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An assessment of the sectional soot model and FGM tabulated chemistry coupling in laminar flame simulations

Abhijit Kalbhor*, Jeroen van Oijen

*Corresponding author.
E-mail address: a.j.kalbhor@tue.nl (A. Kalbhor).

1. Introduction

Clean combustion has become a primary agenda in present combustion research, and stricter regulations have been introduced to lower soot emissions, especially from aircraft engines. In addition to soot concentration, the central focus of the future emission norms will be on regulating the soot particle size and number density. Hence, to comply with increasingly stringent regulations on emissions, the role of accurate predictive soot models in the design and development of futuristic combustion systems has become critical than ever. Therefore, in recent years, there is a great interest in the advancement of detailed soot models to capture the accurate size distributions of soot particles in combustion processes. Nevertheless, the complexity of the soot formation process, and gaps in the present understanding of it, make the development of accurate, computationally-efficient soot models very challenging.

In particular, the accurate description of the collision phenomenon associated with the soot particles is a critical and computationally intensive aspect of detailed soot modeling. To effectively capture particle dynamics, several numerical techniques such as the method of moments (MOM) [1–3], discrete sectional methods [4–8], Monte Carlo (MC) [9,10] have been proposed in the literature. In the discrete sectional method, which is central to the present study, a continuous particle size distribution function (PSDF) is discretized into a finite number of sections having representative particle sizes. The accuracy of the sectional soot model, thus, can be improved by increasing the number of sections used. However, it is obvious that adding more sections also increases the number of transport equations to be solved for soot variables (mass fraction/number density). Hence, the trade-off between computational cost and approximate accuracy becomes an
important factor while discretizing particle size distribution in sectional models.

Besides particle dynamics, the detailed modeling of soot formation processes demands accurate predictions of gas-phase soot precursors. Therefore, to correctly predict the concentrations of soot precursors and other the major species associated with the soot growth and oxidation processes, detailed kinetic schemes involving hundreds of species and thousands of elementary reactions are required in combustion simulations. However, solving combustion chemistry by transporting such a large number of species poses a principal challenge in terms of computational cost. Therefore, the use of detailed kinetic mechanisms is practically not feasible in large-scale combustion simulations considering their computational grid requirements. Moreover, a wide range of time scales associated with the formation of chemical species and fluid mixing introduces additional complexities in their numerical treatment. Hence, several chemistry-reduction strategies are generally available to optimize the overall CPU time by replacing the use of detailed gas-phase chemistry. In this context, the flamelet-based tabulated chemistry approaches have proved to be effective and widely used strategies to simulate sooting flames in terms of reduced computational cost and relatively good precision level.

Over the past decade, in many of the numerical simulations investigating soot formation, various approaches of modeling flamelet-based tabulated chemistry with semi-empirical and detailed soot models have been proposed [3,11-15]. In an earlier study, Dorey et al. [11] adopted FPI (Flame Prolongation of ILDM) tabulated chemistry coupled with a semi-empirical, two-equation soot model for estimating soot formation in ethylene premixed flames. In their approach, the mass exchange between the soot and gas phase was incorporated by transporting additional equations for the species involved in the soot model. Dupoirieux and Berrier [13] later validated this strategy in LES of the DLR burner used by Geigle et al. [16]. In LES of swirl-stabilized ethylene-air flames, Franzelli et al. [17] compared the performance of the full tabulation approach and the hybrid method proposed by Leccoq et al. [18], using a two-equation soot model. Besides steady flamelet based models, unsteady/interactive flamelet based techniques [5,19] have also been explored for modeling sooting flames and have shown good agreement with experiments. However, many of the existing flamelet-based tabulated chemistry approaches employed for soot modeling, have been examined under turbulent flame conditions. Therefore it is of interest to assess the performance of tabulated chemistry-based soot modeling methods in more simplified canonical configurations by isolating the influence of additional complexities induced by turbulence. Such analysis is particularly important to identify the limitations of specific approaches and underlying mechanisms behind the disparity in the level of accuracy associated with them before extending to turbulent conditions. Concerning detailed soot models, Mueller and Pitsch [3,20] applied an integrated kinetics-based LES approach with the Hybrid Method of Moments (HMOM)-based soot model, and Radiation Flamelet Progress Variable (RPFPV) tabulated chemistry for soot prediction in an aircraft gas turbine combustor. In their strategy, an additional transport equation was introduced to model the PAH during the soot formation by assuming a quasi-steady state of PAH dimers. This lumped PAH transport approach is widely used when considering tabulated chemistry models for sooting flames. It has also been extended to the sectional methods-based soot model by Rodrigues et al. [15]. Recently, Wick et al. [21] conducted a combined a-priori and partial a-posteriori analysis of errors in soot predictions associated with the flamelet-based combustion model FPV [22], using Direct Numerical Simulation (DNS) data of a temporally evolving turbulent jet diffusion flame. The quantitative uncertainties in soot predictions were found to be mainly related to the accuracy of the chemical mechanism in capturing the PAH formation and modeling of the PAH source term for PAH-based processes (nucleation and condensation). It was further observed that, lumped PAH transport approach is particularly sensitive to appropriate scaling relations that are applied to the dimer formation rate to minimize the errors associated with PAH-based processes.

Therefore, although many of the flamelet based techniques have been applied for modeling sooting flames and even widely used, there is still no general agreement about the best approach. Hence, it is of interest to devote further efforts in identifying alternate strategies to accurately model the contributions of soot-related processes with somewhat different tabulated chemistry methods. In this regard, modeling sooting flames with FGM tabulated chemistry would be of interest. There have been limited studies on coupling FGM tabulated chemistry with soot models for applications to diffusion flames [23,24] and spray combustion [25]. Notably, Zimmer [24] explored various strategies for storing soot information in the FGM database for application to laminar diffusion flames. The approach of storing the rates associated with individual soot processes showed a good qualitative agreement between the FGM and detailed chemistry solutions. However, these studies were based on semi-empirical soot models and thus, emerge as inadequate in terms of capturing particle size distributions. The coupling of the FGM tabulated chemistry with a sectional method-based soot model has not been fully explored. The authors are aware of the only work by Hoerle [20] on this subject, which essentially motivates the current study.

This work aims to present different approaches for coupling the FGM tabulated chemistry with the sectional soot model and assess their performance for the accuracy of soot prediction and the overall CPU time. In this context, the one-dimensional laminar counterflow flame provides a simplified platform for analyzing contributions of various soot processes at a fundamental level by avoiding complexities arising from turbulence-flame interactions. The soot characteristics obtained from proposed FGM-DSM strategies are compared against the experimental measurements from the literature and corresponding detailed chemistry solutions to examine their predictive capabilities for the soot formation in laminar counterflow flames. Furthermore, the accuracy of FGM-DSM coupling approaches is evaluated for variations in strain rate and reactants composition. In turbulent conditions, flames typically encounter fluctuating strain rates. Hence, to examine if the FGM database built from steady-state flamelets can be extended to evaluate the dynamic response of soot formation to the unsteady conditions, the counterflow flames are subjected to time-dependent strain rates, and the transient evolution of soot is investigated for various FGM-DSM approaches. The present study thus serves as a preliminary step towards the application of FGM for modeling soot formation in complex, unsteady, and turbulent combustion environments.

The paper is organized as follows. The numerical methodology, sectional soot model, and FGM-DSM coupling approaches are briefly explained in Section 2. In Section 3, the results for the assessment of FGM-DSM approaches under the steady conditions are presented. In Section 4, the transient response of soot formation to unsteady conditions is studied for different FGM-DSM approaches. The computational performance of the FGM-DSM approaches is analyzed in Section 5. Conclusions are summarized in Section 6.

2. Numerical modeling

2.1. Governing equations and sectional soot model

Numerical simulations are performed by using the in-house developed fully implicit solver CHEM1D [27,28]. The set of
one-dimensional governing equations representing conservation of mass, momentum, energy, chemical species, and transport of soot mass fractions are solved.

The sectional soot model employed in the present study is based on the work of Hoerlle and Pereira [29]. In the soot model, the continuous soot PSDF is divided into \( n_{\text{sec}} \) discrete sections with representative particle sizes [4,5]. In each section \( i \), a standard transport equation for the soot mass fraction \( Y_{\text{s},i} \) is solved along coordinate \( x \) perpendicular to the flame surface as:

\[
\frac{\partial (\rho Y_{\text{s},i})}{\partial t} + \frac{\partial (\rho u Y_{\text{s},i})}{\partial x} = -\frac{\partial}{\partial x} \left( \rho D_{\text{s},i} \frac{\partial Y_{\text{s},i}}{\partial x} \right) + \rho_s (Q_{\text{nuc},i} + Q_{\text{cond},i} + Q_{\text{coag},i} + Q_{\text{sg},i} + Q_{\text{migr},i}) \quad i = 1, 2, \ldots, n_{\text{sec}}
\]

where \( u \) is the gas velocity, \( V_t \) is the particle thermophoretic velocity (computed using Friendlander et al. [30] method), \( D_{\text{s},i} \) is the molecular diffusion coefficient of soot (assumed to be 1% of the average gas diffusion), \( \rho_s \) is the soot density (equivalent to the density of solid carbon). The sectional source terms \( Q \) are evaluated from the contributions of physical and chemical growth processes associated with soot particles including soot nucleation (nuc), PAH condensation (cond), coagulation (coag), surface growth (sg), and soot oxidation (ox). In the model, a fixed interval of volume from 3.43 \( \times 10^{-22} \) cm\(^3\) (carbon-equivalent volume of two soot precursors molecules) to 5.23 \( \times 10^{-10} \) cm\(^3\) is discretized into 60 sections using a geometric progression specified in Hoerlle and Pereira [29]. A constant soot volume fraction density distribution is assumed within each section [14,31].

Nascent soot particles are presumed to nucleate through the dimerization of two gas-phase PAH species (pyrene molecules (A4) here). The condensation of PAHs on the surface of soot particles is modeled through Brownian collisions between PAH and soot particles following Smoluchowski equation with unity condensation efficiency [32]. The particle–particle coagulation process between the sections is evaluated using the model proposed by Kumar and Ramkrishna [33]. The soot surface growth is modeled by the standard hydrogen-abstraction-C\(_2\)H\(_2\)-addition (HACA) mechanism [34]. Soot oxidation through surface reactions of OH and O\(_2\) is included. The fraction of the surface sites available for chemical reactions, \( \alpha \) is chosen as a function of temperature and particle size following Appel et al. [34]. For simplicity, the radiation effects of soot have not been included in the model. In the case of detailed kinetics, the consumption of gaseous species as a result of soot formation is accounted for in the species conservation equations through a two-way coupling. Simultaneously, the contribution of soot into the thermodynamic properties of the system (e.g., \( \rho, h \)) is also included. Details of the soot model and validation can be found in Hoerlle and Pereira [29].

#### 2.2.2. Manifold generation

The first step consists of building a manifold from flamelets. In FGM, the choice of representative flamelets and control variables depends on the combustion system to which it is being applied. Therefore, for non-premixed flames, the mixture fraction \( Z \), and reaction progress variable \( \gamma \) are considered as the most relevant control variables, describing mixing and chemical reaction processes, respectively. The mixture fraction \( Z \) is expressed in terms of the element mass fractions following Bilger’s [39] definition

\[
Z = \frac{Z_0 - Z_{\text{fu}}}{Z_{\text{sec}} - Z_{\text{fu}}}, \quad Z_{\text{sec}} = 2Z_{\text{H}_2} + \frac{1}{2}Z_{\text{H}_2} - Z_O
\]

where \( Z_s \) and \( W_s \) denote, respectively, the elemental mass fractions and atomic weights of element \( j \). The subscripts \( fu \) and \( ox \) represent quantities for pure fuel and oxidizer, respectively. For the reaction progress variable \( \gamma \), the necessary constraint is that it should be monotonous from unburned state to chemical equilibrium state to facilitate an unambiguous mapping of dependent variables. Hence, for the present study, a suitable progress variable is chosen as a linear combination of species mass fractions (\( \gamma \)) with

\[
\gamma = \frac{Y_{\text{H}_2} - Y_{\text{H}_2,0}}{W_{\text{H}_2}} + \frac{Y_{\text{CO}} - Y_{\text{CO},0}}{W_{\text{CO}}} - 0.6 + \frac{Y_{\text{CO}} - Y_{\text{CO},0}}{W_{\text{CO}}} - 2\frac{Y_{\text{A4}} - Y_{\text{A4},0}}{W_{\text{A4}}} - 0.1 \frac{Y_{\text{O}} - Y_{\text{O},0}}{W_{\text{O}}}
\]

It is worth noting that, in the FGM simulations, the choice of progress variable is of great importance as it essentially determines the soundness of the mapped thermodynamic state of the mixture in the manifold. In the present study, the definition of progress variable \( \gamma \) (Eq. (3)) is obtained through a guess and check method, mainly based on previous experience. In general, the standard definitions of \( \gamma \) are expressed by the major combustion product species such as \( \text{H}_2\text{O}, \text{CO}, \text{CO}_2, \text{H}_2 \). However, since A4 is the PAH species in the current soot model, the contribution A4 is considered in the present progress variable definition to improve the mapping of its chemical source term evolution. Besides, the inclusion of \( \text{C}_2\text{H}_2 \) species responsible for the soot surface growth is also tested while choosing the \( \gamma \) definition (although not included after the assessment) following [26]. The weight factors for \( \text{H}_2 \) and \( \text{O}_2 \) in \( \gamma \) definition are chosen through trial and error approach to reproduce the concentrations of important gas-phase species and the shape of soot volume fraction profiles against their detailed chemistry counterparts. The current choice of \( \gamma \) facilitates the unique mapping of the thermo-chemical variables in the \( Z-\gamma \) space. The influence of \( \gamma \) definition on the accuracy of soot prediction by FGM-DSM approaches is highlighted in Section 3.3.

The manifold is constructed from the solutions of steady and unsteady adiabatic counterflow diffusion flamelets. During this, the steady flamelets are simulated by varying the applied strain rate (\( a \)) from very low value (\( a \approx 1 \) s\(^{-1}\)) representing the composition state closer to chemical equilibrium until an extinction limit (\( a \approx 2800 \) s\(^{-1}\)). Since the counterflow diffusion flamelets extinguish beyond the extinction strain rate, the composition space between the most strained flamelet and mixing limit is covered by using an unsteady formulation of flamelets. The time-dependent solutions of flamelets are thus stored until the solution reaches the mixing limit (where there is no reaction). The FGM database is stored using a 401 \times 401 rectangular discretization of the \( Z-\gamma \) space. Important thermo-chemical parameters \( \mathcal{W}(Z, \gamma) \) of the system are then mapped on to these two control variables using linear interpolation. For simplicity and robustness, a bilinear interpolation procedure is used to lookup the thermo-chemical variables from the manifold during simulation run-time.

#### 2.2.2. Coupling with soot model (FGM-DSM)

After the creation of the manifold, FGM tabulated chemistry is linked to the CHEM1D code. During the run-time, first, the FGM
database is loaded into memory. Then, transport equations are si-
multaneously solved for the control variables, together with con-
tinuity, momentum, and sectional soot mass fractions. Therefore,
the application of FGM strategy essentially reduces the total num-
ber of equations by up to \( N_t +1 \), since the governing equations for all
\( N_s \) species, and energy can be avoided by transporting two control
variables (\( Z \) & \( Y \)) and tabulating temperature. The transport equation for a control variable (\( \phi = \sum \alpha_i Y_i \)) can be expressed in a
general form as:

\[
\frac{\partial (\rho \phi)}{\partial t} + \frac{\partial (\rho u \phi)}{\partial x} = \frac{\partial}{\partial x} \left( \alpha \frac{\partial \phi}{\partial x} \right) + \sum_{i=1}^{N_s} \alpha_i \dot{\omega}_i,
\]

with

\[
D = \left[ \left( -\sum_{i=1}^{N_s} \rho \alpha_i U_i \right) - \frac{\partial \phi}{\partial x} \right]
\]

from flamelets

where \( U_i, \alpha_i, \dot{\omega}_i \) denote diffusion velocity, weight coefficient (se-
lected to obtain a unique mapping of control variable), and source
term for species \( i \), respectively. The first term on the right-hand
side represents the preferential diffusion flux and is only a func-
tion of the gradient of the main control variable, \( \phi \) (mixture frac-
tion here). For sooting flame simulations, the concentrations of es-
cessential species involved in soot formation processes, and thermo-
chemical parameters (e.g. \( \rho, c_p, \lambda \)) required for the transport of
Eq. (4) are retrieved from the FGM database. The retrieved species
concentrations and thermo-chemical parameters are subsequently
provided as an input to the soot model for computations of the
source terms associated with sectional soot transport equations
(Eq. (1)).

Since the soot particles are formed from the processes involving
contributions from various gas-phase species, the concentrations of
associated gas-phase species also get affected as a result of soot
formation. Therefore, including this two-way coupling of the soot
and gas phase is a critical aspect of soot modeling in combustion
simulations. In the case of detailed chemistry, transport equations for
the gas-phase species are solved. Thus, the exchange of mass
between the solid and gas phase associated with the soot forma-
tion can be straight forwardly incorporated in the chemical source
terms of their transport equations. On the other hand, in a tabu-
lated chemistry approach such as FGM, the species data is looked-
up from a pre-computed manifold. Consequently, a special treat-
ment is required to include the soot-gas phase coupling, which can
influence the predictive capabilities of the soot model. Concerning
this, five different approaches of FGM-DSM coupling, listed in the
Table 1, are investigated for their predictive accuracy and computa-
tional performance. The key features of these approaches can be
summarized as:

- **FGM-Unc**: In the FGM-Unc approach, the manifold is gener-
ated using gas-phase flamelets without soot formation follow-

| Coupling type | Contribution of soot to gas phase | Soot processes included in manifold generation | \( Y_t \) transport |
|---------------|-----------------------------------|-----------------------------------------------|-----------------|
| Detailed      | Fully included during the run-time| Not applicable                                | Yes             |
| FGM-Unc      | Not included                      | None                                          | Yes             |
| FGM-Trans     | Partially included by transporting PAH| Contribution from soot nucleation included | Yes             |
| FGM-Nuc      | Partially included in the manifold | All soot formation processes included         | Yes             |
| FGM-Full      | Fully included in the manifold     | All soot formation processes included         | No              |

solved for sectional soot mass fractions. However, the coupling
between the soot and gas-phase for the mass exchange of con-
cerned species is not considered. It is interesting to note that,
since the nucleation process is modeled through PAH dimeriza-
tion, the soot prediction is particularly sensitive to the concen-
tration of PAH. The unaccounted consumption of gas-phase PAH
associated with soot nuclelation and condensation results in its
surplus, and can lead to overprediction of soot.

- **FGM-Trans**: DNS study of Bisetti et al. [40] revealed that the
unsteady mixing tends to affect the PAH concentrations signif-
icantly. Therefore the dependence of PAH on turbulence-
chemistry interactions needs to be appropriately modeled.
Species with slower chemistry, such as PAH, show delayed re-
sponse to rapid changes in the turbulent flow field. As a result,
the slower time scales of PAH formation compared to other gas-
phase species often violate the fast chemistry assumption in
flamelet-based tabulated chemistry approaches [21]. To model
the unsteady effects, an additional transport equation is gen-
ernally solved for PAH species following the approach proposed by
lhe and Pitsch [41] for NO chemistry. The standard PAH trans-
port equation is given by:

\[
\frac{\partial (\rho u \dot{\omega}_{PAH}^{\text{TRA}})}{\partial t} + \frac{\partial (\rho u \dot{\omega}_{PAH}^{\text{TRA}})}{\partial x} = \frac{\partial}{\partial x} \left( \alpha \frac{\partial \dot{\omega}_{PAH}^{\text{TRA}}}{\partial x} \right) = \dot{\omega}_{PAH}^{\text{TRA}}
\]

where \( \dot{\omega}_{PAH}^{\text{TRA}} \) distribution is computed from the mixture-
averaged diffusivity of PAH species. The chemical source term of
the PAH transport equation can be generally expressed as:

\[
\dot{\omega}_{PAH}^{\text{TRA}} = \dot{\omega}_{PAH}^{\text{Gas}} + \dot{\omega}_{PAH}^{\text{Soot}}
\]

where the first term on the right-hand side, \( \dot{\omega}_{PAH}^{\text{Gas}} \), indicate the
contribution from gas-phase reactions, and the second term,
\( \dot{\omega}_{PAH}^{\text{Soot}} \), denotes the consumption of PAH though soot forma-
tion processes (nucleation and condensation). The \( \dot{\omega}_{PAH}^{\text{Gas}} \) term
is computed by using tabulated production (\( \dot{\omega}_{PAH}^{\text{Tab}} \)) and consump-
tion (\( \dot{\omega}_{PAH}^{\text{Tab}} \)) rates as per expression:

\[
\dot{\omega}_{PAH}^{\text{Gas}} = \left[ \dot{\omega}_{PAH}^{\text{Tab}} - \frac{Y_{PAH}^{\text{TRA}}}{Y_{PAH}^{\text{TRA}}} \dot{\omega}_{PAH}^{\text{Tab}} \right]
\]

where \( \dot{\omega}_{PAH}^{\text{Tab}} \) is linearized with the tabulated (\( Y_{PAH}^{\text{Tab}} \)) and trans-
ported (\( Y_{PAH}^{\text{TRA}} \)) values of PAH (A4) mass fraction since A4 con-
sumption via gas-phase reactions usually scales linearly with
A4 concentration. The term, \( \dot{\omega}_{PAH}^{\text{Gas}} \), representing contribution
from soot processes can be treated in different ways. In some of the
studies [3,15,21], tabulated rates of dimer formation have been used as an additional consumption term in the PAH trans-
port equation to include the influence of PAH-based soot
growth processes. However, in the present study, we compute
\( \dot{\omega}_{PAH}^{\text{Soot}} \) during the simulation run-time as done in the detailed
chemistry case. The \( \dot{\omega}_{PAH}^{\text{Soot}} \) term, thus, include the contributions
from the soot nuclelation and PAH condensation processes. Note
that the number density of PAH required for the compu-
tations of nucleation and condensation rates is calculated based
on transported PAH mass fractions. Therefore, the PAH trans-
port equation is solved to model PAH itself. The linearization is
not applied for $\xi_{\text{PAH}}$ term as it is computed on-the-fly. For the FGM-Tran approach, the manifold is built from the gas-phase flamelets without soot formation (same as the one employed in the FGM-Unc case).

**FGM-Nuc**: Solving additional transport equations to account for the soot-gas coupling can lead to increased computational cost. Therefore, the FGM-Nuc approach is proposed in this study to partially include the effect of gas-phase PAH extraction in the soot model. It is observed that the change in PAH concentration due to soot processes is substantially higher compared to other major species such as $C_2H_2$, $O_2$, $H_2O$. Hence, to avail the advantage in terms of model accuracy at a lower computational cost, the consumption of PAH through the soot nucleation process is accounted for in the manifold generation step. Accordingly, for the FGM-Nuc approach, the contribution of soot nucleation to PAH and $H_2$ concentrations are included in their respective transport equations while simulating flamelets for the manifold creation. Subsequently, the resulting PAH mass fractions are stored in the FGM table. During run-time, the tabulated PAH concentration is directly used in the soot model for the computations of nucleation and condensation rates. Since the soot nucleation source term only depends on gas-phase, soot modeling is not required in the manifold generation stage. It is to be noted that in addition to nucleation, the condensation process also leads to the consumption of PAH. However, the condensation rates also depend on soot parameters such as soot number density. Its inclusion, thus, increases the duration of manifold generation, as additional transport equations need to be solved during flamelet creation to compute related soot parameters. Therefore, the contribution from soot nucleation alone is considered, for the computational simplicity of the manifold generation stage.

**FGM-Full**: In the FGM-Full approach, the contribution of soot in the consumption of all the relevant gas-phase species is included in the manifold generation step. Therefore, in addition to gas-phase species, transport equations for soot mass fractions are also solved during the flamelet simulations with the same soot model parameters as employed for the detailed chemistry simulations. The concentrations of important gas-phase species related to the soot formation process are then tabulated in the FGM table. Since the soot model is included in its entirety during the flamelet generation, the tabulated concentrations of gas-phase precursors species reflect their more accurate prediction. During the FGM simulation run, the tabulated concentrations of these species are looked-up from the FGM database and used as an input to the soot model for the computations of soot source terms. With the FGM-Full approach, the accuracy of soot prediction is expected to improve as the gas-phase species concentrations are predicted with a higher level of accuracy. However, since contributions from all the soot processes are included in the flamelets, the manifold generation process becomes computationally intensive to a certain extent.

**FGM-Read**: In the FGM-Read approach, flamelets are solved with the complete sectional soot model (as done in the FGM-Full case), and in addition to relevant gas-phase species, the values of sectional soot mass fractions, $Y_{s,i}$, are stored in the manifold. Due to the inclusion of the full soot model in the manifold generation stage, the concentrations of gas-phase species relevant to the soot model are well predicted in FGM-Read, as their consumption through soot formation processes is incorporated in the manifold. Moreover, during FGM simulation run-time, the values of sectional soot mass fractions ($Y_{s,i}$) are looked-up directly from the manifold. Since the soot mass fraction values are explicitly retrieved from the manifold, no additional transport equations are required to be solved for sections when the FGM-Read approach is applied. As a result, a significant reduction in computational time can be achieved. The comparable soot quantities such as soot volume fraction, number density, particle size are derived from the sectional soot mass fractions in the post-processing step by applying appropriate relations specified in [29].

### 2.3. Flames investigated

In total, six ethylene counterflow flames are simulated for the assessment of FGM-DSM coupling strategies. The details of the studied flames are summarized in Table 2. The chosen target flames also facilitate the variation of strain rate and the change in reactants composition for additional verification of the FGM-DSM coupling approaches. The detailed kinetic mechanism, KAUST Mechanism 2.0 (KM2) [42], consisting of 202 species and 1351 elementary reactions, is used for all the simulations reported in the present study. This mechanism has been validated for modeling the soot formation in premixed/non-premixed ethylene flames in previous works [29,43,44]. A mixture-averaged approximation of Hirshfelder and Curtiss [45] is used for modeling species diffusion transport during flamelet generation as well as sooting flame simulations.

### 3. Assessment of FGM-DSM coupling for steady conditions

#### 3.1. Simulations with the detailed chemistry

Prior to the assessment of FGM-DSM coupling, we investigate the predictive capabilities of the soot model with detailed chemistry for the target flames. In Fig. 1, the measured [LE/LS technique] and computed profiles of soot volume fraction ($f_v$), number density, and average particle diameter are compared for the WRC15 and WC16-1-3 flames. In the experiments by Wang et al. [46] the strain rate variation for WC16-1-3 flames was achieved by changing the nozzle exit velocity. However, in the present simulations, assuming potential flow in the far-field, this variation is represented via applied strain rate ($\alpha$), after comparing the simulated temperature and velocity profiles reported in the reference [46]. An increase in strain rate lowers the flame temperature as well as PAH.
concentration, which leads to reduced soot nucleation rates, and consequently, the overall soot formation is decreased. In Fig. 1(a), a good agreement can be observed for the computed soot volume fraction profiles against their measured counterparts. Although the present soot model could not capture all the quantitative parameters of soot perfectly, the qualitative trends of number density and particle size with strain rate variation are predicted reasonably well in Fig. 1(b) and (c).

Similarly, the computed profiles of soot volume fraction for WC16-N2 and XYZ18 flames are compared against the experimental measurements in Fig. 2. The profiles for WRC15 flame are plotted as a baseline case, to highlight the difference in soot concentration as a result of changes in fuel/oxidizer composition. The reduction in soot volume fraction for the WC16-N2 flame can be attributed to the dilution effect of N₂. This is because N₂ dilution causes a drop in flame temperature and consequently PAH concentration, which eventually diminishes the soot formation. On the other hand, soot formation is enhanced for the XYZ18 flame, in which the oxygen concentration in the oxidizer is increased compared to the WRC15 flame. The enhancement in soot concentration stems from the fact that a higher oxygen concentration elevates the flame temperature, which leads to increased levels of PAH formation, and eventually, higher soot surface growth rates. In Fig. 2, the qualitative trends of the soot volume fraction are well predicted for WC16-N2 and XYZ18 flames, indicating the encouraging capability of the present soot model to capture thermal and dilution effects associated with the variation in reactants composition. The detailed validation of soot model can be found in [29,49].

3.2. Simulations with FGM-DSM coupling

3.2.1. Manifold verification

Before discussing the results for FGM-DSM coupling approaches, the important features of thermo-chemical parameters stored in FGM manifolds are illustrated. Figure 3 presents the progress variable source term \( \dot{\phi}_s \), temperature, \( C_2H_2 \), and A4 mass fractions stored in the manifold as a function of \( Z \) and \( Y \) for the WRC15 flame. The contours of temperature and progress variable source terms show peak near the oxidizer side (\( Z \) close to zero), whereas the A4 concentration is more prominent near the sooting zone (\( Z \sim 0.3-0.5 \)) for flamelets at very low strain rates. Besides, the flamelets for the strain rates ranging from 1 s⁻¹ to 1000 s⁻¹ demonstrates the monotonicity of the progress variable necessary for the unique mapping of stored variables in the \( Z-Y \) space. The higher concentrations of \( C_2H_2 \) within strain rates 1 s⁻¹ to 100 s⁻¹ denote the composition space pertinent to a higher amount of soot formation. Note that the A4 mass fraction distribution is very narrow in the FGM composition space, which indicates that the evaluation of PAH-based processes is particularly sensitive to the accurate retrieval of A4 mass fractions during simulation run-time.

Furthermore, a-priori analysis is conducted to verify the solutions of control variable transport equations obtained from the FGM approach. Accordingly, the profiles of mixture fraction (\( Z \)), progress variable (\( Y \)), and temperature obtained from FGM solutions for WRC15 flame (without soot) are compared against the detailed chemistry counterparts in Fig. 4. It can be observed that the FGM simulations reproduce the detailed chemistry solutions for the control variables (\( Z, Y \)) and temperature with good accuracy. Only a marginal deviation is observed in detailed and FGM
profiles of progress variable near $x \sim -0.15$ cm, as the mixture composition approaches pure fuel conditions ($Z \sim 1$). This essentially underlines the good capability of the FGM in predicting the thermo-chemical composition space of the manifold relevant to the soot formation.

### 3.2.2. Comparison of different FGM-DSM approaches

In this section, we assess the performance of various FGM-DSM coupling strategies described in Section 2.2.2 for the accuracy of soot prediction in the target flames. First, we evaluate the results obtained through the FGM-Unc coupling approach for the WRC15 flame. In Fig. 5, the computed profiles of soot volume fraction, number density, and particle size for the FGM-Unc approach are presented along with the detailed chemistry solution and experimental measurements. It can be observed that the soot volume fractions are overpredicted by almost an order of magnitude compared to their measured values and detailed chemistry counterparts. Such a drastic deviation in the predicted soot concentration is expected since the consumption of gas-phase species (PAH in particular) due to soot formation is not included in the FGM-Unc approach. The overpredicted PAH concentration significantly increases the soot nucleation rate, number density, and con-
measurements. Both FGM-Tran and FGM-Nuc approaches show an overestimation of soot quantities. Besides flamelet induced errors, such quantitative disparity is mainly a consequence of intrinsic differences in strategies employed for including soot-gas coupling in them. Note that, in the FGM-Nuc approach, the consumption of PAH, and production of H$_2$ due to nucleation is accounted a-priori during the manifold generation step. On the other hand, in FGM-Tran, the consumption of PAH through soot nucleation and condensation introduced during the run-time by solving the PAH transport equation. However, the FGM-Nuc approach appears to show a better prediction of soot concentration compared to the FGM-Tran approach. The important factors responsible for such behavior can be explained through mass fraction distributions of A4 and C$_2$H$_2$ species involved in the soot model (refer Fig. 7).

From Fig. 7(a) it can be observed that the concentration of PAH (A4) predicted by FGM-Tran is somewhat higher than the one obtained in FGM-Nuc. To identify the extent of disparity in the PAH concentrations predicted by FGM-Tran and FGM-Nuc, the results for A4 mass fractions, obtained for detailed chemistry and FGM-Tran cases without the inclusion of PAH condensation process, are compared in Fig. 8. Note that in FGM-Tran, the contributions of both nucleation and PAH condensation are included in the chemical source term of the PAH transport equation (Eq. (7)). Therefore FGM-Tran is expected to predict the PAH concentration relatively well as compared to FGM-Nuc, wherein only the contribution of soot nucleation is included. However, it can be observed from Fig. 8 that the concentration of PAH in FGM-Tran after consumption through both the PAH related processes (Nucleation and PAH condensation) is slightly higher than the PAH concentration obtained in FGM-Nuc. It essentially indicates that solving a transport equation for the PAH, although provides a partial coupling for gas and soot phase, its accuracy of the PAH prediction is somewhat inferior compared to when the entire process of soot nucleation is computed during the flamelet generation stage. In addition to the numerical accuracy in the retrieval of various thermo-chemical parameters associated with the PAH transport equation under the FGM chemistry, the discrepancies in the prediction of PAH for FGM-Tran could be associated with the following factors: i) In FGM-Tran, the $\omega_{PAH}$ is computed from the transported PAH mass fractions. Therefore, the effect of PAH consumption through the soot process on the detailed kinetics associated with its formation in the gas phase can not be well captured. In other words, the effect of PAH consumption through soot processes on its gas-phase dynamics can not be accurately reproduced with tabulated chemical production and consumption rates. Considering the fact that the consumption of PAH also affects its gas-phase kinetics, the proposed FGM-Tran thus only ensures partial two-way coupling, which results in the overestimation of PAH compared to detailed chemistry results. ii) The mere application of the linear scaling approach $\omega_{PAH} = \frac{Y_{PAH}}{Y_{PAH}^{Tab}} \omega_{PAH}^{Tab}$ for PAH consumption rate can not fully reproduce their concentrations as the production rates of large aromatics (such as PAH) are found to be proportional to smaller aromatics which exhibit transient effects [50]. Therefore, the assumption of constant production rates for PAH, employed in FGM-Tran during tabulation, may not be entirely appropriate. As a consequence, the simplified approach used in the PAH transport equation for modeling the chemical source term could not reproduce the correct PAH concentration. On the other hand, for FGM-Nuc, since the complete model of soot nucleation is solved in the flamelet generation stage, the change in PAH species concentration through its detailed gas-phase kinetics, and soot-to-gas mass exchange, is well captured. The existing overprediction of PAH in FGM-Nuc compared to detailed chemistry is a direct consequence of neglecting the contribution of PAH-condensation.

\[ \text{Fig. 5. Measured and computed profiles of soot volume fraction (a), soot number density (b), and soot particle diameter (c) for the WRC15 flame with the FGM-Unc approach.} \]
The effect of predicted species concentrations on various processes related to soot formation can be elucidated through their rate distributions in Fig. 9. A slightly higher A4 concentration in FGM-Tran over FGM-Nuc is reflected in enhanced soot nucleation rates since the nucleation rate is proportional to the square of A4 concentration in the present soot model. The higher nucleation rate in FGM-Tran causes an increment in the number density of soot particles within the region $x \sim 0.05-0.1$ cm in Fig. 6(b). However, near the flame stagnation region ($x \sim 0.0-0.02$ cm), the A4 concentration is found to be higher for the FGM-Nuc approach compared to FGM-Tran. Therefore, the soot number density is seen to be overpredicted for the FGM-Nuc approach near the stagnation region, which is essentially reflected in the deviation of the power-law mode of the PSDF in Fig. 6(d). The overestimation of PAH concentration and soot number density, lead to increased condensation rates for the FGM-Tran approach against the FGM-Nuc, as observed in Fig. 9(b). Furthermore, higher soot surface growth rates are observed in Fig. 9(c) for the FGM-Tran approach compared to FGM-Nuc. The profiles of $\text{C}_2\text{H}_2$ mass fractions (Fig. 7(c)) responsible for surface growth under the HACA mechanism show no substantial deviation for FGM-Tran or FGM-Nuc. Therefore the observed difference in surface growth rates can be attributed to the effect of the increased surface area of soot particles as a consequence of increased number density induced by higher PAH prediction. In brevity, both FGM-Tran and FGM-Nuc approaches, show quantitative discrepancies in the accurate prediction of soot characteristics, due to ad-hoc techniques employed for the inclusion of mass feedback from soot processes to gas-phase. However, the findings discussed above suggest that the proposed FGM-Nuc strategy serves equally well (even better) in comparison with the FGM-Tran approach while predicting the soot concentration.

We now assess the performance of FGM-Full and FGM-Read approaches. From Fig. 6 it can be observed that the FGM-Full approach shows a good agreement with the detailed chemistry solutions for various soot parameters, including PSDF. The accurate prediction of soot quantities stems from the fact that, for the FGM-Full approach, a soot model is included in its entirety during the flamelets generation, as discussed in Section 2.2.2. Therefore, the mass exchange between the soot and gas phase, associated with the sub-processes akin to the soot formation, is implicitly included in the manifold. As a result, the concentration profiles of soot precursor species in Fig. 7 are found to be accurately reproduced by the FGM-Full approach. With the correct prediction of gas-phase species, the rates of various sub-processes associated with the soot formation are also predicted with a great level of accuracy in Fig. 9, as expected. To that end, the FGM-Full stands out as a superior strategy in terms of accuracy of soot prediction amongst the FGM-DSM coupling approaches investigated.

The FGM-Read approach is the most trivial and the simplest of all since no additional transport equations are solved for soot mass fractions in the FGM application stage. Instead, the values of sectional soot mass fractions are stored in the manifold and retrieved directly during the simulation. Fig. 6 shows that the FGM-Read approach captures the trends in soot number density and particle size considerably well. However, it slightly overpredicts the soot formation compared to the detailed and FGM-Full solutions. The deviation in the soot volume fraction profile can be attributed to flamelet-induced errors in the retrieval of soot mass fractions in larger sections. To illustrate this, the values of peak sectional soot mass fractions $\phi_{\text{max}}$ predicted by FGM-Read are compared against the detailed chemistry results in Fig. 10.

---

Fig. 6. Measured and computed profiles of soot volume fraction (a), number density (b), average particle diameter (c), and particle size distribution function (PSDF) (d) for the WRC15 flame with different FGM-DSM coupling approaches. PSDF is taken at the maximum $t_f$ position.
It can be observed that the mass fraction values for higher sections ($i > 20$) are not very well reproduced. The overestimation of soot mass fractions for large-sized sections have a negligible influence on the soot number density, but, it can directly reflect in the overprediction of the overall soot volume fraction. Such discrepancies in the retrieval are commonly encountered for species that evolve at a slower rate when compared to other species in the manifold [24]. A close observation of stored sectional mass fractions in the FGM database showed steep gradients for their values in the $Z$-$Y$ composition space, especially for higher sections, where soot production is more prominent. Such steep gradients in stored values of sectional mass fractions also lead to interpolation errors during their retrieval. It is further observed that by modifying the progress variable definition marginally, the prediction of soot under FGM-Read, can be improved to some extent for certain flames (Refer Section 3.3). However, such improvements in the accuracy of FGM-Read are only limited to steady conditions and do not reflect in the dynamics of soot evolution in unsteady simulations. Therefore it is reasonable to state that the extremely minor difference in $Z$-$Y$ mapping of the sectional soot mass fractions influences the accuracy of a direct look-up based approach such as FGM-Read. Hence, solving transport equations for soot mass fractions would be the ideal choice to improve the accuracy of results, as we noticed in the case of the FGM-Full approach. Nonetheless, soot number density is better predicted by the FGM-Read as compared to FGM-Tran or FGM-Nuc approaches, for obvious reasons.

### 3.3. Remark on the influence of progress variable on the performance of FGM-DSM

As highlighted in Section 2.2.1, the definition of progress variable $Y$ used in the reported FGM-DSM simulations (Eq. (3)) is determined based on trial and error approach. Accordingly, an appropriate $Y$ definition has been chosen based on preliminary analysis, which leads to a unique mapping of the thermo-chemical variables in the $Z$-$Y$ space to recover their tabulated quantities. The results for six different progress variable definitions are analyzed to examine the influence of the progress variable on the prediction of global soot quantities. A general expression for $Y$, as the linear
A combination of species mass fractions, can be given by:

\[ \gamma = \sum_{i=1}^{N_s} \alpha_i Y_i \]

with \( \alpha_i \) as a weight factor for species \( i \). Table 3 summarizes the weight factors used for the progress variable definitions selected for the analysis.

To demonstrate the effect of various \( \gamma \) definitions on the overall soot prediction, the variation of normalized peak soot volume fractions for WRC15 flame under different FGM-DSM approaches are compared in Fig. 11. It can be observed that the accuracy of FGM-Tran, FGM-Nuc, and FGM-Full approaches, in which transport equations are solved for the soot mass fractions, is significantly affected with the choice of progress variable \( \gamma \). However, the progress variable definition has a substantial impact on the accuracy of the FGM-Read approach in which sectional soot mass fractions are directly looked-up from the manifold. Reducing the weight factor of \( O_2 \) by order of magnitude changes the accuracy of FGM-Read drastically.

From the performance of various \( \gamma \) definitions, it can be observed that by modifying progress variable definitions to \( \gamma_5 \) the prediction for FGM-Read becomes slightly better than compared to \( \gamma_6 \) (which is used currently). Therefore, it is noticed that the
look-up of sectional soot mass fractions is particularly sensitive to its mapping in Z-Y space in the FGM table. Given the fact that FGM-Read results can be improved with a better Y definition to a certain extent, its applicability is limited only to the steady-state flames. For the unsteady conditions, FGM-Read could not capture the dynamics of soot evolution for any of the suitable Y choice, as will be discussed in the later sections.

3.4. Assessment of FGM-DSM coupling for variation in strain rates and reactants composition

We further extend the assessment of FGM-DSM approaches to evaluate their performance for different flames characterized by the variation in applied strain rates and changes in the composition of reactants.

3.4.1. Variation in strain rates

To demonstrate the effect of strain rate variation, WC16-1, WC16-2, WC16-3 flames are simulated under different FGM-DSM coupling approaches. Since the reactant composition in the WC16 series flames remains identical to the WRC15 flame, the same manifolds generated in the WRC15 flame cases are used for the FGM chemistry simulations in WC16 flames. In Fig. 12, the soot characteristics for the WC16 series flames with various FGM-DSM approaches are compared against their detailed chemistry solutions and measured values.

It can be observed from Fig. 12 that the qualitative trends of reduction in soot volume fraction, number density, and soot particle size with strain rate are captured considerably well with all the FGM-DSM approaches. The predictive capability of FGM-DSM coupling approaches under varied strain rate conditions is, thus, encouraging. Furthermore, the FGM-Nuc approach shows a relatively better prediction of soot concentration compared to FGM-Trans at all the strain rates, as noticed earlier for the WRC15 flame. The FGM-Full approach is found to capture the trends in soot quantities with great accuracy, as expected. However, the FGM-Read approach shows a significant overprediction of soot in WC16-2 and WC16-3 flames, whereas, for WC16-1 flame, the distribution of soot volume fraction is well captured. Such large discrepancies in the prediction of soot volume fractions essentially suggest that the accuracy of the FGM-Read approach is influenced by the strain rates. However, it is primarily a consequence of its sensitivity to the mapping of sectional mass fractions in the FGM database for a particular choice of progress variable.
3.4.2. Variation in reactants composition

Next, to assess the capabilities of FGM-DSM coupling approaches for variation in mixture composition, WC16-N2, and XY18 flames are simulated. Separate manifolds are generated for WC16-N2 and XY18 flames as the reactant compositions in them are different from that of WRC15 flame. In Fig. 13, the soot volume fraction profiles for various FGM-DSM coupling approaches are compared against the detailed chemistry solutions and experimental values in WC16-N2, and XY18 flames. In the XY18 flame, the oxygen concentration in oxidizer is increased, thus overall soot volume fraction is increased compared to WRC15 flame. This increment is qualitatively captured well by all the FGM-DSM approaches. Similarly, in the WC16-N2 flame, ethylene is diluted with N$_2$ causing a reduction in soot concentration over the WRC15 flame. This reduction in soot volume fraction is also captured with FGM-DSM coupling approaches in Fig. 13(b).

As identified earlier, in both the flames, FGM-Train and FGM-Nuc approaches overpredict the soot concentration, whereas the FGM-Read approach gives a more accurate prediction of soot parameters. In contrast, the FGM-Read approach shows a significant deviation in soot prediction for the WC16-N2 flame, while the prediction is relatively better for the XY18 flame. It is important to note that the reported observations for FGM-Read are related to the definition of progress variable chosen in the analysis. Therefore, the conclusions regarding the level of accuracy for FGM-Read can not be generalized. Nevertheless, the modeling results indicate that the FGM-DSM coupling qualitatively captures the trends in the distribution of soot volume fractions when the reactive system is subjected to changes in mixture composition, although quantitative discrepancies still exist.

To quantify the precision levels of the different FGM-DSM coupling approaches, the values of peak soot volume fractions in the target flames obtained from simulations ($f_{\text{exp}}^{\text{num}}$), are compared against their measured ($f_{\text{exp}}^{\text{num}}$) counterparts in Fig. 14. In FGM-Train and FGM-Nuc approaches, the peak $f_{\text{exp}}$ values for the modeled flames are overpredicted by approximately 30–50%, with FGM-Nuc being relatively more accurate. The accuracy of FGM-Read is recognized to be susceptible to large deviations for the prediction of lower values of soot volume fractions ($f_{\text{max}} \sim 0.5$ ppm), whereas, the same is found to be improved for a higher amount of soot concentration ($f_{\text{max}} \sim 1.0$ ppm). However, although not presented extensively in the current study, with a somewhat different choice of progress variable, the accuracy of soot prediction for FGM-Read can be found improved for flames yielding lower soot concentrations. Therefore, the rationale behind better accuracy of FGM-Read at higher soot volume fractions is the result of the chosen progress variable and may not correspond to a particular physical phenomenon associated with the soot formation. Such discrepancies can be recognized as the main drawback of direct look-up based method such as FGM-Read. On the other hand, in all the target flames, the FGM-Full approach showed an excellent agreement with the detailed chemistry results. The precision level of the FGM-Full approach is found to be almost equivalent to the detailed chemistry solutions.

While understanding the quantitative inconsistencies in the soot prediction by FGM-DSM approaches against the detailed chemistry, it is important to recall that the accuracy of soot prediction strongly depends on local concentrations of the gaseous species and soot precursors such as C$_2$H$_2$, H$_2$, A4, etc. Since these values are read from a pre-computed manifold, multiple factors including choice of progress variables, method of soot-gas phase coupling, interpolation errors during lookup can influence the overall soot prediction in FGM chemistry approaches. Therefore, quantitative discrepancies between detailed and tabulated chemistry results are not surprising. Nevertheless, it is important to highlight that the FGM-DSM approaches can predict the amount of soot within the factor of 1.5 from the detailed chemistry results, which can still be considered as acceptable for many practical applications considering the complexities of the soot formation process.
4. Assessment of FGM-DSM coupling for unsteady conditions

Up to this point, we investigated the performance of different FGM-DSM approaches in predicting soot formation under steady-state conditions. However, the main purpose of employing tabulated chemistry is to facilitate computationally efficient chemistry reduction in large scale turbulent conditions. In turbulent reacting flows, the flamelet structures are subjected to time-dependent strain rates as they evolve in the flow field. Hence, it is essential to assess the capabilities of FGM-DSM approaches for soot prediction under the influence of unsteady hydrodynamics. To illustrate this, four unsteady scenarios are considered as follows:

- **Case 1**: Unsteady simulations at a constant strain rate, with a steady-state gas-phase flame solution as an initial condition \( \alpha = a_0; Y_{ij}(t = 0) = 0 \)
- **Case 2**: Unsteady simulations at a time-dependent strain rate, with a steady-state gas-phase flame solution as an initial condition \( \alpha = f(t); Y_{ij}(t = 0) = 0 \)
- **Case 3**: Unsteady simulations at a time-dependent strain rate, with a steady-state sooting flame solution as an initial condition \( \alpha = f(t); Y_{ij}(t = 0) = Y_{ij}^* \)
- **Case 4**: Unsteady simulations at a sinusoidally varying time-dependent strain rate, with a steady-state sooting flame solution as an initial condition \( \alpha = f(t); Y_{ij}(t = 0) = Y_{ij}^* \)

### 4.1. Case 1: constant strain rate with steady-state no-soot initial condition

In the first case, unsteady simulations are carried out at a constant strain rate of 120 s\(^{-1}\) for all the FGM-DSM approaches. To capture the dynamics of soot evolution, all the simulations are initialized with the gas-phase flame solution without soot. In Fig. 15 the distributions of soot volume fraction in the WRC15 flame at different time instants for various FGM-DSM approaches are compared against the detailed chemistry solutions. In soot formation (SF) type flame such as WRC15, the soot particles nucleate in the region with higher PAH concentration on the fuel side of the flame and are convected by the flow towards the flame stagnation location. During this process, soot particles grow in size through surface reactions and particle-particle coagulation, such that their number density decreases. As a result, the shape of soot volume fraction profiles becomes somewhat skewed towards the flame stagnation location. Figure 15 reveals that the temporal evolution of soot from its inception to the steady-state is qualitatively captured well by FGM-Trans, FGM-Nuc, and FGM-Full approaches. The FGM-Full approach predicts the progress of soot formation under unsteady conditions with great accuracy when compared with the detailed chemistry response. The FGM-Trans and FGM-Nuc approach on the other hand show quantitative overprediction in instantaneous soot concentration expected from detailed chemistry results, with the latter being relatively better. All the approaches, except FGM-Read, predict the correct dynamics of soot formation. The FGM-Read approach fails to capture the trends in soot formation for obvious reasons. This is because, in FGM-Read, the soot mass fractions are directly looked-up from the manifold based on distributions of control variables. Therefore, the time-dependent response of soot formation is not captured well in FGM-Read, as transport equations for their evolution are not explicitly solved. It emerges as the major limitation of FGM-Read in predicting soot formation under unsteady conditions.
4.2. Case 2: time-dependent strain rate with steady-state no-soot initial condition

In the second case, a time-dependent variation of strain rate is introduced under unsteady conditions. For demonstration, the applied strain rate at the oxidizer boundary is varied from the initial value of \( a_0 \) to final strain rate \( a_f \) over a simulation time \( t_f \) using:

\[
a = a_0 + (a_f - a_0) \left[ 1 - e^{-\left(\frac{t}{t_f}\right)^3} \right] \tag{9}
\]

where \( a_0 = 120 \text{ s}^{-1} \), \( a_f = 200 \text{ s}^{-1} \) and \( t_f = 10 \text{ ms} \) are chosen. The initial strain rate of \( a_0 \) corresponds to the strain rate of gas-phase flame solution, whereas the value \( a_f \) is taken arbitrarily for illustration. The timescales of soot formation are generally in order of milliseconds [51], accordingly, the chosen \( t_f \) value is expected to be reasonable to capture the response of soot evolution to unsteady strain rate variation. The instantaneous distributions of soot volume fraction for different FGM-DSM approaches are presented in Fig. 16 along with detailed chemistry results for the WRC15 flame.

All the cases (Detailed and FGM-DSM) are initialized from the gas-phase flame solutions without soot formation. From the computed soot volume fraction profiles, it can be noticed that the soot concentration initially increases as soot begins to form from gas-phase species. However, as the strain rate increase with time, the process of soot formation gets affected, and the overall soot concentration tends to decrease towards the end of the simulation time \( (t = t_f) \). The reduction of soot concentration is evident from the soot volume fraction profiles at 7.5 and 10 ms. Moreover, from Fig. 16, it can be observed that except for FGM-Read, the quantitative trends of soot evolution are predicted well by the FGM-DSM approaches. The FGM-Full approach shows a very close quantitative agreement with the detailed chemistry results whereas, FGM-Trans and FGM-Nuc somewhat overpredict the peak soot concentration in line with the previous test cases. The FGM-Read approach, on the other hand, fails to capture the unsteady effects, as expected.

For clarity, the temporal evolutions of peak soot volume fraction in various FGM-DSM approaches are compared in Fig. 17. The increasing and decreasing trend of peak soot volume fraction under strain rate variation witnessed in a detailed chemistry case is
also captured by FGM-DSM solutions (except for FGM-Read). The results, therefore, indicate that the response of soot formation to the unsteady effects associated with the time-dependent strain is reasonably well captured with the FGM-DSM coupling strategies, although quantitative discrepancies exist.

4.3. Case 3: time-dependent strain rate with steady-state soot initial condition

In turbulent flows, the strain rates can change rapidly, and their fluctuations could vary by order of magnitude [52]. However, the soot evolution occurs at a relatively slow time scales. Hence it is imperative to investigate the capabilities of various FGM-DSM coupling approaches in capturing the dynamic response of soot formation to rapid strain rate fluctuations. For this reason, a time-dependent variation is introduced to the applied strain rate in sooting flames as per the function:

\[ a = a_0 + \left( \frac{a_f - a_0}{2} \right) \left[ 1 + \tanh \left( \frac{t - t_0}{\xi \tau_f} \right) \right] \quad (10) \]

with \( a_0 = 120 \text{ s}^{-1}, \ a_f = 720 \text{ s}^{-1}, \ t_f = 20 \text{ ms}, \ t_0 = t_f/2 \) and \( \xi \) is the profile parameter. Two different values, \( \xi = 0.01 \) and \( \xi = 0.1 \) are chosen for the profile parameter to mimic the rapid and gradual variation, respectively, in the temporal distribution of the strain rate.

In this case, the steady-state sooting flame solutions corresponding to the respective FGM-DSM approaches are used as initial conditions for unsteady simulations. To understand the overall response of soot formation to temporal variation of strain rate, the time evolution of the peak soot volume fractions in various FGM-DSM approaches are compared in Fig. 18. The qualitative trends of decrease in the peak soot concentrations for rapid and gradual increments in strain rates are captured well by FGM-Trans, FGM-Nuc, and FGM-Full approaches. The FGM-Full approach shows a good agreement with the detailed chemistry solutions as expected. FGM-Read approach, on the other hand, could not reproduce the trends in the soot volume fraction evolution well. Note that for the \( \xi = 0.01 \) case, the strain rate increases drastically (within 1ms). Therefore, the sharp change in the evolution of soot volume fractions can be noticed at 10ms. Furthermore, relatively slow time scales associated with the soot formation cause a lag between the response of soot to the strain rate variation. FGM-Read approach, however, could not capture this behavior since the trends shown by FGM-Read qualitatively follow the course of progress variable evolution considering that the soot mass fractions are retrieved directly from the FGM database as a function of the progress variable. The analysis hence justifies that the FGM-DSM approaches (except for FGM-Read) can qualitatively reproduce the dynamic response of soot formation to the drastic strain rate variations.

4.4. Case 4: sinusoidal strain rate variation with steady-state soot initial condition

In turbulent combustion systems, under high Reynolds number conditions, eddies with a wide range of length and time scales exist, which can introduce fluctuations in the strain rates. The strain rate fluctuations begin to induce unsteadiness on the flame structure. The flame unsteadiness further influences the formation of soot precursors and affects the overall sooting characteristics. The transient response of non-premixed flames to the unsteadiness is often modeled by imparting harmonic oscillations to the flow field in laminar counterflow diffusion flames [51,53]. Following this feasible approach, sinusoidal oscillations are introduced to the applied strain rate with varying frequencies and amplitudes. The response of flame to the strain rate fluctuations is analyzed in terms of induced oscillations of the peak soot volume fractions for various FGM-DSM approaches. The strain rate oscillations are assumed to have the following form:

\[ a = a_0 + \Delta a \cdot \sin(2\pi ft) \quad (11) \]

where \( \Delta a \) is the semi-amplitude, \( f \) is the frequency of the imposed oscillations and \( a_0 = 120 \text{ s}^{-1} \) is the strain rate of steady-state flame solution. Since the soot formation is usually a slow process, the soot field could not respond to the imposed oscillations at higher frequencies (\( > 100 \text{ Hz} \)) [54]. For illustration, at two different values of frequencies (\( f = 50 \text{ Hz} \) and 100 Hz) and semi-amplitudes (\( \Delta a = 20 \text{ s}^{-1} \) and 50 s\(^{-1} \)) are chosen. Similar to Case 3, all the cases (Detailed and FGM-DSM) are initialized from their corresponding steady-state solutions at \( a_0 \) with soot formation. The numerical results obtained for the detailed chemistry and FGM-DSM cases in terms of the evolution of peak soot volume fractions against the number of oscillation cycles are shown in Fig. 19.

It is evident that when low-frequency oscillations are imposed, the induced amplitudes for soot field become large, and the symmetry around their mean value is lost [51]. For the same frequency, with an increase in the amplitude of the imposed oscillations, the value of the observed amplitude in soot volume fractions also increases. On the contrary, at higher frequencies, the amplitude of induced oscillations is reduced for an identical amplitude of imposed oscillations. These characteristics can be witnessed from
the dynamic response of peak soot volume fractions to the strain rate oscillations in Fig. 19. The FGM-Full coupling shows a good agreement with the detailed chemistry results, whereas FGM-Tran and FGM-Nuc predict the qualitative trends reasonably well. The departure from the detailed-chemistry values for peak soot volume fractions follows the sequence: FGM-Tran > FGM-Nuc > FGM-Full > Detailed. On the other hand, the FGM-Read fails to capture the qualitative trends of soot evolution. Besides, a significant phase-lag exists in the temporal evolution of peak soot volume fractions. Such a tendency is mainly observed in FGM-Read since the soot mass fractions are retrieved directly from the table. As a result, time-scales associated with the transport of soot mass fractions are not captured, which leads to a phase-lag in their temporal evolution for the imposed sinusoidal oscillations of the strain rates.

It is interesting to note that, for imposed sinusoidal fluctuations, the response of FGM-Full shows a slight phase deviation compared to detailed chemistry results. To elucidate this, in Fig. 20, the temporal evolutions of normalized maximum progress variable, PAH (A4) mass fractions, and soot volume fractions are presented for the detailed chemistry and FGM-Full simulations. From detailed chemistry results, it can be noticed that the phase-lag arises between the progress variable and PAH as a consequence of the time scales associated with the slower chemistry of PAH formation. In contrast, since PAH mass fractions are directly looked-up from the manifold in FGM-Full, the phase difference between the PAH and progress variable can not be captured. Given that the PAH concentration directly influences the overall soot formation process, the phase-difference associated with the progress variable and PAH also reflects in the response of soot volume fractions. The time-scales associated with soot formation are, however, significantly slower than the progress variable transport.

The present analysis also shows that although the dynamics of PAH species is slightly slower compared to the progress variable, its influence on the overall dynamics of soot leads to a phase-lag of less than 10% of the period for the detailed chemistry and FGM-DSM approaches (except for FGM-Read). Therefore it is more crucial to capture the dynamics of soot evolution compared to the dynamics of PAH when the FGM chemistry is employed. Accordingly, including sectional transport equations for soot prediction in FGM-DSM approaches is critical (as opposed to FGM-Read). Hence, it can be argued that as long as the concentration of PAH is accurately predicted, the FGM-DSM approaches characterized by the transport of soot mass fractions, can yield considerably good accuracy of quantitative and qualitative soot prediction.

It is worth mentioning that since the goal of the present study is to assess the performance of FGM-DSM coupling approaches, the mechanism associated with the dynamic response of soot forma-
tion to harmonic strain rate fluctuations is not addressed. Based on the presented analysis, it can be argued that the influence of unsteady hydrodynamics on soot formation is qualitatively captured by FGM-DSM approaches, with FGM-Full being relatively accurate, although quantitative discrepancies exist. The assessment of FGM-DSM coupling under a range of unsteady conditions, provide confidence in their relevance for turbulent conditions.

5. Computational performance of FGM-DSM coupling approaches

The most important aspect of chemistry-reduction methods such as FGM is the computational advantage gained compared to detailed kinetics. Hence, it is imperative to analyze the computational performance of different FGM-DSM modeling approaches for studied cases. In sectional methods, increasing the number of sections for capturing particle size distribution enlarges the CPU time since additional transport equations are required to be solved. In this context, the total CPU time needed to perform time-dependent simulations for a period of 1 ms is compared in Fig. 21(a) for a varied number of sections in different FGM-DSM approaches. The simulations are performed with identical soot model parameters on Intel(R) Core(TM) i7-8700 CPU @ 3.20 GHz processor. In the simulations, a fully implicit time integration with an adaptive time step is used. Each of the FGM-DSM cases is initialized with an identical gas-phase flame solution at a strain rate of 90 s$^{-1}$. The initialization from a flamelet at a lower strain rate is done intentionally, to provide slightly different initial values of thermo-chemical variables from their desired final solution. The total CPU time, thus, encompasses two important aspects. First, the transport of sectional soot mass fractions and second, the adjustment to the flow field strain. The chosen initial conditions, thus, inherently control the extent of the computational advantage that can be attained with the FGM-DSM approaches compared to detailed chemistry.

From Fig. 21(a), it can be observed that the total CPU time reduces by approximately an order of magnitude when the FGM-DSM coupling strategy is applied. This speed-up is mainly caused by a drastic reduction in the number of transport equations needed to be solved for the evolution of gas-phase species. This is because, in CHEM1D, the CPU time for matrix factorization generally scales with $N^3$, while the overall solution time scales approximately as $N^2$ for $N$ transported variables. Furthermore, the FGM-DSM strategy provides the advantage of larger time steps since smaller time scales associated with the gas-phase chemistry are eliminated. Therefore, the total computational time of FGM-DSM coupling approaches is found to be significantly lower than the detailed chemistry.

On comparing different FGM-DSM approaches, relative differences can be seen in the evolution of their CPU time with the number of sections. In the FGM-Tran approach, an additional transport equation is solved for PAH. As a result, the CPU time for FGM-Tran is higher compared to FGM-Nuc. The FGM-Tran approach is found to be about 10 equivalent sections slower compared to FGM-Nuc and FGM-Full. This stems from the fact that
the simulation time-steps in FGM-Tran become relatively smaller as a consequence of the PAH transport equation, leading to an increment in the number of iterations and CPU time. The results also suggest that simulations with FGM-Full approaches progress somewhat comparable to FGM-Nuc. Only a marginal reduction in CPU time is observed for a higher number of sections. On the other hand, the CPU time for the FGM-Read approach is significantly lower and hardly affected by an increment in the number of sections. For a higher number of sections, the FGM-Read approach gives a faster solution by almost two orders of magnitude compared to detailed chemistry. Such a strong reduction in CPU time is expected since no additional equations are solved for sectional soot mass transport in the FGM-Read approach.

The extent of CPU time reduction can be further elucidated through the analysis of the total time required per time-step. In Fig. 21(b) the ratios of CPU time per time-step in detailed ($t_{\text{CPU}}^{\text{Det}}$) and FGM ($t_{\text{CPU}}^{\text{FGM}}$) chemistry cases are plotted against the number of sections. It is observed that the iteration time of the FGM-DSM computations (except FGM-Read) can be reduced by approximately 6 to 40 times depending on the number of sections employed. Note that in the present analysis, an implicit time integration scheme is used, and the computational advantage is expected to improve further for explicit time integration methods [35].

It is obvious that constructing an FGM table for one-dimensional simulations is rather inefficient, considering the total time taken in manifold creation. However, the reduction in CPU time achieved by using FGM in large-scale, multidimensional combustion simulations, could be orders of magnitude larger than the time required for manifold construction. Moreover, for a given reactants composition, the manifold needs to be created only once and can be used in multiple simulations. The use of FGM thus facilitates design optimization.

6. Conclusions

To facilitate the use of FGM tabulated chemistry for soot modeling in turbulent flames, it is important to validate and assess the suitable strategies of coupling FGM with soot model over a range of conditions in a canonical, laminar one-dimensional flame configuration. In this regard, five different approaches for coupling FGM tabulated chemistry with the sectional model namely FGM-Unc, FGM-Tran, FGM-Nuc, FGM-Full, and FGM-Read were examined in six target flames to evaluate their performance in terms of accuracy of soot prediction and computational advantage. The main conclusions of the present assessment are:

1. The prediction of soot properties with the FGM-Nuc and FGM-Tran approaches is encouraging, considering the expected discrepancies associated with the lack of full coupling of the gas-soot phase. Both FGM-Nuc and FGM-Tran approaches predicted soot volume fractions within a factor of 1.3–1.5 against the experimental values for investigated flames. However, the FGM-Nuc emerged as a superior approach compared to the FGM-Tran in terms of accuracy and computational time.

2. The FGM-DSM coupling approaches recover the distributions of the soot parameters with reasonable accuracy against detailed chemistry solutions and experimental measurements when mass-exchange from the soot processes to gas-phase species is appropriately incorporated. Accordingly, the FGM-Full approach, in which full soot model solutions were used for the manifold generation, showed an excellent agreement with the detailed chemistry results. On the other hand, the accuracy of the FGM-Read approach, in which soot mass fractions are retrieved directly from the manifold, is found to be influenced by the mapping of sectional soot mass fractions in mixture fraction and progress variable space.

3. Numerical results revealed that the FGM-DSM coupling approaches predict the qualitative trends in soot formation for variations in strain rate and reactant compositions reasonably well. Moreover, the proposed FGM-DSM coupling approaches (except FGM-Read) can capture the dynamic response of soot evolution under unsteady conditions considerably well. The accuracy of the FGM-Read approach is found to be significantly compromised in unsteady conditions, making it unsuitable for use in turbulent combustion simulations.

4. Computational time analysis suggests that the FGM-DSM modeling strategies can reduce the CPU time by 1 to 2 orders of magnitude compared to the detailed chemistry. For unsteady simulations under the implicit time integration method, the FGM-DSM approaches facilitated up to 40 times reduction in the CPU time per time-step, based on the number of sections used.

Even though numerical analysis in one-dimensional flames can not exactly predict the actual performance of FGM-DSM coupling in simulations of turbulent flames, it can be concluded that FGM-DSM is a promising technique for soot prediction in non-premixed flames with a reasonable level of accuracy at enormously reduced computational efforts. Especially, for approaches in which transport equations are solved, the unsteady dynamics of soot formation is captured reasonably well. This essentially highlights the encouraging capabilities of FGM-DSM approaches to characterize the flame conditions that are different from the steady flamelets used for building the FGM database. Therefore, except for FGM-Read, the FGM-DSM approaches proposed would be suitable for predicting soot formation under more complex environments.

It should be noted that based on the complexity of combustion phenomena, FGM chemistry relies on user knowledge for the suitable choice of the flamelets, and it is sometimes required to increase the dimensions of the manifold to account for additional physics (e.g., heat loss effects). Therefore, it is reasonable to state that the effectiveness of FGM-DSM approaches for the soot prediction in more complex cases will also depend on the elements that are considered during the manifold generation.

However, it is unexplored at this point how the FGM-DSM approaches can be integrated into the simulations involving subgrid-scale modeling (LES, RANS). Therefore the extension of conclusions obtained for the performance of different FGM-DSM approaches under laminar flame investigations to more complex, turbulent conditions is rather intuitive. The comprehensive assessment conducted in this paper is essentially a preliminary step towards the use of FGM in modeling soot formation for turbulent flames, and further research is required to understand the influence of turbulence on model accuracy.

Declaration of Competing Interest

The authors declare that they have no known competing financial interests or personal relationships that could have appeared to influence the work reported in this paper.

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References

[1] M. Frenklach, H. Wang. Detailed modeling of soot particle nucleation and growth, Symposium (International) on Combustion, 23, Elsevier (1991), pp. 1559–1566.
[2] A. Kazakov, M. Frenklach, Dynamic modeling of soot particle coagulation and aggregation: implementation with the method of moments and application to high-pressure laminar premixed flames, Combust. Flame 114 (3–4) (1998) 301–312.

[3] M.E. Mueller, G. Blanquart, H. Pitsch, Hybrid method of moments for modeling soot formation and growth, Combust. Flame 156 (6) (2009) 1143–1153.

[4] E. Gelbard, Y. Tsimhoni, J.H. Seinfeld, Sectional representations for simulating aerosol dynamics, J. Colloid Interface Sci. 76 (2) (1980) 541–556.

[5] K. Netzel, H. Lehtiniemi, F. Mauss, Calculating the soot particle size distribution function in turbulent diffusion flames using a sectional method, Proc. Combust. Inst. 31 (1) (2007) 667–674.

[6] Q. Zhang, H. Gao, F. Liu, C. Smallwood, M. Thomson, Modeling of soot aggregate formation and size distribution in a laminar ethylene/air coflow diffusion flame with detailed PAH chemistry and an advanced sectional aerosol dynamics model, Proc. Combust. Inst. 32 (1) (2009) 761–768.

[7] T. Blacha, M. Di Domenico, P. Gerlinger, M. Aigner, Soot predictions in premixed and non-premixed laminar flames using a sectional approach for PAHs and soot, Combust. Flame 159 (1) (2012) 181–193.

[8] D. Aubagnac-Karkar, Soot sectional Model for Diesel RANS Simulations, Chaléney-Malabry, Ecole centrale de Paris, 2014 PhD. thesis.

[9] A. Violi, Modeling of soot particle inception in aromatic and aliphatic premixed flames, Combust. Flame 139 (4) (2009) 279–287.

[10] M. Baltasar, M. Frenklach, Detailed kinetic modeling of soot aggregate formation in laminar premixed flames, Combust. Flame 140 (1–2) (2005) 130–145.

[11] L.-H. Dorey, N. Bertiér, L. Tessé, F. Duponrieux, Soot and radiation modeling in laminar ethylene flames with tabulated detailed chemistry, Comptes Rendus Mécanique 339 (12) (2011) 756–769.

[12] Y. Xuan, G. Blanquart, Numerical modeling of sooting tendencies in a laminar co-flow diffusion flame, Combust. Flame 160 (9) (2013) 1657–1666.

[13] F. Duponrieux, N. Bertiér, Methodology for the numerical prediction of soot formation in turbulent reactive flows and application to aircraft engine combustors, Int. J. Sustain. Aviat. 2 (1) (2016) 15–33.

[14] D. Aubagnac-Karkar, J.-B. Michel, O. Colin, P.E. Vervisch-Kljakic, N. Darabahi, Soot sectional model coupled to tabulated chemistry for diesel RANS simulations, Combust. Flame 162 (8) (2015) 3081–3099.

[15] P. Rodrigues, B. Franzelli, R. Vicqcelin, O. Ciquel, N. Darabahi, Coupling an LES approach and a sectional model for the study of sooting turbulent non-premixed flames, Combust. Flame 190 (2018) 477–499.

[16] K.P. Goëge, M. Köhler, W. Olsoglin, W. Metz, Investigation of soot formation in pressurized swirl flames by laser measurements of temperature, flame structures and soot concentrations, Proc. Combust. Inst. 35 (3) (2015) 3373–3380.

[17] B. Franzelli, E. Ribé, B. Cuenot, M. Ihme, Numerical modeling of soot product in aero-engine combustors using large eddy simulations, ASME Turbo Expo 2015: Turbine Technical Conference and Exposition, American Society of Mechanical Engineers Digital Collection (2015).

[18] G. Lecocq, D. Potrau, I. Hernández, F. Duchaine, E. Ribé, B. Cuenot, A methodology for soot prediction including thermal radiation in complex industrial burners, Flow Turb. Combust. 92 (4) (2014) 947–970.

[19] T. Kim, Y. Kim, Interactive transient flamelet model for soot formation and oxidation processes in laminar non-premixed jets, Combust. Flame 162 (5) (2015) 1660–1678.

[20] M.E. Mueller, H. Pitsch, LES model for sooting turbulent nonpremixed flames, Combust. Flame 159 (6) (2012) 2166–2180.

[21] P.A. Wick, A. Attia, F. Bisetti, H. Pitsch, DNS-driven analysis of the flamelet/progress variable model assumptions on soot inception, growth, and oxidation in turbulent flames, Combust. Flame 214 (2020) 437–449.

[22] C. D Pierse, P. Moen, Progress-variable approach for large-eddy simulation of non-premixed turbulent combustion, J. Fluid Mech. 594 (2004) 175–203.

[23] M.R. Busupally, A. De, Numerical modeling of soot formation in a turbulent CH4/air diffusion flame, Int. J. Spray Combust. Dyn. 8 (2) (2016) 67–85.

[24] L. Zimmer, Numerical Study of Soot Formation in Laminar Ethylene Diffusion Flames, Universidade Federal do Rio Grande do Sul, 2016 Ph.D. thesis.

[25] H.Y. Akargun, B. Akkurt, N.G. Deen, Extending the flamelet generated manifold for soot and NOx modeling in diesel spray combustion, The Proceedings of the International Symposium on Diagnostics and Modeling of Combustion in Internal Combustion Engines 20173.1. The Japan Society of Mechanical Engineers, 2017. p. A105.

[26] C.A. Hoerle, Modelling of Soot Formation Based on the Discrete Sectional Method: CO Effects and Coupling With the TGM Technique, Ph.D. thesis, University Federal do Rio Grande do Sul, 2020.