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Experimental investigation of the natural gas confined flames using the OEC

Alex Álisson Bandeira Santos [a,b,*], Ednildo Andrade Torres [b,c], Pedro Afonso de Paula Pereira [b,d]

[a] SENAI CIMATEC – Integrated Center of Manufacturing and Technology, Avenida Orlando Gomes, 1845, Piatã, 41650-010 Salvador, Bahia, Brazil
[b] CiEnAm – Interdisciplinary Center of Energy and Environment, Federal University of Bahia, Rua Professor Aristides Novis, n 2 - Escola Politécnica, 3 Andar - Federação, Salvador, BA, Brazil
[c] Laboratory of Energy and Gas, Department of Chemical Engineering, Federal University of Bahia, Rua Professor Aristides Novis, n 2 - Escola Politécnica, 3 Andar - Federação, Salvador, BA, Brazil
[d] Institute of Chemistry, Federal University of Bahia, Rua Barão de Geremoabo, s/n, Campus Universitário de Ondina, Salvador, Bahia, Brazil

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ABSTRACT

The concept of environmental efficiency in equipment is increasing with the unfolding of global warming. In terms of industrial equipment, it is the burners which have a major impact in this discussion because of industrial combustion. Demand for environmentally more efficient burners with a reduction in emissions is essential for the proper use of fossil fuels during the transition between this energy and alternative energy sources for the next fifty years or more. This study experimentally evaluates the technique of oxygen-enhanced combustion – OEC – and its interaction with soot formation and thermal radiation in natural gas confined flames. The literature shows that the OEC technique – an important technique for improving the thermal efficiency of combustion – causes under certain conditions an increase in soot formation. Soot, as an important participant in radiant heat transfer, can increase the thermal efficiency of burners, implementing heat transfer from the flame to the heating areas, thereby reducing fuel consumption, the temperature of the flame, and consequently a reduction in the emission of NO_x. In the experiment was used low enriched with oxygen, which does not require significant existing equipment changes. This technology can play an important role in preparing particularly the oil and gas industry for the technological challenge of reducing global warming.

1. Introduction

Glassman (1987) [1] defined soot as carbonaceous particulates formed in the gas phase of combustion processes. These particulates consist mainly of carbon and contain up to 10% hydrogen on a molar basis. According to Turns (1996) [2], soot formation and evolution proceeds in a four-step sequence: (i) formation of precursor species, (ii) soot particle inception, (iii) surface growth and particle agglomeration, and (iv) particle oxidation. The emission of soot from combustors, or from flames, results from competition between soot formation and oxidation. Soot emission occurs when fuel is burnt in insufficient oxygen. The phenomenon of soot formation is still not fully explained due to the fact that the formation process is not slow enough to allow the precise observation of each step.

Oxygen-enhanced combustion (OEC), mentioned by Baukal (1998) [3] can improve the combustion process by producing improved flame characteristics (larger inflammability limit, better ignition, stability and shape control); smaller combustion gas volumes; increased productivity and thermal efficiency (larger heat transfer process efficiency, improved product quality, reduction in fuel consumption, cost reduction in raw material, reduced costs of new equipment and possibly production increase in existing equipment).

Atmospheric air contains about 21% oxygen in volume. Low levels of enrichment of the combustion air with oxygen, corresponding to an O_2 index below 30%, are usually used in retrofit applications in which only small modifications to the existing equipment are necessary.

Data about soot, including the use of chemical additives to control its formation, have been obtained mostly from studies performed with elementary flames, as in the present work. These flames are usually defined as either premixed, partially premixed, or non-premixed (diffusion) flames.

In a diffusion flame the reactants are initially separated, and reaction occurs only at the interface between the fuel and the oxidizer, where mixing and reaction both take place. The addition of oxygen in diffusion flames can be carried out by direct addition to the fuel, or to the combustion air in a burner with an annular oxidizer, parallel or counterflow.

* Corresponding author. SENAI CIMATEC – Integrated Center of Manufacturing and Technology, Avenida Orlando Gomes, 1845, Piatã, 41650-010 Salvador, Bahia, Brazil. Tel./fax: +55 7132839808.
E-mail address: alex.santos@cimatec.fieb.org.br (A.Á. Bandeira Santos).

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Table 1
General information – coflow burner tests.

Authors	Flames characteristics	O_2 in oxidizer [%]	Fuel
Yaccarino and Glassman, 1980 [13]	Open flame laminar	9–50	Ethylene
Lee et al., 2000 [14]	Open flame laminar	50; 100	Methane
Zelepouga et al., 2000 [15]	Open flame laminar	21; 35; 50; 100	Methane
Hwang and Gore, 2002 [16]	Open flame	21; 67	Methane
Wang et al., 2002 [17]	Open flame turbulent	21–100	Propane Natural gas Ethane/methane
Wang et al., 2005 [19]	Confined flame turbulent	40	Propane
Santos et al., 2009 [25]	Open flame laminar	21; 23; 25	Acetylene
Present work	Confined flame turbulent	21; 23; 25	Natural gas

The direct addition of oxygen to a methane diffusion flame has been studied by Saito et al. (1986) [4] and Gülder (1995) [5]. Wey et al. (1994) [6], Hura and Glassman (1988) [7], Du et al. (1990) [8], Leung and Lindstedt (1991) [9], and Gülder (1995) [5] studied the addition of it to propane and butane diffusion flames. Hura and Glassman (1988) [7], Du et al. (1990) [8], Leung and Lindstedt (1991) [9] and Hwang et al. (1998) [10] studied the addition of oxygen to ethane diffusion flames. Kent and Bastin (1984) [11] studied the addition of oxygen to free turbulent diffusion acetylene flames over a wide range of velocities and nozzle sizes.

Oxygen enhancement of the air side of a methane counterflow diffusion flame was studied by Beltrame et al. (2001) [12]. They verified that with an increase in the oxygen content in the oxidizer jet, soot formation was enhanced. The range of oxidizer oxygen content tested was 21–100%. They also verified the influence of the interaction between soot and NO_x.

Literature about the addition of oxygen to combustion air in a burner with a parallel annular oxidizer flow includes Yaccarino (1980) [13], Lee et al. (2000) [14], Zelepouga et al. (2000) [15], Hwang and Gore (2002) [16], Wang et al. (2002) [17] and Wang et al. (2005) [19].

Yaccarino and Glassman (1980) [13] studied the influence of the O_2 concentration on ethylene flames. The O_2 index was varied between 9 and 50% by the authors. They observed that soot formation reached a minimum around 24%. This was explained by competition between fuel pyrolysis and soot oxidation in the process domain.

Lee et al. (2000) [14] studied the influence of O_2 enrichment in laminar methane diffusion flames for conditions of 50 and 100% O_2. The authors found a reduction in soot production in both enrichment conditions, with a larger reduction for 100% O_2.

Zelepouga et al. (2000) [15] also examined the influence of O_2 enrichment on the air side of methane laminar diffusion flames, for 35, 50 and 100% O_2. The evaluation parameter was the integrated radial soot concentration. The authors observed a reduction in soot formation in all three situations and predicted that the soot concentration was smaller for flames with the larger O_2 index due to smaller flame lengths and consequently smaller residence time available for soot particle growth. In 2002, Hwang and Gore [16] investigated the radiation intensity of a methane/oxygen flame in comparison with a methane/air flame. A laser-induced incandescence technique was used to visualize the instantaneous and average soot distribution in the flames. Different combinations of central or annular fuel-oxygen supplies were studied to find the best arrangement to increase the thermal radiation intensity. The results showed that an oxygen-enhanced inverse diffusion flame

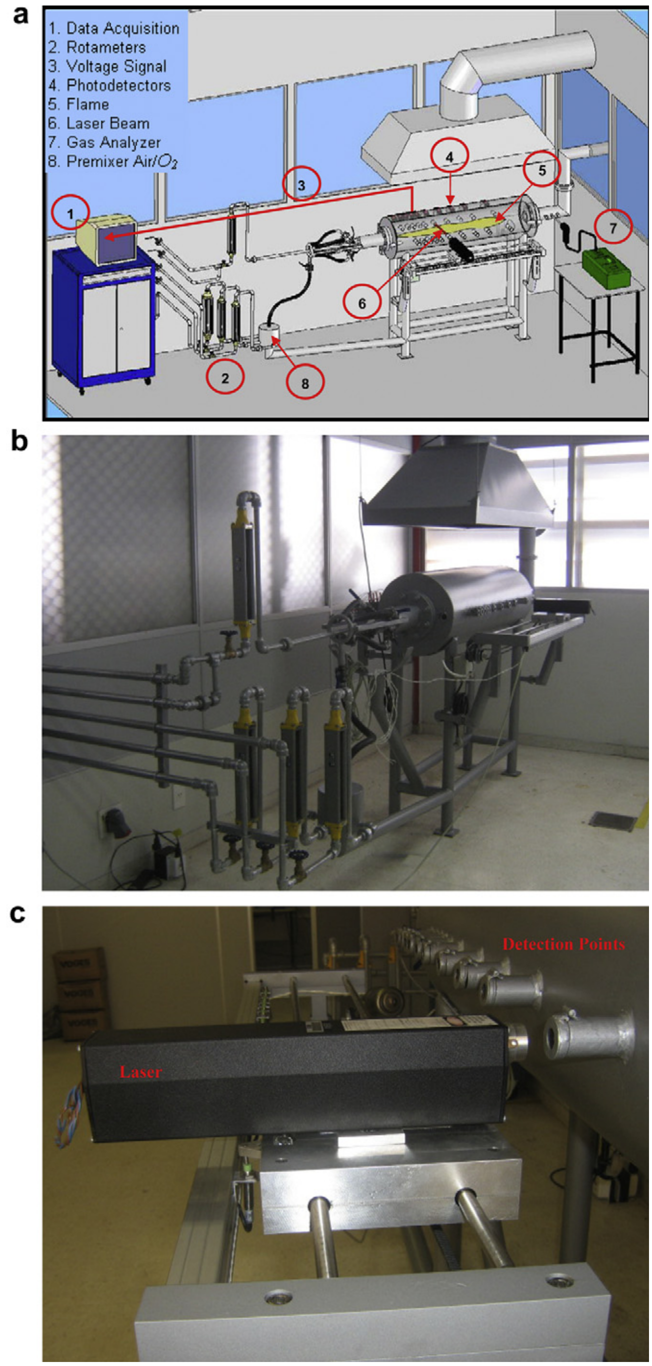

Fig. 1. Experimental setup; (a) Schematic of installation. (b) View of combustion chamber and rotameters. (c) View of laser system and ten points for soot and thermal radiation measurement.

(when the diffusion direction is opposite to that in the normal diffusion flame, where fuel flows from the central tube into still air) was very effective in increasing thermal radiation compared to a normal oxygen diffusion flame. This would happen due to the increased soot production in the inverse oxygen diffusion flame. The authors also found a more uniform spatial distribution of soot in the methane/oxygen flames compared to methane/air flames.

Furthermore, in 2002, Wang et al. [17] studied the influence of the oxygen index on soot, radiation and NO_x formation characteristics of turbulent jet flames for a range of oxygen indices from 21% (air) to 100% (pure O_2). The jet flame rig used in the experiments

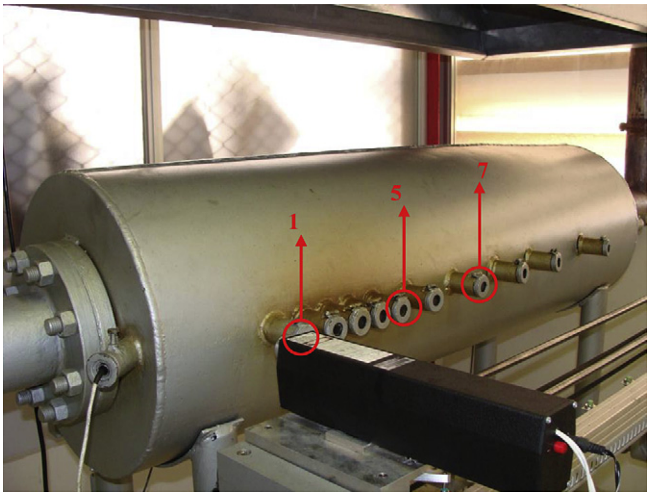

Fig. 2. Details view of cylindrical combustion chamber.

Table 3
Conditions used in the tests.

O_2 index	φ
21%	1.3
	1.1
	1.0
	0.9
	0.7
23%	1.3
	1.1
	1.0
	0.9
	0.7
25%	1.3
	1.1
	1.0
	0.9
	0.7

was designed to produce a vertical jet flame in a nearly quiescent air–oxygen coflow. The burner consisted of a 3 mm i.d. fuel tube centered in a 220 mm i.d. stainless steel flame chamber. Before entering the chamber, the air-oxygen oxidizer flow passed through a glass bead bed and a ceramic honeycomb, producing a uniform, laminar coflow.

The oxidizer flow was 4–6 times the stoichiometric flow. The fuel jet to oxidizer-coflow velocity ratios ranged from about 40 to 450. The combination of maintaining low coflow velocities and supplying in excess of the stoichiometric oxidizer requirements resulted in conditions close to a free flame. The fuel types used were natural gas, a methane/ethane blend, and propane.

The authors observed that soot quantities for all flames increased with the initial oxygen enhancement and then decreased as the oxygen content was further increased. The highest soot values occurred in the range of 30–40% oxygen index. As for the effect of the fuel type on the flame, the propane flame produced much more soot than the methane/ethane blend flame, which produced slightly more soot than the natural gas flame. The fuel jet velocity had a significant influence on soot formation and its dependence on oxygen index through residence time.

Goldstein Jr. et al. (2002) [18] verified the influence of the O_2 index on the oxidizer side of a partially premixed acetylene/air flame. The flame was submerged in atmospheric air, and surrounded by a N_2 shield. It was verified that soot formation increased in the flame with the shield, which was justified by the lack of O_2 available to intensify the oxidation process.

In 2005, Wang et al. [19] presented a comprehensive computational fluid dynamics (CFD) model which integrated detailed chemistry, soot formation and oxidation, radiation and NO_x formation for a propane-fueled, oxygen-enriched, turbulent, non-premixed jet flame. The results, compared with the experimental data available, gave an indication of the level of modeling that would be necessary.

Table 2
Natural gas composition and characteristics used in the tests.

Methane	88.82% vol.
Ethane	8.41% vol.
Propane	0.55% vol.
N_2	1.62% vol.
CO_2	0.60% vol.
HHV (kJ/m^3)	39 329.60
LHV (kJ/m^3)	35 564.00

Kumfer et al. (2006) [20] explored the criteria for soot inception in oxygen-enriched laminar coflow flames. In these experiments an axial height in the coflow flame is selected at which to identify the sooting limit. The sooting limit is obtained by varying the amount of inert until luminous soot first appears at this predefined height. The sooting limit flame temperature is found to increase linearly with stoichiometric mixture fraction, regardless of fuel type. To understand these results, the relationships between flame structure, temperature, and local C/O ratio is explored through the use of conserved scalar relationships. Analysis of experimental results suggests that soot inception occurs when the local C/O ratio is above a critical value. The values for critical C/O ratios obtained from the analysis of experiments using several fuels are similar in magnitude to the corresponding C/O ratios for premixed flames. In addition, temperatures and polycyclic aromatic hydrocarbons (PAH) fluorescence were measured to identify regions in these flames most conducive to particle inception. Results indicate that the peak PAH concentration lies along a critical iso-C/O contour, which supports a theory that soot particles first appear along this critical contour, given sufficient temperature.

Ergut et al. (2007) [21] carried out a study into the evolution of products of incomplete combustion (PIC) emitted from one-dimensional, laminar, atmospheric-pressure ethylbenzene flames in the vicinity of the soot onset threshold. The objective of this study was to identify the role of the fuel-to-air equivalence ratio in the evolution of PAH and other PIC as soot precursors just prior to and subsequent to soot onset in premixed flames. Temperature measurements and product sampling were conducted at various heights above the burner. Collected samples were analyzed for soot, PAH, oxygenated species, fixed gases, and light hydrocarbons. The results indicated that the soot onset limit is not a function of flame temperature alone; i.e. while the maximum measured flame temperatures was kept fairly constant, the flame could be either sooting, at the sooting limit or non-sooting depending on the equivalence ratio.

Kumfer et al. (2008) [22] studied the combination of oxygen enrichment and fuel dilution on diffusion flames which results in an increase in the stoichiometric mixture fraction, Zst, and alters the flame structure, i.e. the relationship between the local temperature and the local gas composition. Increasing Zst has been shown to result in the reduction or even elimination of soot. In the present work, the effects of variable Zst on soot inception are investigated in normal and inverse coflow flames, using ethylene as the fuel.

Ferrières et al. (2008) [23] studied the oxidation of laminar premixed natural gas flames experimentally and computationally with variable mole fractions of hydrogen (0, 20, and 60%) present in the fuel mixture. All flames were operated at low pressure (0.079 atm) and at variable overall equivalence ratios ($0.74 < \varphi < 1.0$) with constant cold gas velocity. At the same global equivalence ratio, there is no significant effect of the replacement of natural gas by 20% H_2.

Andersson et al. (2008) [24] evaluated the OEC influence in soot formation in propane-confined flames. The O_2 was premixed in the air combustion with gas recirculation. The results showed that OEC use associated with CO_2 recirculation increased the soot formation and in addition to the implementation of thermal radiation.

Santos et al. (2009) [25] evaluated the OEC influence in soot formation in acetylene diffusion flames. The results suggested that the simultaneous variation of the oxygen content and of the oxidizer velocity can provide control of the soot formation and distribution along the flame. Table 1 summarizes the general information about coflow burner tests with the OEC.

The soot control and OEC have been studied to improve energy and environmental efficiencies of thermal equipments and processes (see [24,26–30]).

Evaluating the described aspects, control of the formation of soot can be an important factor for a more rational implementation of OEC. With this control, the transferred thermal radiation in heating processes can be monitored and the formation of NO_x controlled. This aspect can be a factor in the use of the technology, and its peculiarities require further research.

The effects of the process variables, such as oxidizer oxygen content, fuel jet shape, diameter and velocity on soot formation and distribution are complex and coupled. In almost all the articles presented here, the work was with burners open to the atmosphere.

The objective of the present work is to explore the correlations and interactions between the oxygen content in the oxidizer of the combustion, soot concentration, thermal radiation and NO_x along the length of a natural gas diffusion confined flame produced in a cylindrical combustion chamber with a parallel annular coaxial oxidizer flow, such that the natural gas discharge is surrounded by a flow of air, or oxygen-enriched air. The applied enrichment levels were 23 and 25% O_2 and they were used in retrofit applications where only small modifications in the existing equipment are required, furthermore with poor reported in the literature.

2. Experimental setup and methods

The experimental setup is shown in Fig. 1. The flame was generated in a horizontal cylindrical combustion chamber, which consisted of a burner with two concentric tubes: a 5 mm i.d. central tube, and a 100 mm i.d. external tube, and a chamber 1.35 m in length. Natural gas flowed up through the internal tube while air, or enriched air, flowed through the annular region between this tube and the larger diameter concentric tube. The illustration of cylindrical combustion chamber is presented in Fig. 2. Gas flow rates were controlled by valves and metered by rotameters. Diffusion air and oxygen were premixed before being fed into the combustion chamber.

Soot concentrations were measured along the flame length using the laser light extinction technique. The laser system was mounted on a step-motor driven horizontal translation table, which allowed the beam coming from laser to reach the flame at any desired level. The laser was of He–Ne, with a wavelength of 632.8 nm. Since the power output from the laser was only about 3 mW, background radiation was blocked from the flame by narrow band pass interference filters at the laser wavelength. The light was transformed into a voltage signal by the photodetectors and registered by data acquisition.

The thermal radiation was measured at the same points where the soot concentration was also measured, through a radiometer in the narrow band of soot radiation influence between 0.6 and 3 μm. The uncertainty of the thermal radiation measurements was in the order of 3%.

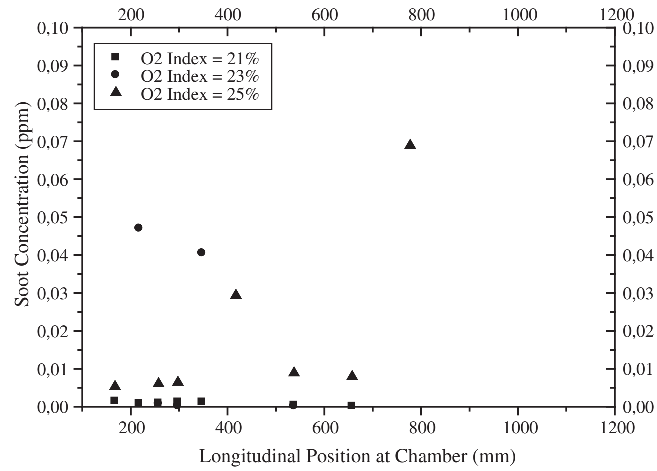

Fig. 3. The longitudinal distribution of soot concentration in combustion chamber at equivalence ratio $\varphi = 0.7$.

The NO_x concentration was measured in the exhaust gas on the chamber exit through the portable exhaust gas analyzer. The detection limit of analyzer has a maximum of 1000 ± 5 ppm.

Soot volume fraction (or concentration) (ppm) was calculated from the laser light extinction data using the Rayleigh limit of the Mie theory, so that:

$$\phi = \frac{\lambda}{6\pi \mathrm{Im}\left|\frac{m^2-1}{m^2+2}\right|} K_{abs} \tag{1}$$

where,

$$K_{abs} = \frac{1}{L}\ln\left(\frac{I_o}{I_L}\right) \tag{2}$$

λ, is the laser wavelength, L is the optical path length, I_o and I_L are the laser beam intensities, before and after traversing the flame, and m is the refractive index, adopted as $m = 1.90–0.55i$, according to Lee and Tien (1981) [31], Hulst (1981) [32] and Iuliis et al. (1998) [33].

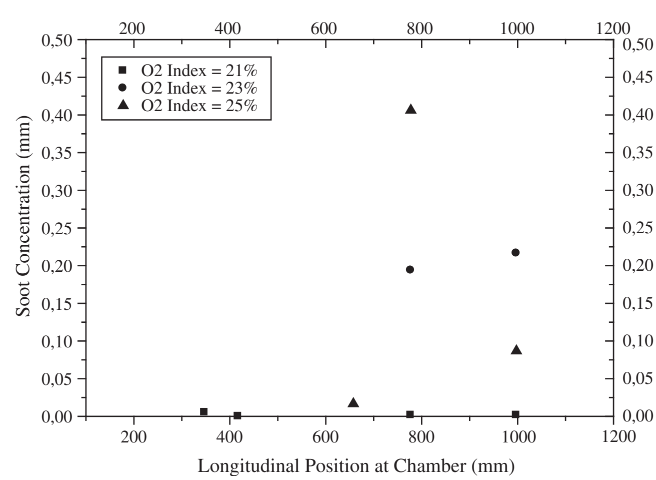

Fig. 4. The longitudinal distribution of soot concentration in combustion chamber at equivalence ratio $\varphi = 1.0$.

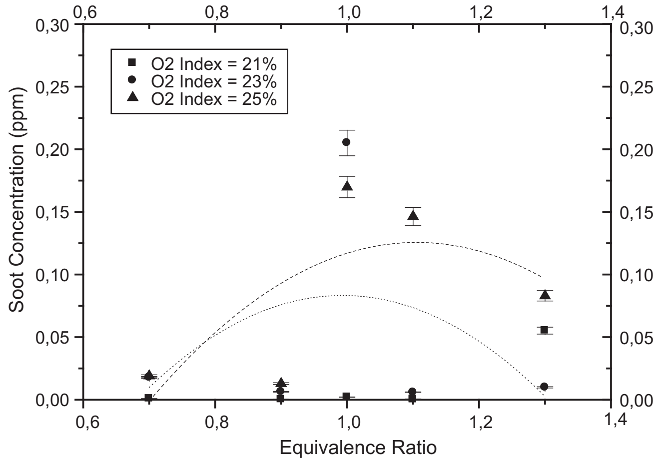

Fig. 5. Soot concentration along combustion chamber.

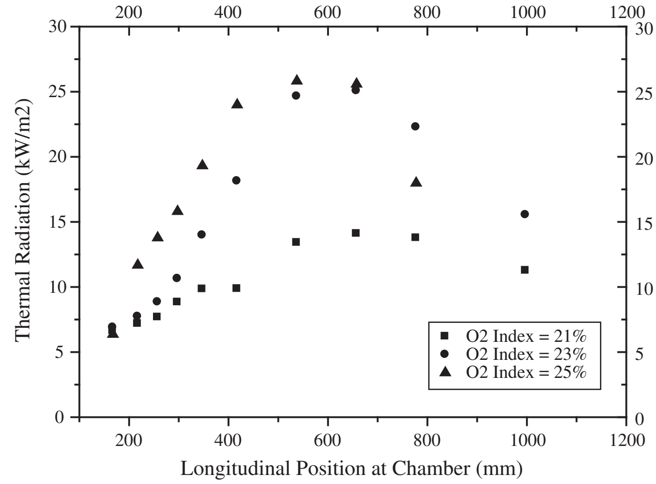

Fig. 7. The longitudinal distribution of thermal radiation in combustion chamber at equivalence ratio $\varphi = 0.7$.

According to the scattering theories of Mie and Rayleigh, the radiative properties of soot can be evaluated. From this, the soot concentration – established in Eq. (1) – was determined on the basis of its absorption coefficient defined in those theories, linking the complex index of refraction and soot concentration. Once the equation of soot concentration is established, it was possible to estimate indirectly the soot concentration over the flame from the attenuation of laser, established in Eq. (2). The higher the attenuation of the laser is, the higher is the soot concentration, thereby indicating a formation of particulate.

Some tests were performed in order to check the repeatability of results. In this work, the average uncertainty of the soot concentration measurements was in the order of 1%.

To examine the effect of the oxygen content of the combustion air, tests were performed comparing experiments with 23 and 25% O_2 to experiments with plain air (21% O_2). In the tests the equivalence ratios (φ) were maintained over a wide range (1.3–0.7). The natural gas flow was 0.0003 m^3/s (18 L/min), referred to 20 °C and atmospheric pressure. The burner power was 9.76 kW. Table 2 presents the natural gas composition and characteristics used in the tests. Table 3 summarizes the conditions used in the tests. The gas flow was in transition regime (laminar to turbulence).

3. Results and discussion

The analysis of the soot formation in the process was based on the values found for soot concentration located in the flame and also by its luminous soot characteristics (radiation indicator). The analysis of the thermal radiation followed the same methodology.

Figs. 3 and 4 present the longitudinal distribution of soot in the combustion chamber at equivalence ratio equal to 0.7 and 1.0. The soot formation increased with OEC.

As stated in Eq. (1), regions with higher soot concentration are those with the highest attenuation of laser. In Fig. 3, it was verified that using the OEC, the region of initiation of the flame provided a higher soot concentration.

Fig. 4 also showed an increase in the soot concentration with the OEC, and this increase was identified in the posterior region of the flame, near the combustion chamber exit, indicating a higher absorption of the laser signal from the agglomerates particles formed in this region.

Fig. 5 presents the soot concentration in the tested conditions using the average value found from the performed measurements at the ten points shown in Fig. 1(b), in which the measurement was possible, representing the tendency of the tested condition. The possibility of non-detection of the concentration is caused by the frequent instability of the confined flame, as well as the possibility of non-absorption of the power of the laser by very small particles due to flames which did not encourage soot formation. Therefore, further analysis of the luminous soot becomes important.

Increased soot formation using OEC was found in particular 25% O_2 index compared to 23% and plain air. This possibly occurs because of the increased production of radicals which are precursors of the

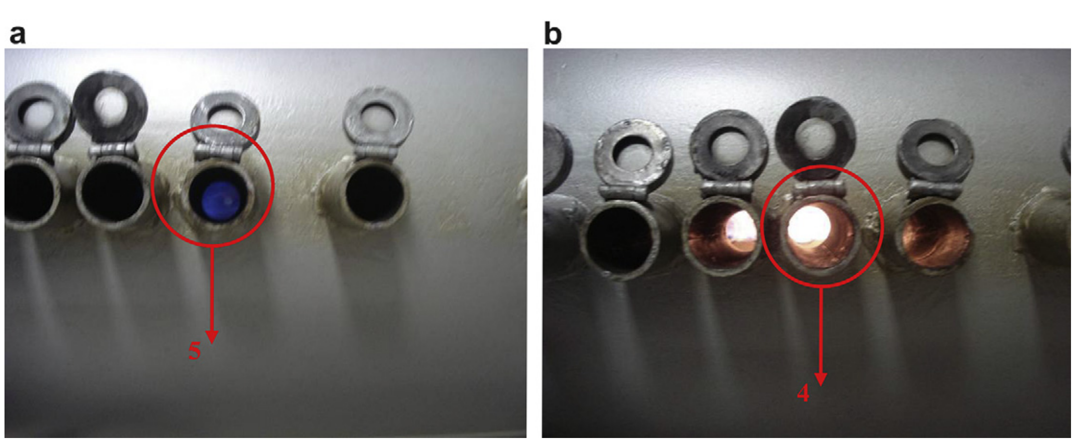

Fig. 6. Visual Aspects of Flames with OEC; (a) Flame without OEC utilization. (b) Flame with OEC utilization.

soot in the presence of O_2 in the pyrolysis of natural gas, as well as the best meeting between fuel and oxidizer in the confined flames without the influence of the external environment. Increasing the oxygen concentration increases the stoichiometric flame temperature which in turn increases the fuel pyrolysis and soot formation rates. This occurs in all equivalence ratios. The variation of soot concentration with the equivalence ratio becomes the variation of temperature with the equivalence ratio.

Detection problems were observed during conditions of flame instability in confined situations. Therefore the characteristics of luminous soot in the tested conditions were also evaluated (mechanism also evaluated in Andersson et al. [24]). Using the OEC with 23% or 25% O_2 index, a stronger yellow light, typical feature due to a greater soot concentration was verified. In all equivalence ratios, this was identified. On the other hand, the flames with air as oxidant have a typical blue color under the tested conditions. The typical aspects of tested flames are shown in Fig. 6.

Figs. 7 and 8 present the longitudinal distribution of thermal radiation in the combustion chamber at equivalence ratio equal 0.7 and 1.0. The thermal radiation was increased with the OEC.

Fig. 9 presents the average value of thermal radiation in the combustion chamber. There is a thermal radiation implementation using the OEC in the band of the soot influence. With the use of the OEC there was an average increase of 59% in the transferred energy by radiation compared to the flame with the plain air for all equivalence ratios.

The results suggest possible improvements in thermal equipments such as boilers and furnaces. Increasing the soot concentration perceived using the OEC coupled with augment of the radiative heat flux, can bring an increase in thermal efficiency of these devices, increasing productivity.

The OEC has a tendency to increase the flame temperature and consequently increases the available energy for thermal radiation transfer. If the flame is composed of components with a higher tendency to transfer heat by radiation, more energy will be transferred to heat any kind of surface or load. Therefore the temperature of the flame can be reduced. With OEC, the formation of soot was increased (increase is shown in Fig. 9), which caused an increase in the transferred energy from the flame (Fig. 9), decreasing its temperature.

Fig. 10 presents the NO_x formation at the combustion chamber exit. There is a slight reduction in NO_x using the OEC. OEC use causes the NO_x concentration to remain close the values that are found on burning with plain air.

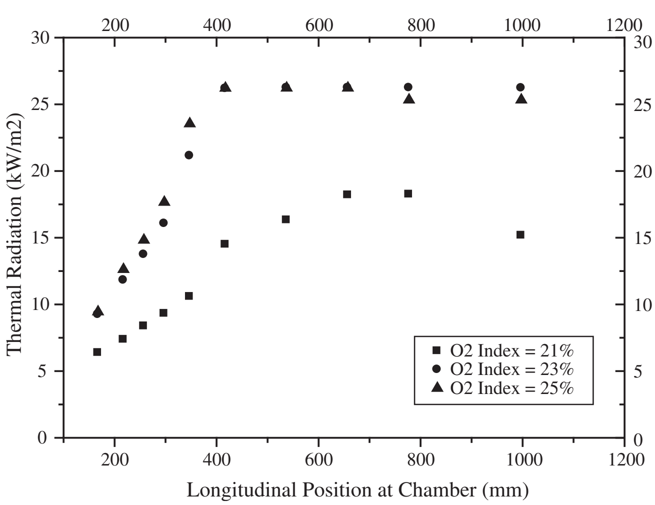

Fig. 8. The longitudinal distribution of thermal radiation in combustion chamber at equivalence ratio $\varphi = 1.0$.

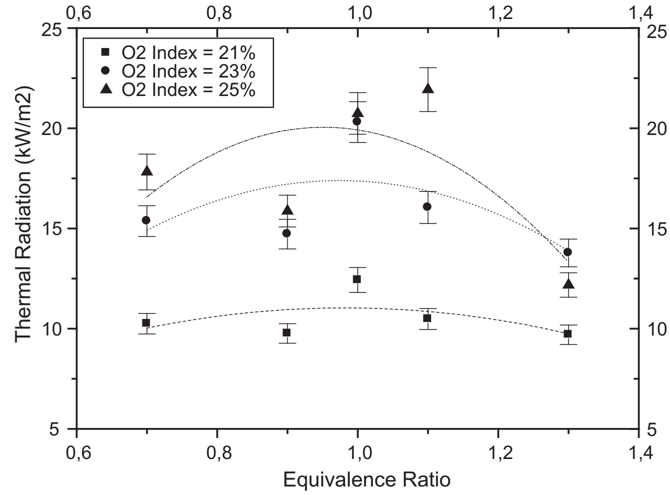

Fig. 9. Thermal radiation along combustion chamber.

OEC tends to increase the flame temperature consequently increasing the thermal formation of NO_x. A slight reduction and stabilization close to the values with plain air was found in most of the tested conditions. This can be explained by the increase in soot formation using OEC. This is illustrated in Figs. 5 and 6, which increases the potential for radiation heat transfer of the flame – soot increases the thermal radiation of flames – decreasing its temperature. It can be seen in Fig. 9 because of the physical trend of NO_x reduction.

The correlations between soot formation, thermal radiation and NO_x concentration can be indirectly seen through the temperature of the exhaust gas at the combustion chamber exit. In Fig. 11 the temperatures in question are identified.

Even though OEC tends to increase the flame temperature, the temperature of the exhaust gas also tends to increase. In the tested conditions, the temperature of the exhaust gas remains close to the values that are found when burnt with plain air; which confirms the influence of the soot in the reduction of flame temperature through the thermal radiation transfer resulting in NO_x reduction.

Temperature stabilization under the tested conditions was verified as being between 600 and 800 K (in most tested conditions).

A tendency of temperature stabilization justifies the concentration of NO_x in a narrow band (120–160 ppm – in most tested conditions).

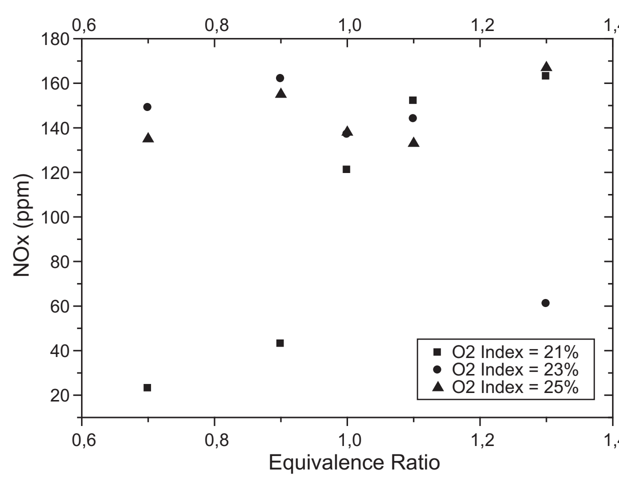

Fig. 10. NO_x concentration at combustion chamber exit.

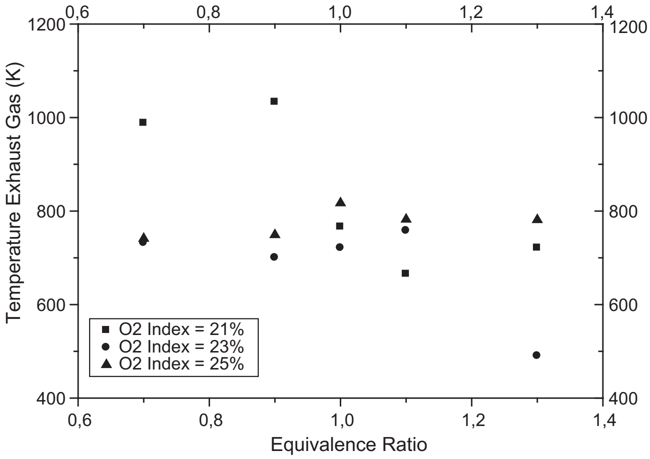

Fig. 11. Temperature of exhaust gas at exit combustion chamber.

The results of the NO_x concentration found in the geometry tested were lower than those required by the regulation of atmospheric emissions in Brazil (180–1440 ppm). They are also below the levels of emission of equipment without NO_x control, Borman and Ragland (1998) [34] state that these levels are between 1000 and 4000 ppm.

It is noticed that besides the increased thermal efficiency, the use of OEC by correlations between soot formation, thermal radiation and NO_x concentration, can bring a reduction of NO_x emissions in industrial combustion systems, increasing the environmental efficiency of these systems since NO_x is an important pollutant regulated for this type of industrial application.

4. Conclusions

In this present work the effect of the oxygen index on the oxidizer of combustion on soot, thermal radiation and NO_x concentration along the length of a natural gas diffusion confined flame was investigated. The levels of air enrichment applied were 2% and 4%, which can be applied in retrofit burners where only small modifications in existing equipment are required.

The results for the average concentration recorded at the measuring points have a tendency to increase soot formation by using the OEC, in particular at the level of 4% enrichment. With the use of OEC, there was also an increase of luminosity in the flame, indicating the qualitative increase in soot concentration.

The enrichment levels tested using the OEC, showed that thermal radiation has increased the heat flux. The NO_x emission was reduced when compared to the case of flame with atmospheric air. These results suggest that the use of OEC in natural gas confined flames produces an increase in thermal radiation coupled with significant reductions in NO_x formation.

These results may have application in industrial equipment such as boilers and furnaces that need high capacities for radiative heat transfer to ensure production efficiency. Using the OEC, the soot formation can be expanded and, under its influence, thermal radiation flux is also increased. Consequently, a reduction in flame temperature and NO_x formation through the thermal mechanism are provided. This occurs prevalently in combustion systems, which becomes an environmental benefit in industrial heating equipment.

OEC therefore proves to be a promising technique to promote an increase in radiation heat transfer, thermal efficiency while simultaneously reducing emissions of nitrogen oxides, which constitute an important regulated environmental pollutant.

Acknowledgments

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Nomenclature

φ: equivalence ratio
ϕ: soot concentration
λ: laser wavelength
HHV: higher heating value
LHV: low heating value
L: optical path length
i.d.: internal diameter
I: laser beam intensity
m: refractive index
K_{abs}: absorption coefficient