Numerical prediction of soot emissions in ethylene flames based on DQMOM

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The main aim of the present work is to understand and determine the formation and evolution of soot particles and its effect on heat transfer from the system. The Direct Quadrature Method of Moments (DQMOM) is employed to solve in a computationally efficient manner the Population Balance Equations (PBE). Numerical simulations of turbulent ethylene-air nonpremixed flames are carried out. Gas-phase chemistry is modeled with PDF (Probability Density Function) mixture-fraction method combined with equilibrium chemistry. As many as 20 chemical species are considered in the equilibrium chemistry calculations. Turbulent-chemistry interactions are through presumed β-PDF. The particle phase is computed with PBE.

The evolution of the soot particles is described by nucleation, surface growth, aggregation and oxidation, the most important processes in the evolution of soot.The kinetic models for nucleation, growth, oxidation are shown below along with aggregation model.

Nucleation : In the present numerical approach acetelyne is considered responsible for soot nucleation. The corresponding rate equation from Moss et al. [1995] is:

$$J = N_A \rho^2 T^{\frac{1}{2}} 6 \times 10^6 \exp\left(-\frac{46100}{T}\right) X_{C_2H_2} \tag{1}$$

Molecular growth : acetylene molecules are considered to be responsible for the molecular growth and the corresponding rate expression is given by Liu et al. [2003]:

$$G_{mg} = \frac{6}{D_f \rho_s} \left(\frac{R_c}{R_{c0}}\right)^{\frac{3-D_f}{3}} 2M_s \cdot 6 \exp\left(-\frac{6038}{T}\right) [C_2H_2] \tag{2}$$

Oxidation : Oxidation kinetic model proposed by Said et al. [1997] is used.

$$G_{ox} = \frac{P}{D_f \rho_s} T^{-\frac{1}{2}} 6.5 \exp\left(-\frac{26500}{T}\right) Y_{O_2} \tag{3}$$

Aggregation : Widely used Fuchs interpolation formula of Brownian kernel is considered for the aggregation (see Zucca et al. [2006]).

$$\beta_{12} = 4\pi(D_1 + D_2)(R_{c1} + R_{c2}) \left[\frac{R_{c1} + R_{c2}}{R_{c1} + R_{c2} + (g_1^2 + g_2^2)^{\frac{1}{2}}} + \frac{4(D_1 + D_2)}{R_{c1} + R_{c2} + (c_1^2 + c_2^2)^{\frac{1}{2}}}\right]^{-1} \tag{4}$$

Where, $c_i = \sqrt{\frac{8k_bT}{\pi m_i}}$, $D_i = \frac{k_bT}{6\pi\mu R_{ci}} \left[\frac{5+4Kn_i+6Kn_i^2+18Kn_i^3}{5-Kn_i+(8+\pi)Kn_i^2}\right]$ $l_i = \frac{8D_i}{\pi c_i}$, $g_i = \frac{(2R_{ci}+l_i)^3-\left(4R_{ci}^2+l_i^2\right)^{\frac{3}{2}}}{6R_{ci}l_i} - 2R_{ci}$

Here, X is mole fraction, Y is mass fraction, k_b is Boltzmann constant, m is particle mass, M_s is soot molecular weight, N_A is Avogadros number and $[C_2H_2]$ represents the acetelyne concentration.

Mono-variate population balance modeling is considered with particle diameter as the internal co-ordinate. With the DQMOM approach one needs to solve a limited number of scalar transport equations (typically 4-6) coupled with the Navier-Stokes equations. It is relatively easy to implement DQMOM in commercial CFD solvers (here *Ansys-Fluent*) and it can be extended for multivariate PBE. The DQMOM algorithm is coupled to *Ansys-Fluent* through a complex User Defined Function (UDF). In the literature, many experimental results can be found concerning soot formation in turbulent nonpremixed flames burning ethylene. Three different flames have been chosen for our simulations, involving different flow conditions and a wide range of Reynolds numbers as shown in Table 1.

Flame	Fuel	Re	D (mm)	u_i (m/s)	Reference
I	C_2H_4	14600	3.0	52.0	Kent and Honnery [1987]
II	C_2H_4	13500	4.56	25.8	Yang and Koylu [2005]
III	C_2H_4	5700	9.0	6.3	Coppalle and Joyeux [1994]

Table 1: Different experimental test conditions considered for simulation

The comparison of simulation results with the experimental data demonstrates that the developed modeling approach can describe accurately the formation and evolution of soot particles in turbulent flames. Analysis of results shows that heat loss from the system is highly significant and there is a clear need for better radiation models in future investigations. Further tests involving other conditions and different fuels will be carried out in the near future.

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Introduction

- Due to the adverse effects of soot on human health (lung cancer) and on environment (global warming) it has attracted great attention of researchers from many years. The research still has high significance due to many open issues.
- The complexities are there starting from understanding the inception of soot to its modeling and finding affordable solution methods.
- The detailed analysis of literature on modeling of soot formation in turbulent diffusion flames can be divided into 4 major parts. i) Gas phase chemistry, ii) Turbulence and chemical interactions iii) Soot evolution mechanisms and its representation iv) Solution methodology.

Objectives

- Quantitative prediction of soot particle formation in ethylene flames using numerical simulation: Computational Fluid Dynamics (CFD) coupled with Population Balance Equations (PBE).
- Analyse different factors affecting soot-particles formation.
- Assess detailed physical and chemical processes involved.

Soot evolution

There are four important mechanisms involved in soot formation and its subsequent evolution.

1. Nucleation
2. Molecular growth
3. Aggregation and
4. Oxidation

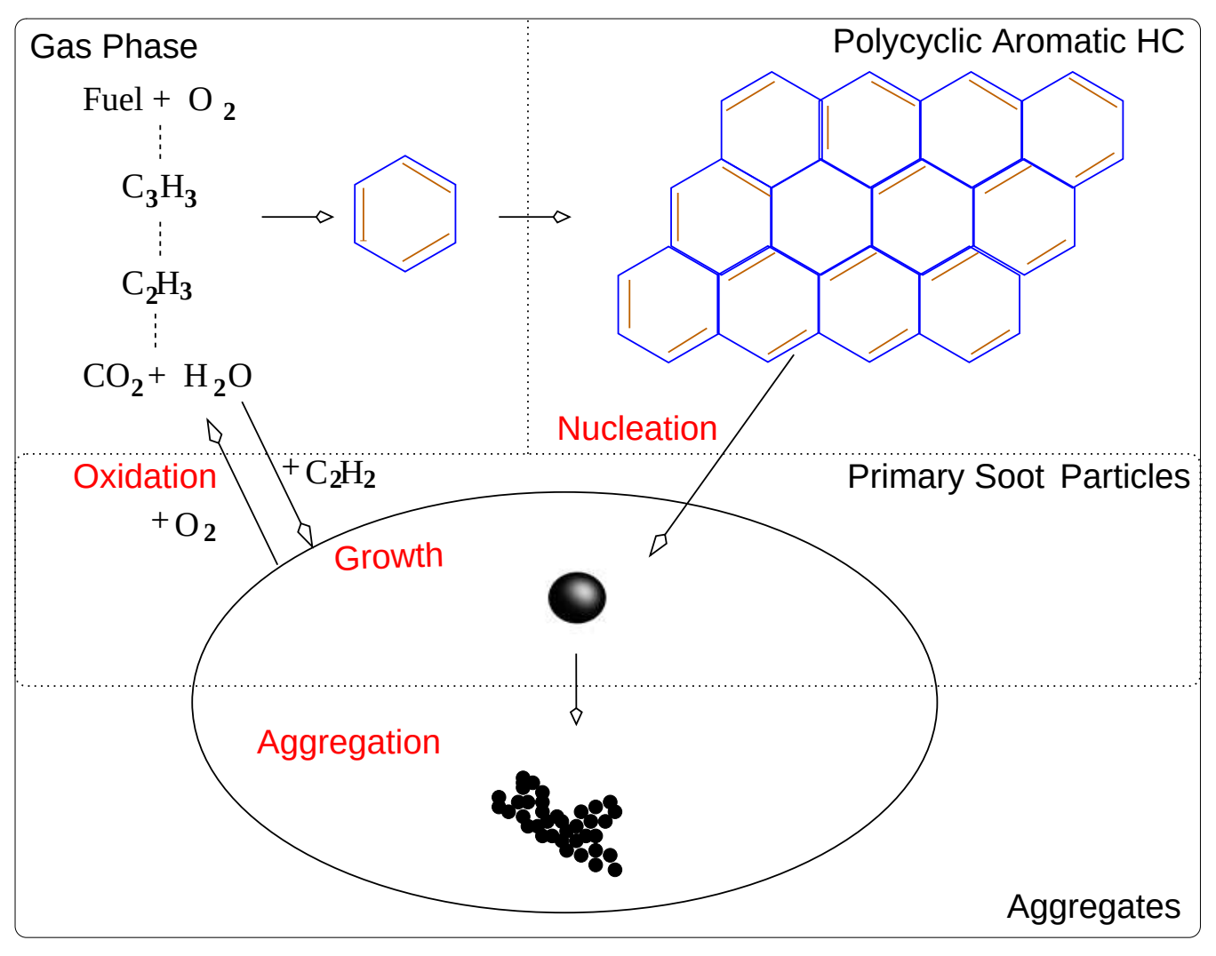

FIGURE 1: Soot particle formation and evolution

Governing equations and solution procedure

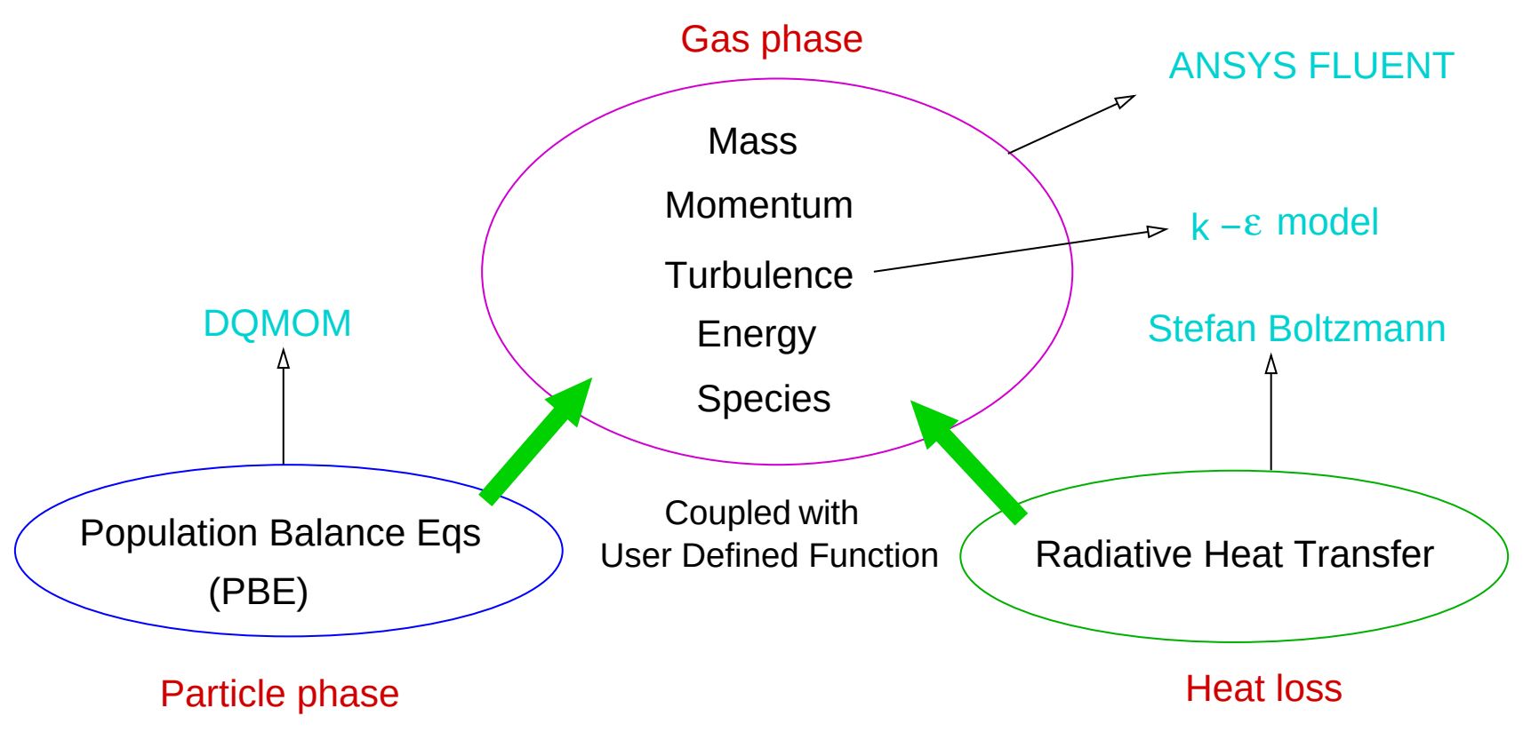

FIGURE 2: Governing equations and solution procedure

Population Balance Equation (PBE)

- The PBE for the particle number density function, NDF (f) is shown below.

$$\frac{\partial f(\boldsymbol{\xi};\mathbf{x},t)}{\partial t}+\frac{\partial}{\partial x_i}\left[\langle u_i|\xi\rangle f(\boldsymbol{\xi};\mathbf{x},t)\right]-\frac{\partial}{\partial x_i}\left[(\Gamma+\Gamma_t)\frac{\partial f(\boldsymbol{\xi};\mathbf{x},t)}{\partial x_i}\right]=S(\boldsymbol{\xi};\mathbf{x},t) \qquad (1)$$

- Here the internal co-ordinates are represented with $\boldsymbol{\xi}$, $\langle u_i|\xi\rangle$ is the mean velocity conditioned on the property value ξ, and $S(\boldsymbol{\xi};\mathbf{x},t)$ is the source term.
- Source term includes all the transformations such as nucleation, molecular growth, aggregation and oxidation, etc which are responsible for change in particle concentration, $f(\boldsymbol{\xi};\mathbf{x},t)$.
- Nucleation, molecular growth and oxidation are modeled with kinetic rate expressions, which are available in the literature. Aggregation is evaluated through the Brownian aggregation kernel (from Fuchs interpolation).

Direct Quadrature Method of Moments-DQMOM

- DQMOM is a moment-based method to solve PBE.
- In the DQMOM approach the distribution function is represented as a summation of a finite number, N of multi-dimensional Dirac delta functions [2] as shown below.

$$f(\boldsymbol{\xi};\mathbf{x},t)\approx\sum_{\alpha=1}^{N}w_\alpha(\mathbf{x},t)\,\bar{\delta}\left[\boldsymbol{\xi}-\langle\boldsymbol{\xi}\rangle_\alpha(\mathbf{x},t)\right] \qquad (2)$$

- Here, N is the number of delta functions; $w_\alpha(\mathbf{x},t)$ is the weight of node α.

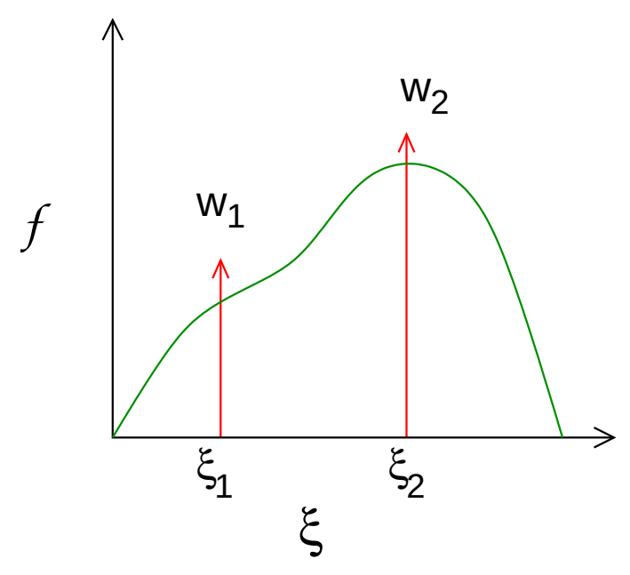

FIGURE 3: Representation of NDF f with two nodes ($N=2$)

- Substituting Eq. (2) in Eq. (1) and after subsequent manipulations and moment transformation, one obtains greatly simplified coupled equations.

$$(1-k)\sum_{\alpha=1}^{N}\langle\xi\rangle_\alpha^k a_\alpha+k\sum_{\alpha=1}^{N}\langle\xi\rangle_\alpha^{k-1}b_\alpha=\bar{S}_k+\bar{C}_k \qquad (3)$$

$$\frac{\partial w_\alpha}{\partial t}+\frac{\partial}{\partial x_i}\left(\langle u_i\rangle_\alpha w_\alpha\right)-\frac{\partial}{\partial x_i}\left((\Gamma+\Gamma_t)\frac{\partial w_\alpha}{\partial x_i}\right)=a_\alpha \qquad (4)$$

$$\frac{\partial w_\alpha\xi_\alpha}{\partial t}+\frac{\partial}{\partial x_i}\left(\langle u_i\rangle_\alpha w_\alpha\xi_\alpha\right)-\frac{\partial}{\partial x_i}\left((\Gamma+\Gamma_t)\frac{\partial w_\alpha\xi_\alpha}{\partial x_i}\right)=b_\alpha \qquad (5)$$

- For $N=2$, monovariate PBE results in 4 transport equations (easy to implement in CFD solver) and set of 4 algebraic equations which can be solved easily.

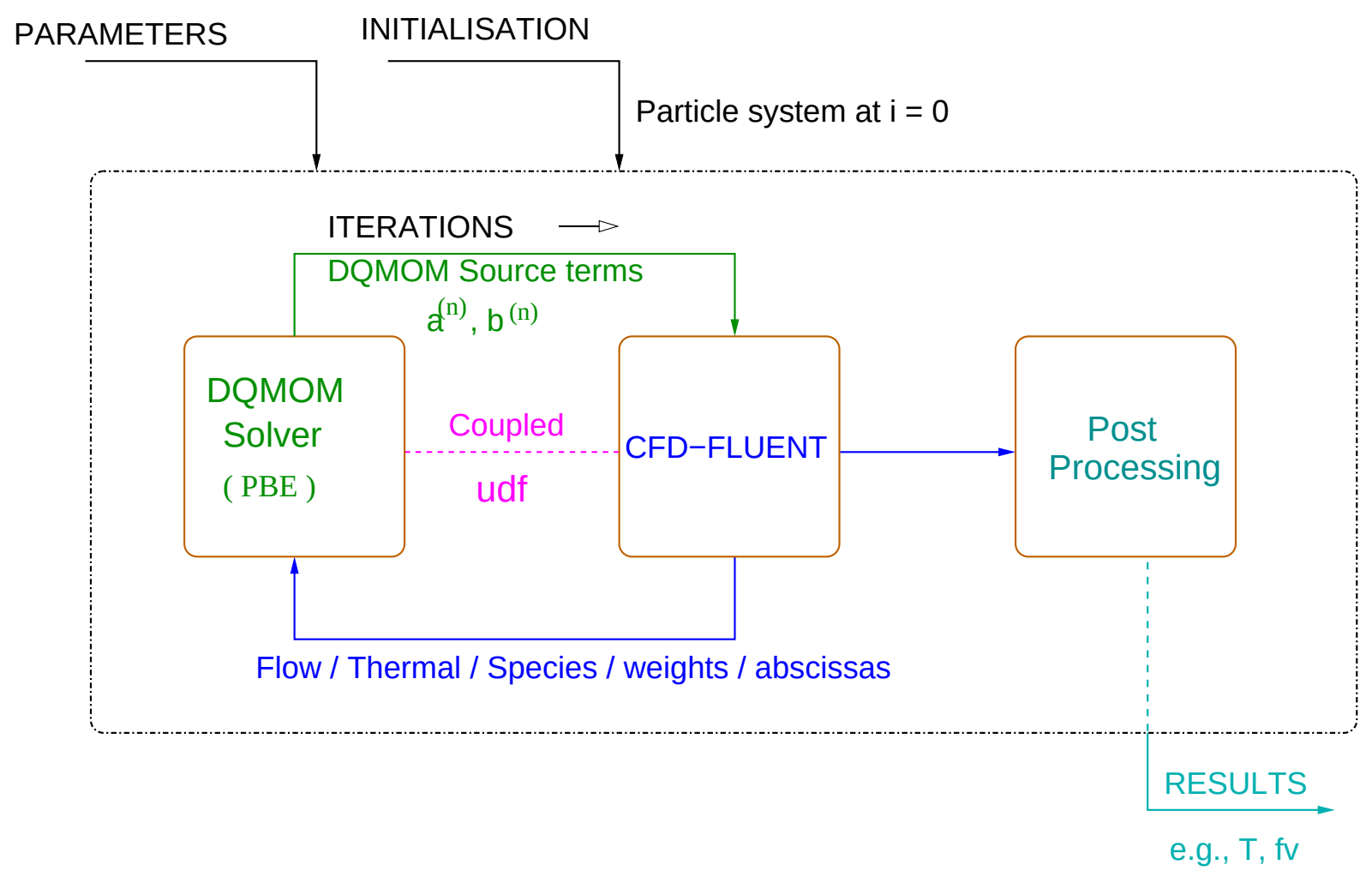

FIGURE 4: Schematic representation of DQMOM implementation in CFD solver (ANSYS FLUENT)

Computational Domain

- Pure ethylene enters vertically into the quiescent atmospheric air through a nozzle.

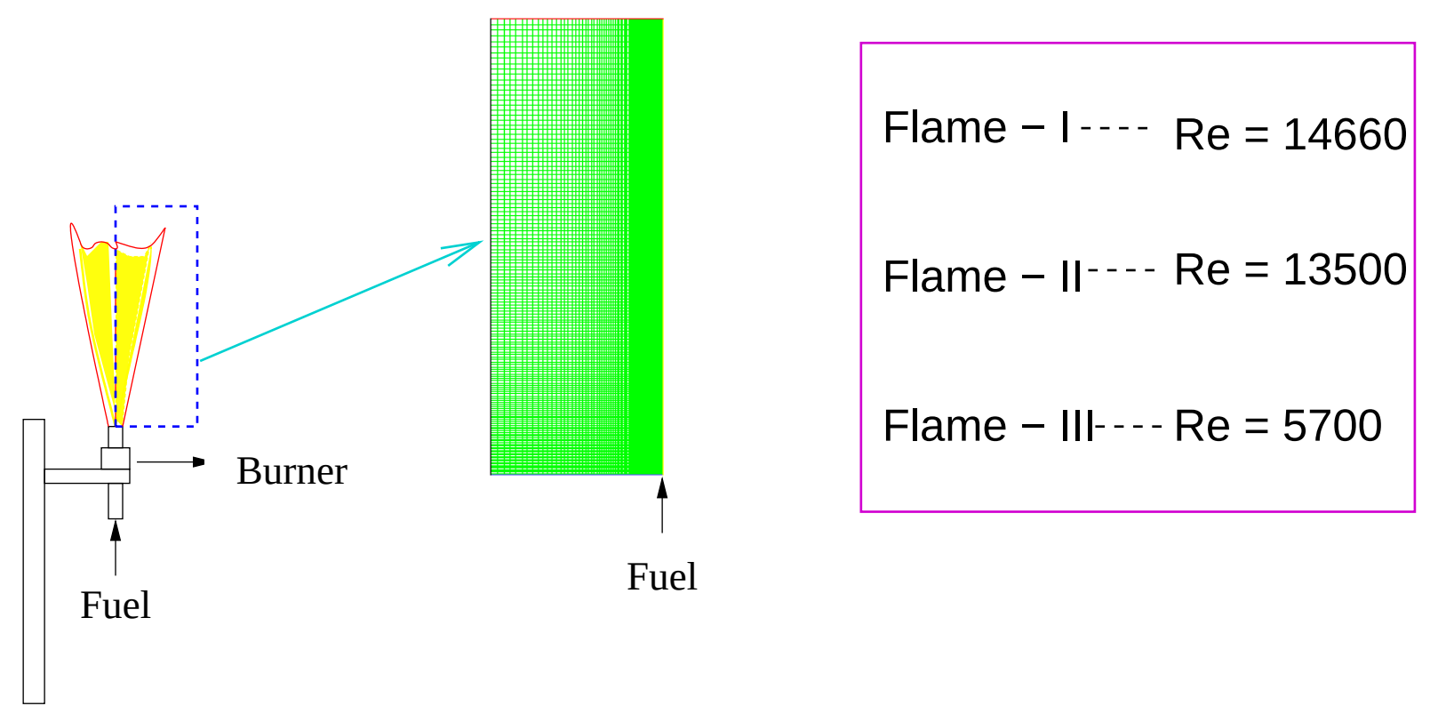

FIGURE 5: Computational domain (Symmetry)

- Three different ethylene-air diffusion flames have been chosen for our simulations, involving different flow conditions and a wide range of Reynolds numbers.

Results and Discussions

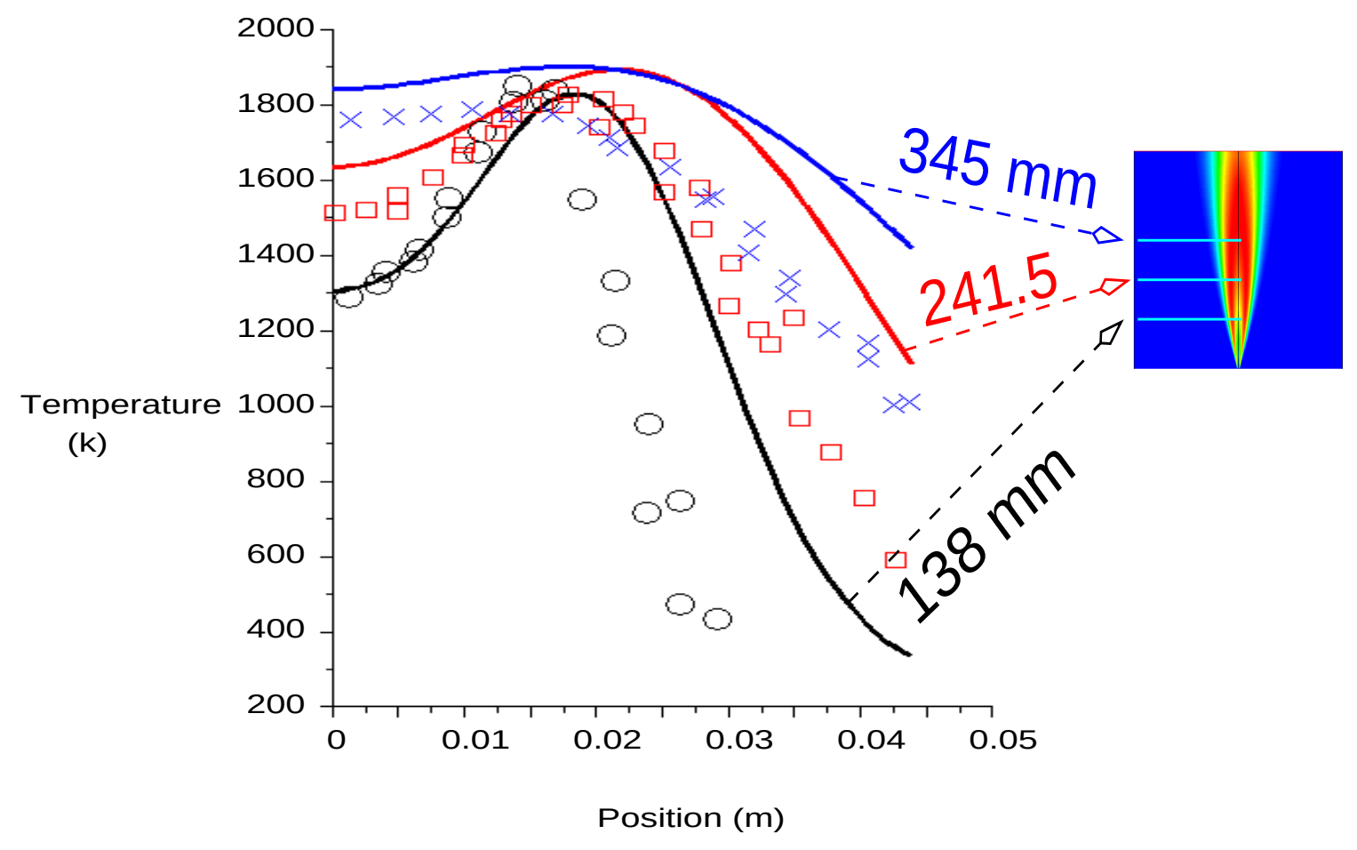

FIGURE 6: Comparison of radial profiles of temperature from CFD (lines) with experimental data points [1] (Flame-I) at different heights from burner

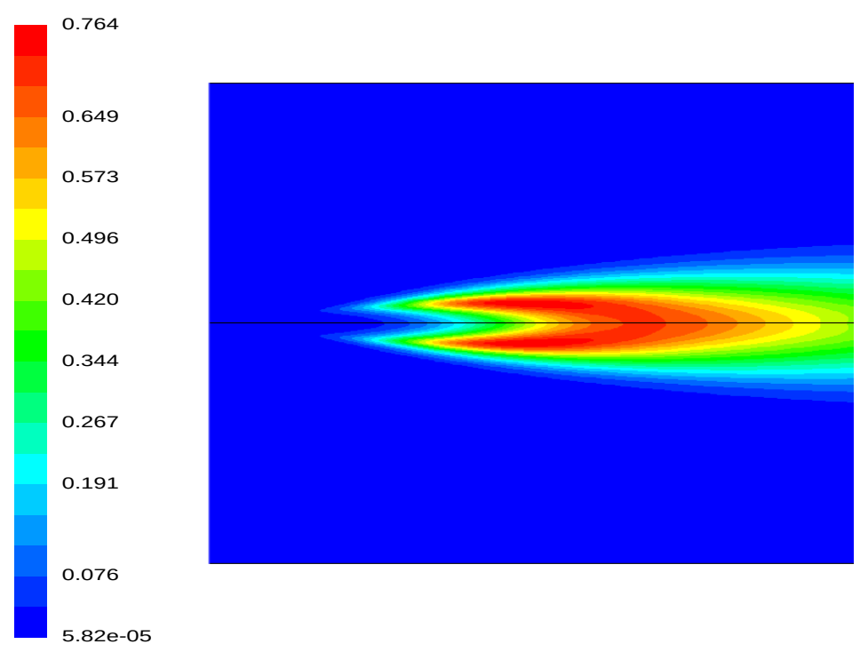

FIGURE 7: Contours of computed soot volume fraction (f_v) for the Flame-II conditions [3]

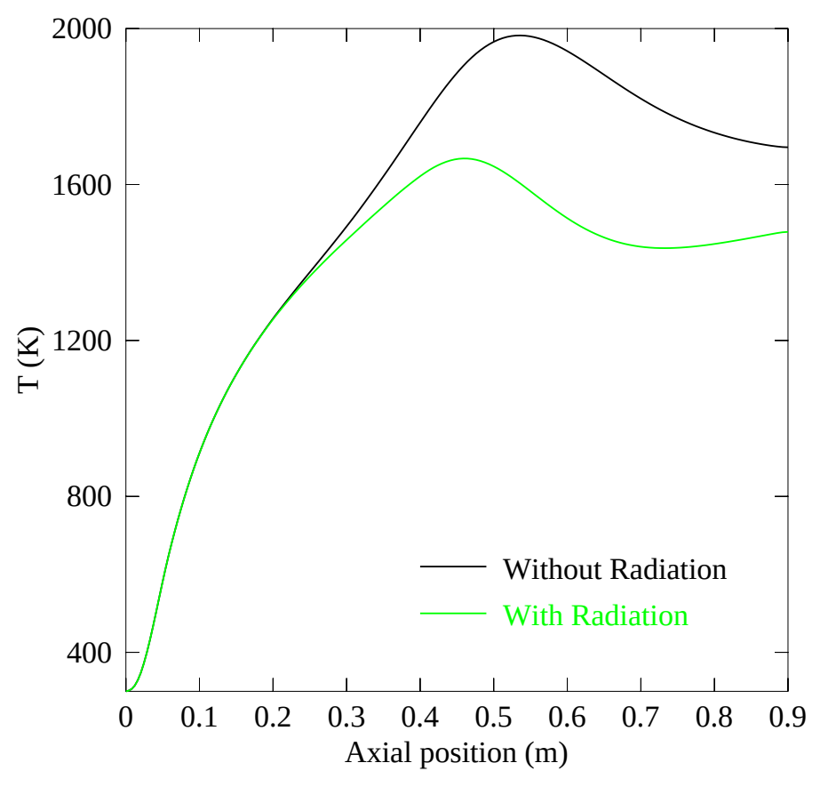

FIGURE 8: Comparison of centerline axial profiles of temperatures computed with and without radiation for the Flame-II conditions [3]

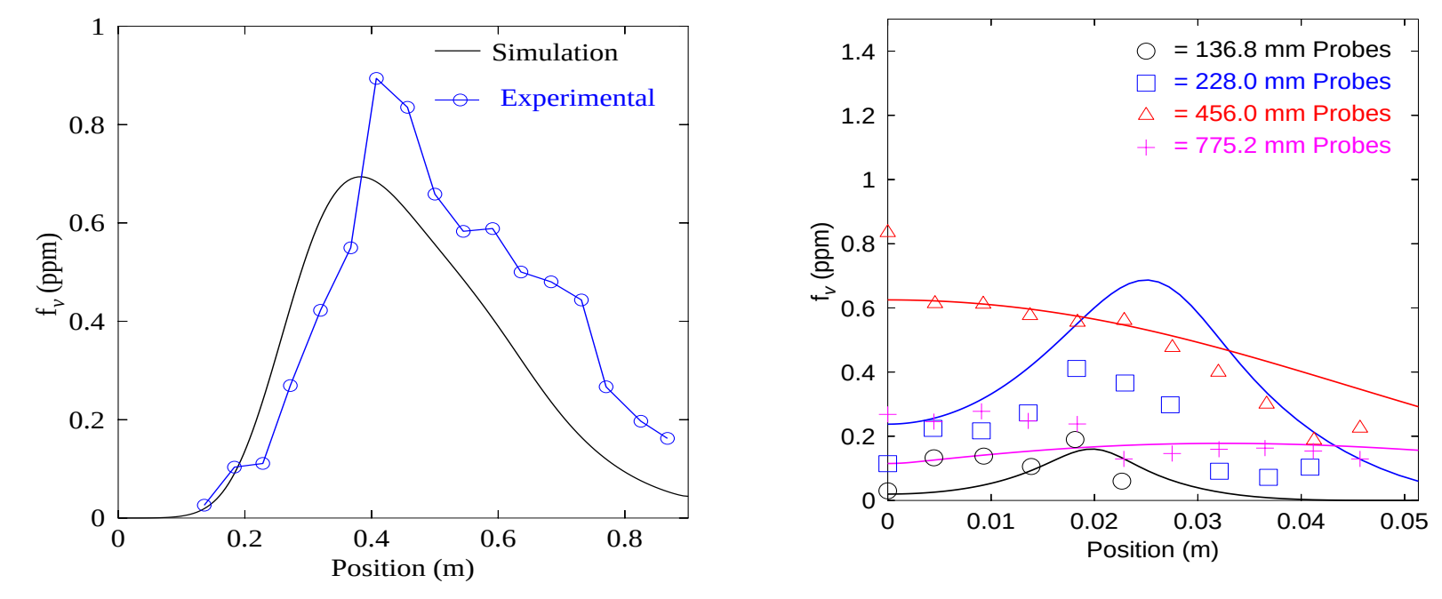

FIGURE 9: Comparison of profiles of soot f_v with the experimental data points [3] (Flame-II) in axial and radial direction

Conclusions

- Due to the adverse effects of soot on human health and environment its detailed understanding is necessary.
- Coupled processes leading to soot are extremely complex.
- Considerable heat-loss due to soot radiation.
- Results demonstrates potential of DQMOM in solving PBE at acceptable computational cost.
- Study of individual mechanisms on soot production shows that nucleation and aggregation are the most important mechanisms.
- First comparisons with experimental data are acceptable, but not perfect.

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