COMBINING MULTISECTIONAL (MUSE) METHOD AND CONDITIONAL MOMENT CLOSURE (CMC) FOR SOOT MODELLING IN TURBULENT FLAMES

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Abstract

Future soot modelling in combustion aims to predict total soot particle loading, sizes, morphology and chemical composition. Detailed modelling of soot particles in turbulent flames remains mostly unexplored also because of the lack of experimental data in well controlled conditions. The timescales of soot formation and oxidation generally overlap with fluid-dynamic ones strongly reducing the possibility to treat this problem by using the Stationary Laminar Flamelet Model (SLFM) approach. Conditional Moment Closure (CMC) coupled with detailed models has given encouraging results for soot predictions in turbulent flames [1].

In this work we use a CMC method coupled with a MultiSectional (MuSe) method in which the mixture fraction is used as a scalar parameter in the transport equation. The transport terms for the mixture fraction and its stochastic fluctuations are taken into account. The "first order" CMC is used, in which the temperature is used in the closure term and strongly depends on the mixture fraction. The MuSe method coupled to CMC represents an evolution of the previous sectional method for soot modelling developed in laminar premixed and diffusion flames. Discretization has been operated on carbon atoms, carbon/hydrogen ratio and morphology of particles [2]. Molecular growth, particle inception, particle agglomeration and oxidation are considered [2].

A turbulent axisymmetric diffusion flame of methane has been modelled. Particle volume fractions and temperature are available for this flame [3]. Discrete transfer method has been used for accounting the radiation term with coefficients adjusted for matching the experimental temperature. Turbulence is closed by the k-ε model using typical coefficients for axisymmetric flow. The flame is a jet flame of methane in air operated with a fuel jet velocity of 22m/s (Re=3500) [3].

Results obtained for this flame show a general agreement of the model with the experimental data. In particular the flame structure and the main zone of soot formation are well predicted. Further information about the H/C ratio have been obtained with the use of the multi sectional method.

Introduction

Turbulent combustion represents a particular difficult problem due to the overlap between the kinetic timescale and the turbulent mixing timescale. This makes almost impossible, with some exceptions, the decoupling of these two aspects of the problem.
The last twenty years have raised great advances in the understanding the processes that lead to the formation and the oxidation of soot particles in non-turbulent conditions. Recently, studies conducted on the effect of particles on human health and climate change have pointed the attention not only on the total amount of particles produced in combustion but also on their size distribution, chemical composition and morphology. These aspects have become an issue for the scientific community that is involved in the study of the formation and evolution of soot particles. Most of the studies have been conducted in well-controlled reactors at laboratory scale. The insights gained with several experimental techniques have helped in the building up models which are able to predict with details the evolution of soot particles in these experimental conditions. Modeling attempts to predict the formation of soot particles in turbulent flame remain pioneering, and sooting turbulent flames are largely unexplored. .
On the other side, turbulent fields have been explored both experimental and numerical with advanced techniques. Experimentally Ultras-Fast laser have been increasingly used for the investigation of reactive and non-reactive turbulent flow, to explore the mixing properties and the velocity field. Numerically, Large Eddy Simulation (LES) has been established as reliable tool to predict transient phenomena. Directly Numerical Simulation (DNS), although seems to be very promising, remains almost unexplored due to time consuming. RANS (Reynolds-Averaged Navier Stokes) simulation provides only the mean values of the turbulent fields. It is a simple approach to the turbulence issue but it allows to have a reduced computational cost and explore the use of larger number of species.
Several approaches have been developed to couple the kinetic and the turbulence field. Soot kinetic are generally slow compared with main oxidation pathways and thus are not reproduced well by steady-state flamelet modeling. Several approach to account for explicit models for formation and oxidation rates have been proposed. Generally, due to increasing computational cost as the number of species increases, when the PDF method is used inception and growth processes are treated with a very simple approach [4-6].
Here turbulent reaction is modeled by using Conditional Moment Closure (CMC) [7] in which a full chemical kinetic scheme for gas and particle phases is used. This approach is much less intensive computationally and therefore allows solution of a large number of species within reasonable time two-dimensional flows [1].

Numerical Approach

CMC approach

The governing equations for unconditional and conditional-average quantities are steady-state, axisymmetric and the treatment follows Cleary and Kent [9]. Transport of momentum in axial and radial directions, mixture fraction and its variance, standardised enthalpy, together with turbulent kinetic energy and dissipation are solved as unconditional quantities. Turbulence is closed by the k-ε model using typical coefficients for axisymmetric flow [10]. Heat loss by radiation is modelled broadband by the discrete transfer method [11]. The absorption coefficients are dominated by soot where present and they are adjusted so that predicted temperatures match measurements.
Conditional Moment Closure (CMC) [7] is used to obtain mean species and density of the mixture. Modelling of fluctuation of the reaction rate uses species conditioned upon the mixture fraction, ξ. Mixture fraction is considered ξ=0 for a pure fuel mixture. Mixture fraction is conditioned upon a certain value, η.
Clipped Gaussian PDF's, P(η), are used here to calculate the unconditional average species mass fraction. The PDF transport equation for P(η) is an adjoint equation to the conditional species transport equation and both must be satisfied for the model to be valid.
Density and mass fraction transport equation can be rewritten in steady-state axisymmetric coordinates for the conditional quantity. By using the Favre and Reynolds average correlations, the transport equation can be rewritten as for unconditional quantities.
The first-order closure in CMC is based on the conditional temperature. Which strongly affects conditional reaction rates. The conditional temperature profile is scaled from a corresponding adiabatic profile. The scaling is pivoted at $\eta = 0$ and $\eta = 1$, the air and fuel temperatures.
Central differencing is used for diffusive terms and first-order hybrid upwind differencing for the advection terms in the finite-volume discretised equations for conditional quantities. A set of linearized equations is obtained where conditional mass fraction is given in terms of spatial and η neighbours. More details on CMC can be found elsewhere [1,7-8].
At each spatial location conditional mass is solved simultaneously for all species and all η using a modified Newton-Raphson scheme. The Jacobian includes all conditional species at the spatial location. An initial solution is obtained from an SLFM library. The criterion for convergence is that the mean absolute residual for all conditional species, normalised by the maximum value of the species, is about 10^{-6}-10^{-5}.
The spatial grid expands in direction of increasing axial and radial distance from the nozzle and has a finest resolution of about 0.7mm in the radial direction and

about 5mm in the axial direction. There are 40 points in η with closer increments around stoichiometry. The unconditional grid covers 1m axially × 0.3m radially. The CMC grid only covers about 0.85m × 0.05m radially this being the region of significant interest defined by the mixture fraction field.

Kinetic Scheme

A Multi-Sectional (MuSe) method is used for the modelling of aromatic growth and particle inception. It is based on a previously developed kinetic mechanism of particle formation with a single discretization of the particle phase in terms of C atoms with a fixed H/C ratio. A successive discretization has been introduced to take into account variable H/C ratios. The upper and the lower limit of this discretization are fixed by physical constraints. H/C ratio can reach 0 a bottom limit value, e.g. structures containing just C-atoms, on the other hand H/C ratio equal to 1 representing the value of benzene. The present model also distinguishes between different particle structures based on their state of aggregation, i.e. high molecular mass aromatic molecules (Molecules), clusters of molecules (Clusters) and agglomerates of particles (Aggregates) [2,12]. This allows to follow not only the mass of the formed particles, but also their hydrogen content and internal structure. Two coagulation reactions have been taken into account: coalescence and aggregation. Coalescence leads from molecules to cluster and from clusters to larger clusters; aggregation leads from clusters to aggregates or from aggregates to large aggregates
Oxidation-induced fragmentation is also considered. It can be viewed as the inverse of the coagulation process. During oxidation, the carbon extraction from the high-molecular mass structures could affect the internal structure of soot species, depending on the position of the C-atom that is oxidized. For internal C-atoms, oxidation leads to a breaking up of the particle producing smaller fragments. Since the model separately accounts for aggregates and primary particles different fragmentation processes have to be considered. Aggregates can form smaller aggregates or eventually primary particles (clusters) and clusters can explode forming smaller clusters. Fragmentation is considered forming two new entities of equal mass. Oxygen is considered the only species able to not react on the surface and diffuse towards the points of contact of the primary particles.

Results

The model is applied to a turbulent non-premixed methane flame. The methane turbulent flame [3] issues from a 2.5mm diameter nozzle at 22m/s ($\tau \approx 10$ms) and it has a disc around the nozzle to assist in stabilising the flame. The flame normalised stoichiometric length Xst/D is about 90. The flame has high scalar dissipation rates near the nozzle which rapidly decay and in the main body of the flame to $N < 1$s-1. To prevent extinguishment in the model near the nozzle and consequent

extinguishment of the whole modelled flame the scalar dissipation rate is limited to 10s-1. The limit only exerts its influence near the nozzle for X/D < 10.
The methane flame particulate volume fraction predictions and data are shown in Figure 1 (bottom). This is a low-sooting fuel where data is obtained by laser-induced emission to detect nanometric particles of organic carbon (NOC) characterized by a size range of 1-10 nm and soot particles with sizes in the range 10-100 nm by fluorescence and incandescence [3].

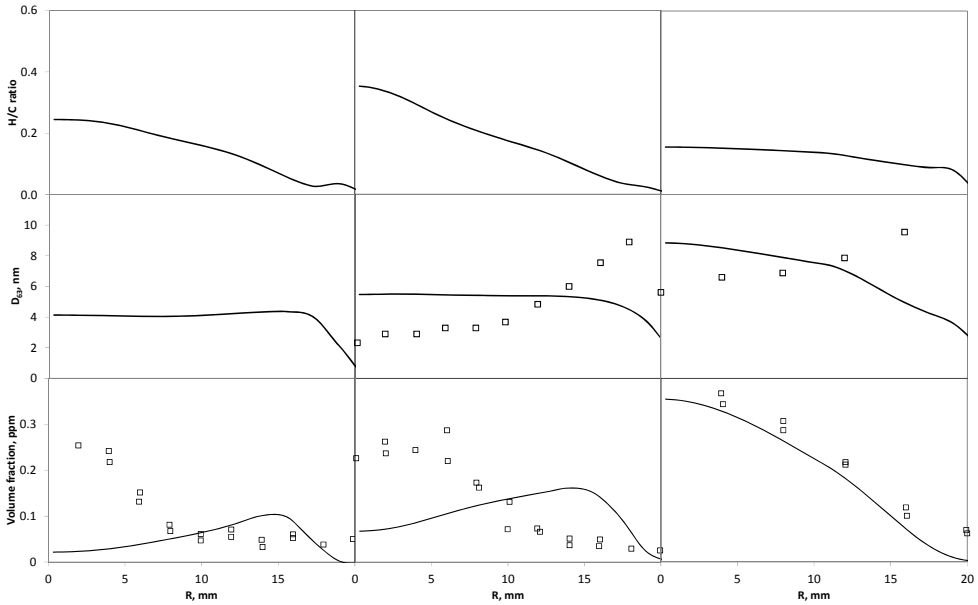

Figure 1. Particulate volume fraction (bottom), d63 (middle), H/C ratio (top) in turbulent methane flame for X/D = 28 (left), X/D = 40 (centre), X/D = 80 (right). Data (dots) are taken from [3], model predictions are reported as solid lines.

The adiabatic conditional temperature profile based on the local conditional species concentrations. Figure 1 (middle) shows D63 particle diameter predictions and here reveals a good agreement against data. The measurements [3] showed that there is negligible loading as soot in the larger size range for the methane flame and the predictions by scaling the adiabatic temperature profile are also strongly biased to the small size particulates.
Finally figure 1 (top) shows the modelled H/C ratio for the particulate matter along the radial coordinates for three different axial locations. It is possible to see that the values of H/C ratio result quite low. Although a further validation with experimental data is necessary, the code seems to suggest a strong graphitization process occurring also in the early region of soot formation.

Conclusions

In this work a MultiSectional method has been used with a CMC approach to couple kinetic and turbulence in a RANS simulation in a sooting turbulent methane flame. The model shows a good capability to reproduce the volume fraction of particulate matter measured by optical techniques. Model also reproduces, with good agreement the mean size of the particles. The small sizes of the particles suggest that methane flame in the investigated conditions only produces small primary soot particles. Model analysis confirms this trend because no aggregates are found. Finally the model also furnishes details on H/C ration of the particle produced. Although no experimental data are available, model suggest that the first nuclei of few nanometers produced in the flame undergo a rapid graphitization process and finally the particles produced have a very low H/C ratio typical of large soot aggregates.

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