Influence of radiative property models on soot production in laminar coflow ethylene diffusion flames

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Abstract. Two axisymmetric laminar coflow non-smoking and smoking ethylene diffusion flames are studied numerically in order to assess the influence of different radiative property models on the soot formation and oxidation processes. Simulations are carried out by considering the Steady Laminar Flamelet (SLF) concept and a modified two-equation acetylene-based model to describe the soot nucleation, surface growth and oxidation processes. Several radiative property models are considered: the simple Optically Thin Approximation (OTA), the Weighted-Sum of Grey-Gases (WSGG), the Grey-Wide-Band model (GWB), the Statistical Narrow Band Correlated-k model (SNBCK) and the Full Spectrum Correlated-k model (FSCK). Comparisons between calculations carried out with the SNBCK model and experimental data show a reasonable agreement. Model results show that the choice of the radiative property models influence largely the soot prediction, especially in the upper part of the flame where oxidation occurs. Simulations show that the reabsorption of spectral gas and soot is an important feature and thus the commonly used OTA or grey models introduce large discrepancies. The GWB model leads to improved solutions, but it should be avoided if accurate predictions are desired. The FSCK provides equivalent results as compared to the SNBCK model with a substantial gain in CPU time.

1. Introduction
An important topic of research is to predict the sooting characteristics of a specific fuel or understanding how the soot is produced and consumed in order to effectively control it. Great efforts have been done in understanding the mechanisms of soot inception, growth and oxidation. Detailed models involving several hundreds of elementary reactions and comprehensive models for both the soot-particle dynamics and the soot kinetics were successfully applied to compute ethylene coflow diffusion flames [1]. Simplified semi-empirical models [2-6], describing the main features of soot formation processes, were also developed. One characteristic that is well recognized is the important coupling between soot kinetics and thermal radiation. For laminar coflow diffusion flames the radiative heat transfer is of vital importance to predict the soot kinetics, since it significantly affects the temperature and therefore the complex reactions associated with the soot processes. This coupling
is generally treated in a simple manner using the Optically Thin Approximation (OTA) [7–9], despite some studies which concluded that a detailed modelling of thermal radiation is necessary for accurately predict the flame structure, species concentrations and formation of soot [10–13]. In one of these studies, Liu et al. [11] showed that the use of the OTA induces significant temperature underpredictions in the upper part of coflow laminar ethylene diffusion flames, affecting directly the prediction of soot volume fractions. Despite these efforts, an extensive comparison of several radiative properly models on soot predictions has not been reported.

The aim of the present study is to assess the influence of different radiative property models over the soot formation/oxidation processes in axisymmetric laminar coflow ethylene flames. To accomplish this task, a Non-Smoking (NS) and a Smoking (S) flame, previously investigated experimentally by Santoro et al. [14] and Megaridis and Dobbins [15, 16], are simulated by considering different radiative property models. The selection of the radiative models follows a bibliographic survey, choosing those that are most representative and commonly used. The Steady Laminar Flamelet (SLF) model is used as combustion model and a modified version of the two-equation soot model proposed by Leung et al. [2] is employed to predict soot nucleation, surface growth and oxidation.

2. Model formulation and numerical method

The numerical simulation of the laminar flames includes the solution of the overall continuity equation, the Navier-Stokes equations in a low Mach number formulation and transport equations for the mixture fraction ($\zeta$), the soot mass fraction ($Y_S$), the soot number density per unit mass of mixture ($N_S$) and the total enthalpy ($h$).

Chemical interactions are modelled by using the Steady Laminar Flamelet concept. This model allows obtaining state relationships for the scalar properties of the medium as function of the mixture fraction, the scalar dissipation rate ($\chi = 2D_{th} \nabla^2 \zeta$) and an enthalpy defect parameter ($X_r = (h - h_{ad})/(h_u - h_{ad})$). In the last expressions $D_{th}$ is the thermal diffusivity and $h_{ad}$ and $h_u$ are the adiabatic and unburned enthalpies, respectively. The flamelet library was generated by using the GRI-Mech 3.0 chemical reaction mechanisms [17]. The enthalpy defect parameterization is introduced by using the methodology described by Carbonell et al. [18].

2.1. Soot model

The soot kinetics is modelled using a semi-empirical acetylene-based two-equation model proposed by Leung et al. [2] and modified later by Liu et al. [11]. This model accounts for the contributions of soot nucleation, surface growth and oxidation. Thermophoretic velocities of soot are expressed as $V_{Th,i} = -0.55\mu/(\rho T) \partial T/\partial x_i$, where $\mu$, $\rho$ and $T$ represent the viscosity, the density and the temperature of the gas mixture, respectively. $x_i = r, z$ represents the radial and axial directions.

Acetylene ($C_2H_2$) is assumed to be the main gaseous species involved in the nucleation ($r_1$) and surface growth ($r_2$) processes. These reactions and their reaction rates are assumed as follows:

$$C_2H_2 \rightarrow 2C(S) + H_2 \quad \text{with} \quad r_1 = 2 \times 1.7 \exp(-7548/T)[C_2H_2] \quad (1)$$

$$C_2H_2 + nC(S) \rightarrow (n + 2)C(S) + H_2 \quad \text{with} \quad r_2 = 2 \times 5.0 \exp(-6038/T)A_S[C_2H_2] \quad (2)$$

In these reactions $C(S)$ represents carbon in its solid form. $A_S = \pi d_S^2 (\rho N_S)$ and $[C_2H_2]$ represent the total soot surface area and the molar concentration of acetylene. $d_S = (6 Y_S/\pi N_S \rho_S)^{1/3}$ is the diameter of the (assumed) spherical soot particles and $\rho_S = 2000$ kg/m$^3$ is the soot density.

The soot oxidation process is attributed to OH, $O_2$ and O reactions. Soot oxidation by $O_2$ is given by the NSC model, while soot oxidation by OH and O is based on the Fenimore and Jones model. These reactions and their reaction rates are assumed as follows:

$$0.5O_2 + C(S) \rightarrow CO \quad \text{with} \quad r_3 = 120 \left[ k_a X_{O_2} \frac{1}{1 + k_x X_{O_2}} + k_p X_{O_2} (1 - x_1) \right] A_S f_{O_2} \quad (3)$$

where $k_a$, $k_p$, $k_x$ are the reaction rate constants and $x_1$ is the mole fraction of acetylene.
\[ \text{OH} + C(S) \rightarrow CO + H \quad \text{with} \quad r_4 = 1270 \varphi_{OH} T^{-1/2} X_{OH} A_{sOH} \quad (4) \]

\[ O + C(S) \rightarrow CO \quad \text{with} \quad r_5 = 55.4 \varphi_0 T^{-1/2} X_0 A_S \quad (5) \]

In the latter expressions \( X_i \) represents the mole fraction of the \( i \)-th species, whereas the collision efficiency factors, \( \varphi_{OH} \) and \( \varphi_0 \), are fixed to 0.05 and 0.2 respectively. \( X_1 \) is expressed as 
\[ X_1 = (1 + k_t / k_b X_{O_2})^{-1} \]
with \( k_a, k_b, k_z \) and \( k_t \) defined from [19]. The rate constants used in \( r_4 \) and \( r_5 \) are obtained from [20] and [21] respectively. The commonly used NSC model and the OH soot oxidation rates are modified in this study by introducing two temperature dependent correction factors proposed by Liu et al. [11].

\[ f_{O_2} = \left( 1 + \exp\left[ \frac{-(T - 1650)}{80} \right] \right)^{-1} \quad (6) \]

\[ f_{OH} = \begin{cases} 
0.1824\left( 1 + \exp\left[ \frac{-(T - 1600)}{85} \right] \right)^{-1}, & T \leq 1600 \text{ K} \\
(1 + \exp\left[ \frac{-(T - 1675)}{50} \right])^{-1}, & 1600 \text{ K} < T < 1675 \text{ K} \\
(1 + \exp\left[ \frac{-(T - 1650)}{80} \right])^{-1}, & T \geq 1675 \text{ K} 
\end{cases} \quad (7) \]

Finally, the source terms of the transport equations for the soot number density \( (S_N) \) and soot mass fraction \( (S_m) \) are defined as

\[ S_N = \frac{N_A}{c_{min}} r_1 \quad \text{and} \quad S_m = \left( r_1 + r_2 \right) W_S - r_3 - r_4 - r_5, \quad (8) \]

where \( N_A \) is the Avogadro’s number \((6.022 \times 10^{23}\) part/mol), \( c_{min} \) is the number of carbon atoms in the incipient soot particle \((700)\) and \( W_S \) is the soot molecular weight \((12.011 \text{ g mol}^{-1})\). In the source term of the soot number density (see equation (8)), the agglomeration process is neglected based on the experimental data of Megaridis and Dobbins [15, 16], which established that soot aggregates consist of more or less identical primary particles and obtained an almost constant primary soot particle density in the soot growth region. Therefore the primary soot particle coalescence can be neglected [10].

2.2. Radiation models

Several radiative property models are assessed in the present study. A brief description of every model is presented below. More details can be found in [22].

In this study the soot particles are assumed to attain the Rayleigh scattering limit. In this regime the scattering is negligible and the spectral soot absorption coefficient, at wavelength \( \lambda \), is given by

\[ \kappa_{s,\lambda} = 5.5 \frac{f_S}{\lambda}. \]

The soot volume fraction \( f_S \) is defined as \( f_S = \rho Y_S / \rho_S \). Depending on the radiative model, a Planck-averaged absorption coefficient for the soot can be required. This coefficient is given by the following expression [23]

\[ \kappa_{s,\text{Planck}} = 5.5 \left( 3.83 / 14388 \times 10^{-6} \right) f_S T \quad (9) \]

In this study only \( \text{CO}_2 \) and \( \text{H}_2\text{O} \) are considered as the participating gases, neglecting the other combustion species. A previous study showed that neglecting the contribution of CO has only a minor influence on the divergence of the radiative flux [24].

2.2.1. Optically Thin Approximation model (OTA). This model is widely used as a first approximation to estimate the divergence of the radiative flux. It neglects the gas and soot reabsorption, assuming that the medium is optically thin. In the present study the Planck-mean absorption coefficients for the combustion gases are evaluated from the correlations given in [25], whereas the Planck-mean absorption coefficient for the soot is computed from equation (9).

2.2.2. Planck model. This grey model solves the Radiative Transfer Equation (RTE) with the Planck-mean absorption coefficients computed as described by the OTA.

2.2.3. Statistical Narrow Band Correlated-k model (SNBCK). This model is based on the assumption that the spectrum can be divided into narrow bands (NB), in which the blackbody intensity and soot
properties can be considered as constant. The erratic spectral absorption coefficient is then reordered on each NB to form a monotonically increasing function, called the k-distribution, which can be easily integrated. A 7 points Gauss-Legendre quadrature scheme is used for this integration. NB k-distributions are generated from the 43 NB database of Soufiani and Taine [26] and mixed using the scheme of Modest and Riazzi [27]. To speed up the solution a mixed NB database of k-distributions is pre-generated for 15 values of \( R = X_{CO_2}/X_{H_2O} \), between 0.01 and 2.2, and 23 temperatures in the range between 300 and 2500 K. This database was generated by considering a fixed H_2O concentration of \( X_{H_2O} = 0.1 \). This procedure implies that the weak dependence of spectral line broadening on the species mole fraction is neglected [28], allowing to express the absorption coefficient of the mixture as:

\[
\kappa_{mix}(R, T, X_{H_2O}) = \frac{X_{H_2O}}{X_{H_2O}^0}\kappa_{mix}^0(R, T)
\]

where \( \kappa_{mix}^0(R_i, T) = \kappa_{H_2O}(X_{H_2O}^0, T) + \kappa_{CO_2}(R_i \times X_{H_2O}^0, T) \) 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 \( T \). Introducing this database has a minor influence as compared to direct SNBCK calculations. Soot absorption coefficients for each NB are estimated as \( \kappa_{mix} = 5.5 f_s(r, z) \), where \( r \) is an effective absorption coefficient and \( f_s \) is the Planck-mean absorption coefficient. \( f_s \) is determined to match the total radiative intensity leaving a uniform medium of path length \( l \) bounded by black walls at 0 K. The path length is taken as \( l = 2\tau_f = 8.13(D_0^2 l_f / g)^{1/4} \), where \( \tau_f \), \( D_0 \), \( l_f \) and \( g \) are the characteristic radius of the flame, the diffusion coefficient, the flame height and the acceleration of gravity, respectively. This model is denoted as SNBCKgs hereafter.

2.2.4. The Weighted-Sum of Grey Gases (WSGG). This model is based on expressing the total gas emissivity as the weighted-sum emissivity of four grey gases. In this study a non-grey formulation of this model is considered, working directly with the absorption coefficients and emission weighted factors provided by the model. Then the RTE is solved directly for the four “grey gases”. Emission weighted factors and absorption coefficients for each grey gas are calculated from the polynomial relationships of Smith et al. [29]. The coefficients involved in the polynomials are tabulated for particular ratios of partial pressure \( p_{H_2O}/p_{CO_2} \). In the present study, parameters tabulated for \( p_{H_2O}/p_{CO_2} = 1 \) are used, corresponding to the stoichiometric ratio for the complete combustion of ethylene in air. The soot absorption coefficient is considered as grey (equation (9)) in this model.

2.2.5. Grey-Wide-Band model (GWB). This model considers that the spectrum can be divided into 6 bands and in each one a Planck-mean absorption coefficient is considered. This model is implemented in the CFD code Fire Dynamic Simulator (FDS) [30]. As in FDS, the absorption coefficient of each wide band is computed from the RADCAL database. It should be noted that Planck coefficients computed from RADCAL and from the database of Soufiani and Taine are in close agreement [31, 32]. As a consequence, the use of these different databases to compute radiative properties should not be a great source of discrepancies. In addition, the part of the spectrum considered in the RADCAL database has been restricted to be consistent with that of Soufiani and Taine. In FDS a grey formulation is also implemented. This radiative model was used for the calibration of new soot models based on the concept of the smoke point [5, 6]. In this case the absorption coefficient is computed as \( \kappa = \min(\kappa_e, \kappa_p) \), where \( \kappa_e \) is an effective absorption coefficient and \( \kappa_p \) the Planck-mean absorption coefficient. \( \kappa_e \) is determined to match the total radiative intensity leaving a uniform medium of path length \( l \) bounded by black walls at 0 K. The path length is taken as \( l = 2\tau_f = 8.13(D_0^2 l_f / g)^{1/4} \), where \( \tau_f \), \( D_0 \), \( l_f \) and \( g \) are the characteristic radius of the flame, the diffusion coefficient, the flame height and the acceleration of gravity, respectively. This model is denoted as GWBg hereafter.
2.2.6. **Full Spectrum Correlated-k model (FSCK).** This model is an extension of the SNBCK model, in which the reordering is carried out into the entire spectrum at once. This allows an important decrease in the number of resolutions of the RTE and then in the computational resources. The Full Spectrum (FS) k-distributions are assembled from the same 43 mixed-NB database used for the SNBCK model. The assembly scheme chosen is the one presented by Modest and Riazzi [27]. Two different procedures to generate the FS k-distributions are implemented and compared in this study: in the first, non-grey soot is added to the pre-databased mixed-NB k-distributions and then the k-distributions are assembled (FSCK MNB). In the second, mixed-FS k-distributions are pre-databased (assembled from the mixed-NB database) and then a Planck-mean soot absorption coefficient is directly added (FSCK MFS). The latter procedure is much more efficient on a computational point of view as the assembly procedure is not carried out during the sequence of calculations, but this formulation requires considering soot particles as grey. For both formulations the integration of the FS k-distributions is carried out considering a 7 points Gauss-Legendre quadrature scheme. One characteristic of this model is the need to define a reference state \( (T, X_{CO_2}, X_{H_2O}, f_s)_{ref} \) to treat non-homogeneous non-isothermal conditions. A simple average over the flame volume is considered for the gas species and soot volume fraction, while an emission-weighted temperature is used as the reference temperature [22, 24].

2.3. **Numerical method**

The governing equations are discretized and solved in cylindrical coordinates using a control volume method on a staggered grid. For the convective terms the ULTRASHARP approach is applied, while a second-order central difference scheme is used for the diffusion terms. The pressure-velocity coupling is solved using the Iterative PISO algorithm. The tridiagonal matrix algorithm is used to solve the transport equations. The overall computational domain is \( 4.5 \) \( cm \) \( r \) \( \times \) \( 13.12 \) \( cm \) \( z \). A non-uniform mesh with \( 89(\ r \) \times \) \( 400(\ z \) \) cells is used, with a mesh size of \( 0.1875 } cm \) \times \( 0.375 } cm \) in the flaming zone. Further refinement of the computational mesh has negligible effects on the results. The radiative transfer equation is solved by the finite volume method using the special mapping developed by Chui et al. for axisymmetric configurations [33]. Computations are carried out using the first-order UPWIND spatial discretization scheme and an angular mesh with 12×16 control angles. Convergence is achieved when the maximum relative error for all the computed variables is lower than 10^{-4}.

3. **Results and discussion**

The different radiative property models have been used to simulate the Smoking (S) and Non-Smoking (NS) ethylene coflow flames studied experimentally by Santoro et al. [14], Megaridis and Dobbins [15, 16], Kennedy et al. [3] and Smyth [34]. Experiments used a burner consisting in two concentric tubes of 11.1mm and 101.6mm inner diameter respectively. Pure ethylene flows through the inner tube, while air passes through the annular region between the two tubes. The mean velocities for the fuel and air streams are of 3.98cm.s^{-1} and 8.9cm.s^{-1} for the NS flame and 5.05cm.s^{-1} and 13.3cm.s^{-1} for the S flame. A parabolic profile is imposed for the fuel injection zone while a flat profile is considered for the oxidant injection zone. A small solid wall of 0.55 mm separates both injection zones. Fuel and oxidizer are injected at ambient temperature, neglecting the effects of preheating. Ambient temperature and pressure are fixed to 300 K and 1atm.

3.1. **Comparison with available data**

A first comparison is carried out in order to assess the main characteristics of the simulations and have an overall idea of the accuracy obtained. Computations carried out with the SNBCK model are compared to the experimental data of Santoro et al. [14] and Megaridis and Dobbins [15, 16]. Figures 1a and 1b present the radial profiles of temperature and soot volume fraction at three heights of the NS flame. Overall predictions present an acceptable agreement with the measurements, especially for the temperature. The locations of the temperature peaks as well as their magnitudes are well predicted at \( z = 7 \) mm and 20 mm. The preheating of the fuel caused by conduction with the pipe flow is disregarded.
in this study, which may explain the underprediction observed for the centreline values (see figure 1a).

Figure 1b shows that the predicted profiles of soot volume fraction at these two heights present wider peaks, with values slightly lower than the experimental values. The results at $z = 70$ mm show that the numerical simulations still predict the peaks of soot volume fraction and temperature in an annular region whereas the experiments exhibit maximum values along the centreline (see figures 1a and 1b).

Figure 1c presents the soot primary particle number density and soot primary particle diameter along the path line exhibiting maximum soot volume fraction for both flames. The experimental data show that the values of soot primary particle number density in both flames are quite similar in most of the soot growth region. Numerical results are in reasonable agreement with the experimental data for both flames. The soot primary particle diameter also presents the same behaviour as observed experimentally. For both flames an excellent agreement between simulations and experiments is achieved within the first 4cm. Above this height the diameter is somewhat overpredicted, showing that the model underestimate the oxidation process. Figure 1d presents the integrated soot volume fraction along the flame height, experimental values from Santoro et al. [14].

Figure 1. Comparisons between experimental data and predictions obtained with the SNBCK model. Radial profiles of (a) temperature and (b) soot volume fraction at different heights for the NS flame, experimental values from Santoro et al. [14]; (c) soot primary particle number density and soot primary particle diameter along the path line exhibiting the maximum soot volume fraction for the NS and S flames, experimental values from Megaridis and Dobbins [15, 16]; and (d) integrated soot volume fraction along the flame height, experimental values from Santoro et al. [14].
A comparison between the experiments and the numerical results shows that the integrated soot volume fraction is slightly underestimated in the lower part of both flames. At the peaks this tendency is inverted and a more important overestimation is observed. The predicted peak locations are shifted by a few millimetres away from the burner as compared with the experiments.

3.2. Influence of the radiative models

The accuracy of the different radiative property models is assessed by using the SNBCK model as reference for the S flame. It is important to mention that only Line-by-Line (LBL) calculations provide ‘exact’ solutions of the radiative heat transfer. However, the use of the LBL method in multidimensional combustion problems requires prohibitive computational resources and is beyond the scope of this paper. The SNBCK assumes a statistical description of each line of the spectrum and for non-isothermal non-homogeneous medium it is assumed that the absorption coefficient is correlated. Previous numerical studies have demonstrated that the SNBCK model, in the absence of LBL solutions, is accurate enough to be used as the reference solution. It has been shown also that the 43 NB database provides solutions in close agreement with the 367 NB database (both from Soufiani and Taine) [22].

Local relative errors between the current model and the reference are estimated as

$$E_{r,i}(\phi) = \frac{\phi_{model,i} - \phi_{ref,i}}{\phi_{ref,i}}$$

where $i$ represents the current mesh point and $\phi$ is the variable considered. In order to avoid interpreting discrepancies encountered in regions where $\phi$ is close to zero, points where $\phi_{ref,i} < 0.02 \times \max_{i=1,N} \phi_{ref,i}$ are excluded from the error analysis. Mean and maximum errors are defined as

$$E_{mean} = \frac{1}{N} \left( \sum_{i=1}^{N} |E_{r,i}| \right)$$

and

$$E_{max} = \max_{i=1,N} |E_{r,i}|,$$

respectively.

Figures 2a, 2b and 2c present the temperature, the divergence of the radiative flux (radiation source term for the energy equation) and the soot volume fraction along the path of maximum soot volume fraction for the SNBCK model. In these figures the temperature difference and the local relative error are also presented for all the radiative property models studied. Figure 2a shows that along the path line exhibiting the maximum soot volume fraction the temperature increase rapidly in the first 3 cm and then reaches an approximately constant value in the range of 1500-1600 K; at around $z = 9.5$ cm the temperature starts to decrease linearly until it attains a temperature of 1300 K at $z = 15$ cm. The zone of constant temperature is characterized by very large radiative losses (see figure 2b); the large soot volume fraction observed in this region is the main responsible of this intense radiation (see figure 2c). Figure 2b shows that the radiative source term decreases after this zone due to a reduction in both temperature and soot volume fraction (see figures 2a and 2c). Figure 2d shows the integrated soot volume fraction along the flame height for the SNBCK model and the relative errors obtained with the different radiative model considered. Between $z=6$ cm to $z=12$ cm the dominant mechanism is the soot oxidation. The results show that the oxidation process is quite sensitive to the radiative property model, more sensitive than the nucleation and the surface growth of the soot. This is directly related to the temperature attained for every model, conditioning the oxidation rates in each simulation. This effect produces important relative errors between the models, especially in the oxidation region (see Figure 2). Therefore the choice of a radiative property model largely affects the amount of soot released by the flame.

Table 1 summarizes the mean and maximum relative error, or temperature difference, obtained for all the radiative models presented in figure 2. The first important result is that the OTA leads to very poor predictions, with a maximum underprediction of the temperature of about 139 K. Maximum errors are higher than 100% for all the variables considered and the amount of soot released from the flame is largely overestimated. In addition the OTA also affects significantly the soot production in the soot growth zone, where the soot volume fraction is around 15% lower as compared to the SNBCK.
These comparisons show clearly that reabsorption by gas and soot should be considered. If reabsorption is taken into account by considering the simple grey model based on Planck-mean absorption coefficient (Planck Model), Table 1 shows that temperature differences and relative errors are similar to those obtained with the OTA. The grey model implemented in FDS (GWBg) improves the predictions but discrepancies are still high. The influence of the grey approximation for soot can be isolated by considering non-grey gases, modelled by the SNBCK, and grey soot modelled by a Planck-mean absorption coefficient. Table 1 shows that this approximation (SNBCKgs) should also be avoided, demonstrating that the spectral nature of soot must be taken into account. Although temperature is relatively well predicted with a maximum difference of 29 K as compared to the reference, it produces substantial mean errors of about 8.5, 12.5 and 14.3% for the radiative source term, the soot volume fraction and the integrated soot volume fraction respectively.

GWV and WSGG take into account the spectral dependence of combustion gases and soot in a simplified manner. The GWB model presents similar discrepancies as the GWBg discussed previously with a maximum temperature difference of about 50 K. The parameters of the WSGG model, i.e. $a_j$.

Figure 2. Comparisons between radiative property models for the S flame. The solution predicted by SNBCK model acts as reference. (a) Temperature and temperature differences $\Delta T = T_{\text{model}} - T_{\text{ref}}$, (b) divergence of the radiative flux and (c) soot volume fraction along the path line exhibiting the maximum soot volume fraction; and (d) integrated soot volume fraction along the flame height. Relative error is calculated with equation (11). The reference curve is represented on the left axis whereas the relative errors for the other methods are represented on the right axis.

(see figure 1b). These comparisons show clearly that reabsorption by gas and soot should be considered. If reabsorption is taken into account by considering the simple grey model based on Planck-mean absorption coefficient (Planck Model), Table 1 shows that temperature differences and relative errors are similar to those obtained with the OTA. The grey model implemented in FDS (GWBg) improves the predictions but discrepancies are still high. The influence of the grey approximation for soot can be isolated by considering non-grey gases, modelled by the SNBCK, and grey soot modelled by a Planck-mean absorption coefficient. Table 1 shows that this approximation (SNBCKgs) should also be avoided, demonstrating that the spectral nature of soot must be taken into account. Although temperature is relatively well predicted with a maximum difference of 29 K as compared to the reference, it produces substantial mean errors of about 8.5, 12.5 and 14.3% for the radiative source term, the soot volume fraction and the integrated soot volume fraction respectively.

GWV and WSGG take into account the spectral dependence of combustion gases and soot in a simplified manner. The GWB model presents similar discrepancies as the GWBg discussed previously with a maximum temperature difference of about 50 K. The parameters of the WSGG model, i.e. $a_j$. 
and \( k_{r} \), are obtained for fixed ratios of \( H_{2}O \) and \( CO_{2} \) partial pressures. In the present study, a ratio of \( P_{H_{2}O}/P_{CO_{2}} = 1 \) is considered, resulting from the complete combustion of ethylene in air. It should be noted that this condition is never encountered in the computational domain as a consequence of incomplete combustion and differential diffusion. This may explain the large discrepancies observed for this model.

The FSCK model using the mixed-NB database and spectral soot (FSCK MNB) predicts temperatures that are quite close to the reference, with only mean and maximum differences of around 5 and 18 K, respectively. This model also present the lowest relative errors among all the radiative models tested, with mean and maximum errors lower than 4.4 and 16%. These discrepancies may be explained by the assumption of a correlated absorption coefficient, which is introduced for both NB and FS k-distributions in the case of a non-homogeneous non-isothermal medium. This assumption is less restrictive on a NB basis than on a FS basis. By considering the mixed-FS database and grey soot (FSCK MFS) the solution are obviously less accurate. Nevertheless, the errors are globally lower than the SNBCKgs and closer to the FSCK MNB.

Table 1 also summarizes the CPU time ratio between the current model and the SNBCK (\( CPU_{ratio} = t_{ref}/t_{model} \)). It is important to mention that each calculation differs only by the radiative model considered. The FSCK MNB model, which present the best results compared to the SNBCK, provides a solution with a CPU time reduced by a factor of 4, making it attractive for this kind of calculation.

4. Conclusions

A numerical study of two axisymmetric laminar coflow smoking and non-smoking ethylene diffusion flames was carried out in order to assess the influence of different radiative property models on soot formation and oxidation processes. The SLF concept was used as combustion model and a modified two-equation acetylene-based model was used to describe the soot nucleation, surface growth and oxidation processes. The main conclusions of this study can be summarized as follows:

1. Comparisons between the experimental data and the numerical solutions obtained with the SNBCK model show a fairly good agreement.
2. Predicted soot volume fractions are very sensitive to the radiative property model used, especially in the zone where the soot is oxidized. In particular the amount of soot released by the smoking flame is highly dependent of the radiative model. As a consequence numerical study concerning soot formation/oxidation processes should consider an accurate radiation model.
3. Gas and soot reabsorptions are crucial features for accurately estimating the radiative losses, and then temperature and soot volume fractions. Consequently the OTA leads to inaccurate predictions.
4. The spectral nature of gas and soot is a crucial feature to obtain reliable predictions. Therefore the use of grey models and GWB models should be avoided.
5. The FSCK provides similar results as compared to the SNBCK with a substantial gain in CPU time. Therefore it is a good alternative to be implemented in order to study soot formation processes.

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