

## SOOT AND CH\* EMISSION SPECTROSCOPY IN AN ACOUSTIC EXCITED JET DIFFUSION FLAME OF LPG.

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**Abstract.** *This work presents an experimental study of the emission of soot and instable radicals ( $CH^*$ ,  $C_2^*$ ) in an acoustic excited jet diffusion flame of LPG (Liquefied Petroleum Gas), at various frequencies (500 Hz to 600 Hz) and acoustic pressures (up to 2800 Pa). The spectra were taken at several positions of the flame, using a medium-resolution spectrometer (0.3 nm) coupled to an optic fiber. The emission bands of the  $CH^*$  (387 nm and 430 nm) and  $C_2^*$  (463 nm, 516 nm, 563 nm) have been observed; soot emission has been observed at 620 nm. Acoustic excitation promotes air entrainment in the fuel region. Spectroscopy measurements have shown an enhancement of the radical emission on the burner exit for high amplitudes of the applied acoustic wave. It has also been found that for 575 Hz and 600 Hz acoustic frequencies the flame showed the highest radical relative concentration (most effective combustion). It suggests that the characteristic frequency of the system is near those frequencies. Some acoustic excitations have caused a local soot increase due to local temperature enhancement and low  $O_2$  concentration to complete the combustion process. It was observed that the curves of relative concentration of radicals  $C_2^*$  and  $CH^*$  as a function of the position in the flame follow the same trend for all the flames studied.*

**Keywords:** *emission spectroscopy, acoustic excited flame, soot,  $CH^*$  radical,  $C_2^*$  radical.*

### 1. INTRODUCTION

The pulsating flame is a kind of combustion that takes place in an environment where the physical parameters of the combusted mixture (pressure, density and velocity) change periodically depending on the time between the two limits of opposite polarity. The pulsating combustion can be obtained by pressure oscillations produced by a loudspeaker in a burner. The acoustic actuation enhances the mixing rate between fuel and air, increasing turbulence in the flame region, and decreasing CO, soot and unburned hydrocarbon (UHC) emissions (Lefebvre, 1983).

The process has been identified as one of the technologies that can meet the objectives of clean combustion and good fuel economy. Pulsating combustion offers low emissions of pollutants, high heat transfer and efficient combustion. The benefits of the pulsating combustion depend on several factors, such as combustible used, type of mixture, equivalence ratio, flow (laminar or turbulent) and characteristics of the burner. The disadvantages of this process are the intensity of the sound (noise) and the danger due to stability loss of the flame and metal destruction by fatigue.

Various applications of pulsating combustion can be found, mainly as drying and heating devices, of which the latter also have had commercial success. Due to the lack of a well-founded theory of the phenomena, it is difficult to predict the operating frequency, the heat release etc, of a particular device. Research concerning control over the combustion process is essential for developing high efficiency pulsating combustors with low pollutant emissions.

The emission spectroscopy in the ultra-violet and visible part of the spectra can provide valuable insights into flame processes. In this spectrum range the emissions are chemiluminescent and come from excited radicals. In the flame, the presence of radicals evidences the occurrence of chemical reactions and the position of the flame front. The spontaneous emission from flames can be continuous or discontinuous. The continuous spectrum is observed, for example, in the sooty region of rich hydrocarbons flames. In this kind of spectra, the emitted energy is distributed in a continuous manner between all wavelengths within a certain domain, and usually presents a maximum. On the other hand, in the discontinuous spectrum the energy is confined around certain narrow wavelengths and is mainly attributed to isolated atoms or molecules. Molecules present the lines arranged in groups called band.

Chemiluminescent emissions in the ultra-violet and visible areas have considerable intensity for some unstable radicals produced in the reaction zone of the flame. The primary combustion zone of hydrocarbon flames emits mainly  $CH^*$  bands (387 nm and 432 nm) and  $C_2^*$  bands (436 nm, 516 nm and 563 nm) besides the ultra-violet  $OH^*$  bands (283 nm and 308 nm) (Leandro, 2004).  $CN^*$ ,  $HCO^*$ ,  $CO^*$ ,  $NH^*$ ,  $NH_2^*$  emission bands can also be observed. If there is oxygen deficiency (rich flames) the flame becomes yellow to white luminous as a result of the thermal continuous emission of unburned soot particles (Gaydon, 1957, Zizak, 2000).

There is no general description for the emission of the flames because the radiation is sensible to many factors, as temperature, fuel-oxidant mixture, equivalence ratio, fuel pureness, burner construction, combustion regime (laminar or turbulent), flame region and the spectrometer resolution. The intensity relations of the bands in a spectrum are of

greatest importance for practical spectroscopy. The intensities emitted by a flame are the integral along the observation path and some of the emitted photons can be absorbed inside the flame. As a consequence the beam emitted at a given point is weakened during its travel towards the edge of the flame.

Due to the short lifetime of the excited radicals it is possible to associate the local chemiluminescence with the real zone of its formation. Quantitative relation between emission intensities and concentration is difficult. This intensity, besides the radical concentration, depends on the flame temperature, collisions with the excited radical and other species that causes non-radiative energy transfer, possible absorptions of the emitted radiation, etc.

The purpose of this work is to perform a global chemiluminescent emission investigation of diffusion LPG (Liquefied Petroleum Gas) flame in a cylindrical tube under acoustic excitation. The chemiluminescent of three active chemical species have been measured and analyzed: soot (at 600 nm), CH\* (430 nm) and C<sub>2</sub>\* (516 nm).

## 2. EXPERIMENTAL

The experimental setup is shown in Fig. 1. It consists of a burner system, a signal generation and data collection system and an optic fiber spectrometer for the collection of chemiluminescent emissions. The burner system consists of a fuel supply system (LPG) and a cylindrical tube burner (dimensions shown in Fig. 1). Internally, the burner presents gratings and three small ducts that induce a homogeneous flow. The LPG volumetric flow rate was regulated by a control valve and measured by a calibrated flow meter (Cole-Parmer, model FM014-96ST rotameter). The experiments were conducted with a  $(2641 \pm 36)$  mL.min<sup>-1</sup> LPG volumetric flow rate, a  $(35.8 \pm .9)$  cm.s<sup>-1</sup> jet fuel speed and a  $519 \pm 8$  Reynolds number, calculated by the hydraulic diameter.

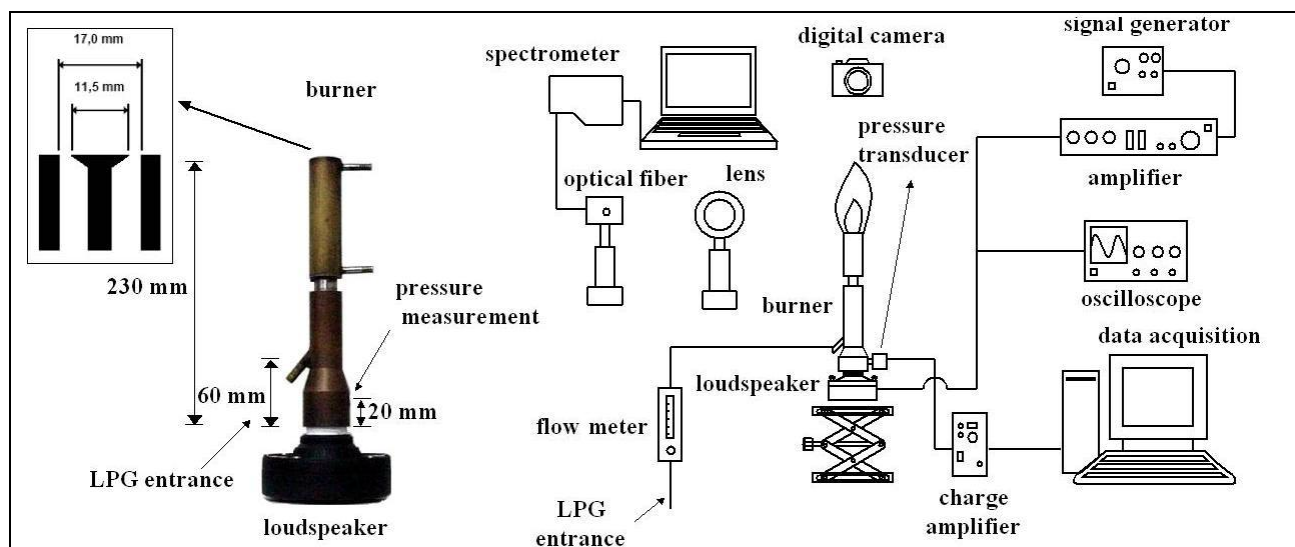


Figure 1. Experimental set-up and burner details.

The sine wave frequency employed is obtained through an Instek, model IT 100K signal generator. A Luxman, model SQ 700x amplifier controls the electrical signal amplitude. The amplified electrical signal is monitored by a Kenwood, model CS-5375 oscilloscope and sent to the loudspeaker. The acoustic oscillations propagate through the burner and the acoustics pressure is taken  $20.0 \pm 0.5$  mm above its base using a quartz transducer (Kistler, type 7261). The captured analogical signal is sent to an amplifier (Kistler, type 5006). The amplified signal is sent to a signal conditioning (National Instruments, model SCB-68) coupled to a data acquisition device (National Instruments, model PCIMIO-16E-4). The software in LabView 7.1 language controls the system and monitors simultaneously the frequency and the acoustic pressure via the fast Fourier transform (3000 samples by second). The obtained measurements are corrected on the basis of comparative calibration process performed by Flugel (2006).

The spectra are obtained with an Ocean Optics HR2000+ spectrometer (0,35 nm optical resolution). It is able to continuously transfer spectra in milliseconds time delays. A 10 mm focus quartz lens collects the light through a path centered at the middle of the burner (light spot at this position show a diameter around 3 mm), as shown in Fig. 1. A longitudinal scan along the flame is carried by a support of adjustable height attached to the burner-loudspeaker system.

In this work, experiments were performed for different conditions of frequency, amplitude of wave acoustic pressure and longitudinal position of the produced flames. The flames were analyzed from the burner exit up to  $50.0 \pm 0.5$  mm in intervals of  $10.0 \pm 0.5$  mm. During previous experiments, it was observed that the flame behavior was more sensitive to frequencies above 500 Hz. The flame was insensitive to frequencies above 700 Hz. The measured frequencies were 500 Hz, 525 Hz, 550 Hz, 575 Hz and 600 Hz. This interval was chosen because at this range it was

possible to study the influence of more acoustic pressures. A photo of each flame was obtained through a digital camera (Sony, model MVC-FD88, 1.3 mega pixels). Acoustic pressures between 490 Pa and 3000 Pa were employed.

The spectra were obtained using an integration time between 0.50 s and 2.50 s and a media of 4 measurements. The integration times were varied to avoid detector saturation in some conditions due to intense soot emission and to guarantee the best signal to noise relation for each spectrum. The spectra were subsequently normalized to a 2.00 s integration time.

### 3. RESULTS

Figure 2 shows the band attributions for the region of experimental spectra. Simulated spectra at 2000 K were obtained for CH\* radical (Luque, 2002) and C<sub>2</sub>\* radical (Gonçalves, 2009). The software GASEQ (Morley, 2005) gives an adiabatic temperature (maximum possible for the LPG flame) of about 2350 K for propane or butane burning into stoichiometric mixture with air.

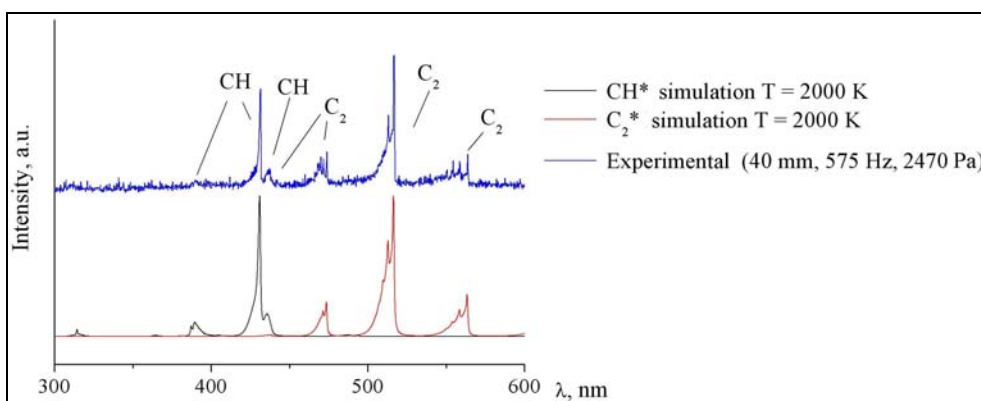


Figure 2. Band attribution (Luque, 2002; Gonçalves, 2009).

In order to obtain band intensities, the region of CH\* and C<sub>2</sub>\* radical emissions were baseline corrected (see Fig. 3, left) and the band intensity was measured at the center of the most intense band of each radical (430 nm for CH\* and 516 nm for C<sub>2</sub>\*). Soot emissions intensities were measured at the maximum of the continuous spectra (about 620 nm) as shown in Fig. 3 (right).

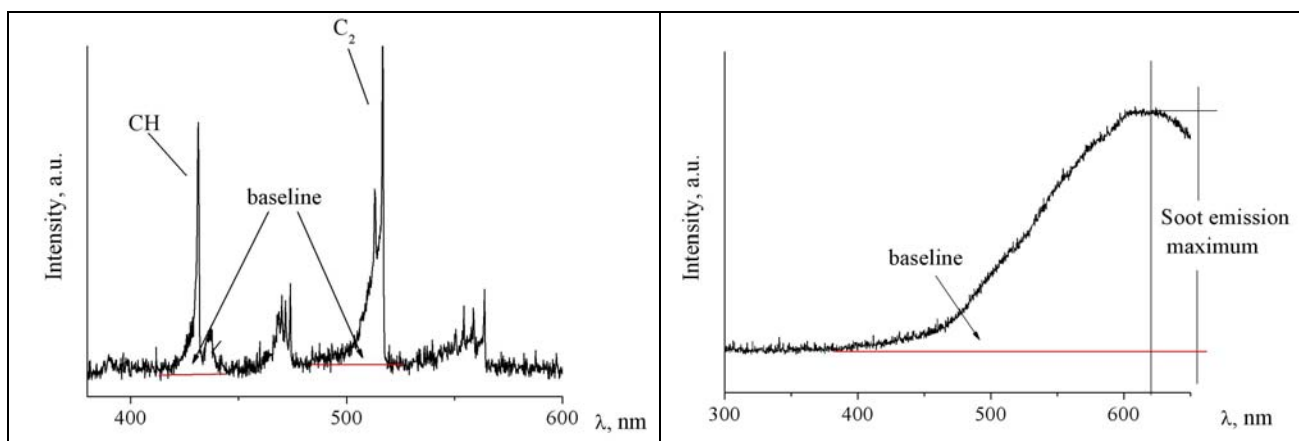


Figure 3. Baselines considered to measure (left) band height for CH\* and C<sub>2</sub>\* radicals, (right) maximum soot emission.

In the absence of an acoustic field, it was noticed that the flame was almost completely yellow and irregular, reaching an apparent height of about 30 cm (Fig. 4). Immediately above the burner, it is possible to observe the presence of CH\* (emission at 431 nm) and C<sub>2</sub>\* (emission at 516 nm) radicals; at this position, the flame was blue. From the exit to higher heights, these signals decrease and the curve of soot (continuous spectrum) increases. According to Rocha (2007), a flame in the absence of diffusive acoustic field displays O<sub>2</sub>, although in small quantity, only in the region near the exit of the burner, a phenomenon was attributed to air entrainment. Thus, the CH\* and C<sub>2</sub>\* radicals formation must be linked to the presence of O<sub>2</sub>. The yellow color is associated with the incandescence of the particles of unburned carbon (soot).

Figure 5 exemplifies the results of the acoustic action on the flame (575 Hz, various acoustic pressures). The photos show that the increase of oscillation amplitude changes the yellow color of a typically non-premixed flame to a blue color, showing the transformation in a (partially) premixed flame, by the acoustic field. The increase of oscillation amplitude reduces the flame length. Emission spectra show  $\text{CH}^*$ ,  $\text{C}_2^*$  and soot intensities variations with the acoustic pressure, and flame height.  $\text{C}_2^*$  radical emission variations at 516 nm are similar to the  $\text{CH}^*$  at 430 nm.

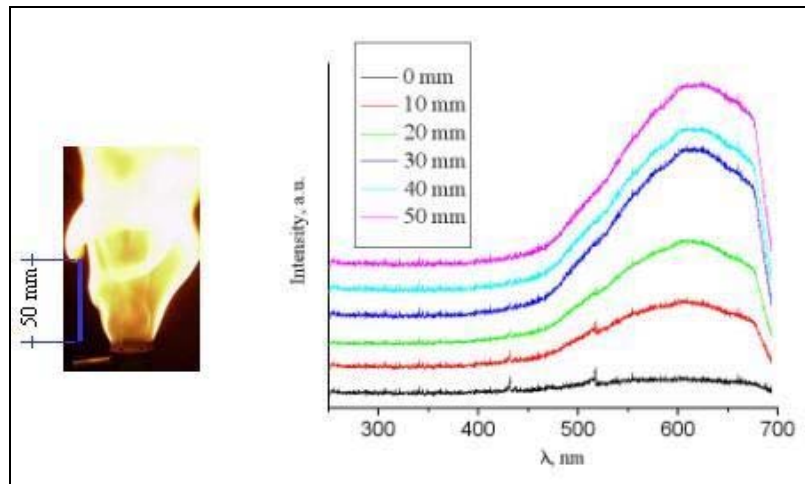


Figure 4. Emission spectra obtained without acoustic excitation at various positions above the burner exit. The minimums of the spectra are displaced to improve the visualization but the relative intensities are in real scale. Integration time: 2 s.

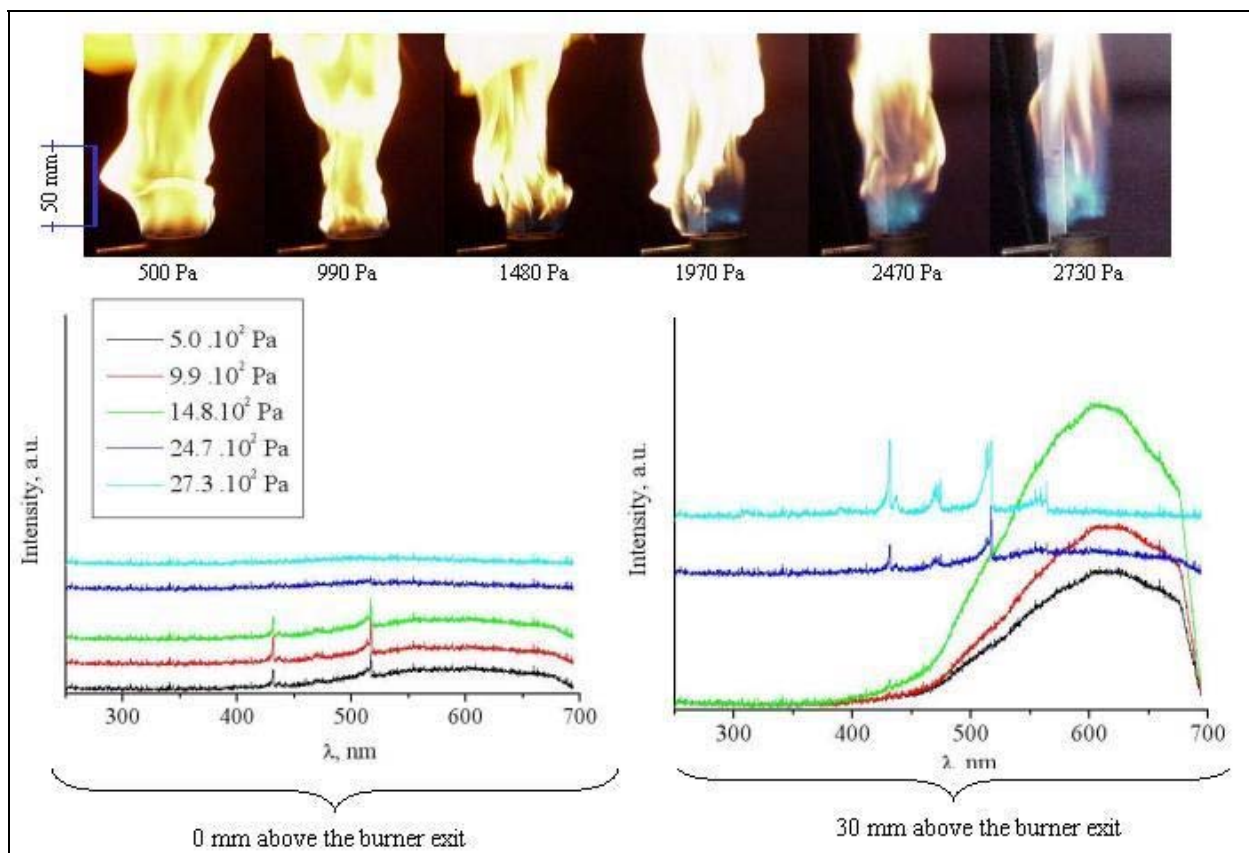


Figure 5. Emission spectra obtained for 575 Hz condition. The minimums of the spectra plotted together are displaced to improve the visualization but the relative intensities are in real scale. Integration time: 2 s.

Figure 6 through Fig. 10 show the CH\* and soot intensities variations with the acoustic pressure and flame height. The main results for each acoustic excitation condition are discussed below.

### 500 Hz (Fig. 6)

The maximum acoustic pressure achieved for this frequency with the employed loudspeaker was about 1500 Pa. The flames excited at 500 Hz showed a small increase in the blue area at their bases with the increase of sound pressure for the range considered, as shown in Fig. 6. The blue color in the flame indicates improved fuel-oxidant mixture and decreased soot at the particular flame region.

On the burner exit (0 mm height), it is possible to note only a slight increase of CH\* emission and a slight decrease of soot emission with the pressure. On the other hand, for higher heights (10 mm, 20 mm, 30 mm), there were clear increases of soot emission with the acoustic pressure.

Considering the same acoustic pressure and different heights, the results show that the greater the height, the greater the soot emission is and lower the CH\* emission is for almost all experiments.

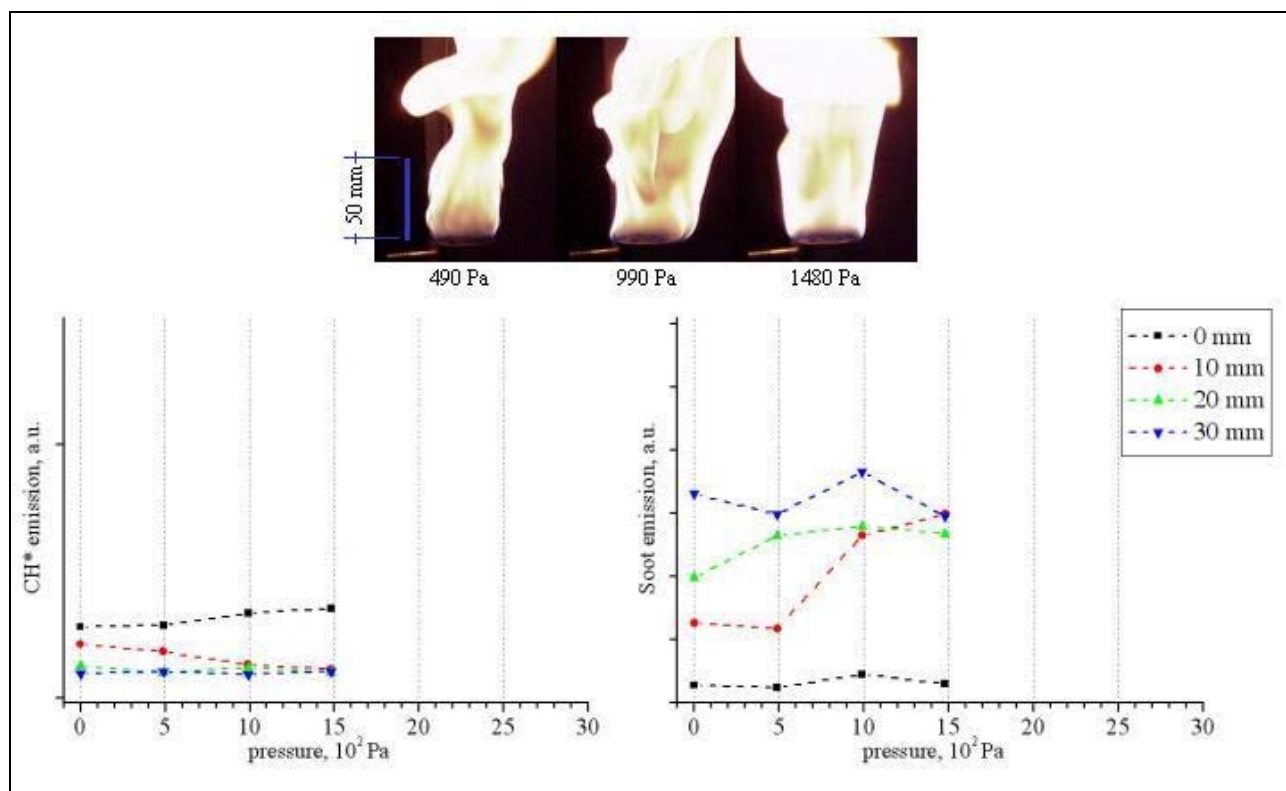


Figure 6. CH\* emission at 430 nm and soot emission at 620 nm,  $f = 500$  Hz. Each color corresponds to a particular height above the burner exit.

### 525 Hz (Fig. 7)

The general behavior is similar to that observed for the 500 Hz condition. At the bottom of the flame, there is an increase of CH\* production and a decrease in soot emission with the pressure increase. At 10 mm height it is observed an increase of soot emission up to 1480 Pa and a decrease at 2010 Pa. Soot emission increase for higher pressures (the opposite occurs with CH\* emission). At 20 mm height the soot emission increases up to 520 Pa and is almost constant for higher pressures. At 30 mm height the soot emission increases up to 1000 Pa and decreases for higher pressures. A decrease of soot emission is always accompanied by CH\* emission increases and vice versa. Considering the same acoustic pressure, the higher the height, the higher the soot emission and the smaller the CH\* emission.

More expressible instabilities on the flame bottom were first observable for the condition of 525 Hz and 2010 Pa acoustic excitation.

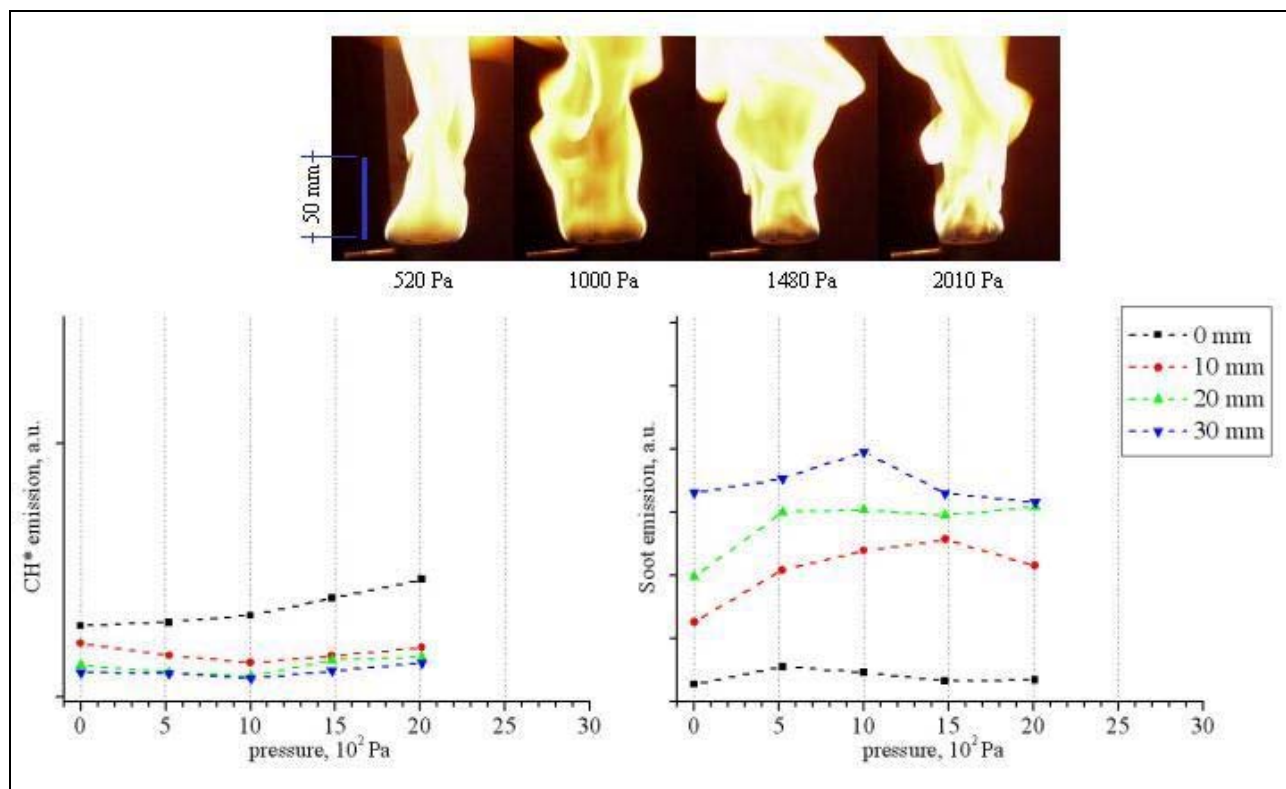


Figure 7. CH\* emission at 430 nm and soot emission at 620 nm,  $f = 525$  Hz. Each color corresponds to a particular height above the burner exit.

### 550 Hz (Fig. 8)

In this condition frequency, the maximum sound pressure achieved is 2420 Pa. Small disturbances in the base of the flames appeared for 1970 Pa acoustic pressure, in addition to the detachment of the flame (*lift-off*) on 2420 Pa.

CH\* radicals emission is only observed in considerable intensity at the flame bottom and at 10 mm height (2420 Pa). It is important to observe that the decrease of CH\* emission at the bottom for 2420 Pa occurs because of the flame detachment (there are no species at this position). Above 990 Pa the soot increases with the acoustic pressure for 30 mm, 40 mm and 50 mm height. At 10 mm, the soot emission increases until 1470 Pa and then decreases. At 20 mm height the soot intensity increase up to 490 Pa, decrease to 1480 Pa, increase up to 1970 Pa and, finally, decrease.

### 575 Hz (Fig. 9)

At 575 Hz, it is observed the most intense effects of acoustic excitations in the LPG flame. Pressures up to 2730 Pa were studied. Above 990 Pa there is the flame detachment and the CH\* emission at the bottom decreases (there are few species at this region). The flame detachment is also responsible for the CH\* radical decrease at 100 mm and 2920 Pa acoustic pressure. CH\* emission increases (soot emission decreases) considerably with the acoustic pressure increase at 20 mm, 30 mm and 40 mm height.

Above 2730 Pa, it is observed the flame blowout. Maximum CH\* radical emission is observed at 40 mm height, 2730 Pa acoustic pressure.

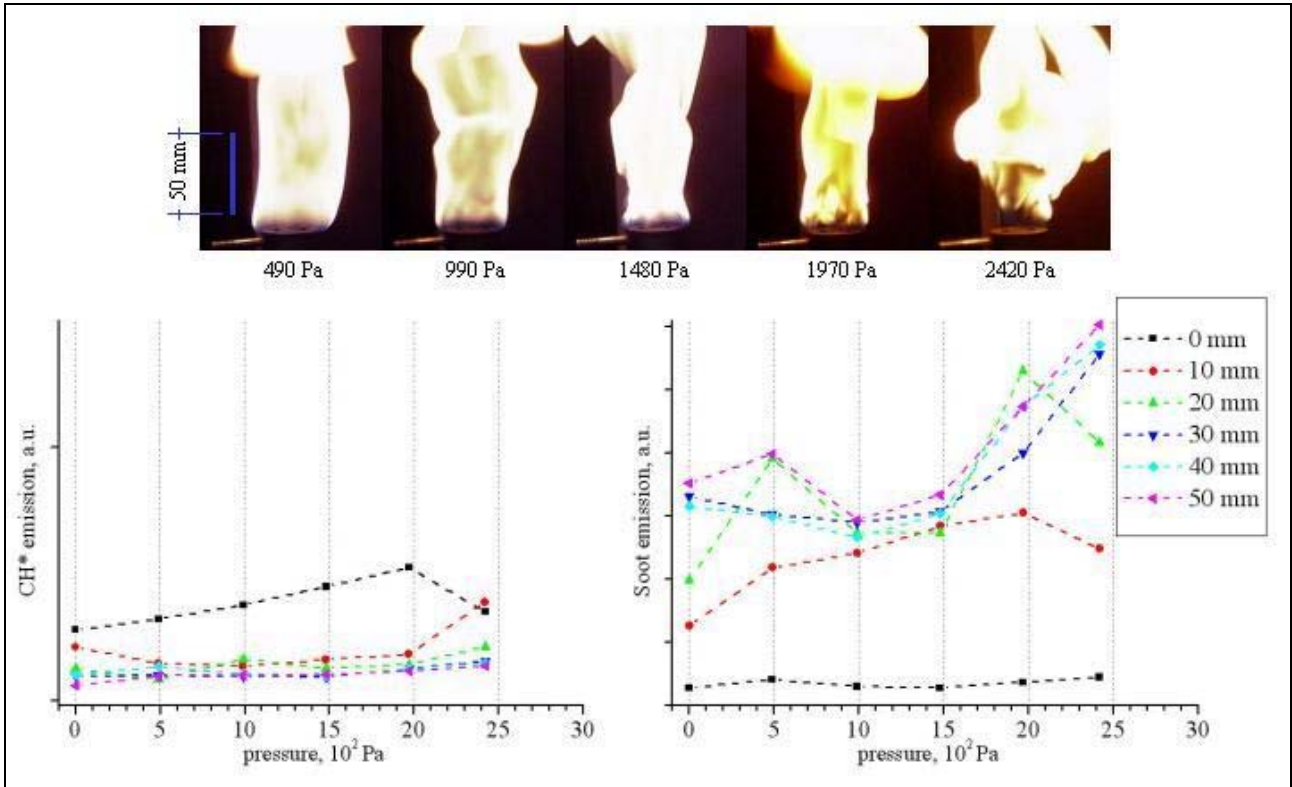


Figure 8. CH\* emission at 430 nm and soot emission at 620 nm,  $f = 550$  Hz. Each color corresponds to a particular height above the burner exit.

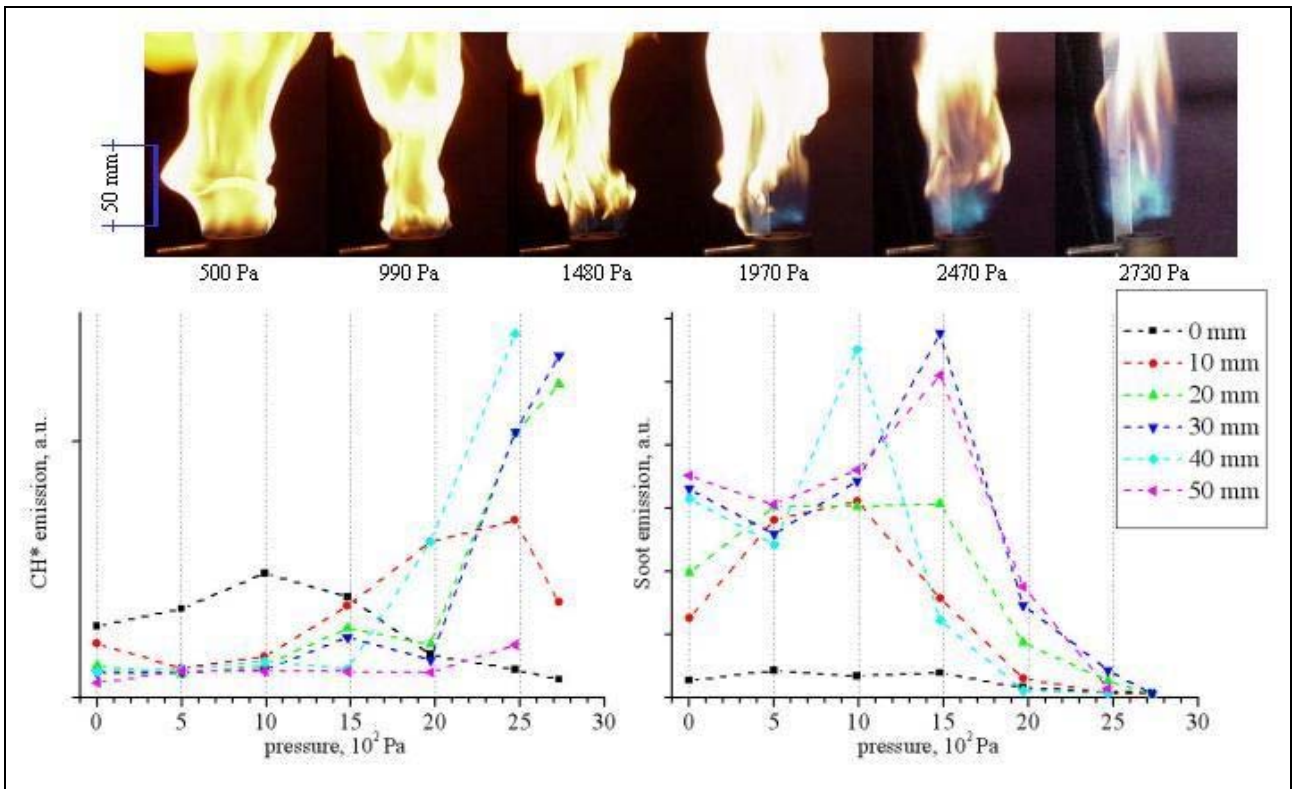


Figure 9. CH\* emission at 430 nm and soot emission at 620 nm,  $f = 575$  Hz. Each color corresponds to a particular height above the burner exit.

### 600 Hz (Fig. 10)

For conditions of 600 Hz, the flame detachment begins at 1970 Pa. Above 2170 Pa the flame blowout. Maximum CH\* radical emission is observed at 20 mm height, 2170 Pa acoustic pressure and maximum soot emission, at 40 mm height, 2170 Pa.

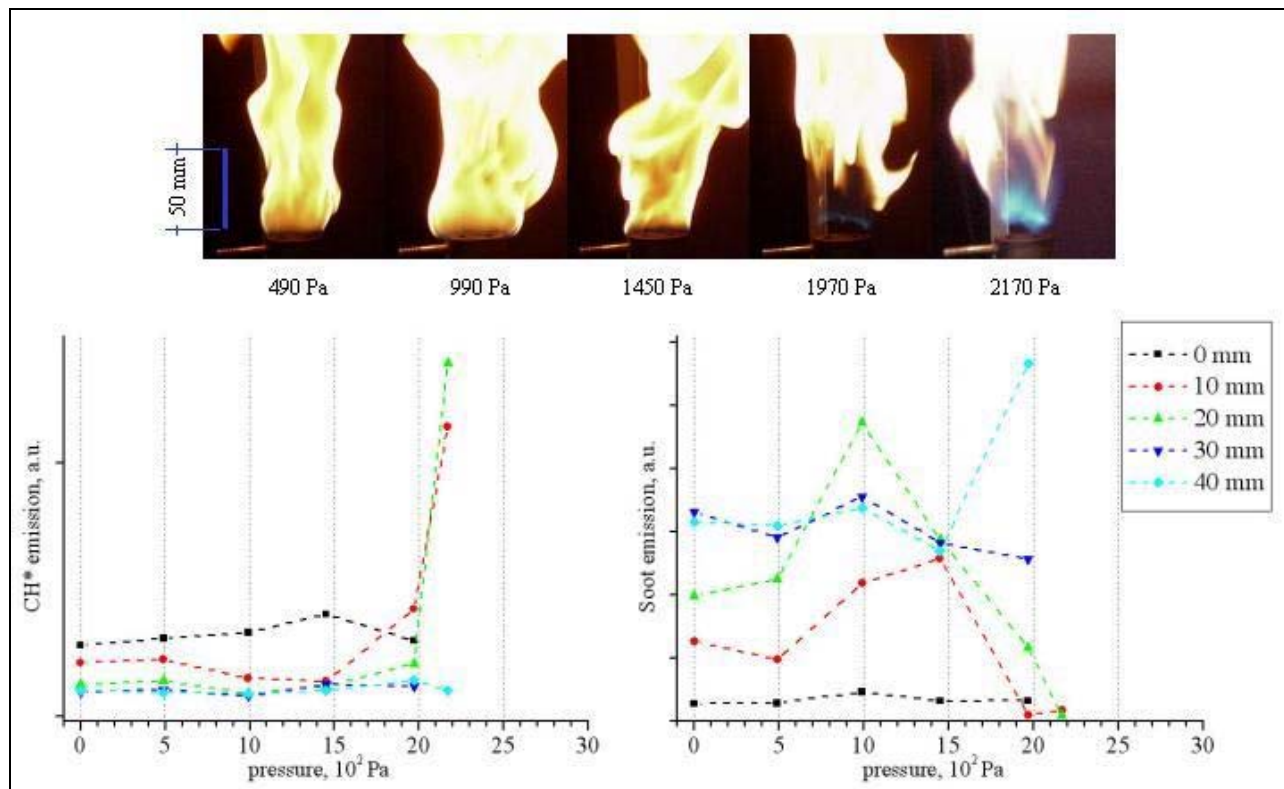


Figure 10. CH\* emission at 430 nm and soot emission at 620 nm,  $f = 600$  Hz. Each color corresponds to a particular height above the burner exit.

## 4. DISCUSSION

The acoustic excitation can change the flame structure drastically and, consequently, the emission spectra, as shown in Fig. 5. For some oscillating frequencies, the increase of oscillation amplitude changes the yellow color of a typically non-premixed flame to a blue color, showing the transformation in a (partially) premixed flame by the acoustic field. The oscillation amplitude increase reduces the flame length. These effects can be observed mainly at 575 Hz and 600 Hz acoustic excitations frequencies (see Fig. 6 to Fig. 10). Comparing the CH\* and soot intensities variations it is possible to note that sharp decreases of soot emission with the acoustic excitation is frequently accompanied by CH\* emission increases.

Martin (2002) noticed that the formation of soot was almost extinguished in the highest noise intensities imposed on a laminar acetylene flame. Oliveira (2007) reported that the effect of sound field in the reduction of soot was more pronounced in the resonant frequency. These results suggest that the presence of acoustic oscillations is able to pre-mix the fuel and the air. According to Rocha (2007), when sound field is present the diffusive flames has a high concentration of O<sub>2</sub>, especially in more remote regions of the base of the burner, reinforcing the idea that the acoustic field increases the air entrainment.

The most effective acoustic influences were observed for 575 Hz and 600 Hz wave frequencies. According to Rocha (2007), the transformations that occur in a diffusive flame subjected to an acoustic field are related to the phenomenon of resonance between the wave sound frequency and burner natural frequency. The author noticed that largest amplitudes of oscillation were obtained in the natural frequency of the burner. It is, therefore, possible that the frequency around 575 Hz – 600 Hz is the natural frequency of the burner used in this work.

A local soot increase observed at various flame heights for some acoustic conditions can be understood considering that the soot formation depends on the local temperature and on a lack of oxygen to complete de combustion process. The acoustic excitation near a burner natural resonance promotes air entrainment. In some conditions this air entrainment causes an increase of combustion reactions (increasing the local temperature) but the O<sub>2</sub> is not enough to



complete the process favoring the soot formation. When the acoustic excitations become more effective the flame turns into a partially premixed one (high  $\text{CH}^*$  and  $\text{C}_2^*$  concentrations).

## 5. CONCLUSION

The presence of an acoustic actuation can drastically change the structure of the flame produced in the cylindrical burner. The following observations were made: a) the increase of oscillation amplitude reduces the flame length; b) the pulsating flame changes the combustion regime and soot formation.

The biggest signals of the radicals  $\text{CH}^*$  and  $\text{C}_2^*$  emission are associated with the blue zone of the flame. Some acoustic frequencies (575 Hz and 600 Hz) had demonstrated a more efficient action. The biggest sensitivity of the flame to the determined frequency must be associated to the phenomenon of resonance between the wave frequency and the natural frequency of the burner.

A number of extensions of this work could be made. PIV measurements at a number of phase steps would allow an improved visualization of the acoustic action on the flame at the studied burner. It is interesting to obtain the temperature profiles along the flame in the presence of soot, oxygen, gaseous pollutants such as CO, SO<sub>x</sub> and NO<sub>x</sub>, and distribution of. Thus, the investigation of the influence of the acoustic field in the type of burner used in this work would be more detailed.

## 6. ACKNOWLEDGEMENTS

This research has been financially supported by CNPq (Conselho Nacional de Desenvolvimento Científico e Tecnológico).

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