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An experiment on the effect of oxygen content and air velocity on soot formation in acetylene laminar diffusion flame produced in a burner with a parallel annular coaxial oxidizer flow☆

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ABSTRACT

The effect of oxygen content and of the combustion air velocity on soot formation was studied in acetylene diffusion flames. These flames were produced in a burner with a parallel annular coaxial flow of oxidizer. The effect on the flame axial temperature profile was also evaluated. The soot volume fraction was calculated by the laser light extinction methodology. The oxygen content in the combustion air was smaller than 30%, which does not require significant retrofit of existent equipment when the combustion conditions are varied. The results suggest that the parallel manipulation of the oxygen content and of the oxidizer velocity can provide means for managing soot formation and distribution. The formation of soot in industrial combustion systems is of interest in engineering, because the presence of soot in the flame enhances the heat transfer from the combustion gases by thermal radiation, increases the need for burner maintenance, and constitutes an environmental problem when emitted in the atmosphere.

1. Introduction

Glassman [1] 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 [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 the competition between soot formation and oxidation. Soot emission occurs when fuel is burnt in insufficient oxygen. The phenomenon of soot formation still is not fully explained, due to the fact that the formation process isn't slow enough to allow the precise observation of each step.

The enrichment of combustion air with oxygen, mentioned by Baukal [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; fuel consumption reduction, raw material costs reduction, reduced costs of new equipments and, possibly, production increase in existing equipments).

Atmospheric air has about 21% of 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 that only small modifications are necessary in the existent equipment.

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 into either premixed, partially premixed, or nonpremixed (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 part. 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.

The direct addition of oxygen to a methane diffusion flame was studied by Saito et al. [4] and Gülder [5]. Wey et al. [6], Hura and Glassman [7], Du et al. [8], Leung and Lindstedt [9], and Gülder [5] studied the addition to propane and butane diffusion flames. Hura and Glassman [7], Du et al. [8], Leung and Lindstedt [9] and Hwang et al. [10] studied the addition of oxygen to ethane diffusion flames. Kent and Bastin [11] studied the addition of oxygen to free turbulent diffusion acetylene flames over a wide range of velocities and nozzle sizes.

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Nomenclature

Alphabet symbols

I	laser beam intensity
K	absorption coefficient
L	optical path length
m	refractive index of soot
V	velocity

Greek symbols

Φ	soot volume fraction
λ	laser wavelength

Subscripts

O	initial
L	optical path length
abs	absorption
g	gas
a	air

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

The literature about addition of oxygen to the combustion air in a burner with a parallel annular oxidizer flow includes Yaccarino [13], Lee et al. [14], Zelepouga et al. [15], Hwang and Gore [16], Wang et al. [17] and Wang et al. [18].

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

Lee et al. [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 larger reduction for 100% O_2.

Zelepouga et al. [12] 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 of soot formation in all three situations, and predicted that soot concentration was smaller for flames with the larger O_2 index, due to smaller flames length and, consequently, smaller residence time available for soot particle growth.

In 2002, Hwang and Gore [16] investigated experimentally the radiation intensity of a methane/oxygen flame in comparison with a methane/air flame. A laser-induced incandescence technique was utilized 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 for increasing 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 would happen 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.

Also, in 2002, Wang et al. [17] studied the influence of the oxygen index on soot and radiation 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. 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.

In 2005, Wang et al. [18] presented a comprehensive CFD model, which integrated detailed chemistry, soot formation and oxidation, and radiation, for a propane-fueled, oxygen-enriched, turbulent, nonpremixed jet flame. The results, compared with the experimental data available, gave indication of the level of modeling that would be necessary.

Goldstein Jr, et al. [19] verified the influence of the O_2 index on the oxidizer side of a partially premixed acetylene/air flame. The flame

Table 1
General information — coflow burner tests.

Authors	Configuration	O_2 in oxidizer [%]	Fuel
Yaccarino and Glassman [13]	The fuel and oxidizer were supplied through two concentric tubes. A 10 mm i.d brass fuel tube was surrounded by a glass tube of 100 mm diameter and 1000 mm length. Laminar	9–50	Ethylene
Lee et al. [14]	The fuel and oxidizer were supplied through two concentric tubes of 11.1 and 101.6 mm i.d., respectively. Laminar	50; 100	Methane
Zelepouga et al. [15]	The burner consists of an 11.0 mm i.d., 300.0 mm long brass fuel tube centrally positioned in the 82.0 mm diameter stainless steel oxidizer tube. Laminar	21; 35; 50; 100	Methane
Hwang and Gore [16]	Burner consisting of three concentric tubes. The central tube had a 4 mm i.d., while the first concentric outer tube had 6.5 mm i.d. and the external tube 9.5 mm i.d.	21; 67	Methane
Wang et al. [17]	The burner consisted of a 3 mm i.d. fuel tube centered in a 220 mm i.d. stainless steel flame chamber. Turbulent jet	21–100	Propane natural gas ethane/ methane
Wang et al. [18]	The burner consisted of a 3 mm i.d. fuel tube centered in a 220 mm i.d. stainless steel flame chamber. Turbulent jet	40	Propane
Present work	Burner consisted of two concentric vertical tubes: a 23 mm i.d. central tube, 1 mm thick, and a 26 mm i.d. external tube. On the top of the central tube it was placed a perforated cap, with 0.5 mm i.d. holes. Laminar	21; 23; 25	Acetylene

was submerged in atmospheric air, and involved 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.

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. Table 1 summarizes the information from the literature about the different configurations for the addition of oxygen to combustion air in a burner with a parallel annular oxidizer flow, which was tested by the authors.

The objective of the present work was to explore the effect of the oxygen content and the combustion air velocity on the soot concentration along the height of an acetylene laminar diffusion flame produced in a vertical axis burner with a parallel annular coaxial oxidizer flow, such that the acetylene discharge was surrounded by the flow of air, or oxygen-enriched air. The applied enrichment levels were 23 and 25% O_2, used in retrofit applications, where only small modifications in the existent equipment are required.

2. Experimental apparatus and methods

The experimental setup is shown in Fig. 1. The flame was generated in burner QM1, which consisted of two concentric vertical tubes: a 23 mm i.d. central tube, 1 mm thick, and a 26 mm i.d. external tube. On the top of the central tube it was located a perforated cap, with 0.5 mm holes. Acetylene 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 VI, V2, V3 and metered by rotameters R1, R2 and R3. Diffusion air and oxygen were premixed in PM1, before being fed to the burner QM1, whose main dimensions are shown in Fig. 2.

Soot concentration was measured along the flame height using the laser light extinction technique. The burner was mounted on a step-motor driven vertical translation table, which allowed the beam coming from laser L1 to reach the flame at any desired level. The laser L1 was of He–Ne, with a wavelength of 632.8 nm. Since the power output from the laser was only about 1 mW, background radiation was blocked from the flame by a narrow band pass interference filter Fl, at the laser wavelength. The light was transformed in an electrical current signal by the photodiode FT1, and registered by the electrometer ET1.

Flame temperatures were measured by an uncoated type S thermocouple Tl (Pt–Pt/10%Rh) along the central axis of the flame, and the signals were registered by the temperature meter RG1. The thermocouple tip was cleaned out before every temperature reading. Preliminary work was required to find the flow rates which provided a steady flame, avoiding flash back and flame lift.

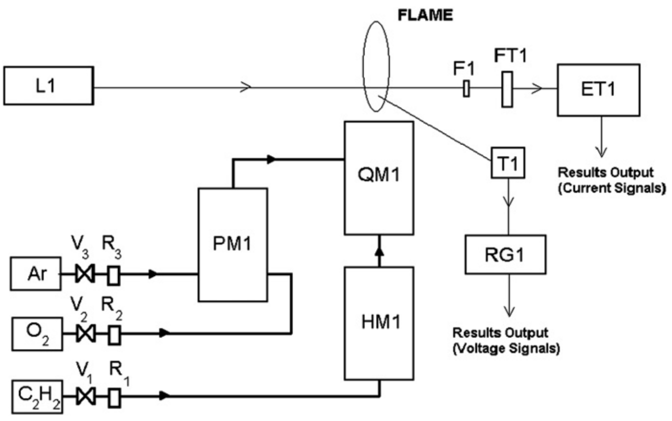

Fig. 1. Experimental setup.

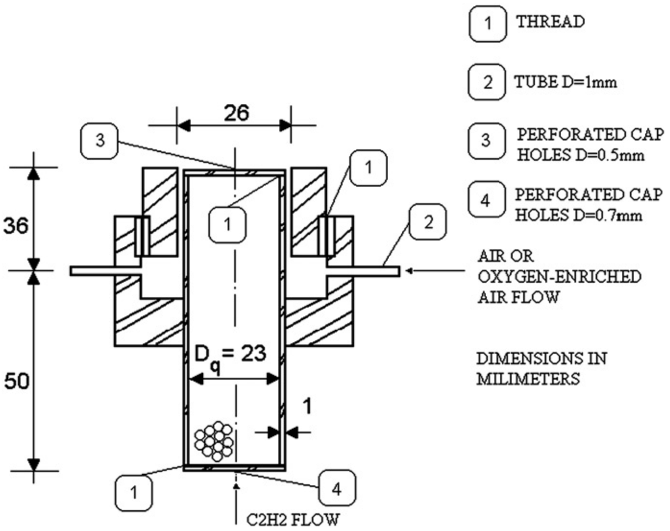

Fig. 2. Burner QM1.

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

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

where

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

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 [20], Hulst [21] and Iuliis et al. [22]. In this work, the average uncertainty of the soot volume fraction measurements was in the order of 8.5%. Both flows were in the laminar regime.

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), stagnant or flowing. The air velocities, V_a, were 0.10, 0.15, 0.85 and 1.39 m/s, and the acetylene velocities, V_g, were 0.22 and 0.36 m/s, referred to 20 °C and atmospheric pressure. The burner power was 0.72 and 1.16 kW. Table 2 summarizes the combination of fluid velocities used in the tests.

3. Results and discussion

Figs. 3 and 4 present the soot volume fraction and the temperature along the flame height for experiments where $V_a < V_g$, while Figs. 5 and 6 present the data for $V_a > V_g$, respectively. The position along the flame was established by a dimensionless ordinate.

To facilitate data comparison, the soot and temperature distributions for plain air (21% O_2), stagnant or flowing, were included in all figures, as reference.

When $V_a < V_g$ the combustion air is accelerated by the gas jet, and the contact between the fluids varies as a function of $(V_a - V_g)$. Mixing

Table 2
Velocity ratio V_a / V_g.

V_g (m/s)	V_a (m/s)						Power (kW)
	0	0.10	0.15	V_g	0.85	1.39	
0.22	0	0.45		1	3.86		0.72
0.36	0		0.42	1		3.86	1.16

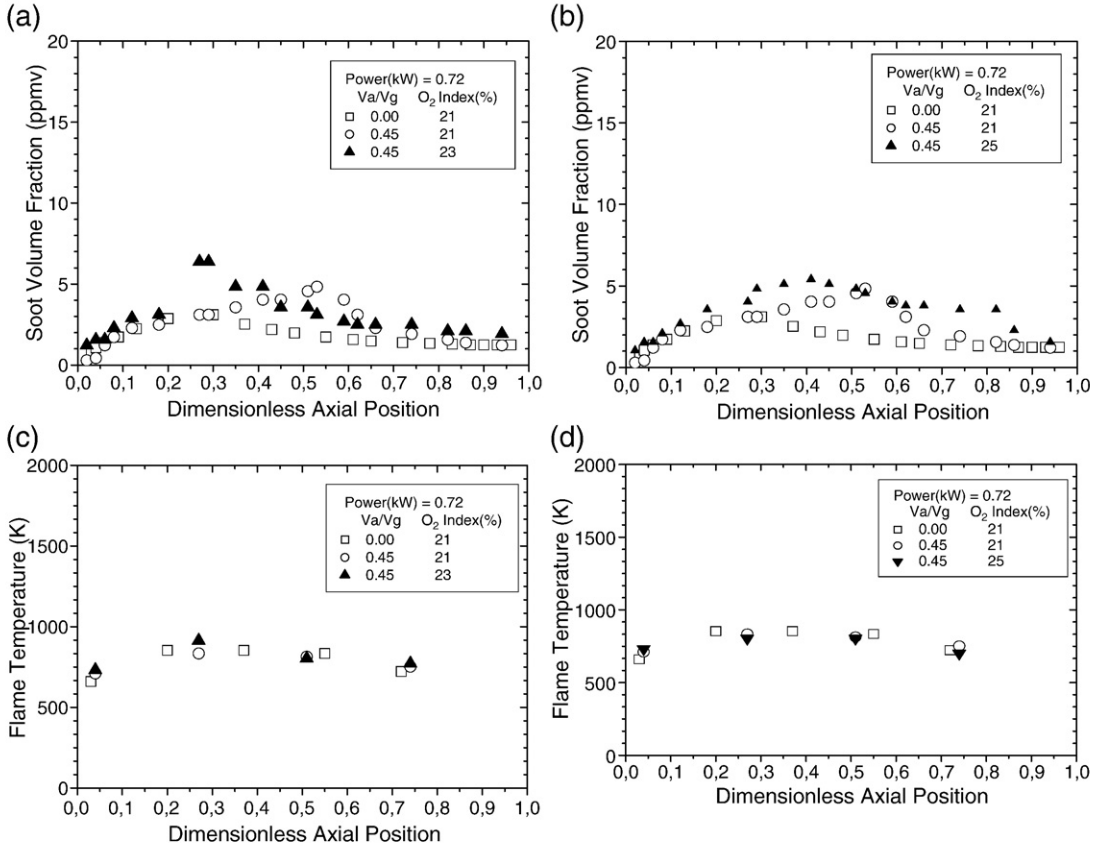

Fig. 3. Soot volume fraction (a,b) and temperature (c, d) along the flame height $V_a / V_g = 0$ and 0.45; power = 0.72 kW.

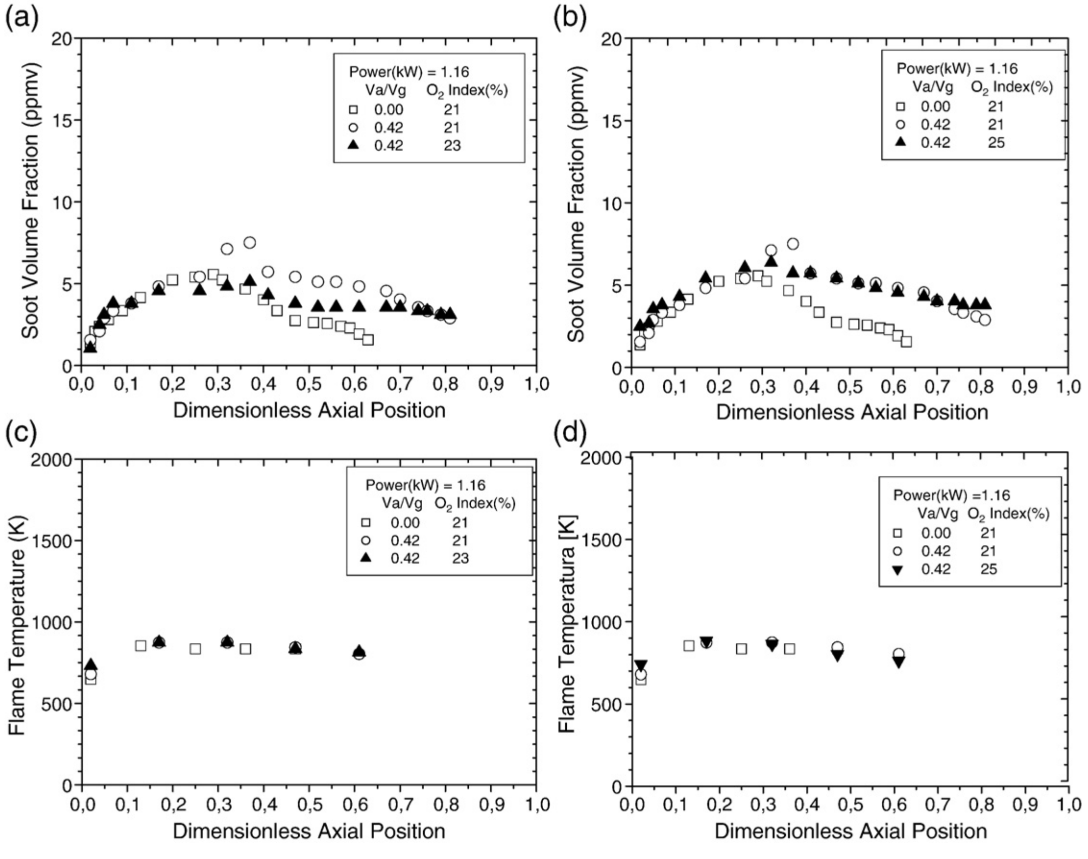

Fig. 4. Soot volume fraction (a,b) and temperature (c,d) along the flame height $V_a / V_g = 0$ and 0.42; power = 1.16 kW.

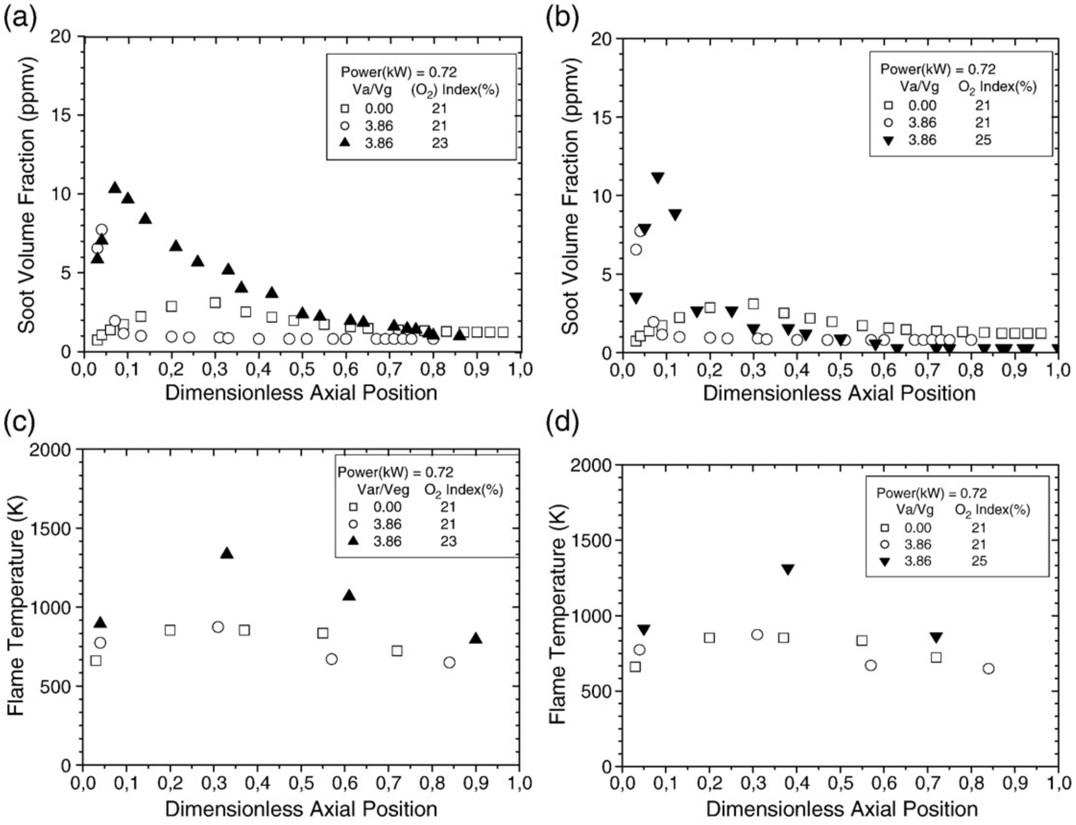

Fig. 5. Soot volume fraction (a,b) and temperature (c,d) along the flame height $V_a/V_g = 0$ and 3.86; power = 0.72 kW.

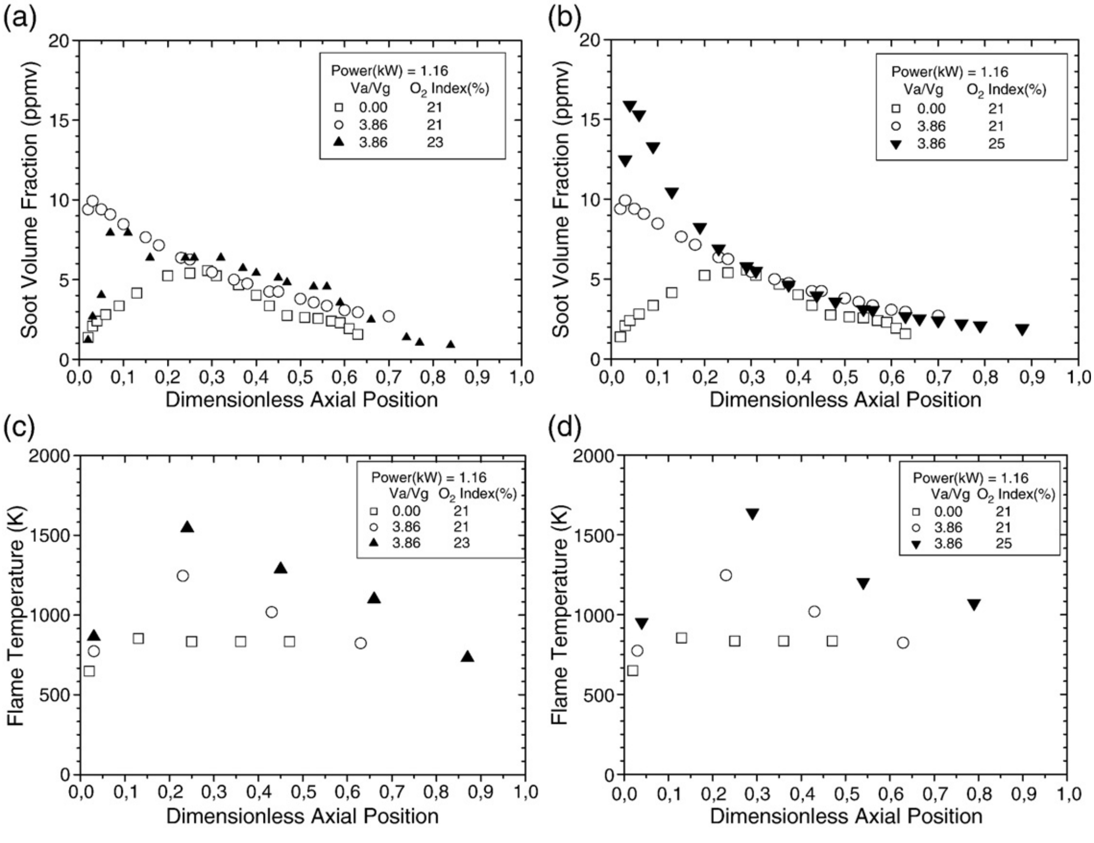

Fig. 6. Soot volume fraction (a,b) and temperature (c,d) along the flame height $V_a/V_g = 0$ and 3.86; power = 1.16 kW.

decreases as the relative velocity decreases, so that the situation with the least fuel-oxidant mixing and, consequently, large soot concentration, is experienced when $V_a - V_g = 0$.

Figs. 3 and 4 show the data obtained for $V_a \approx 0.45V_g$, for burner powers of 0.72 kW and 1.16 kW, respectively.

The results show that the movement of plain air alone ($V_a \neq 0$) could by itself increase soot concentration, relatively to the stagnant air case ($V_a = 0$), due to the decreased air entrainment in the flame.

It can be observed in Fig. 3 a and b that when there was a relative velocity between the fluids as, for example, when $V_a \approx 0.45V_g$, the O_2 enrichments — from 21 to 23% and from 21 to 25% O_2, did not decrease the concentration of soot along the flame axis.

As the burner power was increased from 0.72 kW to 1.16 kW, it can be observed in Fig. 4 a and b that there was an increase in soot concentration for the 23 and 25% O_2 enrichment conditions, comparatively to the stagnant air condition, but this rise was not bigger than for plain air flowing with the same velocity. A possible explanation for it is that the zones of formation and oxidation of soot in the flame are diversely affected by the O_2 enrichment. According to Yaccarino and Glassman [13], changing the oxygen concentration has two different and competing effects on the tendency of a fuel jet to soot. Increasing the oxygen concentration increases the stoichiometric flame temperature which in turn increases the fuel pyrolysis and soot formation rates. However, increasing the oxygen concentration also increases the particle burn-up rate in the vicinity of the flame. The initial effect dominates at high oxygen concentrations and the latter at low concentrations.

In the tests performed the flame temperature increases close to its basis and then stays basically constant along the height. The effect of the increase in the O_2 concentration is smaller than the effect of combustion air moving.

The temperature distributions presented are just indicative of the real flame temperatures. The readings uncertainties were commented by McEnally and Pfefferle [23,24], and Melton et al. [25], and include the influence of soot.

Figs. 3 and 4 show that, in the range of velocities studied, the soot volume fraction increased with temperature up to a certain height in the vertical flame, in which the increase of soot deposited on the thermocouple, introduces a growing thermal resistance that does not allow the temperature readings to increase. The temperatures of the enriched flames should be higher than the non-enriched flames but, in general, were smaller or equal to them.

Figs. 5 and 6 show the results which were obtained for $V_a / V_g = 3.86$, with burner power of 0.72 kW and 1.16 kW.

As the burner power augmented an increase in soot formation was observed, probably caused by the higher temperatures achieved by the flame.

At this air velocity level, a strong increase in soot concentration can be observed at the flame basis when enriched air flow, with oxygen contents of 23 and 25%, was imposed. The soot concentration level increased when the air was enriched, and increased with the oxygen content. This might have been caused by the strong fuel-air mixing, which increases the pyrolysis rate and, as a consequence, soot formation. On the other hand, soot formed in this process is subsequently reduced by oxidation, which is intensified by the oxygen enrichment of the combustion air.

4. Conclusions

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.

This work explored the effects of the oxygen contained in the oxidizer flow and of the combustion air velocity on the concentration of soot along the height of an acetylene laminar diffusion flame produced in a vertical axis burner with a parallel annular coaxial oxidizer flow, such that the acetylene flow was surrounded by the flow of air, or oxygen-enriched air. The applied enrichment levels were 23 and 25% O_2, used in retrofit applications, where only small modifications in the existent equipment are desired.

The results suggest 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 to attend the retrofit load.

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