effects of N2 gas on preheated laminar LPG jet diffusion flame”. *Energy Conversion and Management*, V. 51, p. 2144-2149.

Santoro, R. J., Semerjian, H. G. and Dobbins, R. A., 1983. “Soot particle measurements in diffusion flames”. *Combustion and Flame*, V. 51, p. 203-218.

Turns, S. R., Myhr, F. H., Bandaru, R. V. and Maund, E. R., 1993. “Oxides of nitrogen emissions from turbulent jet flames: part II – fuel dilution an partial premixing effects”. *Combustion and Flame*, V. 93, p. 255-269.

Turns, S. R., 2000. *An introduction to combustion: concepts and applications*. 2ª. ed. Boston: McGraw-Hill, 2000, ISBN 0072300965.

Schulz, C., Kock, B.F. and Hofmann, M., 2006. “Laser-induced incandescence: Recent trends and current questions”. *Appl. Phys., B 83, 333-354.*

Vander Wal, R. L. and Weiland, K. J., 1994. “Laser-induced incandescence: development and characterization towards a measurement of soot-volume fraction”. *Appl. Phys*., 445.

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