Soot formation in turbulent non premixed flames

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A coupled radiation/flamelet combustion model is applied to the simulation of a turbulent diffusion flame fed with natural gas. The major species and temperature fields are calculated using the Steady Laminar Flamelet and the Eddy Dissipation Concept models. The formation of soot particles is predicted using the method of moments (MOM), coupled with simple semi-empirical models of soot nucleation, growth, oxidation and aggregation. The interaction of turbulence and soot chemistry is described by a probability density approach based on the laminar flamelet formulation.

The scope of this work is mainly to investigate the accuracy and reliability of available soot semi-empirical models by comparison with experimental data in turbulent flames. Particular attention is devoted to the effects of turbulent fluctuations on the closure of soot source terms in the moment equations.

The predicted soot amount in the turbulent flame investigated is found relatively insensitive to the nucleation models. On the contrary, growth and oxidation models significantly influence soot formation.

1. Introduction

The problem of soot formation in combustion devices is gaining rising importance due to its negative effects on human health and for the increasingly stringent limitations concerning the emissions of pollutants from combustion devices. Moreover, soot formation affects the radiation heat transfer in furnaces and various practical applications.

Large detailed kinetic models can be successfully used to help identify the conditions that reduce soot formation. However, the direct coupling of detailed kinetic schemes and CFD (Computational Fluid Dynamics) is a very difficult task, especially when considering the typical dimensions of the computational grids used for complex geometries and industrial applications. Moreover, turbulent flows are characterized by strong interactions between fluid mixing and chemical reactions, which cannot be neglected in most cases. Therefore, the modeling of soot formation in turbulent flames requires the formulation of simple but reliable models for describing the nucleation, growth, aggregation and oxidation of soot particles. Simplified models consider only

Table 1. Semi-empirical models for nucleation (N), growth (G), oxidation (O) and aggregation (A) of soot particles.

Name	N	G	O	A	Fuel	Reference
Liu, 2001	X	X			Ethylene	(Yeoh and Yuen 2009)
Liu, 2002	X	X			Methane	(Liu et al. 2002)
Moss, 1999	X	X			Methane	(Brookes and Moss 1999)
Wen, 2003	X	X			Kerosene	(Wen et al. 2003)
Lindstedt, 1994	X	X			Ethylene/Propane	(Yeoh and Yuen 2009)
Leung, 1991	X	X			Ethylene	(Yeoh and Yuen 2009)
Lee			X			(Yeoh and Yuen 2009)
NSC			X			(Yeoh and Yuen 2009)
Neoh			X			(Yeoh and Yuen 2009)
Smoluchowski				X		(Yeoh and Yuen 2009)
Moss				X		(Brookes and Moss 1999)

the phenomena essential for obtaining sufficiently accurate predictions of soot concentrations and reliable CFD calculations of radiation. If the thermal field and most chemical species can be successfully modeled using non-equilibrium chemistry through flamelet libraries and presumed probability distribution functions (PDF), the same approach is not able to describe soot formation, mainly due to its comparatively slow chemistry (Kent and Honnery 1987).

In the present work the soot volume fraction in a turbulent non-premixed flames fed with methane (Grosschmidt et al. 2007) is predicted using a two-equation model for the soot number density and the volume fraction. The source terms of soot equations are calculated using several semi-empirical models to take into account nucleation, surface growth, aggregation and oxidation phenomena. Two different approaches for the closure of soot source terms in the transport equations are used and compared.

2. Numerical modeling of soot formation

Two additional transport equations are solved in a CFD code to predict soot formation and evolution: the first equation accounts for the transport of particle number density m_0 (which is the moment of zero order of the particle size distribution); the second equation describes the transport of the soot mass density M (which is related to soot volume fraction f_V through the moment of order three of the particle size distribution).

The particles are assumed to be spherical and the size distribution simply represented by an average diameter d_P. For numerical convenience two density weighted variables are introduced: $\phi_N = m_0 / \rho N_{AV}$ for the particle density and $\phi_M = \rho_{soot} f_V / \rho$ for the soot mass fraction (Brookes and Moss 1999). The transport equations for these variables are given by:

$$\bar{\rho} \frac{\partial \tilde{\phi}_N}{\partial t} + \bar{\rho} \tilde{u}_i \frac{\partial \tilde{\phi}_N}{\partial x_i} = \frac{\partial}{\partial x_i} \left(\Gamma_t \frac{\partial \tilde{\phi}_N}{\partial x_i} \right) + \tilde{S}_{m0} \tag{1}$$

$$\bar{\rho} \frac{\partial \tilde{\phi}_M}{\partial t} + \bar{\rho} \tilde{u}_i \frac{\partial \tilde{\phi}_M}{\partial x_i} = \frac{\partial}{\partial x_i} \left(\Gamma_t \frac{\partial \tilde{\phi}_M}{\partial x_i} \right) + \tilde{S}_M \tag{2}$$

where $\bar{\rho}$ and $\tilde{u}_i$ are the gas phase density and velocity respectively, Γ_t is the turbulent viscosity and $\tilde{S}_{m0}$ and $\tilde{S}_M$ the soot source terms.

2.1 Soot source terms

The source terms can be written as a sum of different contributions:

$$S_{m0} = \left(S_{m0}\right)\big|_{nucleation} + \left(S_{m0}\right)\big|_{coagulation} \tag{3}$$

$$S_M = \left(S_M\right)\big|_{nucleation} + \left(S_M\right)\big|_{growth} + \left(S_M\right)\big|_{oxidation} \tag{4}$$

Several semi-empirical models for each of these contributions were studied and applied in the present work. They are summarized in Table 1.

2.2 Effects of turbulent fluctuations

For turbulent flames the effects of turbulent fluctuations should be taken into account in the evaluation of these source terms. The simplest approach, which has been followed by many authors (Wen et al. 2003; Zucca et al. 2006), solves the transport equations and neglects the effects of turbulence. In the present work this simplified approach (called *mean approach*) is compared with a different closure model, which accounts for the effect of turbulent fluctuations on the soot source terms and which will be referred as the *uncorrelated approach* in the following. In general the mean source term can be expressed using the proper joint-PDF $P\left(\xi, \chi_{st}, \phi_H, m_0, f_V\right)$, where ξ is the mixture fraction, χ_{st} the scalar dissipation rate and ϕ_H the enthalpy defect:

$$\tilde{S} = \int_0^{+\infty} \int_0^{+\infty} \int_{-\infty}^{+\infty} \int_0^{+\infty} \int_0^1 S\left(\xi, \chi_{st}, \phi_H, m_0, f_V\right) \cdot P\left(\xi, \chi_{st}, \phi_H, m_0, f_V\right) \cdot d\xi \cdot d\chi_{st} \cdot d\phi_H \cdot dm_0 \cdot df_V \tag{5}$$

This joint-PDF is unknown, both experimentally and theoretically, and therefore simplified approaches must be used to obtain the mean source term. The uncorrelated approach (Brookes and Moss 1999) assumes that ξ and ϕ_H are uncorrelated with soot properties (m_0 and f_V) and that ξ, χ_{st} and ϕ_H are statistically independent:

$$P\left(\xi, \chi_{st}, \phi_H, m_0, f_V\right) \cong P\left(\xi, \chi_{st}, \phi_H\right) \cdot P\left(m_0\right) \cdot P\left(f_V\right) \tag{6}$$

The single PDFs for m_0, f_V and ϕ_H are assumed to be Delta Dirac functions. A β and log-normal PDFs are considered for the ξ and χ_{st} respectively. A flamelet library storing the soot source terms was built at different strain rates, from equilibrium conditions to the extinction value χ_{ext}. Additional details can be found in (Cuoci et al. 2008).

3. Main results

This work analyses a turbulent, non premixed flame, experimentally investigated in (Grosschmidt et al. 2007). The experimental rig consists of a H_2 piloted burner with a nozzle diameter of 15 mm. The fuel mixture consists of 95% CH_4, 3% C_2H_6, 2% N_2
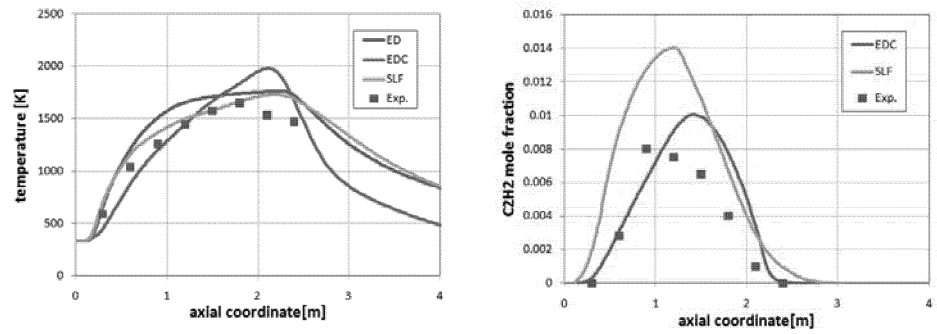
Figure 1. Experimental (Grosschmidt et al. 2007) and calculated temperature and acetylene mole fraction profiles along the axis and radial directions.

(natural gas), fed at the velocity of $u_0 = 82$ m/s ($Re = 60,000$). Velocity fields, soot concentrations and gas-phase concentrations are available in the experimental data set.

The flow field solutions are obtained by using the FLUENT 6.3 commercial code (Fluent Inc. 2006). A 2D steady-state RANS simulation of the physical domain was considered because of the axial symmetry of the system. A structured numerical grid was used on a rectangular 4125x1125 mm domain.

The Favre averaged Navier-Stokes equations together with the standard κ-ε model are employed to calculate the reactive flow. The buoyancy effect has been taken into account in the turbulence model. For the spatial resolution the First-Order Upwind Scheme was adopted. The segregated implicit solver was used with the SIMPLE procedure for the pressure-velocity coupling. It is well known that the standard κ-ε model tends to over predict the dissipation rate of axially symmetric jets, so several turbulence models were tested and compared with experimental measurements of gas velocities. The best agreement was obtained with a corrected κ-ε model, in which a value of 1.60 was assigned to the $C_{1\varepsilon}$ constant. Additional details are reported in (Patriarca 2010).

3.1 Temperature and composition fields
The interaction between chemistry and turbulence was taken into account through different approaches: the Eddy Dissipation (ED), the Eddy Dissipation Concept (EDC) and the Steady Laminar Flamelet (SLF) models. For ED simulations a two-step global mechanism for methane combustion was applied (Westbrook and Dryer 1984). The EDC simulations were performed using a skeletal kinetic mechanism (Glarborg et al. 1992). The lookup tables needed for the application of the SLF model were constructed using a detailed kinetic scheme describing the combustion and pyrolysis of methane, involving ~70 species (Ranzi et al. 2005) .

Figure 1 compares the predicted axial temperature and acetylene mole fraction profiles with the experimental measurements as numerically obtained using the approaches mentioned above. The agreement with the experimental measurements can be considered satisfactory and it is pretty evident that the best results were obtained using the SLF model, both along axial and radial directions (not here reported). Only in the
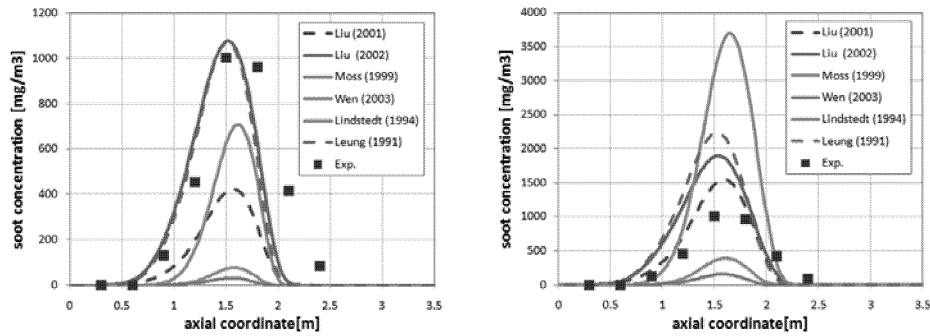
Figure 2. Experimental (Grosschmidt et al. 2007) and calculated soot concentration profiles: (a) mean approach; (b) uncorrelated approach. The curves refer to the NSC oxidation and to the Smoluchoski aggregation models.

tail of the flame the temperature is slightly over predicted. However, the experimental measurements show that soot formation mainly occurs along the axis in the region between 0.5 and 2.5 m, were the temperature predictions can be considered satisfactory. Acetylene is particularly important because the soot model described in the previous section assumes this species as the main precursor. Its numerical prediction must be as accurate as possible, in order to expect good results in terms of soot formation. From Figure 1 it is evident that both EDC and SLF models give a satisfactory prediction of acetylene, both along the axial and radial directions.

3.2 Soot predictions

Soot formation was numerically predicted using both SLF and EDC results. Figure 2a shows a comparison of soot volume fraction along the axis as predicted through the *mean approach* using the semi-empirical models described in the previous section. It is quite evident that these models lead to very different predictions (more than 1 order of magnitude in terms of peak values). This is not so surprising, because the kinetic constants in these models were tuned on the particular kind of fuel and for the operating conditions for which they were formulated and applied. The best agreement with experimental measurements was obtained with the (Liu, 2002) and (Leung, 1991) models. The peak values and the shape of the whole profile are well described, even if soot oxidation seems to occur too early. It is interesting to observe that these two models have a growth velocity which depends on the square root of soot particle surface area (A_s), while all the remaining models have a linear dependence with A_s. Sensitivity analyses (not here reported) showed that the nucleation model has only a marginal role for soot predictions in this flame. On the contrary, the large differences between predicted profiles in Figure 2a are mainly related to the growth velocity.

The application of the uncorrelated approach required the construction of a look up table, storing the soot source terms evaluated according to the Equation (1). Figure 2b shows the soot concentration profiles as obtained using the uncorrelated approach. By comparison with Figure 2a, the first observation is that the main effect of turbulent fluctuation corresponds to an increase of soot formation for every semi empirical model.

This effect is less important for (Liu, 2002) and (Leung, 1991) models, because of their dependence on the square root of A_s. Also in this case these two models give a satisfactory prediction of soot formation, even if the peak value is over estimated. The effect of turbulent fluctuations are particularly strong for the (Lindstedt, 1994) model. The (Moss, 1999) and (Wen, 2003) models give very poor results, both for the *mean* and the *uncorrelated* approaches. The soot particle mean diameters predicted by the different models are quite similar. In particular, in the region around the peak value, the models predict a mean diameter between 20 and 30 nm.

4. Conclusions

The principal objective of this work is to demonstrate the application of a two-equation model for soot predictions in a turbulent non-premixed flame, coupled with a EDC or a SLF model for the description of thermal and gaseous species fields. Simple semi-empirical models for the description of soot nucleation, growth, oxidation and coagulation were adopted using two different closure methods for the source terms in the transport equations of soot.

The adopted models showed very different results in terms of soot concentrations (more than 1 order of magnitude in terms of peak values), but most of them were able to give satisfactory predictions. The predicted soot amount in the turbulent flame is relatively insensitive to the semi-empirical model adopted for the nucleation process. On the contrary, growth and oxidation models significantly influence soot formation.

A deeper sensitivity analysis to the growth, oxidation and aggregation models could be very useful in a future work to investigate the role and effect of individual soot formation rates to determine the optimal parameters for a two-equation soot model to be used in the simulation of turbulent non-premixed flames.

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