A Comprehensive Parametric Investigation of efficient Soot Modeling using High Fidelity Method of Moments

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Abstract

Soot is one of the most regulated pollutants in all the combustion applications like automotive engines, aviation engines, and industrial furnaces. Modeling soot formation is quite challenging due to the complex processes involved in the formation of the soot particulates. Empirical formulation based on two-equation soot models is commonly employed. However, they always need tuning for a given application and operational range. On the other hand, the sectional soot model is though highly accurate, has limitations in terms of calculation speed for industrial applications. The method of moment model offers an optimal combination of the efficiency of two-equation soot models and the fidelity of the sectional models. In this model, the soot particle size distribution (PSD) is modeled using the method of moments. In addition to the PSD, the other crucial aspect is the accurate modeling of the subprocess of soot formation like nucleation, surface growth, and oxidation. The modeling of each subprocess involves sensitive modeling parameters like the choice of nucleation precursors, the soot site density, the sticking coefficients, and the number of moments.

In this work, a comprehensive investigation is carried out for modeling soot formation using the method of moments in Ansys Fluent software. The objective of the current investigation is to develop an optimal soot modeling strategy that minimizes the need for case-based tuning and applicable for a wide range of operating conditions and different fuels. In the current work, the soot is modeled using three moments with hydrogen abstraction carbon addition (HACA) based surface growth. The gas-phase mechanism, involving polycyclic aromatic hydrocarbon (PAH), is obtained from Ansys Models Fuel library. The results are compared with experimental data for different flames and modeling guidelines are proposed based on the model performance for the validation cases.

Keywords: PSD, PAH, nucleation, Soot, Radiation, Method of moment

Introduction

Soot formation is a highly complex process and involves various sub-processes like soot inception, coagulation, growth, and oxidation. Accurate and consistent soot modeling requires resolution of these soot sub-processes by detailed chemistry along with the finite rate modeling of the coupled gas-phase combustion. In addition to the chemistry, the sectional model [1-3] with many bins are required to capture the soot particle size distribution. Detailed chemistry along with the sectional soot model [1] has a prohibitive computational cost and hence suitable only for small scale problems or canonical cases. Alternatively, the method of moments [4] along with the reduced-order combustion models like Flamelet Generated Manifold [5-6], can potentially be used to model the soot in an efficient manner and higher accuracy than traditional two-equation semi-empirical models. Although, the soot MoM is not entirely empirical, it still involves modeling constants, and

assumptions for modeling soot sub-processes. These modeling parameters, like nucleation rates, the number of moments, etc., are not universal in nature and require user intervention with change in the fuel or the operating conditions. Therefore, an effort is required to minimize the case based turning of these parameters and develop a general guideline that can be used to model soot with the method of moment for different operating conditions. This is the objective of this work, where canonical flames are used to understand the impact of model parameters for soot method and moment to find an optimal parameter for soot modeling for a range of fuel and operating conditions.

The current work is carried out into two stages, first, a series of fundamental laminar flames are studied with finite rate chemistry and detailed chemical mechanism. Premixed burner stabilized stagnation flames [7] are considered with three different fuels and four different residence time. The convergence rate is used to decide optimal number of moments. A parametric investigation is then carried out for different precursors, and their corresponding molecular weight based sticking coefficients [8]. The outcome of this investigation provided a model setup which is then used in the second part of the validations where turbulent sooting flames are considered.

The combustion in the turbulent flames is modeled using the flamelet generated manifold approach and the turbulence is modeled using two equations RANS based closures. The absorption coefficient is modeled using a weighted sum of grey gas model, including the soot-interaction. In this part of the investigations, the first turbulent sooting flame used is an ethylene turbulent jet flame [3], a target flame in the International Sooting Flame Workshop [9]. For this flame, the simulations are performed for two different inlet velocities. The second turbulent flame in the current work is a turbulent kerosene-air flame. The flame has been experimentally studied by Young et al. [10]. For this flame, the fuel is represented by two-component surrogates involving dodecane and toluene. The simulations for this flame are performed for five different operating pressure to investigate the accuracy and sensitivity of the current soot modeling approach with increasing pressure. The simulation results in this case also compare well with the experimental results. Finally, the current modeling strategy is used for a model 3D combustor to analyze the efficacy of the proposed modeling strategy.

Model Formulation

The r^{th} moment of soot particle distribution is given by

$$M_r = \sum_{i=1}^{\infty} m_i^r N_i$$

Where m_i is the mass of i^{th} class and N_i is the number density of the i^{th} size class, r is the moment number. For r =0, M_0 is the total number density of the particle, while M_1, for r=1, gives the total mass of the soot.

$$M_0 = \sum_{i=1}^{\infty} N_i \quad M_1 = \sum_{i=1}^{\infty} m_i N_i$$

The following generic transport equations are solved for the soot moments

$$\rho \frac{\partial \tilde{M}_r}{\partial t} + \rho u_j \frac{\partial \tilde{M}_r}{\partial x_j} = \frac{\partial}{\partial x_j}\left(\Gamma \frac{\partial M_r}{\partial x_j} \right) + \tilde{S}_r$$

The source term in the soot moment transport consists of the sources and the sinks from different sub-process of soot formation. These sub-processes are modeled in the following manner.

Nucleation : Nucleation is modeled using kinetic theory of collision of two polycyclic-aromatic hydrocarbons (PAH) as precursors. The rates are adjusted according to the choice of precursors.

Coagulation : Coalescent coagulation is using for continuum as well as in free molecular regime. In this work, aggregation of the soot particles is not considered.

Surface Growth and Oxidation : Hydrogen abstraction and carbon addition (HACA) [4] is used to model the soot surface growth, where acetylene is considered as primary surface growth driver. Algebraic formulations are used for the soot surface growth by assuming steady state assumptions in HACA mechanism. The oxidation is considered by O_2 as well as OH. This approach is very efficient as there is no cost associated with the solution of soot chemistry.

Test Cases

In this work, the method of moments for soot modeling is validated for four different cases. The first configuration is a laminar case while the other three cases are turbulent. For the laminar case, the gas phase combustion is modeled using finite rate chemistry with detailed mechanisms derived from Ansys Models fuel library [11]. For all turbulent cases, the gas phase combustion is modeled using Flamelet Generate Manifold (FGM). In FGM, one dimensional counter flow flamelets are generated to create the manifold, the details of the FGM are provided in earlier published works[5, 6]. All simulations in the current work are performed using Ansys Fluent [12].

Results and Discussions

This section reports the findings for each flame. Here the gas phase results are not reported and only global soot quantities like soot volume fraction and the particle number density are compared with earlier published work on with the experiments.

Burner Stabilized Stagnation Flame (BSSF)

BSSF has been used previously for different fuels and operating conditions and extensive soot measurements are available for this configuration [3,13]. This is an attractive case for soot model validations due to its simple CFD configuration, easy to parameterize, and availability of extensive experimental data for different fuels. In this work, the BSSF case has been used with three different fuels, ethylene, n-heptane, and toluene. Ansys Model Fuel Library [12] has been used to generate the chemical mechanism for these fuels. For ethylene flame, a chemical mechanism of 187 species is used, while for n-heptane and toluene cases, the mechanisms contain 217 and 181 species, respectively.

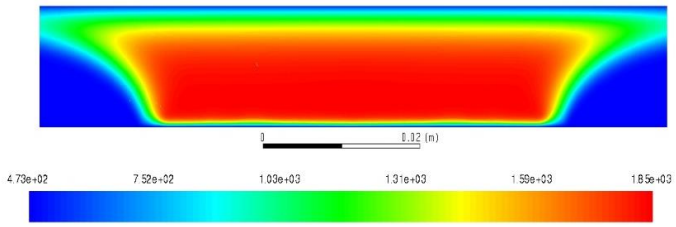


Figure 1. Gas phase temperature for BSSF

The radiative heat transfer is ignored for this case and soot is solved with one way coupling in which the soot does not affect the gas phase mixture properties. Before modeling the soot, an accurate gas phase prediction is important. Figure 1 shows the contours of the temperature for ethylene flame. The temperature and species (not shown here) are found in good agreement for with the measurements and published works[3].

Number of moments and optimal Convergence

To accurately capture the PSD, a larger number of moments are preferable. But the values of successive moments are usually two or more orders of magnitude different from each other and the system of equations becomes stiff for a higher number of moments. This leads to slow convergence and poor numerical stability. After performing numerical experiments with 3 to 6 moments for the BSSF flame, it was found that there is a marginal improvement going from 3 to higher moments. Hence, in this work, all results are reported using three moments. It can be seen from figure 2 the method has an excellent convergence rate with three moments.

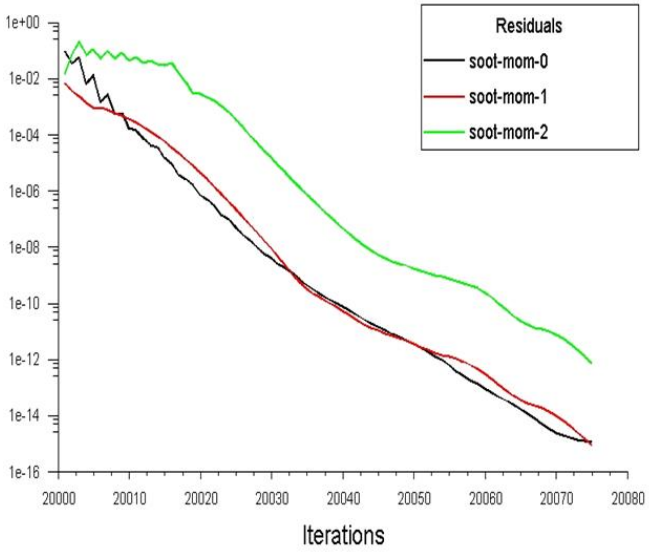


Figure 2. Convergence Rate of method of moments with three moments for BSSF

Table 1. Different fuel and separation distance between inlet and stagnation plane

Fuel	Separation distance or Burner Heights (mm)
Ethylene	6,8,10,12
n-Heptane	7,8,10,12
Toluene	5,6,8,9

PAH Precursor

Choice of soot precursor directly affects the rate of nucleation and eventually the soot yield. The nucleation is modeled using the kinetic theory of collision, which usually overpredicts the rates. The corrections to the nucleation rates are made using sticking coefficients. Therefore, not only the choice of the precursor is important, but a parameter estimation is required for the sticking coefficient for different precursors. In this work, the different precursors have been used to study the BSSF flame and the impact on total soot is investigated for different operating conditions. Table 2 summarizes the different PAH species used as precursors. The value of the sticking coefficient depends on the precursor(s) chosen. Blanquart and Pitsch [8] provided the sticking coefficients for commonly used precursors. Here, and a curve fitting of the sticking coefficient is done as a function of molecular weight from published values of Blanquart and Pitsch.

Table 2. PAH species to be used as precursors for nucleation

PAH	Formula	Sticking Coefficient
Benzene	**C6H6**	**0.00014**
Acenaphthylene	**C12H8**	**0.002**
Pyrene	**C16H10**	**0.00623**

Figure 3 shows the soot volume fraction at the stagnation plane for different fuels and burner heights using different PAH species for nucleation. For very low burner height, the residence time is quite low, and overall soot yield is small, leading to a higher mismatch. For burner height of 8 mm and above, the predictions had good match with measurements for Acenaphthylene as a precursor. Benzene as PAH consistently yields significant over prediction in the soot yield, which is expected as benzene concentration in the gas phase is an order of magnitude higher than the other PAH used. Pyrene is often considered as a reasonable PAH species, but in the current framework of the method of moments, the choice of pyrene consistently under-predicts the soot. Coronene (results not shown here) lead negligible soot.

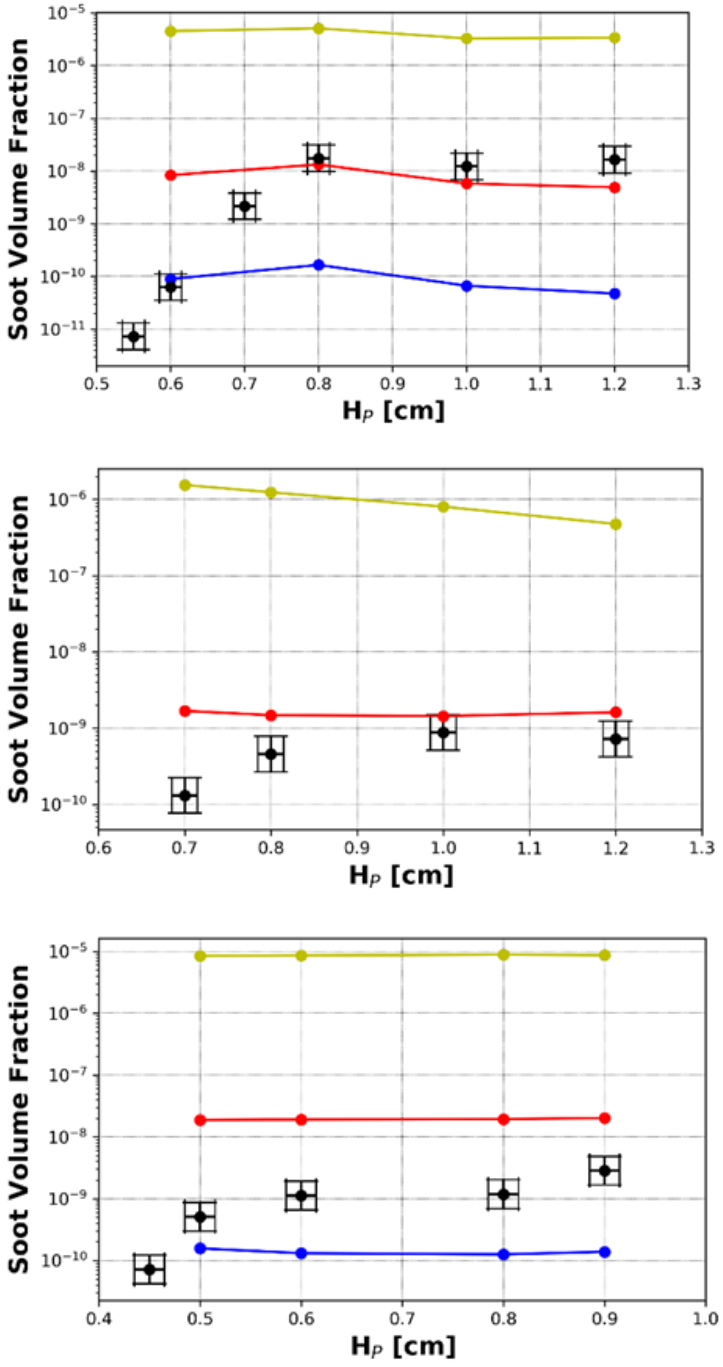


Figure 3. Soot volume fraction with height of stagnation plane with different precursors

The results of the BSSF are encouraging as the MoM solution approach is efficient and used here without any case specific tuning. The efficacy of this approach and modeling parameter is to be tested for turbulent combustion problems. In the next sections, all the cases are turbulent combustion. From BSSF results of 12

different simulation, the choice of PAH is Acenaphthylene with its curved fitting fix value of sticking coefficient [12]. In the remaining work of this paper, all flames use Acenaphthylene as PAH with sticking coefficient in table 2.

Adelaide Jet Flames

Adelaide Jet Flames [9], a series of six different jet flames, have been a target flame in the international sooting flame workshop. These canonical flames also have simple geometry but additional complexity due to turbulence. Therefore, these flames provide a second level of validation case after the laminar flames. In the current work, only two of the six flames have been used to investigate the current soot model. The two flames are flame-2 and flame-5, the first one has Re=15000, while the second one is with Re=8000. The fuel for both flames is a mixture of 64.5% C2H4 by mass, 4.7% H2 with N2. The boundary conditions of these flames are summarized in Table 3.

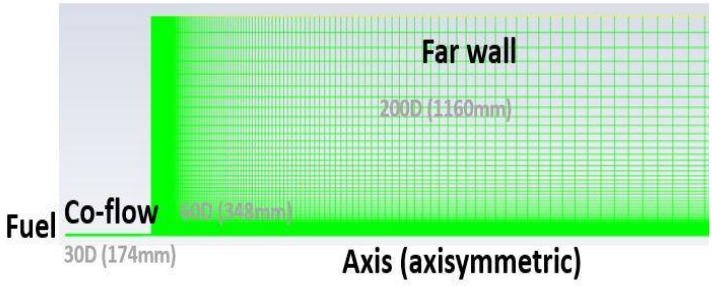


Figure 4. Geometry and Grid for Adelaide Jet Flames

Table 3. Boundary conditions for Adelaide jet flames

Variable	Fuel Inlet	Co-flow inlet
T (K)	294	294
V (m/s)	42.4 (Flame 2) 23.5 (Flame 5)	1.1
Turbulence Intensity	5%	1.5%

To get the correct jet spreading, RNG k-ε model has been used. The combustion is modeled using FGM. Yang et al. [14] have shown the importance of radiative heat transfer for this flame, therefore, in the current work, the radiative heat transfer is also included using P1 radiation model. The absorption coefficient of the gas mixture is modeled using mass-weighted summation of CO2, H2O, CH4 and CO, the data for individual species is taken from Barlow et al. [15]. For the soot absorption coefficient, a linear function of soot mass fraction and temperature is used. The total absorption is the summation of gas and soot absorption coefficients.

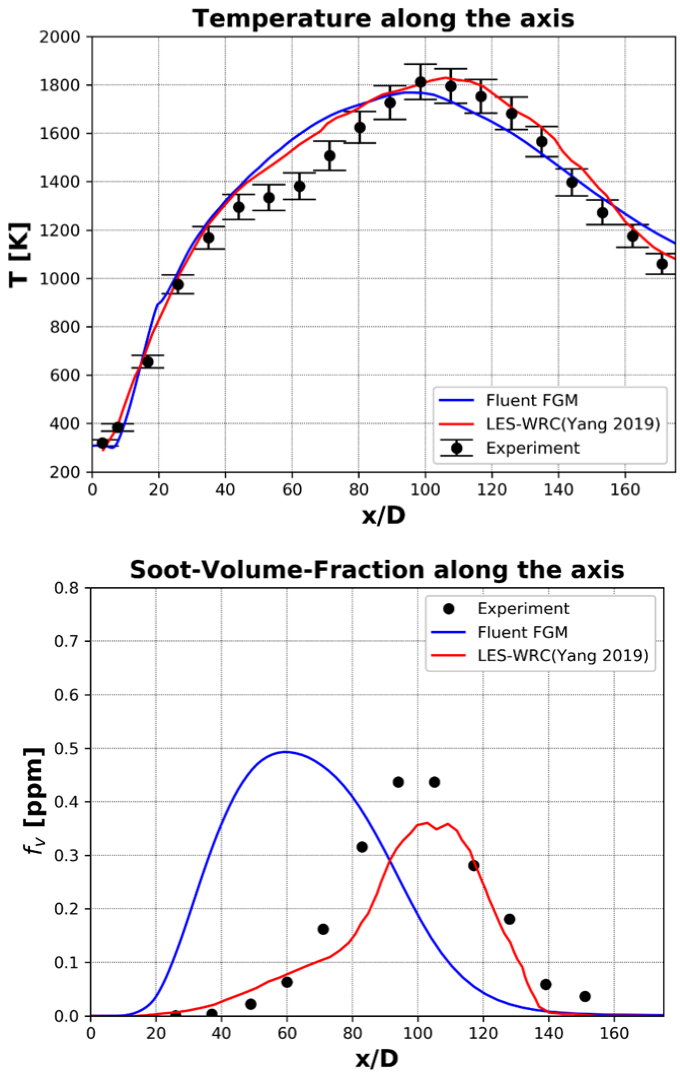


Figure 5. Axial profiles of Temperature and soot volume fraction at centerline for Adelaide Jet Flame-2. Legends: Symbols-experiment, Fluent FGM: Current work, LES-WRC (Yang 2019): Published results of Yang et al.

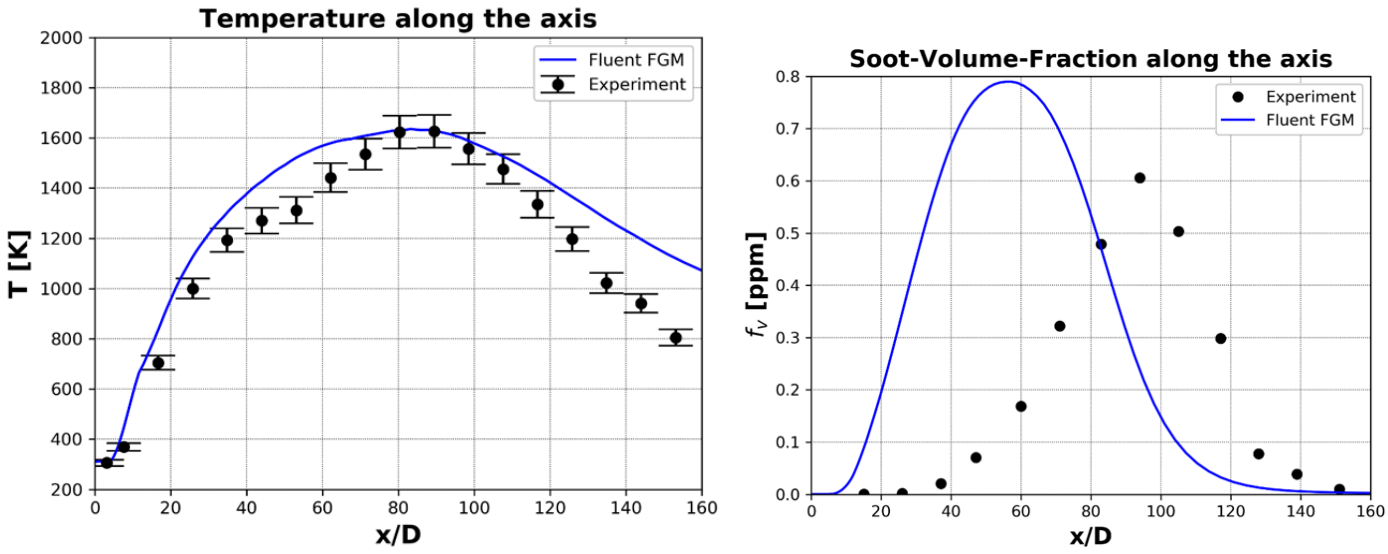


Figure 6. Axial profiles of Temperature and soot volume fraction at centerline for Adelaide Jet Flame-5

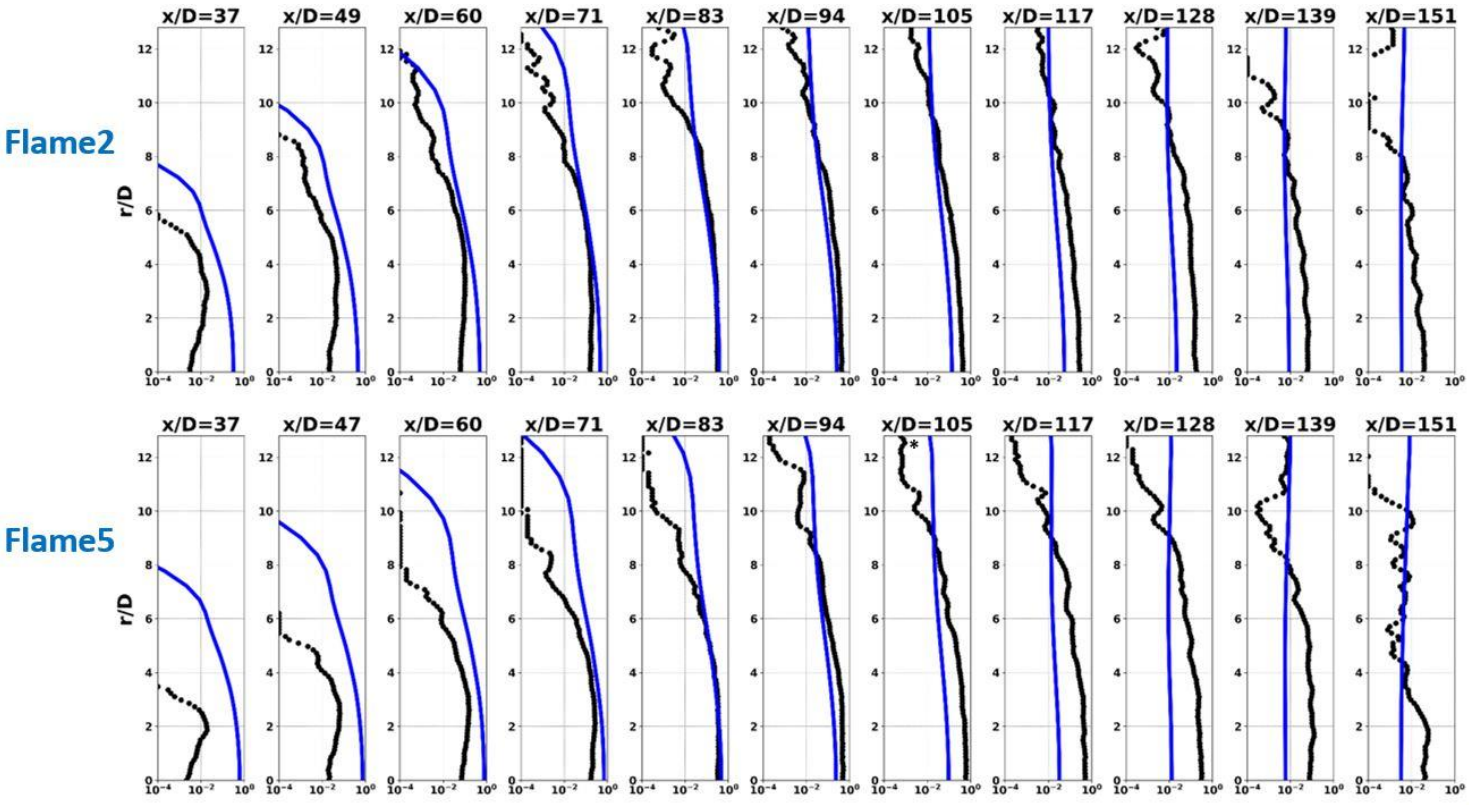


Figure 7. Radial profiles of soot volume fraction at different axial location for Adelaide Jet Flames

Figure 5 shows the temperature profile and soot volume fraction at the centerline along axial direction for Flame-2. The current results are compared with experimental data as well as with earlier published LES results of

Yang et al. [14] using different reaction mechanisms and hybrid method of moment. There is a good match in temperature. The soot volume fraction variation and peak are predicted with good accuracy by the current approach. However, the profile of the soot volume fraction is shifted upstream compared to the LES results of Yang et al. as well with measurements. The same trend is seen in figure 6 for Flame-5, where temperature profile and soot variation are captured well but the upstream shift of soot is seen for this case as well. The upstream shift of profiles is often observed with RANS simulations for round jets. Figure 7 shows the radial profiles of soot volume fraction for different axial locations. The soot trends with the current model are captured well for both flames up to the axial location of 105D. The current model predicts the early burn out of soot due to axial shift of profile as seen in the axial plots.

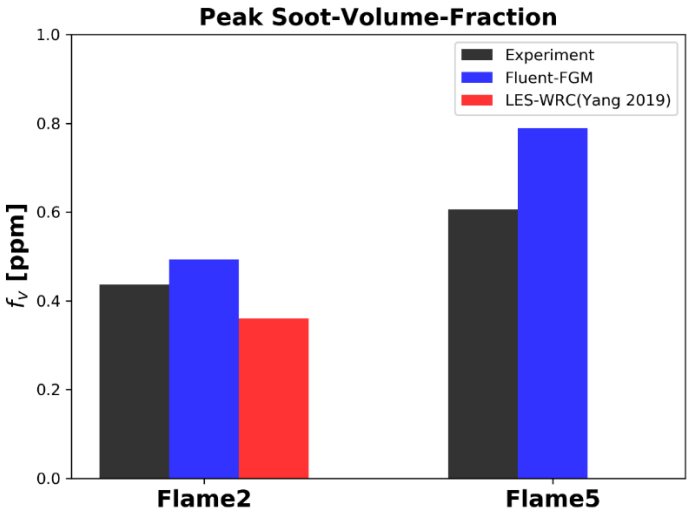


Figure 8. Peak soot volume fraction with change in residence time for Adelaide Jet Flame

Figure 8 shows the change in soot volume fraction with change in inlet velocity from Flame 2 to 5. The current model captures the trend well with the experiments.

Kerosene Flame

The third test in the current work is a kerosene/air flame [3] with a co-flow fuel-air configuration. A 2D axisymmetric model is created with ~31000 grid points. The fuel is represented by a two-component surrogate of Dodecane (~69%) and Toluene (~31%). The current simulations are done for five different pressure. The operating conditions of all five cases are summarized in Table 4. The turbulence here is modeled using standard k-ε model with C1ε=1.6. Figure 9 shows the axial profile of soot volume fraction in PPM along the centerline. It can be seen from the figure the MoM predictions match closely with experiments for all cases except the first one. For the first case, the soot match is only reasonable for lower axial distances, where nucleation happens. This under-prediction has been seen with two-equation semi-empirical models as well [16]. Other than the nucleation, soot formation involves three other subprocesses, coagulation, growth, and oxidation. The lower peak is indicative of under-prediction in coagulation or surface-growth. The detailed investigation of the coagulation process in isolation with a change in pressure is not considered in the current scope of work. In the present work, the primary focus is to optimize the choice of precursor species and its sticking coefficient to capture the trends accurately with a change in operating conditions. Although the current model undergoes from under-prediction for low pressure to the over prediction of soot for higher pressure, it is able to capture the trend of peak soot volume fraction with a change in pressure correctly as can be seen in figure 9. From the laminar flames and these two turbulent flames, the soot MoM with 3 moments and a single precursor consistently captured the impact of operating conditions in the soot formation.

Model 3D Combustor

The case studied in the previous sections were primarily 1D/2D case with simplified flow configuration. In this section, the current soot modeling approach is studied for a 3D swirl combustion operating at high pressure, relevant to gas turbine conditions. This case does not have any experimental data and only used to access the robustness and applicability the current soot MoM settings for practical combustors. The schematic of the combustor is shown in figure 10. The computational domain is a 20-degree sector and meshed with 1.6 M cells. The incoming air is use from primary combustion as well as for dilution. The swirl is generated by a series of vanes, and the fuel is injected in gaseous state from a small triangular slot in the vicinity of the swirler blades. The flame is stabilized by the swirl at a higher radial location as shown in the temperature contours in figure 11.

Table 4. Operating conditions of the Kerosene-air flame for 5 different pressure

Parameter	Flame A	Flame B	Flame C	Flame D	Flame E
Pressure (bar)	1.0	2.03	2.70	3.72	4.81
Fuel flow rate (kg/s)	0.000133	0.000158	0.000208	0.000208	0.000308
Fuel exit temperature (K)	598	633	623	626	630
Air flowrate (kg/s)	0.005400	0.015933	0.015417	0.019883	0.021583
Air exit temperature (K)	288	293	291	292	296

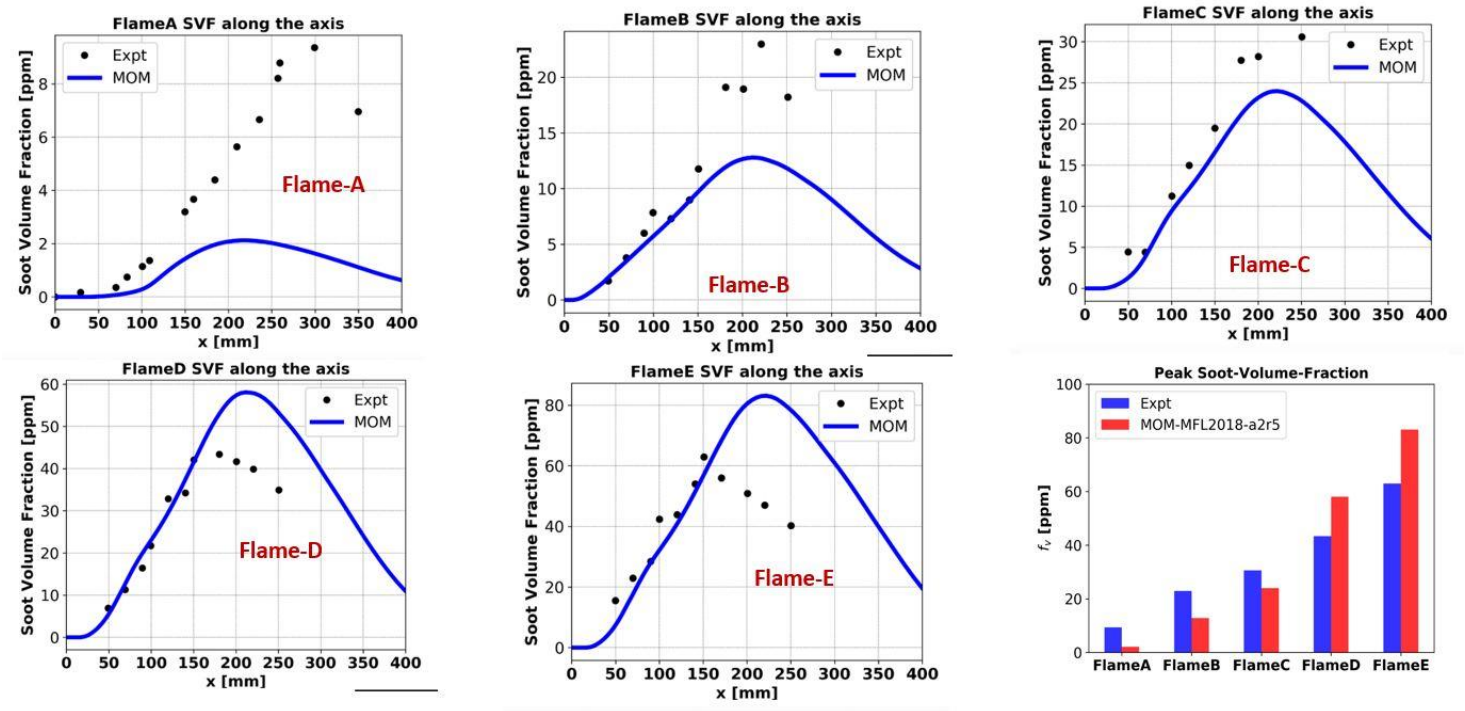


Figure 9. Soot formation with different pressure conditions for kerosene-air flame

The combustor operates at 15 atm with kerosene as fuel. The combustion is modeled using RANS-FGM approach, and the radiation is modeled using Discrete-Ordinate (DO) [12] model. The soot settings are the same as used in the kerosene-air flame in the previous section. Using the same soot modeling settings as used in earlier sections, this case also shows robust and fast convergence for soot number density and the volume fraction as shown in figure 12.

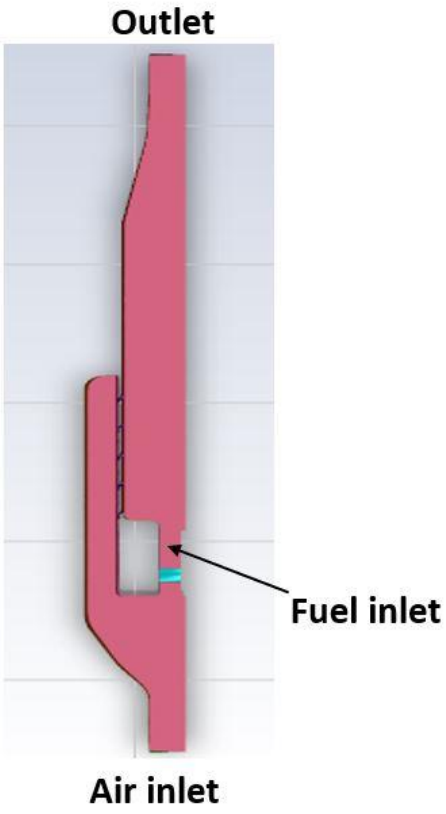


Figure 10. Geometry of the 3D model combustor

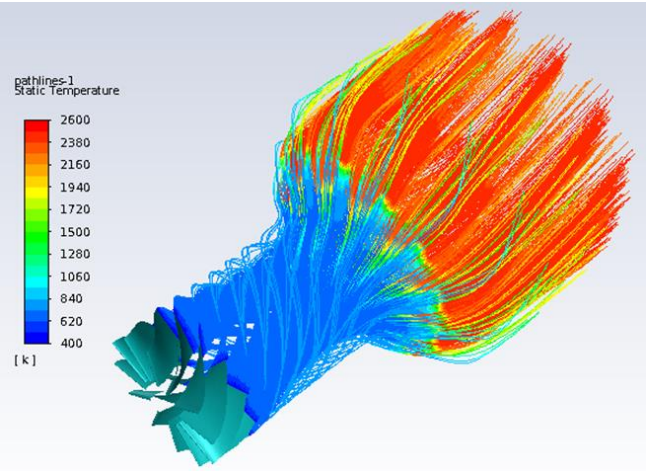
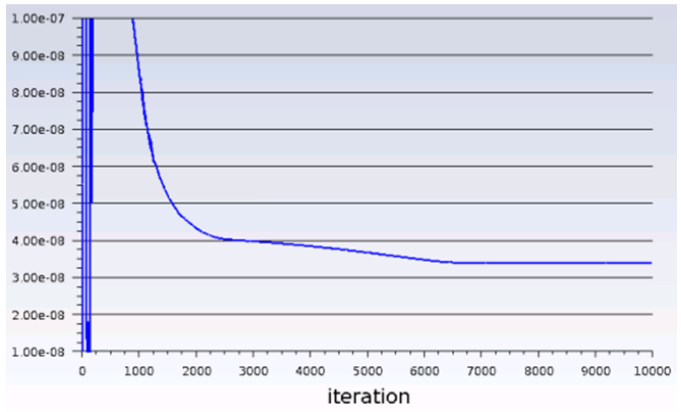


Figure 11. Tempertaure contours of the 3D model combustor



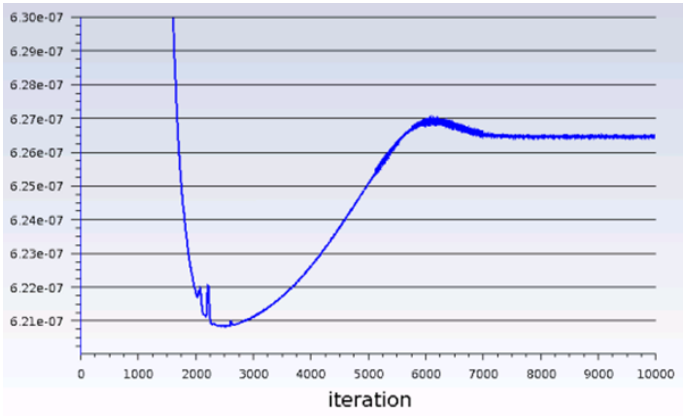


Figure 12. Convergence rate of particle number density (above) and soot volume fraction (below) for high pressure 3D model combustor

Conclusion

The current work proposed a strategy on soot nucleation modeling and the number of moments with the method of moments. The investigation is carried out with a single choice of precursor and its sticking coefficient for one-dimensional flames. The guidelines are established based on the detailed 1D simulations. The soot modeling strategy proposed here has consistently predicted the correct soot trends with a change in a range of operating conditions, like change in fuel type, different residence time and a change in operating pressure for two turbulent flames. The current soot modeling with MoM is done without any case specific tuning of the modeling parameters. The paper outlines an optimal solution strategy and modeling guidelines that allow high fidelity soot modeling in the industrial application in an efficient manner.

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