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EXPERIMENTAL ANALYSIS OF FLAMELESS COMBUSTION FOR REDUCTION OF POLLUTANTS

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ABSTRACT

This report presents a special form of combustion, called flameless combustion. In contrast to the combustion within stabilized flames, temperature peaks can be avoided at flameless combustion. Critical points of this technology are the realization of thermofluodynamic conditions which stabilize the flameless combustion process and a design to reach flameless conditions. Burners for flameless combustion usually show complex geometries which make simplified schemes difficult and inappropriate. In this case, the present paper proposes a study to determine the viability of a flameless burner design for experimental investigation and a combustion chamber on a laboratorial scale able to use high temperature air to reduce pollutant emissions. During the experiments, it will be used natural gas and preheat air. The influence of the field flow, chamber temperature and preheating air temperature on reaction zone volume was examined using techniques capable of analyzing the temperature field in the combustor. The main conclusions were: a) in contrast to the flames, the combustion had low internal temperature gradients; b) the system has promoted changes in the volume characteristics of combustion significantly reducing pollutants: NO_x, CO and UHC (Unburned Hydrocarbons); c) to same conditions no visible flame was observed during the test.

INTRODUCTION

The development of efficient and low polluting combustion systems is the combustion researchers' and combustion equipment manufacturers' main goal. Due to scarcity of resources which adds environmental problems arising from the utilization of energy and extraction of natural products, the concept of recovering thermal energy from combustion has expanded, and solutions have been sought through actions into energy efficiency (better usage of thermal energy with low emissions), greater insertion of biomass fuels, hydrogen usage, concerns with other gases, and greenhouse gases, such as methane and NO_x.

Recently, the control of these emissions, mainly carbon monoxide (CO) and nitrogen oxides (NO_x), has also become the most important target in the design of combustion chambers and industrial burners. Specifically for NO_x case, most control strategies in combustion processes are based on three variables: residence time, temperature and oxygen availability. These strategies are focused on reducing the peak temperature, keeping the residence time, and low oxygen concentration at high temperature zones [1,2].

Despite these existing strategies, the growing global energy demand stimulates the development of combustion systems which are more efficient and pollute less than the old ones.

In this scenario, a technology that has gained interest in the current research as a promising technique for combustion control and minimization of NO_x emissions and other pollutants is the flameless combustion (smoldering visible). This technique is a new type of combustion that changes the characteristics of the reaction zone and consequently the formation of combustion products by process modification.

By now, several groups have investigated flameless combustion under various names, such as MILD Combustion by Cavaliere [2], FLOX by Wünning [3], HiTAC by Gupta [4]. The combustion flameless also consists in a combustion system that promotes the pollutant emission reduction and improves the system thermal efficiency. The main applications of this technique are in the steel and metal industries. Furthermore, other researches and applications have been being performed such as in ceramics and glass industry, chemical industry, power generation among others [3].

Despite being a technique that requires better understanding of its physicochemical operation, currently flameless combustion has been gaining attention among scientists due to the great advantages it offers in comparison to the traditional combustion techniques [1,3]. The main advantages offered by the flameless combustion are: significant reductions in the pollutants formation, especially NO_x and CO, its great thermal efficiency, fuel oxidation and its distribution throughout the volume of the chamber, considerable reductions in the temperature gradients and the species concentration [5,6]. Moreover, it can be applied using a big variety of fuels [3,7]. Recently, Flameless Combustion has been studied by a new method based on Principal Component Analysis (PCA). In this case, experiments are conducted with the mixture of fuels such as CH4/H2 [8]. Due to its operational flexibility, this has been studied in gas turbine combustors [9].

Differently from what occurs in the conventional flames, an important flameless combustion characteristic is that the oxidant does not mix directly with the fuel before the reaction. That is, first the oxidant is mixed to the exhaust gas recirculation into the chamber. After that, the mixing with the injected fuel occurs, in this case, in the center of the chamber, resulting in the zone of distributed reaction. It allows a low temperature flame and partial low pressure of the oxygen inside the reaction zone resulting in a low NO_x formation [10-12].

Regarding NO_x, it is known that its formation is primarily a function of the temperature, oxygen concentration and residence time, and in conventional systems the residence time is usually sufficient to allow the generation of NOx high concentrations. In experimental studies, Medwell [13], investigating the effects of the temperature peak reduction to decrease the NO_x, showed that the temperature profile induced by flameless combustion was relatively flat, thus, the emission of NO_x (influenced by the local temperature of the flame) was significantly reduced. Moreover, by reducing the flame temperature peak, the average temperature level of the zone of the furnace can be increased without leading to a local overheating in the vicinity of the burner.

In NO_x formation, it is known that the thermal Zeldovich mechanism is usually the main factor for its emission in systems with temperatures above 1500 °C. According to Wünning [3] in some cases, the temperature and residence time can have strong influence on the flameless regime; however they do not control the NO_x emissions. Therefore, it is not possible to isolate one or two control parameters because many of them have a comparable magnitude influence. In this case, for the temperature to lower the Fenimore mechanism, NO prompt and / or intermediate N_2O may be important in this formation. The importance of NO prompt in the flameless regime has been little considered [13,14]. In studies conducted by Mancini et al., [13] only 5% of the global NOx emission was considered formed by the NO prompt mechanism. On the other hand, it was observed that the intermediate N_2O mechanism represented 65% of NO_x in the temperature range of 1140-1570 °C.

The geometry of the burner and combustion chamber and the high injection speed are key factors for the internal recirculation of the combustion products in the chamber [5]. The high temperature of combustion products in recirculation (> 800 °C, depending on the fuel and system configuration) is used to initiate and maintain the operation system stability. This temperature must be higher than the auto ignition temperature of the mixture of air and fuel [3,12].

Based in this context, the current work aims to present experimental results about the thermal field and pollutant emissions for a given burner configuration and a chamber, operating in the flameless regime. The results were obtained with temperature measurements inside the chamber and the analysis of the overall gas emission. First, the design and construction of the chamber and the burner are described; subsequently the effects of equivalence ratio in the field of temperature and pollutant emissions are discussed. To direct the output of an exhaust gas, a volute of characteristic size was positioned coupled to the burner. Its frontal side had a circular opening which directed the output of exhaustion gases in the counterflow of the air and fuel injection. In this case, the gases returned from the same input volute face and burner. A chimney, with a rectangular section of 280 mm x 20 mm, was placed in order to recover the gases to the atmosphere

DESIGN AND CONSTRUCTION OF THE EXPERIMENTAL SETUP

Next, it is presented the burner configuration and a laboratory-scale chamber designed and built to operate in the flameless regime. These systems are quite complex because several factors influence the mixture and standard behavior of this combustion regime [2,3,13-15] Generally, its performance can be evaluated in the temperature profile uniformity, possibility of pollutant reduction and operational flexibility [6,16,17].

In this system the front face of the burner consists of a central fuel jet, 4 air jets arranged symmetrically and 56 mm equidistant ones from the center. Each air jet leans about 3 degrees in relation to their central axis. In the air and fuel jets, there is an extension of the characteristic length. The aim of prolonging the jet fuel was to promote the gas release in regions farther from the face of the burner in order to distribute the mixture of the reactants through the chamber volume. The gas capture probe was positioned 300 mm above the center axis of the volute. Figure 1 shows the measures and details of the burner.

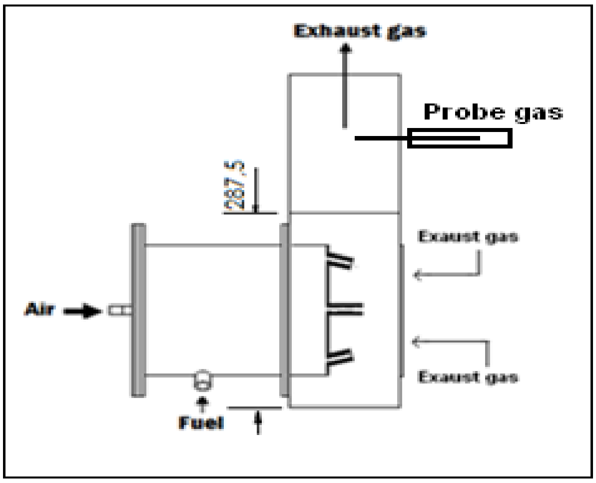

Figure 1. Characteristics and measures (mm) of the burner

Regarding the combustion chamber, the main project parameters were: a) a chamber model and size; b) optical access position; c) thermal insulating; d) thermocouple position. The chamber was built with carbon steel plates in a rectangular shape and projected to be settled horizontally. The chamber internal dimensions are 286 mm x 286 mm of cross section and 540 mm in length.

Figure 2 shows the assembly of the final configuration of the burner and the combustion chamber in the laboratory bench scale for flameless combustion.

The combustion chamber has nine windows for optical access located on three sides along the cross section. Each window has an area of 100 mm x 50 mm. The screens were placed 200 mm, 350 mm, 448 mm from the front of the chamber.

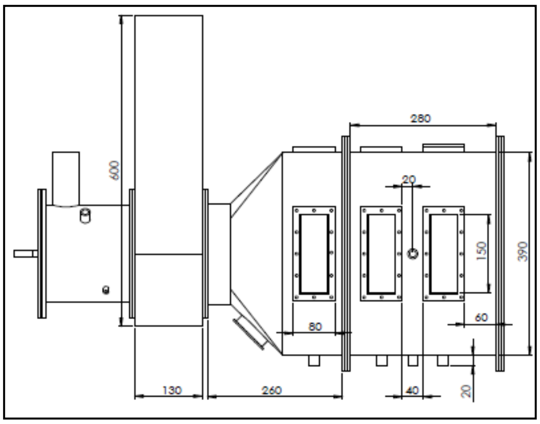

Figure 2 Experimental system for flameless combustion

At the bottom of the chamber, ten thermocouples were installed, nine to measure the profile of the temperature field and one to measure the temperature of the refractory wall. Aiming to minimize heat loss, the chamber walls were coated with a refractory block 80 mm thick.

OPERATION CONDITIONS

The fuel measurement was made through orifice plates "radius tap" type with 5% of maximum uncertainty. For air measurement a mass flow meter of thermal dispersion with 0.02 % full scale maximum uncertainty was used. The gas analyzer was a GreenLine 8000 ECIL with the following accuracy: CO (± 5%: 0-2000 ppm, ± 10%: 2001-10000 ppm) CO_2 (± 0.3% <10% vol. 3%> 10% vol.), NO (± 5ppm: 0-100 ppm), NO_2 (± 5 ppm: 0-100 ppm), O_2 (±0.1%: 025%) and UHC (<100 ppm < 2500 ppm CH_4). For data acquisition, the commercial software LabVIEW ®9.0 was used.

A DBGas2004 program was used to store and manage the collection of the analyzer measures. Monitoring of spatial and temporal variations of temperature was performed using nine thermocouples type K (Chrome / Alumel), diameter of 3.1 mm, with temperature range from 0 to 1300 ºC and an accuracy of +/-1 ºC. Both are positioned at the bottom of the chamber. The ends of the thermocouples were located at half height of the chamber.

In order to make easier the discussion of the results, Fig. 3 shows the arrangement of thermocouples at the bottom of the chamber and displays the chamber volume divided into three different regions in accordance with the position of the thermocouples, and region I: T_1, T_2 and T_3; region II: T_4, T_5 and T_6 and region III: T_7, T_8 and T_9. The thermocouple T_{10} was used to measure the temperature on the chamber wall.

The temperature measurement and gas concentration were acquired simultaneously, and the acquisition of a certain operation condition was performed when the thermal equilibrium was reached. At the acquisition moment the temporal average range of temperature for each thermocouple was close to 1.5 °C/min. The pressure inside the chamber corresponded to the ambient pressure.

The burner operated with fixed gas natural mass flow rate of 0.16 g/s (85% CH_4 and 15% C_3H_8) and with air mass flow rate adjusted according to the equivalence ratio. For the experiment, t was used air flow from 2.5 to 3.5 g/s. The tests were made under various conditions with equivalence ratio between 0.7 and 1.1. The global equivalence ratio is equivalent to the relation of the fuel flow and the oxidant flow used.

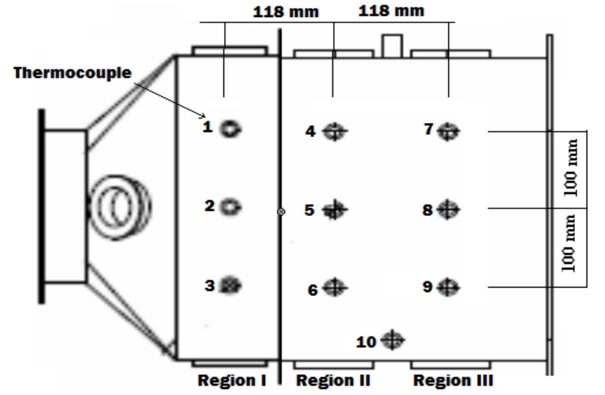

Figure 3 Thermoplate position in the chamber lower face

The operating conditions of reference were: global equivalence ratio of 0.8 and preheated air at 250 °C. In the fuel injection, a nozzle with 4.5 mm of diameter was used whereas for air injection, four nozzles with 4.5 mm of diameter were used. The power of the burner was about 8 kW. The oxidant was heated by a heater in which natural gas was used as a source of thermal energy to heat the air.

In order to start the chamber operation and heating process, a pilot flame was primarily used, and when the chamber temperature was above 850 °C, main fuel input was triggered (flameless). After that, the combustion was kept until the chamber thermal stabilization so that further adjustments were made and data acquisition was initiated. In general, the thermal stabilization occurred after almost 8 hours of operation. The chamber control and safety system were carried out by the data acquisition program. For each working condition the program acquired 40 samples, being each sample a representative average of 1000 measurements. Furthermore, temperature measurement herein represents an average of 30 measurements. Figure 3 shows the sequential steps of operation until the time of system flameless data acquisition.

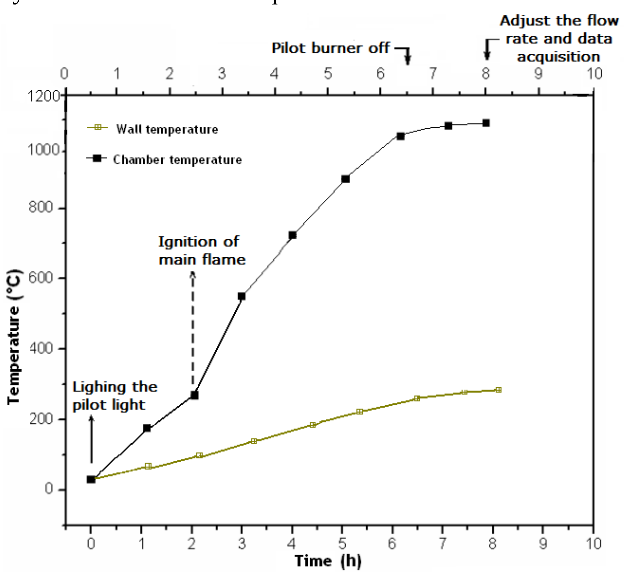

Figure 4. Stages of system operation

RESULTS

Figure 5 shows the temperature profile as a function of the heating time. It also presents the profile of transition from the conventional to flameless regime when the system is changed to the condition of Ø = 1.1.

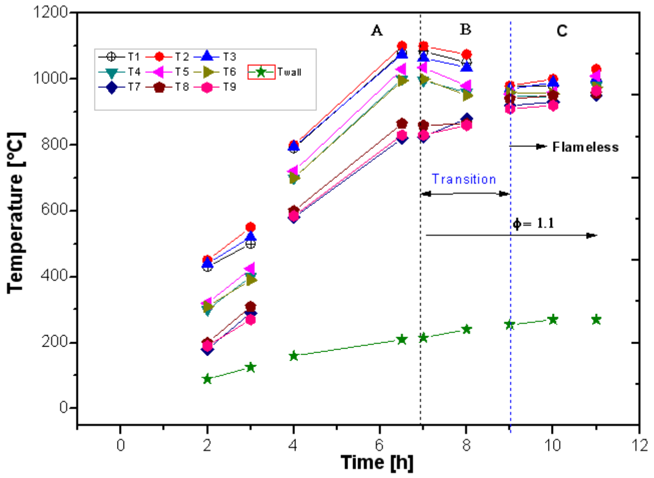

Figure 5 Temperature profiles as a function heating time

Figure 5 shows three situations: region **A** shows the profile of the heating chamber to stabilize the temperature, region **B** indicates the temperature variation after the system starts functioning with Ø = 1.1. In this region it is observed that at a given moment there is a qualitative and quantitative change in the temperature profile and, as it will be shown later, this behavior suggests that this moment is the one when transition occurs for the flameless regime. Region **C** already shows the new temperature profile for Ø = 1.1 with lower spatial difference in temperature. In this new condition, the thermal stability occurred after 1.5 hours of operation. The maximum temperature measured in the chamber during the heating was close to 1040 °C and the wall temperature was 265 °C.

In Figure 5, it is also observed that after one hour, the beginning of temperature measurements located near the inlet (region I (see Fig. 3): T_1, T_2 and T_3) are the highest ones, followed by temperatures located in the chamber midline (region II, T_4, T_5, T_6) and of those located at the end of the chamber (region III, T_7, T_8 and T_9). That is, during the heating range, the temperature gradient among the temperature of the regions is considered. However, the temperature measurement of each individual region has similar values. It is important to notice that these temperatures located longitudinally along the chamber (T_2, T_5 and T_8) have always showed slightly higher values. During heating (i.e. from the start of heating until the end of region A) the maximum temperature was 1100 °C and the wall temperature was 270 °C. Additional reviews of this behavior will be made below.

Temperature field

The system operated until the temperature stabilized, and then data acquisition was made. Figure 6 shows the temperature curves as a function of the equivalence ratios. Here, the objective of these tests was to analyze each operating condition, besides identifying and commenting the changes and situations which characterizes the combustion in the flameless regime.

As can be noticed, the steps of equivalence ratios of 0.7 and 0.8 are qualitatively similar, being, however, the measures for equivalence ratio of 0.8 slightly higher. Ratios in the range of 0.7 and 0.8 equivalent to the temperature increase may only be due to the lower presence of inerts because of the reduced air mass flow rate.

In systems using methane as fuel, the flameless system occurs with the overall chamber temperature above 800 °C, which associated with other factors such as the burner geometry of the input speed of the reactants, promotes a relatively higher homogeneity degree between the temperatures and a considered reduction in the emissions of NOx and CO [3,12].

In conditions with low equivalence ratios 0.7 and 0.8, the average measurement of the minimum and maximum temperatures were 640° and 880°, and 790 °C and 1020 °C, respectively. These values, when compared with other results of the equivalence ratio show that there is a relatively high gradient in the temperature field. This may be, in these conditions, because of the mixture reaction and concentration near the injection point favoring a high temperature upstream of the burner (see Fig. 3). This situation can be enhanced due to the fact that temperatures located upstream (T_1 to T_3) respectively, present a greater value than the temperatures measured in the center (T_4 to T_6) and at the end of the chamber (T_7 to T_9).

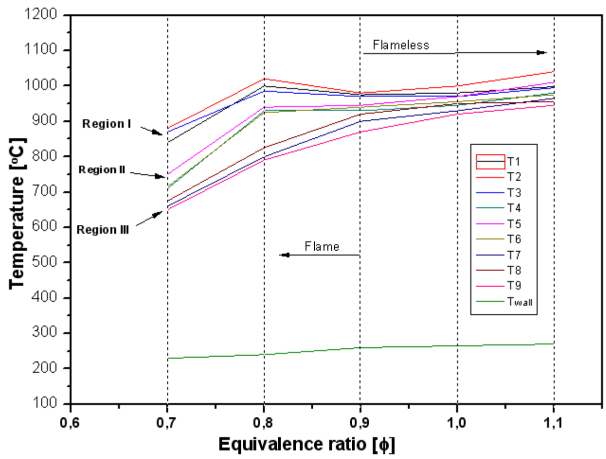

Figure 6 Temperature curves as a function of the equivalence ratio

For lean equivalence ratios, specifically between 0.7 and 0.8, there is a significant difference when comparing the temperatures between regions (region I: T_1, T_2 and T_3, region II: T_4, T_5 and T_6 and region III: T_7, T_8 and T_9), it is important to notice that the temperature measurement for each individual region is approximated, being the temperature measured along the center of each region (T_2, T_5 and T_8) always higher. It occurs because in these conditions there is the presence of stable and visible flames. These thermocouples are located along the centerline of the chamber and are directly influenced by the flame.

In the present study, it was not possible to detect which relevant factors promoted this transition and maintained the stability of the flameless regime in equivalence ratios between 0.9 and 1.1, however, due to the complex interaction of physical-chemical phenomena in the transition from the conventional to flameless regime, this change can be linked to several factors, such as the burner and the camera setting conditions, the use of preheated air, combination of speed, air and fuel flows, and also to the increase in the overall temperature of reaction as well as the wall temperature [5,6,18].

In this paper the use of preheated air (250 °C) compared with other experiments [3-5,18] was very low, but it was useful for the flameless regime for Ø = 0.9 to 1.1. That is, when air was used at ambient conditions or below 250 °C combustion was characterized as conventional. In general, although the preheated air is not necessarily a requirement for the operation of the flameless regime, this system has been important to accelerate and increase the overall temperature within the combustion chamber promoting the reaction support [19,20]. Also, according to Masson [21], the pre-heating air leads to the air jet momentum increase and consequently might increase the conduction of recirculation gases.

In relation to the wall temperature, it is observed that for equivalence ratios of 0.9, 1.0 and 1.1 the temperature was maintained between 260 °C and 265 °C, whereas for equivalence reasons of 0.7 to 0.8 the temperature decreased to a range of 230 °C and 240 °C. As flameless conditions occurred where higher wall temperatures were taken, this means that for this geometry, it was only possible to reach this regime for wall temperatures above 260 °C.

In order to reinforce this statement, it was noticed that during the transition range of Ø = 0.8 and 0.9, some important phenomena were established: a) a pulsating flame was considered, b) the flameless regime was not maintained, because there was always the presence of a flame front, c) as the wall temperature reached 260 °C the flame front was alternatively detached and fragmented until the moment situation in which the flameless regime was characterized and settled.

This behavior suggests that this system which has a minimal wall temperature (260 °C) associated to factors such as reactants temperature and reactant flow and / or the burner geometry may have a significant influence on the thermal stability and the necessary conditions to initiate and maintain the flameless system. Masson and Taupin [21] reported that the reduction of wall temperature by increasing the excess of air changed the combustion dynamics system reducing the ability of flameless operating conditions. In this case, the equivalence ratios between 0.7 and 0.8 is likely to be the one which has occurred, i.e. low temperature wall alter the dynamics of the combustion discouraging the functioning of flameless. In this case, the reduction of the wall temperature can be linked to the increase of the air flow under these conditions, due to its higher volume just by interacting with the walls leading to their cooling.

In the flameless regime, an important characteristic is the difference between maximum and minimum temperature. This difference may indicate the level of homogeneity for each working **condition [3].** In works performed by Szegö [17,19] in the flameless combustion chambers and using preheated air to increase the chamber until 1275 °C, the maximum difference between the temperatures in spatial distribution was 150 °C. Accordingly, the author concluded that low temperature difference in the space field was a result of dilution of the flame, providing a high degree of homogeneity of temperature in the chamber.

In the present research, for equivalent ratios between Ø = 0.7 and 0.8 the maximum and minimum temperature differences were respectively 240 °C and 230 °C; however, for Ø = 0.9, 1.0 1.1 the differences were 110 °C, 80 °C and 95 ºC. Thus, the degree of uniformity was very superior in flameless conditions. Therefore, it enhances the idea that the high degree of uniformity is an important feature of this regime.

Combustion gases analyses

In works performed by Szegö [17] and Dally [18] the relative decrease in the values of NO_x and CO concentration indicates the combustion stability without visible flames.

In this study, Fig. 7 shows the emissions of combustion gases as a function of the equivalence ratio. In general, under standard conditions (Ø = 0.7 and 0.8) the measured values of NO_x and CO were high, but when the flameless regime was reached, there were significant changes. As expected the emissions of unburned hydrocarbons showed the same tendency of the CO emission.

For equivalence ratio of 0.7 the concentration of NO_x, CO and C_xH_y was approximately: 55 ppm, 2550 ppm and 440 ppm. For Ø = 0.8 there was an increase in the concentration of NO_x while the CO decreased.

For the condition ø = 0.9 there is a dramatic drop in gases emissions. For this condition, the emission levels of NOx and CO were 16 ppm and 195 ppm respectively. That is, 70% of NO_x and 92% of CO were reduced respectively. It was also observed that the decrease in oxygen concentration reached values close to zero.

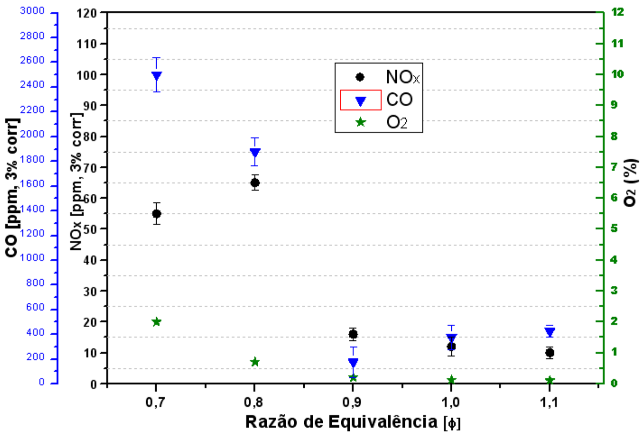

Figure 7 Effect of equivalence ratio of CO, NO_x and O_2 emissions

Under this condition, the reduction of CO can be related to improvement in the increase in the mixing ratio between the reactants. And, moreover, as the air temperature is higher, there is an acceleration of the reactions enabling a faster development in the reaction mechanism reducing the CO emission [20].

For equivalence ratios between 1.0 and 1.1, it can be can be seen a slight trend in the reduction of NO_x emission accompanied by the increase of CO emissions. However, such concentration levels are maintained within a considerably lower range.

Accordingly, the trend of a CO increase may be associated with the reduction of oxygen available to oxidize it. In addition, factors such as residence time and an increase in the recirculation rate of gas in the combustor may also affect its formation.

Regarding the formation of NO_x, in this work the good performance in reducing NO_x may be linked to a lower flame temperature peak and reduced oxygen concentration. As already shown, in flameless conditions the maximum temperature measured by thermocouples in this chamber was 1040 ºC and its distribution was considerably uniform. In this case, the results in Fig. 7 suggest that the NO_x formation is not bounded to this thermal mechanism. In this case, the Fenimore mechanism (NO prompt) and / or intermediate N_2O may be important in this formation. In this study, it was not possible to identify the main mechanism forming NO_x, because in this case, it would be necessary to quantify the contribution of each mechanism. However, it is possible that the formation of NO_x has been changed by various parameters, such as the influence of preheated air, oxygen concentration, dilution and also the kind of fuel. According to Szegö [19] these parameters, besides not being independent on each other, the combined effect of characteristics associated with non-adiabatic temperature and residence time can influence the formation of NO_x in a given system.

According to Wünning [3] in some cases the temperature and residence time can have strong influence on the flameless regime; however they do not control the NO_x emissions. Thus, it is not possible to isolate one or two control parameters, because many parameters have a comparable magnitude influence.

The influence of the air and fuel injection momentum is also very important. The change in the trajectory of the air causes changes with features similar to the model strong-jet/weak-jet of Grandmaison et al., [15]. In the present work it is possible that the momentum in the jet of reactants have favored the mixture of air and fuel with hot combustion products prior to the reaction. However, studies conducted by Szegö [17,19] demonstrated that although the mixture has some influence, it does not control the overall NOx emission because of other factors, such as those due to chemical kinematics, also influence the formation of NO_x. It assumes that the NO_x formation can occur in a condition in which the ratio between the mixing and reaction times are in a close range, resulting in a Damköhler number ($Da = \tau_{mix} / \tau_{chem}$) equal to one unit. However, more accurate studies need to be evaluated to assess more conclusive results for the flameless regime.

CONCLUSIONS

This study aimed to investigate and analyze lab-scale parameters that control the mechanisms of combustion "flameless". Therefore, it was designed and built a chamber and a burner that meets the performance characteristics of this combustion regime. In some operating conditions it was possible to observe the functioning of the flameless combustion. However, one needs deep understanding of the factors (physical and chemical phenomena) that influence the mix and the default behavior of this regime of combustion in a laboratory scale, for more conclusive results. Moreover, it was concluded that:

1. The occurrence of the system occurred with flameless combustion chamber temperatures above 800 ° C, i.e., in the range 870-1040 °C.
2. In flameless conditions considered, the biggest difference between the temperature was 110 ° C, whereas for the other conditions the smallest difference between the temperature was 190 ° C. This indicates a reduction in the thermal field gradient and a general feature of the flameless system;
3. Among the reasons of equity was 0.8 and 0.9 the range of transition from conventional to flameless regime. This assertion can be reinforced in the characteristics of the thermal field and the emission of pollutants in these conditions;
4. For reasons between 0.7 and 0.8 equivalent amount of NOx and CO were high. However, when the system began operating in the flameless regime (equivalence ratios between 0.9 and 1.1) there was a simultaneous reduction of NOx and CO. This phenomenon is different from what occurs in conventional conditions.
5. It is possible that the relative increases in the intensity of the incoming air flow for the preheated case combined with physics and geometry, have had a significant influence to achieve the flameless system. Özdemir et al., [5] concluded that the input intensity of the reactants influences the rate of increase in internal gas recirculation and mixing of air and / or fuel with these gases.

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