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INTERACTION OF SOOT FORMATION AND THERMAL RADIATION IN NATURAL GAS CONFINED FLAMES USING THE OEC WITH ENRICHMENT LOW LEVELS – AN EXPERIMENTAL STUDY

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Abstract. *The concept of environmental efficiency in equipment is becoming increasingly popular with the threat of global warming. Among 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 reduced emissions is essential for the use of fossil fuels during the transition between this energy source and alternative sources which can last more than fifty years. 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 works with the OEC technique –a technique that improves the thermal efficiency of combustion – causing under certain conditions an increase in soot formation. Soot as an important participant in radiant heat transfer. Soot interaction with the OEC can cause an increase in thermal efficiency of burners, implementing heat transfer from the flame to the heating areas, thereby reducing the consumption of fuel, the temperature of the flame, and consequently, reduce the emission of NOx. In the experiment low enriched with oxygen was used, which does not require significant existing equipment changes. This technology can be an important tool for industry in general, particularly in oil and gas, in the technological challenge of reducing global warming.*

Keywords*: Soot, Thermal Radiation, OEC, Natural Gas.*

1. INTRODUCTION

Glassman (1987) defined soot as carbonaceous particulates formed in the gas phase of combustion processes. They consist mainly of carbon, and contain up to 10% hydrogen on a molar basis. According to Turns (1996), 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 the 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), 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; fuel consumption reduction, raw material costs reduction, reduced costs of new equipment and possibly production increase in existing equipment).

Atmospheric air contains about 21% of oxygen in volume. Low levels of enrichment of the combustion air with oxygen, i.e. 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, has 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, parallel or counterflow oxidizer.

The direct addition of oxygen to a methane diffusion flame has been studied by Saito et al. (1986) and Gülder (1995). Wey et al. (1994), Hura and Glassman (1988), Du et al. (1990), Leung and Lindstedt (1991), and Gülder (1995) studied the addition to propane and butane diffusion flames. Hura and Glassman (1988), Du et al. (1990), Leung and Lindstedt (1991) and Hwang et al. (1998) studied the addition of oxygen to ethane diffusion flames. Kent and Bastin (1984) 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). They verified that with an increase in 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 interaction among 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), Lee et al. (2000), Zelepouga et al. (2000), Hwang and Gore (2002), Wang et al. (2002) and Wang et al. (2005).

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

Lee et al. (2000) 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) 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 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 (2002) 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 (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 happens due 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. (2002) 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 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 to 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% to 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) 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 involved by a N_2 shield. It was demonstrated 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. (2005) presented a comprehensive 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.

Kumfer, et al. (2006) 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 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) 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 polycyclic aromatic hydrocarbons (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) studied the combination of oxygen enrichment and fuel dilution for 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) 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% of H_2.

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 effect of the oxygen content in the oxidizer of the combustion on the soot concentration and thermal radiation along the length of a natural gas diffusion confined flame produced in a 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.

2. EXPERIMENTAL SETUP AND METHODS

The experimental setup is shown in Fig. l. The flame was generated in a horizontal cylindrical combustion chamber, which consisted of burner with two concentric tubes: a 5mm i.d. central tube, and a 100mm i.d. external tube, and a chamber 1.35m 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. 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. As the power output from the laser was only about 3mW, background radiation was blocked from the flame by narrow band pass interference filters, at the laser wavelength. The light was transformed into an electrical current signal by the photodetectors, and registered by data acquisition.

The thermal radiation was measured in the same points that the soot concentration was also measured, through radiometer in the narrow band of soot radiation influence between 0.6-3μm.

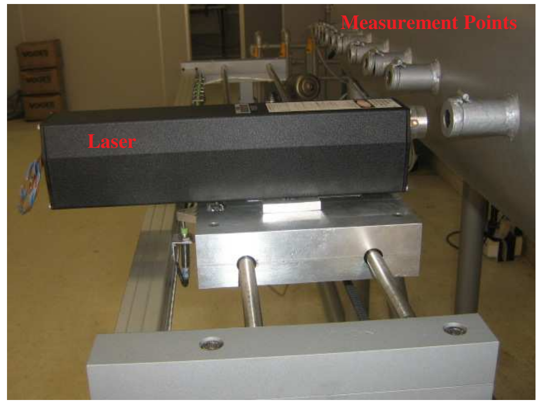
Soot volume fraction (or concentration) (ppm) was calculated from the laser light extinction data using the Rayleigh limit of the Mie theory, so that:

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

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

λ , is the laser wavelength, L the optical path length, I_o and I_L the laser beam intensity, 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), Hulst (1981) and Iuliis et al. (1998). 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%.

(a) View of Combustion Chamber and Rotameters.



(b) View of laser system and ten points for soot measurement.

Figure 1: Experimental Setup.

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 0.0003m^3/s (18 l/min), referred to 20°C and atmospheric pressure. The burner power was 9.76kW. Table 1 summarizes the conditions used in the tests.

Table 1 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

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 characteristics of luminous soot (radiation indicator). The analysis for the thermal radiation followed the same methodology.

Fig. 2 presents the soot concentration in the tested conditions through the average value, which is 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 that are precursors of the 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 tendency occurs in all equivalence ratios. The variation of soot concentration with the equivalence ratio arises from the variation of temperature with the equivalence ratio.

Detection problems were seen by the flames instability in the confined condition therefore the characteristics of luminous soot in the tested conditions were also evaluated. 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 trend 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. 3.

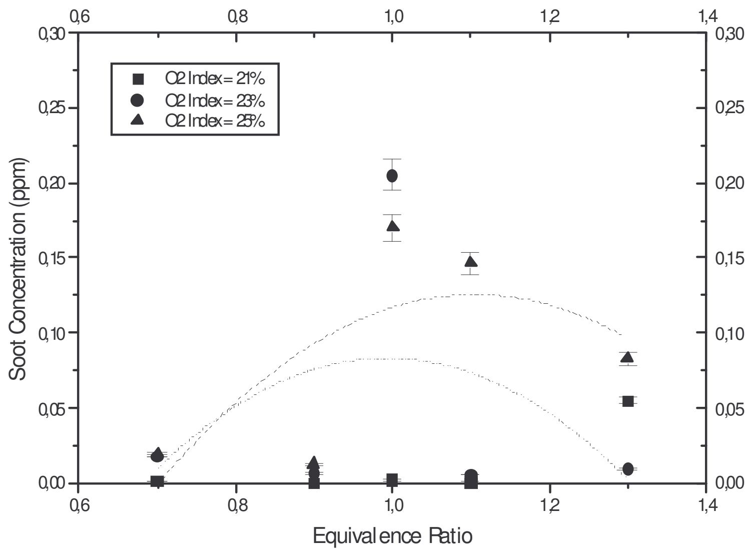


Figure 2: Soot concentration along combustion chamber.

(a) Flame without OEC Utilization.

(b) Flame with OEC Utilization.

Figure 3: Visual Aspects of Flames with OEC.

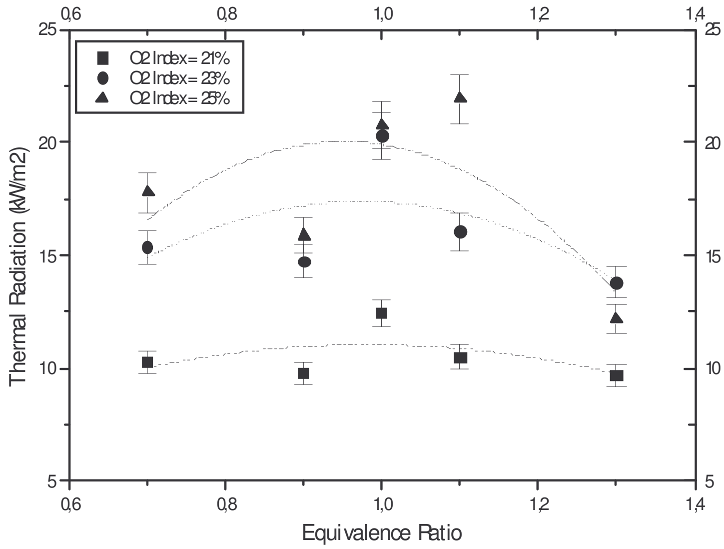


Figure 4: Thermal radiation along combustion chamber.

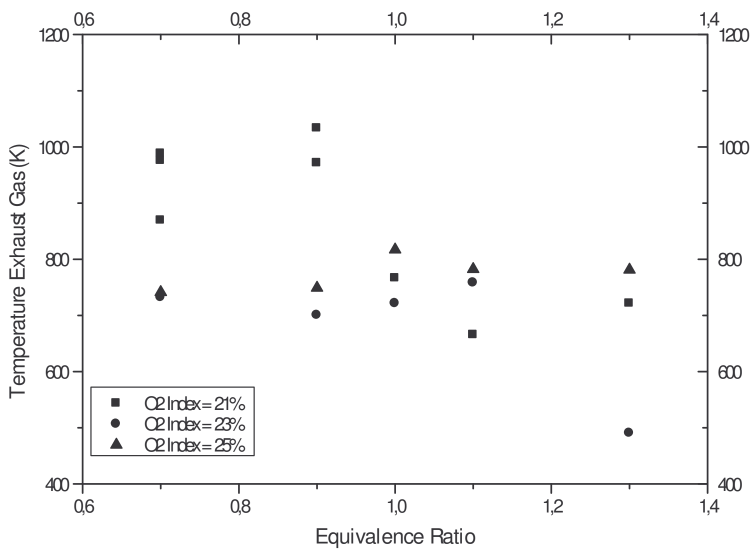


Figure 5: Temperature of exhaust gas at exit combustion chamber.

Fig. 4 presents the thermal radiation at combustion chamber (same points of soot measurement). 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 burning with the plain air for all equivalence ratios.

The OEC has a tendency to increase the flame temperature and consequently increases the available energy for the 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 Figure 2), which caused an increase in the transferred energy by the flame (Fig. 4) lowering its temperature.

This can be indirectly seen through the temperature of the exhaust gas at the combustion chamber exit. In Fig. 5 the temperatures in question are identified. Temperature stabilization under the tested conditions was verified between 600-800K (in most tested conditions).

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

4. CONCLUSIONS

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

The results suggest that the use of OEC in natural gas confined flames produces an increase of soot formation and consequently enhanced thermal radiation. This can be a control tool to implement radiation heat transfer and soot formation in confined burners.

5. ACKNOWLEDGEMENTS

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