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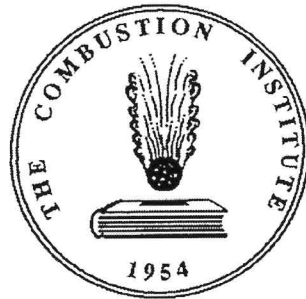
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AND PARTICLE FORMATION IN FLAMES**

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Modelling of turbulent combustion requires an accurate description of turbulence-chemistry interaction in order to obtain reliable temperature and composition profiles; when detailed information on particle formation is needed, the population balance needs to be solved. The stochastic nature of both turbulence and some processes involved in particle dynamics suggests to statistically solve these problems in terms of the fluid probability density function (PDF) and of the particle number density function (NDF).

In this work, two methods based on a similar description of the PDF and the NDF, represented by a finite sum of Dirac delta functions, were successfully employed to solve the problem of turbulence-chemistry interaction in turbulent flames (finite-mode PDF method), and the population balance for modelling soot formation (Direct Quadrature Method of Moments, DQMOM).

Both methods appeared to be versatile and reliable tools, particularly suitable for CFD computation, presenting a good trade-off between accuracy and computational cost.

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INTRODUCTION

The paramount importance of modelling turbulence-chemistry interaction for the appropriate description of turbulent flame properties is well known, and several different approaches have been developed in the past decades to optimally integrate chemistry and fluid dynamics in turbulent reacting flows, in the framework of Computational Fluid Dynamics (CFD). Moreover, it is often important to describe accurately the evolution of combustion-formed particulate, that can be undesired (such as soot) or constitute a tailored material with particular feature (as in the case of flame aerosol synthesis). In a turbulent flow it is important to give an appropriate description of turbulent fluctuations of composition and temperature (i.e., of the related joint temperature-composition probability density function) in order to obtain reliable predictions of the scalar fields of the flame. This is especially important for predictions of intermediate species and products of activated processes (such as NO_x): nevertheless, a detailed modelling of particle dynamics requires to track the distribution of a number of particle properties, identifying for instance size, morphology or composition of the particles, within the evolving population.

In this work, the solution of these problems was approached by novel modelling tools, based on a similar description of the fluid probability density function (PDF) and of the particle number density function (NDF).

In both cases, the distribution is represented by a finite sum of Dirac delta functions, centered in some particular values of the independent variable (nodes) and weighted by some weight functions, representing the related probabilities (in the case of the PDF) or the related particle concentrations (in the case of the NDF).

As far as the modelling of turbulence-chemistry interaction is concerned, the model belongs to the so-called *presumed PDF* methods, and is usually referred to as the Finite-Mode PDF or Multi-Environment model, which has been already successfully employed for modelling precipitation and crystallization in turbulent systems. In this work, the Finite-Mode PDF model has been applied to the study of turbulent non-premixed and partially premixed flames, and compared with other standard models for turbulence-chemistry interaction, already available in commercial CFD codes. The problem of soot formation in turbulent non-premixed flames was then approached by solving the population balance equation (which is a continuity statement in terms of the NDF) by a moment-based method which employs a quadrature approximation for the closure of the problem (Direct Quadrature Method of Moments, or DQMOM), resulting in a 'finite-mode' representation of the NDF. Both a monovariate NDF, in terms of the particle volume or mass, and a bivariate NDF, in terms of particle volume and surface area, in order to obtain more detailed information on particle morphology, were considered in the computation.

The proposed models have been implemented as User-Defined Functions within the commercial CFD code FLUENT 6.1.

GOVERNING EQUATIONS

Modelling of Turbulence-Chemistry interaction

Finite-mode PDF models assume that the presumed joint-composition PDF has the following functional form:

$$f_\Phi(\Psi; \mathbf{x}, t) = \sum_{n=1}^{N_e} p_n(\mathbf{x}, t) \prod_{\alpha=1}^{N_s} \delta\left[\psi_\alpha - \langle \phi_\alpha \rangle_n (\mathbf{x}, t)\right]$$

where N_e is the number of modes or environments, N_s is the dimension of the composition vector, p_n is the probability associated with environment n, and $\langle \Phi \rangle_n$ is the composition vector in environment n [1]. The number of modes N_e is usually variable between 2 and 4, and each mode can be thought of as an environment with particular features. Depending on the modelling choice, one could model the system by two partially mixed and reacting environments (N_e=2), or two unmixed and non-reacting environments, containing each one of the reactants, and a mixed environment where the reaction occurs (N_e=3), or two unmixed environments and two reacting environments (N_e=4). During the mixing process the unmixed environments disappear (i.e. their probability is decreased) while the mixed one(s) are generated (i.e. their probability is increased). In general, a certain degree of freedom is left to the modeller, that yet can accommodate the first three moments of the PDF, forcing in other words the PDF to integrate to one and to have expected value and variance well described.

The resulting transport equations for probabilities and for the weighted compositions in each environment are (repeated indices imply summation):

$$\frac{\partial p_n}{\partial t} + \tilde{U}_j \frac{\partial p_n}{\partial x_j} - (\Gamma + \Gamma_t) \frac{\partial^2 p_n}{\partial x_j \partial x_j} = G_n(\mathbf{p}) + c_n^p,$$

$$\frac{\partial \langle s_\alpha \rangle_n}{\partial t} + \tilde{U}_j \frac{\partial \langle s_\alpha \rangle_n}{\partial x_j} - (\Gamma + \Gamma_t) \frac{\partial^2 \langle s_\alpha \rangle_n}{\partial x_j \partial x_j} = M_{n,\alpha}(\mathbf{p}, \mathbf{s}) + p_n S\left(\langle \varphi \rangle_n\right) + c_{n,\alpha}^s,$$

where $\tilde{U}_i$ is the Favre averaged i-th component of the velocity vector, Γ is the molecular diffusivity, Γ_t is the turbulent diffusivity, $G_n(\mathbf{p})$ is the source term for probability (due to molecular mixing), $\langle s_\alpha \rangle_n = p_n \langle \phi_\alpha \rangle_n$ is the probability-weighted concentration of component α in node n, $M_{n,\alpha}(\mathbf{p}, \mathbf{s})$ is the rate of change of $\langle s_\alpha \rangle_n$ due to molecular mixing, and $p_n S\left(\langle \varphi \rangle_n\right)$ is the average chemical source term, exactly calculated from the local composition in the n-th environment/mode. The terms c_n^p and $c_{n,\alpha}^s$ are diffusive terms that arise due to the finite-mode representation, and are necessary, in non-homogeneous cases, to correctly predict moments of the distribution of order equal to or larger than two.

Modelling of particle dynamics

Let us define a property vector $\xi = (\xi_1, \xi_2, \ldots, \xi_M)$, each element of ξ being a property of the particle, such as volume, temperature, size, surface area, age, fractal dimension; these properties are Cartesian coordinates of the particle state space, and are usually called *internal coordinates* to distinguish them from the usual 'external' coordinates of the physical space. If we introduce the *number density function* $n(\xi; \mathbf{x}, t)$, so that:

$$n(\xi; \mathbf{x}, t) d\xi_1 \ldots d\xi_M dx_1 dx_2 dx_3 = n(\xi; \mathbf{x}, t) d\xi dV$$

is the number of particles contained in the infinitesimal volume dV, with internal coordinates in the range $\left[(\xi_1,\xi_1+d\xi_1);\ldots;(\xi_M,\xi_M+d\xi_M)\right]$, the population balance equation, after Favre averaging, is:

$$\frac{\partial\tilde{n}(\xi;\mathbf{x},t)}{\partial t}+\frac{\partial}{\partial x_j}\langle U_j|\xi\rangle\tilde{n}(\xi;\mathbf{x},t)+\frac{\partial}{\partial x_j}\left[(\Gamma+\Gamma_t)\frac{\partial\tilde{n}(\xi;\mathbf{x},t)}{\partial x_j}\right]=\tilde{S}(\xi;\mathbf{x},t)$$

where the left-hand side contains all the terms related to accumulation and transport of the number density function in the physical space, and the source term in the right-hand side accounts for all the processes that determine the evolution of the population.

Notice that $\langle U_i|\xi\rangle$ is the average value of the particle velocity, conditioned to the property vector, and it is equal to the expected value of the fluid velocity $\tilde{U}_j$ when the particles are so small to be entrained by the fluid, as in the case of soot formation.

The population balance equation can be solved by using DQMOM, which is based on the moment approach, particularly suitable for CFD calculations. The DQMOM uses a quadrature approximation to close any integral involving the NDF, so that the mixed moments of the distribution can be written as [2, 3]:

$$m_{k_1,k_2,\ldots,k_M}=\int_{-\infty}^{\infty}\xi_1^{k_1}\int_{-\infty}^{\infty}\xi_2^{k_2}\cdots\int_{-\infty}^{\infty}\xi_M^{k_M}\tilde{n}(\xi;\mathbf{x},t)\,d\xi_1 d\xi_2\cdots d\xi_M\approx\sum_{\alpha=1}^{N}w_\alpha\prod_{i=1}^{M}\xi_{i\alpha}^{k_i}$$

where $w_\alpha(\mathbf{x},t)$ are the 'weights', $\xi_{i\alpha}(\mathbf{x},t)$ are the 'abscissas' of the quadrature approximation, and k_i is the order of the moment with respect to ξ_i.

The quadrature approximation can be directly tracked by solving the transport equations for weights w_α, and weighted abscissas $\varsigma_{i,\alpha}=w_\alpha\xi_{i,\alpha}$, as explained by Marchisio and Fox [4]:

$$\frac{\partial w_\alpha}{\partial t}+\frac{\partial}{\partial x_j}(\tilde{U}_j w_\alpha)-\frac{\partial}{\partial x_j}\left[(\Gamma+\Gamma_t)\frac{\partial w_\alpha}{\partial x_j}\right]=a_\alpha,$$

$$\frac{\partial\varsigma_{i,\alpha}}{\partial t}+\frac{\partial}{\partial x_j}(\tilde{U}_j\varsigma_{i,\alpha})-\frac{\partial}{\partial x_j}\left[(\Gamma+\Gamma_t)\frac{\partial\varsigma_{i,\alpha}}{\partial x_j}\right]=b_{i,\alpha},$$

where a_α, and $b_{i,\alpha}$ are the corresponding source terms. It is possible to show that such source terms can be easily evaluated solving a linear algebraic system, obtained from the population balance equation after the application of the quadrature approximation and forcing the moments to yield known values [4]. In the bivariate case (two internal coordinates), we obtain the following linear system:

$$\sum_{\alpha=1}^{N}\left[(1-k-l)\xi_{1,\alpha}^k\xi_{2,\alpha}^l a_\alpha+k\xi_{1,\alpha}^{k-1}\xi_{2,\alpha}^l b_{1,\alpha}+l\xi_{1,\alpha}^k\xi_{2,\alpha}^{l-1}b_{2,\alpha}\right]=\bar{C}_{k,l}+S_{k,l}^{(N)}$$

where $\bar{C}_{k,l}$ is a diffusive term which plays the same role as the correction terms c_n^p and $c_{n,\alpha}^s$ in the Finite-mode PDF model, and $S_{k,l}^{(N)}$ is the quadrature approximation with N nodes of the source term for the mixed moment of order k with respect to the first internal, and of order l with respect to the second internal coordinate; such source terms can be easily evaluated once the rates of the involved processes (such as nucleation, aggregation, molecular growth and so on) are defined.

RESULTS AND DISCUSSION

Turbulent combustion

The finite-mode PDF method with three environments (N_e=3) has been applied to the simulation of turbulent non-premixed and partially premixed flames, both with and without swirl. The model has been validated with experimental data provided by the International Workshop on Turbulent Non-Premixed Flames on bluff-body stabilized flames (http://www.ca.sandia.gov/TNF/) fuelled with mixtures of hydrogen and natural gas (1:1 vol., non-swirling flame) or air and natural gas (2:1 vol., swirling flame, with swirl number S=1.59). Finite rate chemistry was implemented, with a simple four-step mechanism for methane combustion [5]. The method was compared to other widely used modelling tools such as the beta PDF model, with the assumption of chemical equilibrium, and the laminar flamelet model (which allows the implementation of detailed chemistry to be used in the regions where chemical equilibrium is not reached, due to aerodynamic strain). Turbulence was closed by the Reynolds-Averaged Navier-Stokes (RANS) equations approach. For the non-swirling flame the k-ε model was found to be a suitable tool, while the swirling flame required the more complex Reynolds Stress Model (RSM) in order to predict properly the flow field of the flame. Simulations were run on two-dimensional axisymmetric conformal grids with a number of computational cells of order 10^4. Results are reported in Fig. 1.
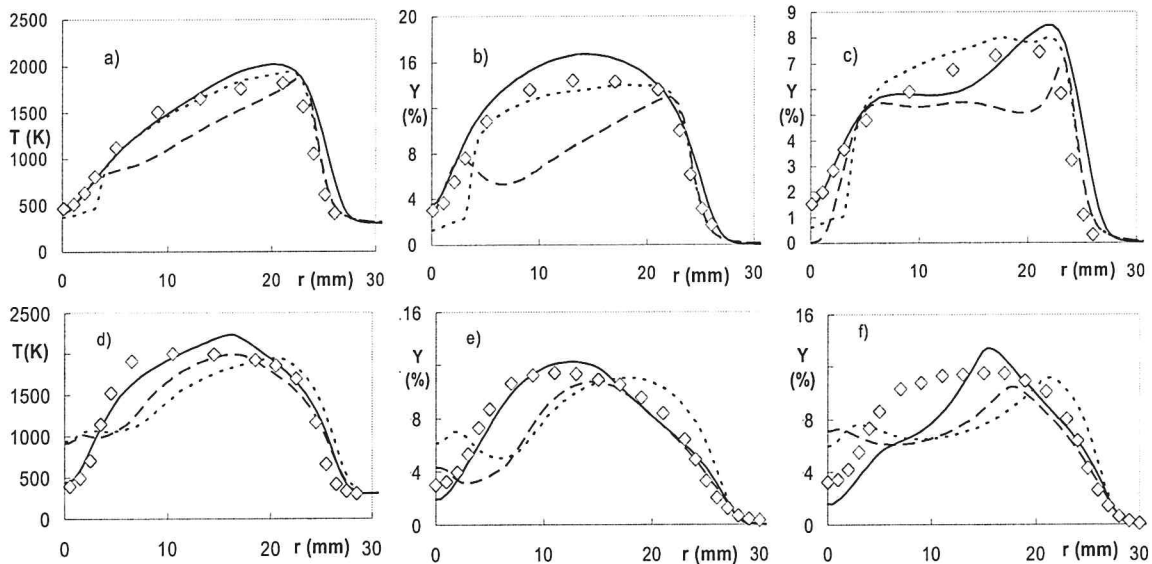

Fig. 1 Comparison betweeen different turbulence-chemistry interaction models for the prediction of temperature [a) non-swirling, d) swirling], H_2O mass fraction [b) non-swirling, e) swirling] and CO_2 mass fraction [c) non-swirling, f) swirling] at 30 mm above the burner. Solid line: Finite-mode PDF with finite rate reduced chemistry. Dotted line: laminar flamelets with detailed chemistry (17 species, 25 reactions). Dashed line: beta PDF model with equilibrium assumption.

It is possible to see that the finite-mode PDF with finite rate chemistry gives the best results, especially where the depart from equilibrium is significant, while the beta PDF model with the equilibrium assumption is not suitable for modelling of the flames under investigation. The laminar flamelet model gives quite good results, thanks to the detailed kinetic scheme implemented, but fails in the region near the central jet, where the flame is strongly stretched and the scalar dissipation rate is very high.

The improvements obtained by the finite-mode PDF model are more significant in the case of partially premixed flames, such as the swirling flame in the present work, for which the hypothesis of local equilibrium is not valid. Further improvements are foreseeable as the composition and temperature PDF will be described with major detail (higher number of reacting environments, better predictions of higher order moments by DQMOM), and as a more detailed kinetic scheme will be implemented. Of course, the increase in detail of the model is paid by an increase in the computational resources required.

Soot formation

The problem of soot formation was considered in the case of a turbulent jet flame of ethylene burning in still air [6]. In this case, the k-ε model was used for turbulence modelling, and the beta PDF approach with the hypothesis of equilibrium was found to be suitable for the simulation of the flame. Particle nucleation, aggregation, surface molecular growth, oxidation and restructuring were taken into account in the model, the rate of each process being evaluated by kinetic expressions available in the literature; further information can be found in [7]. The agreement with experimental radial profiles of soot volume fraction at different axial positions is quite good, as can be seen in Fig 2a.

Particular care was devoted to the description of particle morphology, which is a key issue in the case of soot aggregates. In fact, particle volume V scales with the characteristic radius R with a scaling exponent D_f, or *fractal dimension*, which is in general lower than 3, as stated by the following relationship:

$$V \propto v_0 R^{D_f},$$

where v_0 is the volume of the primary particles constituting the aggregate. If the particle state is characterized by only one internal coordinate (i.e., in the monovariate case), the value of D_f has to be assumed or evaluated by some simplified models [8]. An accurate description of particle morphology requires the solution of the bivariate population balance equation in terms of two geometrical properties of the aggregate (e.g. volume and surface area); in this way, the fractal dimension can be computed from volume and surface area.

In this work, the population balance equation was solved for both the monovariate and the bivariate NDF, and results are compared in Fig. 2b where the volume-area phase plane is used to compare the bivariate simulation (solid line) and the monovariate simulation employing the algebraic model described in [8] to simulate the evolution of D_f. As it is possible to see, the qualitative behaviour is well predicted, but the monovariate simulation seems to overestimate the restructuring process (leading to more compact particles, with higher D_f, in the last part of the flame). Eventually, the importance of the bivariate formulation is well highlighted by Figs. 2c, 2d and 2e, where the profiles of mean volume and surface area are compared, and the projections of the peaks representing the NDF ($N=2$) along each internal coordinate are shown in two locations on the flame axis. The height of each peak is proportional to the corresponding weight (particle number density), so that lower peaks correspond to lower particle concentration.
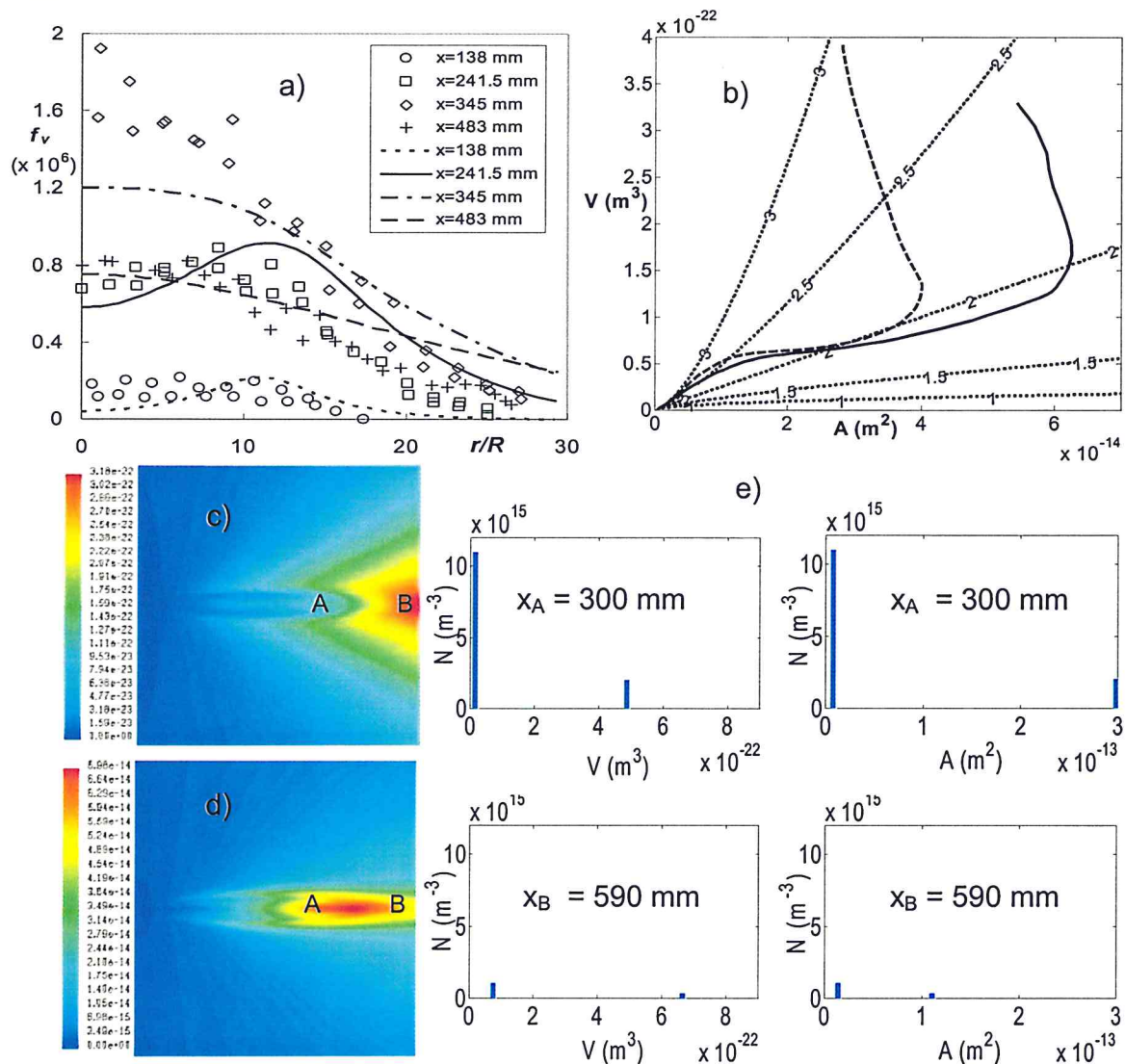
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Fig. 2 a) Soot volume fraction. Symbols: experimental data; Lines: bivariate DQMOM. b) Evolution of particle volume and surface area in the phase plane. Comparison betweeen monovariate (dashed line) and bivariate (solid line). c) Contour plot of particle volume and (d) particle surface area. e) Projections of the nodes (N=2) along each internal coordinate at two locations on the flame axis.

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