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Experimental and Numerical Study of the Effects of Oxygen Index on the Radiation Characteristics of Laminar Coflow Diffusion Flames

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Abstract:

The oxygen concentration in the oxidizer stream ($O_2 + N_2$) of laminar coflow ethylene diffusion flames is varied from 17% to 35% in order to study its influence on the flame height, the soot formation/oxidation processes, the smoke point characteristics and the vertical distributions of radiative flux at a distance of 6.85 cm from the flame axis. Measured values for all the investigated parameters are compared with the predictions provided by a numerical model based on a two-equation semi-empirical acetylene-based soot model and on the statistical narrow band correlated-k model to compute thermal radiation. Predictions are in overall reasonable agreement with the experiments for oxygen indexes in the range 19%-35% whereas all the investigated parameters are significantly underestimated for the oxygen index of 17%. Measured flame heights based on CH* emission below, at, or slightly above the smoke point as well as predicted stoichiometric flame

heights are found to be proportional to $\zeta = V_f [D_{\infty} \ln(1 + 1/S)]^{-1} (T_{\infty}/T_{ad})^{0.67}$ where V_f , D_{∞} , S , and T_{ad} are the volumetric flow rate of ethylene, the diffusion coefficient at ambient temperature, the stoichiometric molecular air to fuel ratio, and the adiabatic flame temperature. Soot formation processes are found to increase with the OI, leading to higher values of the maximum soot volume fraction and the peak integrated soot volume fraction. In addition the latter is reached at a nondimensional height (normalized by ζ) of approximately 0.05, regardless of the oxygen index within the investigated range. Two regimes are evidenced: the first regime, occurring for oxygen indexes lower than 25%, is dominated by soot oxidation and is characterized by an enhancement in both the maximum soot volume fraction and the fuel flow rate at the smoke point with the oxygen index. The second regime, occurring for oxygen indexes greater than 25%, is dominated by soot formation; the rate of increase in the maximum soot volume fraction with the oxygen concentration is lower than in the first regime whereas the fuel flow rate at the smoke point decreases. Finally, the peak of radiative flux increases with the oxygen index but its rate of increase is also found to be considerably reduced for oxygen index greater than 25%.

Key words: Laminar Diffusion Flame, Oxygen Index, Flame Height, Soot, Radiation

Nomenclature

A_{op}	coefficient matrix of the Onion-Peeling deconvolution [m]
A_s	soot surface area [m^{-1}]
C_a	agglomeration rate constant
d_s	soot particle diameter [m]
D	diffusion coefficient [$m^2 \cdot s^{-1}$]
$f(r)$	field variable [m^{-1}]
f_s	soot volume fraction [-]
h_f	flame height [m]
k_B	Boltzmann constant [$J \cdot kg^{-1}$]

k_S	absorptive index of the soot [-]
$k_{S,n}$	soot nucleation Arrhenius reaction rate [s^{-1}]
$k_{S,sg}$	soot surface growth Arrhenius reaction rate [$m \cdot s^{-1}$]
m_S	complex index of refraction of the soot ($m_S = n_S - ik_S$) [-]
N	number of discrete field variables, projection location [-]
N_A	Avogadro number [$part \cdot mol^{-1}$]
NC_{min}	number of carbon atoms in the incipient soot particle [-]
n_S	refractive index of the soot [-]
N_S	soot number density per unit mass of mixture [$part \cdot kg^{-1}$]
OI	oxygen index [-]
$P(y)$	projected area [-]
p_i	partial pressure of species i [atm]
$\dot{q}_R''$	radiative flux [$W \cdot m^{-2}$]
R_f	flame radius [m]
r	radial coordinate [m]
S	stoichiometric air–fuel molar ratio [-]
T	temperature [K]
$\dot{V}_f$	volumetric flow rate of fuel [$m^3 \cdot s^{-1}$]
v_z	axial velocity [$m \cdot s^{-1}$]
W_i	molecular weight of the species i [$kg \cdot mol^{-1}$]
x_i	mole fraction of species i [-]
y	axial location
Y_S	soot mass fraction [-]
z	axial coordinate [m]
α_0	zero order regularization parameter

φ_i	collision efficiency factor of species i [-]
$\kappa_{s,\lambda}$	spectral absorption coefficient for soot [m^{-1}]
λ	wavelength [m]
ρ	density [$kg \cdot m^{-3}$]
$\dot{\omega}_n$	reaction rate for the soot nucleation process [$mol \cdot m^{-3} \cdot s^{-1}$]
$\dot{\omega}_{sg}$	reaction rate for the soot surface growth process [$mol \cdot m^{-3} \cdot s^{-1}$]
$\dot{\omega}_{Ns}$	reaction rate for the soot number density [$part \cdot m^{-3} \cdot s^{-1}$]
$\dot{\omega}_{O_2}$	reaction rate for the soot oxidation process by O_2 [$kg \cdot m^{-3} \cdot s^{-1}$]
$\dot{\omega}_{OH}$	reaction rate for the soot oxidation process by OH [$kg \cdot m^{-3} \cdot s^{-1}$]
$\dot{\omega}_{r_s}$	source term for the soot mass fraction [$kg \cdot m^{-3} \cdot s$]

Subscript

∞	ambient condition
ad	adiabatic
ag	agglomeration
f	formation
n	nucleation
OP	Onion-Peeling
ox	oxidation
R	radiation
sg	surface growth
S	soot
SP	smoke point
u	unburnt

1. Introduction

Mass burning, flame spread and fire growth at hazardous scales are driven by thermal radiation from soot and combustion gases, with the continuum thermal radiation from soot accounting usually for the majority of flame radiation. Real fire occurs generally under vitiated conditions. On the other hand, diffusion flames in oxygen-enriched environments have direct applications in fire safety for spacecraft and planetary surface bases [1]. In addition, enhanced ambient oxygen may be used in bench-scale testing to reach radiative fluxes as severe as those encountered in large-scale well-ventilated fires [2]. As a consequence, how the radiative properties of diffusion flames are affected by varying the oxygen concentration in the oxidizer flow (called oxygen index and referred to as OI hereafter) of non-premixed flames is a crucial issue for the purpose of fire safety. This question is also of practical interest in the design of oxy-fuel burners, applied to a variety of industrial processes in order to improve efficiency and pollution characteristics, where the generation of intermediate soot is desirable for increasing radiant heat transfer [3].

Well-defined laminar flames were used in previous works to investigate the influence of varying the OI on soot production. Laminar flames offer more tractable configurations for analysis and experiments [4-6] than the practical turbulent diffusion flames involved in combustion systems and fires. Moreover their use is justified by the similarities existing between laminar and turbulent diffusion flames and the results can be extrapolated to turbulent flames through approximate approaches like the laminar flamelet concept [4-6]. Experimental studies of counterflow diffusion flames showed that soot production was enhanced with increasing the OI [7-11], thermal effects being the main cause for this enhancement [9, 11]. Encouraging numerical predictions in terms of flame structure and soot concentrations were also reported in oxygen-enriched counterflow configurations by using either a detailed soot formation model [9] or two-equation semi-empirical soot models [10, 12, 13].

Axisymmetric laminar diffusion flames were also experimentally and numerically investigated [3, 14-16]. Bennett et al. [14] provided accurate predictions of oxygen-enhanced axisymmetric laminar methane flames by considering a fully coupled fluid flow/detailed chemistry numerical model.

However, soot formation was ignored. Glassman and Vaccarino [15] investigated experimentally the influence of the OI on smoke point characteristics in ethylene diffusion flames. They found that the ethylene flow rate at the smoke point reached a maximum for an OI of 24%. Lee et al. [16] considered a well characterized methane flame, and two other methane diffusion flames with the same volumetric flow rate of fuel and with oxygen enrichments of 50% and 100%, respectively. A reduction in the mass of soot particles within the flame in both enrichment conditions was observed, with larger reduction for the methane/100% oxygen flame. On the other hand, they found that the peak of soot volume fraction followed a non-monotonic evolution with the OI with a maximum for the methane/50% oxygen flame. Zelepouga et al. [3] used the same methane/air flame as Lee et al. [16] as a reference flame in order to study the influence of the addition of acetylene and polycyclic aromatic hydrocarbons on soot formation. Oxygen concentrations of 35, 50, and 100% were also considered. The authors also observed a reduction in the overall mass of soot particles in the three cases whereas the peak of radially integrated soot volume fraction was found to increase between 21 and 35% and to decrease for OIs greater than 35%.

The aim of this paper is twofold: i) to provide experimental measurements and analysis of flame height, soot volume fraction, smoke point characteristics and radiative flux in laminar coflow ethylene diffusion flames by varying the OI from 17% to 35%. To the authors' best knowledge, such a comprehensive study concerning the effects of varying oxygen index on laminar coflow diffusion flames has not been reported in the past. The experimental conditions differ from those involved in [16] by considering a refined range of OI between 21-35% and extend them by dealing with underventilated configurations (OI in the range 17-21%), ii) to assess the capability of a numerical model based on a direct coupling between fluid flow and detailed gas-phase chemistry, a semi-empirical two-equation acetylene-based soot model, and a state of the art radiation model to predict flame structure, soot formation and radiative flux in such configurations. The choice of a semi-empirical two-equation model is justified by the fact that advanced PAH soot models involving a detailed modelling of soot aerosol dynamics (see [17] as an example) remains probably too

computationally expensive to be implemented in turbulent combustion codes in view of application to real fire situations and industrial problems. The experimental conditions present a severe test for the soot model since soot production is modelled over a wide range of flame temperature and residence time and extend those involved in previous numerical studies [9, 10, 12, 13] by considering a multidimensional scenario and a wider range of OI.

The paper is organized as follows: the experimental setup is described in section 2, whereas section 3 is devoted to the presentation of the numerical model. Section 4 discusses the influence of the OI on the flame heights, soot production, and the vertical distributions of radiative flux. Finally, the conclusions drawn from the present study are summarized.

2. Experiments

2.1. Burner and experimental conditions

Figure 1 shows a schematic representation of the experimental setup. The burner, a replica of Gülder apparatus as described in [18], consists of two concentric steel tubes. The fuel flows through the central tube and the oxidizer is injected through the outer one. The inner diameters are respectively 10 mm for the fuel tube and 100 mm for the oxidizer tube. To obtain a uniform and steady exit flow profile, the oxidizer is injected in four points and flows first through a volume filled with 5 mm glass beads and finally through a porous plate.

The experimental conditions are summarized in Table 1. Ethylene is injected via a mass flow controller (Fig. 1). In most of the experiments, the fuel volume flow rate is fixed at $2.35\text{cm}^3/\text{s}$. For each OI, the fuel flow rate and the corresponding flame height at the SP have been also measured and are given in Table 1. The oxidizer is a mixture of oxygen and nitrogen. Changes in OI can be accomplished by three different ways: the oxygen flow rate can be kept constant and the nitrogen flow rate varied; vice-versa; or the total flow rate is kept constant and the concentrations of the constituents varied. The three procedures have been tested and no differences has been observed on the results in accordance with the conclusions drawn by Glassman and Yaccarino [15]. In the present study, the oxygen flow rate is fixed at 20 L/min, and the OI is varied from 17% to 35% by

decreasing the **nitrogen flow rate** from 97.6 L/min to 37.1 L/min. It has been also verified by using the numerical model described in section 3 that, whatever the OI considered, varying the oxidizer velocity by $\pm 20\%$ has a no influence on the flame height and a weak effect on the soot production. To guarantee the oxygen volume fraction in the oxidizer mixture, both nitrogen and oxygen pass through two independent mass flow controllers. Mass flow controllers from BROOKS corp., for fuel and oxidizer, were related to a data acquisition and a command board (Fig. 1).

2.2. *Optical diagnostics*

A schematic of the optical arrangement used for the extinction measurements is shown in Fig. 1. A CNI 1400 mW diode Laser (MLR) operated at the 670 nm line is used as a light source. An optical system including a THORLABS beam expander is used to expand the laser beam to 100 mm, which passes into the flame. The extinction measurements are obtained materializing the beam in a rotating diffuser (frosted glass). A 12-bit charge couple device (CCD) BOBCAT camera (ICL-B1410) was used to acquire the images with a spatial resolution of 1392×1040 pixels. A band-pass interference filter with peak transmission wavelength at 670nm (10nm FWHM) was mounted in front of the CCD camera to avoid the most common interference sources, mainly radiation from the flame.

During each experimental run, the Laser source is synchronized with the video output signal coming from the camera. The procedure consists in obtaining modulated images of emission and absorption from soot particles, as proposed by Jenkins and Hanson [19]. Reference frames are first obtained from the source under no flame condition. These frames (four frames per second) are taken before flame ignition, providing reference intensities. After flame ignition the same synchronization procedure is used. Four frames are obtained when the source is on (i.e. considering both flame and laser emissions), and four frames are obtained when the laser is off (i.e. considering only flame emission).

Another CCD camera was devoted to CH* chemiluminescence measurements. This camera recorded one side-view frame between each laser pulse and was mounted with a narrow band filter centered at 431nm (10nm FWHM). CH* radicals have proven to be central to many hydrocarbon combustion

chemical pathways [20] as they originate from the reaction between O_2 and C_2H . These excited radicals go back to ground-state either through collisional quenching or through spontaneous fluorescence, whose $A2\Delta \rightarrow X2\Pi$ transition occurs at 431.4 nm. Therefore, CH^* chemiluminescence measurements can be directly related to the burning rate of the primary fuel [20]. A PENTAX f/1.4 (H1214M) 12mm focal length lens was mounted on each camera focused at the frosted glass for the extinction measurements and at the burner plane of symmetry for CH^* measurements. The image spatial resolution was determined by the pixel size in each CCD matrix and the detection optics. The overall result for both techniques in the present experiment was that each pixel corresponds to a region of 0.19mm $\times$ 0.19mm.

The radiation emitted by the flame was measured using a Medtherm 64-0.2-15 Schmidt-Boelter gauge fitted with a 150° angle sapphire window in order to eliminate any convective heating readings (cf. Fig 1). The radiometer was placed parallel to the flame axis at a distance of 68.5mm. Measurements were carried out at different heights separated by a distance of 5mm. The displacement of the gauge between two heights was controlled by placing the radiometer in a Thorlabs motorized stage. This procedure guarantees an absolute accuracy of 50 μ m.

2.3. Determination of soot volume fraction radial profile

The objective of this section is to determine the spectral absorption coefficient distribution from the extinction measurements carried out at a wavelength of 670 nm. The soot volume fraction is then obtained from the soot spectral absorption coefficient by assuming that soot particles are within the Rayleigh regime, which gives

$$k_{s,\lambda} = C_A \frac{f_s}{\lambda} \quad \text{with} \quad C_A = \frac{36\pi n_s k_s}{(n_s^2 - k_s^2 + 2)^2 + 4n_s^2 k_s^2} \quad (1)$$

n_s and k_s are determined by using the correlations of Chang and Charalampopoulos [21], which leads to $C_A = 4.23$ at the wavelength of interest of 670nm.

After some geometric analysis, it can be shown that extinction measurements, $P(y) = -\ln[\tau_\lambda(y)]$, are related to the local soot spectral absorption coefficient $f(r) = \kappa_{s,\lambda}(r)$ by using the Abel's integral equation [22]:

$$P(y) = 2 \int_y^{R_f} \frac{f(r)r}{\sqrt{r^2 - y^2}} dr \quad (2)$$

R_f is the flame radius determined from CDD images. The method considered in the present study consists in solving Eq. (2) directly, by using the Onion-Peeling (OP) algorithm [22, 23]. This method is the simplest and most popular method because it naturally takes advantage of the grid provided by the images of projected data obtained directly by the CCD camera. In the OP method the flame field is divided into N evenly spaced annular rings of thickness $\Delta r = R_f/(N - 1/2)$. In the present study N is taken as being equal to 36. The soot volume fraction and thus the absorption coefficient are considered constant inside each ring. Eq. 2 can be then transformed into the following equation [23]:

$$P_i = 2 \sum_{j=i}^{N-1} f_j \int_{r_j - \Delta r/2}^{r_j + \Delta r/2} \frac{r}{\sqrt{r^2 - y_i^2}} dr \quad (3)$$

where $y_i = i\Delta r$, $r_j = j\Delta r$, $P_i = P(y_i)$, and f_j approximates $\kappa_{s,\lambda}(r_j)$. Writing Eq. (3) for every ring results in a set of linear equations that can be written as $A_{OP,ij}f_j = P_i$ or $A_{OP}f = P$ in a matrix form.

The vector, f , contains the unknown field variables, $f^T = \{f_i, i = 0, 1, \dots, N - 1\}$, and the vector, P , contains the projected data set, $P^T = \{P(r_i), i = 0, 1, \dots, N - 1\}$. A_{OP} is a matrix composed of chord lengths that can be readily calculated from simple geometrical considerations:

$$A_{OP,ij} = \begin{cases} 0, & j < i \\ 2\Delta r[(j + 1/2)^{-2} - i^2]^{1/2}, & j = i \\ 2\Delta r[(j + 1/2)^{-2} - i^2]^{1/2} - [(j - 1/2)^{-2} - i^2]^{1/2}, & j > i \end{cases} \quad (4)$$

As A_{OP} is an upper triangular matrix, f can be readily solved using back-substitution. The details of this method can be found in [22, 23] and references given therein. Abel's integral equation is ill-

posed because the solution for $f(r)$ is highly sensitive to small perturbations in the projected data. The problems caused by the ill-posedness of Abel's integral equation are overcome by using the zero order Tikhonov regularization [23]. The method consists in solving the following matrix equation instead of the set of linear equations resulting from Eq. 3:

$$(A_{OP}^T A_{OP} + \alpha_0 L^T L) \tilde{f}^* = A_{OP}^T P \quad (5)$$

where the regularized solution $\tilde{f}^*$ represents an approximation for f . L is a $N \times (N + 1)$ matrix that approximates the derivative operator in the discrete space and α_0 is the regularization parameter. Details about the form of L can be found in [23].

The regularization parameter, α_0 , is adjusted to determine the optimal trade-off between smoothness and accuracy. Daun et al. [23] used the equations of the OP method to test this regularization method. They obtained the suitable value of α_0 by using an L-curve [23], formed by plotting the norms of solution and residual vectors obtained using different values of regularization parameter α_0 . They suggested a value parameter of $\alpha_0 = 0.1$ that regularizes the solution without significantly affecting the solution accuracy [23]. This value is adopted in the present study.

2.4. Determination of smoke point characteristics

The determination of the SP characteristics was established by means of visible changes in the flame shape and was validated by soot extinction measurements over the flame tip. The transitional fuel flow rates between a non-smoking and a smoking flame for each OI were determined by dichotomy obtaining the fuel flow rate at the SP with a resolution of 0.1 cm³/min. This approach can be time-consuming as it may require several tests to find the transitional fuel flow rate at SP depending on the resolution required.

3. Numerical methods

The numerical simulation of the laminar flame flow fields includes the solution of the overall continuity equation, the Navier–Stokes equations in a low Mach number formulation, and transport equations for gas-phase species mass fraction, for the soot mass fraction, for the soot number density

per unit mass of mixture, and for energy [24]. Additionally, a transport equation for the mixture fraction is solved. The conservation equations are solved in axisymmetric cylindrical coordinates using a finite volume method. Correction diffusion velocities in both r - and z -directions were used to ensure that the mass fractions of gaseous species and soot sum to unity. The thermophoretic velocities of soot in both the r - and z -directions were accounted for, as were the interactions between the gas-phase chemistry and the soot chemistry. Diffusion terms in the transport equations are discretized by the central difference and the convection terms by the power law scheme [25]. The SIMPLE algorithm [25] was used to treat the pressure and velocity coupling. Conservation equations of gas-phase species are solved in a fully coupled fashion at each control volume using a direct solver to ensure the convergence process. All other transport equations are solved using the tridiagonal-matrix algorithm. A full chemical kinetic scheme consisting of 70 species and 463 reactions [26] was considered.

The two-equation acetylene-based soot model proposed by Lindstedt and co-workers is used [27]. The source term, $\dot{\omega}_{r_s} = (\dot{\omega}_n + \dot{\omega}_{sg})W_s - \dot{\omega}_{O_2} - \dot{\omega}_{OH}$, for the soot mass fraction, accounts for the contributions of soot nucleation, surface growth, and oxidation by O_2 and OH. The nucleation and surface growth processes are expressed as:

$$C_2H_2 \rightarrow 2C(S) + H_2 \text{ with } \dot{\omega}_n = 2k_{s,n}(T)[C_2H_2] \quad (mol \cdot m^{-3} \cdot s^{-1}) \quad (6)$$

$$C_2H_2 + nC(S) \rightarrow (n+2)C(S) \text{ with } \dot{\omega}_{sg} = 2k_{s,sg}(T)A_s[C_2H_2] \quad (mol \cdot m^{-3} \cdot s^{-1}) \quad (7)$$

where $k_{s,n} = 0.63 \times 10^4 \exp(-21000/T)$ (s^{-1}) and $k_{s,sg} = 0.45 \times 10^3 \exp(-12100/T)$ ($m \cdot s^{-1}$) [27]. It should be noted that a scaling factor of about 0.6 was applied to the expression for $k_{s,sg}$ derived by Lindstedt [27] to match the experimental peaks of soot volume fraction for an OI of 21%. $[C_2H_2]$ ($mol \cdot m^{-3}$) and A_s (m^{-1}) represent the molar concentration of acetylene and the soot surface area, respectively. Soot oxidation by O_2 is given by the Nagle and Strickland-Constable model [28] while soot oxidation by OH is based on the Fenimore and Jones model [29, 30]:

$$C(S) + 0.5O_2 \rightarrow CO \text{ with } \omega_{O_2} = 120 \left[\frac{k_a p_{O_2} \chi_1}{1 + k_z p_{O_2}} + (1 - \chi_1) k_b p_{O_2} \right] A_S (kg \cdot m^{-3} \cdot s^{-1}) \quad (8)$$

$$C(S) + OH \rightarrow CO + H \text{ with } \omega_{OH} = 1.27 \times 10^3 \varphi_{OH} \frac{p_{OH}}{T^{1/2}} A_S (kg \cdot m^{-3} \cdot s^{-1}) \quad (9)$$

φ_{OH} is assumed to be equal to 0.13. χ_1 is expressed as $\chi_1 = (1 + k_t/k_b p_{O_2})^{-1}$. k_a, k_b, k_z and k_t are taken from [28]. The source term, ω_{N_S} , for the soot number density is given by:

$$\omega_{N_S} = \frac{N_A}{N_{C_{min}}} \omega_n - 2C_a \left(\frac{6W_S}{\pi \rho_S} \right)^{1/6} \left(\frac{6k_B T}{\rho_S} \right)^{1/2} \left(\frac{\rho_Y^L}{W_S} \right)^{1/6} (\rho_{N_S})^{11/6} \quad (11)$$

$N_{C_{min}}$ and C_a are taken as being equal to 60 and 9, respectively [27]. The soot density, ρ_S , is taken as being equal to 2000 kg·m⁻³ [27].

The Statistical Narrow Band Correlated-K (SNBCK) Method is used as a radiative property model [31-33]. The integrated radiation intensity on each narrow band (NB) was calculated with a 7-points Gauss-Legendre quadrature scheme. In our calculations, only CO₂ and H₂O are considered as radiatively participating; the contribution of CO is ignored since it is relatively small [34]. The non-gray soot absorption coefficient $\kappa_{S,\lambda}$ is determined by applying Rayleigh's theory to an ensemble of polydisperse soot particles (see Eq. 1). NB k-distributions are generated from the 43 NB database of **Souffiani and Taine** [31] and mixed by using the scheme of Modest and Riazzi [35]. To accelerate the solution a mixed NB database of k-distributions is pre-generated for 15 values of $R = x_{CO_2}/x_{H_2O}^0$, between 0.01 and 2.2, and 23 temperatures in the range between 300 and 2500 K [36]. This database was generated by considering a fixed H₂O concentration of $x_{H_2O}^0 = 0.1$. This procedure implies that the weak dependence of spectral line broadening on the species mole fraction is neglected [36], allowing to express the absorption coefficient of the mixture as:

$$\kappa_{mix}(R, T, x_{H_2O}) = \frac{x_{H_2O}}{x_{H_2O}^0} \kappa_{mix}^0(R, T) \quad (12)$$

where $\kappa_{mix}^0(R_i, T_j)|_{i=1,1.5;j=1,2,3}$ are the tabulated values. For mixtures of arbitrary species mole fraction ratios and temperature, the absorption coefficient is computed from the database by carrying out linear interpolations on R and spline interpolations on T . The radiative transfer equation is solved by the Finite Volume Method (FVM) using the mapping developed by Chui et al. [37] for axisymmetric configurations. Calculations are carried out with an angular mesh with 36×48 control angles.

The overall computational domain is $0.075\text{m} \times 0.08\text{m}$. A preliminary grid refinement study indicated that a non-uniform mesh with 114×200 cells was adequate to achieve grid independent results. A parabolic profile is imposed for the fuel stream inlet while a flat profile is considered for the oxidizer injection. Fuel and oxidizer are injected at ambient temperature (300K), neglecting the effects of fuel pipe preheating by the flame base. Ambient pressure is fixed to 1atm. The fuel inlet velocity corresponds to the fuel flow rate of $2.35\text{cm}^3/\text{s}$.

4. Results and Discussions

4.1. Flame height and smoke point analysis

Flame height is an important characteristic of coflow laminar diffusion flames since flame height measurements can be used to test models of flame structure [38] and to calculate residence times of soot particles [39]. In particular residence times are known to scale with $\sqrt{h_f}$ in axisymmetric buoyant flames.

Flame height measurements are based on CH^* emission. CH^* intensity presents sharp variations near the reaction zone which allows to determine the “flame” contours from a criterion based on the derivative. For all oxygen conditions the optical arrangement remains unchanged in order to keep the same detection criterion. For oxygen indexes of 17% and 19%, CH^* emission is lower which leads to less-defined CH^* contours, explaining the larger error bars observed in these cases in Fig. 2a. As shown in Table 1, for a fuel flow rate of $2.35\text{ cm}^3/\text{s}$, the flame length is reduced as the concentration of oxygen in the oxidizer stream is enhanced, from about 5.30 cm for the ethylene/17% oxygen

flame to about 2.40 cm for the ethylene/35% oxygen flame. For each OI the flame height measurements for $\dot{V}_f = 2.35 \text{ cm}^3/\text{s}$ can be compared with the corresponding flame height at the SP (see Table 1). It appears that all these flames are below the SP, with the exception of the ethylene/33% oxygen flame and the ethylene/35% oxygen flame.

The analysis of Roper [40] predicts that the evolution of the flame length above the port, h_f , for a circular burner diffusion flame is proportional to $\zeta = \dot{V}_f [D_\infty \ln(1 + 1/S)]^{-1} (T_\infty/T_{ad})^{0.67}$. The adiabatic temperature, reported in Table 1, are computed with STANJAN [41]. Figure 2a shows that the experimental flame heights increase proportionally with ζ as expected from the Roper theory [40]. It can be noted that flame height for the ethylene/33% oxygen flame and the ethylene/35% oxygen flame, which are slightly above the SP, follows the same behaviour as the other measurements.

Predicted flame heights correspond to the location along the flame axis where the mixture fraction reaches its stoichiometric value. Figure 2b shows that the computed flame heights increase also linearly with ζ . Nevertheless, they are about 16% higher than the measurements. The heights corresponding to the maximum axial molar concentration of CH^* are also plotted in Fig.2b. A good accordance with the experimental flame heights is observed. This suggests that the model reproduces correctly the flame structure and that the previously-observed differences between the calculations and the experiments may be attributed to, at least partially, different definitions of flame height.

The evolutions of the fuel flow rate and of the flame height at the SP as a function of the OI are plotted in Fig. 2c. Both fuel flow rate and flame height display a non-monotonic behaviour with maximum values occurring for an OI of about 24%. As a consequence, the tendency to release soot is minimal for an OI of 24% and two distinct regimes, resulting from the competition between distinct mechanisms, are evidenced. The characteristics of these two regimes will be discussed in the next subsection. The evolution of the flow rates with OI agree well with that obtained by Glassman and Vaccarino [15] who also used a 1 cm diameter burner in their measurements. Nevertheless, Fig.

2c shows that the actual measurements are systematically 20% lower than those provided in [15], probably due to different procedures used to determine the onset of smoking.

4.2. Soot production

Increasing the OI is expected to affect the soot production through a competition between several mechanisms: on one hand, the resulting increase in flame temperature (see T_{ad} in Table 1) is expected to lead to higher rates for soot nucleation and growth mechanisms, and thus to higher soot concentrations [9, 11]. On the other hand, the increased presence of oxygen promotes oxidative mechanisms and reduces the flame residence times, which tend to decrease the soot concentrations [16]. Computed flame residence times are summarized in Table 1, showing that they are reduced from about 39.8 ms for the ethylene/17% oxygen flame to about 29.8 ms for the ethylene/35% oxygen flame.

4.2.1. Soot volume fraction

Figures 3a-c show both measured and predicted radial profiles of soot volume fraction for OIs of 21%, 25%, and 29%. For each OI, three axial locations were chosen for the comparison: low in the flame where soot particles undergo nucleation and growth, near the middle of the flame where the maximum soot volume fraction occurs, and higher in the flame where soot is oxidized. Measurements show that soot is first observed to form in an annular region inside the main reaction zone. The peak of soot within this annulus initially increases with height and then decreases as soot is oxidized higher up in the flame. As the height increases the location of the peak is shifted toward the axis since soot particles are transported inwards by the flow. This behavior is typical in buoyant laminar ‘candle-like’ flames. The model captures most of the experimentally-observed trends and the predicted distributions of soot volume fraction are in qualitative agreement with the experiments. The major discrepancies between the predictions and the experiments are: (i) the peak of soot is over-predicted on the whole throughout the flames. The highest relative error is of 34% and is observed at a height of 22 mm for an OI of 29% (see Fig. 3c). (ii) The predicted radial location of the peak is generally shifted outwards as compared with the experiments. This can be clearly observed at

the middle height (28 mm) for the OI of 21%. It can be also noted that the overall agreement concerning this point is improved as the OI increases.

The experimental uncertainties related to soot volume fraction measurements have been estimated by using the error analysis developed in [42]. The two main contributions come from the uncertainties on the complex index of refraction of soot and on the use of non-monochromatic light for extinction measurements, leading to a systematic error of about 26% as illustrated by the error bars in Fig. 3d.

Figure 3d shows also that the evolution of the experimental maximum soot volume fraction, $f_{s,max}$, as a function of the OI, clearly presents two distinct regimes: the first regime is observed for $17\% \leq OI \leq 25\%$ whereas the second occurs for $OI \geq 25\%$. In both cases $f_{s,max}$ increases linearly with OI, but the rate of increase is lower by a factor of about 2.5 in the latter case. These two regimes can be related to those observed for SP data and some characteristics of these regimes can be deduced from Figs. 2b and 3d:

- Fig 3d shows that, for both regimes, the soot formation rate, $\dot{\omega}_f$, increases more rapidly with the OI than the reduction in residence time, leading to higher values of $f_{s,max} \cong \dot{\omega}_f t_{res,f}$ (Fig. 3d). As a consequence, the change in slope observed for $f_{s,max}$ for OI=25% results from a competition between an enhancement in formation mechanisms and a reduction in residence time. Numerical results show that the slowdown in soot volume fraction increases in the second regime is also related to the scrubbing effect of soot forming species (C_2H_2). As more soot is formed, the concentration of C_2H_2 decreases. Further increase in soot can no longer be supported at the same rate as C_2H_2 concentration decreases.

- In the first regime (Regime I) soot oxidation ($\cong \dot{\omega}_{ox} t_{res,ox}$) increases more rapidly with the OI than soot formation ($\cong \dot{\omega}_f t_{res,f}$). On one hand this explains why an increase in fuel flow rate is required to reach the SP as the OI is enhanced (see Table 1 and Fig. 2b). On the other hand this implies that, the increase in the soot oxidation rate, $\dot{\omega}_{ox}$, with OI is also faster than the reduction in time available for oxidation ($t_{res,ox}$).

- In the second regime (Regime II) soot formation mechanisms ($\cong \dot{\omega}_f \tau_{res,f}$) increases more rapidly with the OI than soot oxidation mechanisms ($\cong \dot{\omega}_{ox} \tau_{res,ox}$). This explains why, for a fixed fuel flow rate of 2.35 cm³/s, the flames become smoking for OIs greater than 33% (see Table 1) and why the fuel flow rate at the SP decreases as the OI increases (see Fig. 2b).

The predicted maximum soot volume fraction is also plotted for comparison. Flame blow-off was found to occur for an OI slightly lower than 17% which may explain the significant discrepancies observed between the calculations and the experiments for an OI of 17%. In this case the maximum soot volume fraction is underestimated by a factor of 2.84. The numerical model reproduces the change in slopes observed experimentally for an OI of 25% but the rates of increase in $f_{s,max}$ are significantly overestimated in the two regimes. As a consequence the values of $f_{s,max}$ become significantly overpredicted by factors of 1.62 and 1.67 for the OI of 33 and 35%, respectively. For OI of 21, 25, and 29% this factor is of 0.96, 1.16, and 1.36, respectively.

4.2.2. Integrated soot volume fraction

In order to assess the influence of the OI on the overall soot production, the integrated soot volume fraction, defined as $\beta = 2\pi \int_0^\infty r f_s dr$, is considered. Figure 4a shows the experimental integrated soot volume fraction as a function of the height normalised by ζ . As the normalized height increases the integrated soot volume fraction increases first in the region of soot nucleation and growth, reaches a maximum, and then decreases as soot particles are oxidized. Figure 4a indicates that the normalisation of the height by ζ provides a reasonably good scaling in terms of location of the maximum of integrated soot volume fraction which is reached in the range of 0.043-0.056. This scaling extends that proposed by Santoro et al. for ethylene/air flames [43]. The experimental results in Fig. 4a confirm that the soot formation process occur with higher rates as the OI is increased. Predicted integrated soot volume fractions for the OIs of 21%, 29% and 35% are compared with the experiments in Fig. 4b. Predictions are in reasonable agreement with the experimental data.

The peak of integrated soot volume fraction, β_{max} , as well as the corresponding normalized height at which it occurs, z_{max}/ζ , are plotted as a function of the OI in Fig. 4c. As discussed previously, z_{max}/ζ is in the range 0.043-0.056 and decreases on the whole with the OI. This trend is well reproduced by the numerical model. Both measured and predicted β_{max} increase linearly with the OI over the entire range of OIs considered. This monotonic increase in β_{max} for $21\% \leq OI \leq 35\%$ is consistent with the experimental data reported by Zelapouga et al. [3] for methane flames who observed a higher peak of radially integrated soot volume fraction for an OI of 35% than for an OI of 21%. Figure 4c shows that predicted β_{max} are in satisfactory agreement with the measurements for OI in the range from 21% to 35% with a maximum relative error of 34% for an OI of 21%. For the other OI in this range the relative error does not exceed 16%. For an OI of 17% β_{max} is underestimated by a factor of about 3.16. As mentioned in the previous section, this large underestimation may be related to combustion conditions occurring close to flame blow-off.

4.3. Vertical distribution of radiative flux

Figure 5a shows the vertical distributions of radiative flux at a distance of 6.85cm from the flame axis for OIs of 19%, 21%, 25% and 31%. As the OI increases, the radiative fluxes become higher which can be directly related to the increase in flame temperature and in soot production as discussed in the previous section. The peaks of radiative flux are shifted toward the burner which is consistent with the observed reduction in flame heights. Predicted radiative fluxes are in satisfactory overall agreement with the experimental data. The location of the peak is well captured as well as the changes in the shape of the distribution as the OI is increased.

Figure 5b shows the evolution of the measured peak of radiative flux as a function of the OI. It increases rapidly between an OI of 17% and an OI of 25% from about 370W/m² to about 560W/m². On the other hand, its rate of increase is considerably reduced for $OI \geq 25\%$. This behaviour is well correlated with that observed for the maximum soot volume fraction. Predicted peaks of radiative flux are within $16\% \text{ for } 19\% \leq OI \leq 35\%$ and the asymptotic behaviour observed experimentally

for OI greater than 25% is well reproduced by the model. On the other hand, for $OI = 17\%$ the model underpredicts the experiments by about 30%.

5. Summary

Experimental and numerical investigations were conducted to characterize the effects of the addition of oxygen in the oxidizer stream of laminar coflow ethylene diffusion flames on flame heights, smoke point characteristics, soot production and radiative losses. Oxygen indexes were varied between 17% and 35%. The main results of this study can be summarized as follows:

- 1) The influence on OI on flame heights based on CH^* emission below, at, or slightly above the SP as well as predicted stoichiometric flame heights can be analyzed by using the theory of Roper.
- 2) The peak of integrated soot volume fraction occurs for z/ξ in the range 0.043-0.056, regardless of the value of OI within the range of investigation.
- 3) The evolution of soot formation process with the OI results from a competition between an enhancement of soot formation rate due to higher temperature and a reduction in residence time due to shorter flame length. For the investigated range of OI , the first mechanism dominates and soot formation is found to increase with OI , leading to higher values of the maximum soot volume fraction and of the peak of integrated soot volume fraction. However, the rate of increase in the maximum soot volume fraction is considerably reduced for OI greater than 25%.
- 4) The analysis of smoke point characteristics evidences the existence of two distinct regimes separated also at a value of OI of about 24-25%: the first regime ($OI \leq 25\%$) is characterized by an increase in both fuel flow rate and flame height at the SP with the OI and is dominated by soot oxidation that increases more rapidly with OI than soot formation. The second regime ($OI \geq 25\%$) is dominated by soot formation processes that increase more rapidly with the OI than soot oxidation and both fuel flow rate and flame heights at the smoke point are reduced.
- 5) Radiative fluxes are also enhanced as the OI increases but the rate of increase is also considerably reduced for OI greater than 25%.

6) The numerical model reproduces most of the experimentally-observed trends. The effects of OI on flame structure, integrated soot volume fraction and radiative flux are rather well captured and a reasonable quantitative agreement is observed for OI in the range of 19-35%. However the numerical model underestimates significantly these parameters for an OI of 17% and overestimates severely the rate of increase in maximum soot volume fraction with the OI in the two regimes.

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Table 1. Experimental conditions. The residence times, $t_{res} = \int_0^{h_f} dz/v_z (r = 0, z)$ were deduced from the numerical results. T_{ad} and h_f represent the adiabatic flame temperatures and the measured flame heights, respectively. The last two columns refer to smoke point characteristics.

Oxidizer composition	$\dot{V}_f$ (cm ³ /s)	$\dot{V}_{ox}$ (l/min)	T_{ad} (K)	h_f (cm)	t_{res} (ms)	$\dot{V}_{f,SP}$ (cm ³ /s)	$h_{f,SP}$ (cm)
17%O ₂ + 83%N ₂	2.345	117.6	2169.72	5.30	39.8	2.71	5.91
19%O ₂ + 81%N ₂	-	105.3	2295.54	4.64	38.1	2.98	5.88
21%O ₂ + 79%N ₂	-	95.2	2402.61	4.41	36.3	3.38	6.27
23%O ₂ + 77%N ₂	-	87	2494.65	4.04	35.1	3.55	6.81
25%O ₂ + 75%N ₂	-	80	2574.92	3.58	34.4	3.52	6.22
27%O ₂ + 73%N ₂	-	74.1	2645.91	3.26	33.3	3.18	5.04
29%O ₂ + 71%N ₂	-	69	2709.49	2.99	31.0	2.81	3.98
31%O ₂ + 69%N ₂	-	64.5	2767.06	2.75	31.7	2.41	3.12
33%O ₂ + 67%N ₂	-	60.6	2819.67	2.59	30.2	2.14	2.56
35%O ₂ + 65%N ₂	-	57.1	2868.12	2.40	29.8	1.84	2.07

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Figure 1: Schematic of the experimental apparatus.

Figure 2: a) Measured flame heights as a function of ζ . Open and filled squares correspond to a fuel flow rate of $2.35\text{cm}^3/\text{s}$ and are below and above the smoke point, respectively. Grey deltas correspond to smoke point flame heights, b) Experimental correlation (line), predicted stoichiometric flame heights (filled square) and heights at which the axial molar concentration of CH^* is maximal (open delta) as a function of ζ , c) Experimental flame height (square) and fuel flow rate (delta) at the smoke point as a function of the OI. The continuous line corresponds to the fuel flow rates at the smoke point measured by Glassman and Yaccarino [15].

Figure 3: Radial profiles of soot volume fraction for three OIs: a) 21%, b) 25%, and c) 29%. Symbols and solid lines correspond to the experimental data and the numerical predictions, respectively. d) Maximum soot volume fraction as a function of the OI. Filled and open symbols correspond to the experimental data and the numerical predictions, respectively. The linear fits of experimental $f_{s,max}$ for $OI \leq 25\%$ ($f_{s,max} = 0.278 \times OI - 0.355; R^2 = 0.94$) and for $OI \geq 25\%$ ($f_{s,max} = 0.108 \times OI + 4.126; R^2 = 0.95$) are also plotted. The linear fits of calculated $f_{s,max}$ for $OI \leq 25\%$ ($f_{s,max} = 0.781 \times OI - 11.58; R^2 \approx 1.00$) and for $OI \geq 25\%$ ($f_{s,max} = 0.525 \times OI - 5.26; R^2 \approx 1.00$) are also plotted.

Figure 4: Integrated soot volume fraction: a) vertical distribution as a function of the height normalized by ζ , and b) z_{max}/ζ (square) and β_{max} (delta) as a function of the OI. Filled and open symbols correspond to the experimental data and the numerical predictions, respectively. The dashed line corresponds to the averaged values of z_{max}/ζ over the experimental data ($z_{max}/\zeta = 0.05$).

Figure 5: Radiative flux at a distance of 6.85cm from the flame axis: a) vertical distribution for OIs of 19%, 21%, 25% and 31%, and b) evolution of the peak of vertical radiative flux as a function of

the OI and as function of $f_{S,max}$. Filled and open symbols correspond to the experimental data and the numerical predictions respectively.

Figure 1.

Figure 2.

Figure 3.

Figure 4.

Figure 5.