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Detailed modeling of soot particle formation and comparison to optical diagnostics and size distribution measurements in premixed flames using a method of moments

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ABSTRACT

Recently improved knowledge about key processes for the formation and growth of soot particles lead to very extensive soot models describing several kinetic pathways for particle nucleation and growth in detail. One of these models has been proposed by D'Anna and coworkers (D'Anna et al., 2010, Sirignano et al., 2010). In these original studies, the multivariate formulation was solved with a sectional approach, while a more recent study (Salenbauch et al., 2017) also showed the suitability of moment methods such as the *Conditional Quadrature Method of Moments* (CQMOM) (Yuan et al., 2011). However, being a moment method, CQMOM does not allow to directly access the soot particle size distribution (PSD). This prevents the consistent comparability of CQMOM results to soot measurements based on a scanning mobility particle sizer (SMPS). Furthermore, the comparison to laser-induced fluorescence (LIF) experiments is also limited, as the LIF signal only represents small particles and their concentration is only extractable from the simulation results if the PSD shape is known.

The aim of this study is to extend the previously developed CQMOM soot model by a PSD reconstruction step applying the concept of entropy maximization. Maintaining the efficiency of the CQMOM moment inversion algorithm to close the moment equations, the model extension enables to evaluate the diameter-based PSD in a post-processing step without prescribing a specific shape as input. The updated algorithm is applied to simulate soot formation in two different burner-stabilized premixed $C_2H_4/O_2/N_2$ flames with a very lightly sooting (C/O = 0.67) and a heavily sooting (C/O = 0.77) character. Numerical results are compared to recently published LIF, laser-induced incandescence (LII) and SMPS measurement data. The analysis investigates the model's capability to predict phenomena such as uni- or bimodal distribution shapes and the transition of small nanostructures to agglomerates in the two target flames both of which exhibit very different sooting behaviour.

1. Introduction

In recent years, several experimental and numerical studies advanced the knowledge on soot particle formation in flames significantly [1–5]. This has been accompanied by the formulation of detailed soot models capable to describe processes such as nucleation, growth and oxidation in detail [6–8]. The model development process for soot models happened on two levels.

On the one hand, numerical approaches such as Monte Carlo algorithms, sectional methods and quadrature-based moment methods (QBMM) have been adopted to describe the evolution of soot particle ensembles statistically [9]. While Monte Carlo and sectional models allow to directly compute the soot particle distribution characterizing particles by several independent properties (multivariate models) under laminar conditions, moment methods such as QBMM have been demonstrated to be very suitable for soot simulation in turbulent flames and combustion systems, see [10] for a recent overview.

On the other hand, several groups extended the formulations of the physico-chemical processes involved in the formation and evolution of soot particles aiming at predictive quality. As a result, very detailed models have been formulated and published, e.g. [7,8,11–13]. Among them, the soot model proposed by D'Anna and coworkers [8,13] has been demonstrated to successfully predict soot formation for several conditions including different fuels, equivalence ratios and flame configurations. In the most recent version, it accounts for different types of species such as large PAHs, clusters and agglomerates and includes formulations to describe dehydrogenation and oxidation-induced fragmentation [14]. While the original solution procedure was based on a

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sectional approach, a QBMM based version of the model has been introduced recently in [15] applying the *Conditional Quadrature Method of Moments* (CQMOM) approach [16]. As demonstrated in [15], the CQMOM model is capable to predict soot formation in lightly sooting conditions where the transition from condensed phase nanostructures to soot agglomerates is of minor importance [17]; however, this latter aspect changes at heavily sooting conditions where agglomeration is a dominant process [17].

This study aims to extend and validate the capability of the CQMOM soot model introduced in [15] to predict soot formation not only in lightly but also in heavily sooting conditions. This involves to evaluate the model's capability to predict the transition from nanostructures to soot agglomerates. Recently, these effects were extensively investigated by Sirignano et al. [18] in premixed ethylene flames with different equivalence ratios using a scanning mobility particle sizer (SMPS), laser-induced fluorescence (LIF) and laser-induced incandescence (LII). As will be demonstrated later, the comparison of these measurement results with CQMOM simulations is not directly possible. This is related to the fact that classical moment methods like CQMOM do not predict the number density function (NDF) which describes e.g. the soot particle size distribution. Instead, moment methods focus on the transport of a small set of moments of the distribution. Several studies for different physical systems (e.g. particle formation [19–22], sprays [23], bubbles [24]) have demonstrated that solving only a small set of moments yields results almost as accurate as in reference solutions based on direct solution strategies of the population balance. Thus, moment methods are a very suitable choice both in terms of accuracy and computational efficiency. As a drawback, obtaining information on the shape of the NDF from a small set of moments is a non-trivial, ill-posed problem (Stieltjes problem) and special algorithms are needed to realize a reconstruction of the distribution. Two possible approaches are the *Extended Quadrature Method of Moments* (EQMOM) [25] for univariate systems or the *Extended Conditional Quadrature Method of Moments* (ECQMOM) [26] for multivariate models. Both approaches have already been successfully applied for soot modeling [20,22].

In the present study, we propose a new and complementary procedure based on CQMOM and an additional size distribution reconstruction step using the concept of entropy maximization (EM) [27]. EM has been successfully applied in the field of spray modeling (e.g. [28,29]); however, to the authors' knowledge, it has not yet been transferred and applied for soot formation. Our novel approach proposed here based on a combination of CQMOM and EM is applied to simulate two ethylene/air flames with very different sooting behaviour (very lightly sooting, C/O = 0.67 and sooting, C/O = 0.77) and the results are compared to the measurements of Sirignano et al. [18] mentioned.

2. Soot model

The physico-chemical soot model applied in this work is described extensively in literature [8,13,14], therefore only a brief explanation is given in the following. Starting from PAH formation in the gas phase, the molecular growth continues forming larger PAHs which can either belong to the class of pericondensed aromatic hydrocarbons with a compact and rigid structure and a low H/C ratio such as coronene, or they appear as oligomers of small PAHs linked by aliphatic bonds maintaining a high H/C ratio with a loose structure. Both of these structures can grow via chemical pathways. If the Van der Waals forces are strong enough to hold together these large molecules, a cluster is formed. Clusters can grow through chemical pathway by addition of material from the gas phase. Furthermore, clusters can be subject to physical interaction through coagulation. When the size of clusters is small and their physical state has not reached fully reached a solid structure, coagulation is less effective and it is in fact a coalescence process which preserves the spherical shape of the clusters. When larger clusters are formed, a solid character is established and the coagulation turns to be an aggregation process, giving the chain-like shape which is the final form of soot particles. Furthermore, there is a direct dependency of the sticking efficiency of colliding entities on their chemical structure, which can be related to the respective H/C ratio [13].

The soot model used in this work takes into account all of these aspects as it tracks several properties of the soot precursors and soot particles i.e. their type (high molecular mass aromatic molecule, cluster of molecules or agglomerate of particles), their size (their number of carbon atoms, respectively), their state (radical or stable) and their hydrogen to carbon (H/C) ratio and this is directly considered in the formulation of the kinetic rates. The gas phase model applied includes pyrolysis and oxidation of hydrocarbons and molecular growth pathways to form the first aromatic and successively the rest of PAHs up to pyrene. The model was validated in a wide range of combustion conditions, including different equivalence ratios and different fuels [8,13,14,30–32].

3. Numerical model

Mathematically, the soot model described above can be formulated in terms of a population balance equation (PBE) for the NDF $f(\underline{\xi};\underline{x},t)$. Depending on time t and space $\underline{x}$, the NDF describes the local distribution of characteristical properties $\underline{\xi}$ of the tracked hydrocarbon species. In this work, $\underline{\xi}$ is defined as

$$\underline{\xi} = [\xi_{nc},\xi_{H/C},\xi_{stat},\xi_{typ}]^T, \tag{1}$$

and thus, it represents the species attributes mentioned in Section 2, i.e. the number of carbon atoms ξ_{nc}, the H/C ratio $\xi_{H/C}$, the state ξ_{stat} and the type ξ_{typ}. Since these properties are treated as internal coordinates, the model is quadvariate and the domain of definition for the coordinates is given as follows:

$$\xi_{nc} \in [0,\infty), \tag{2}$$

$$\xi_{H/C} \in [0,1], \tag{3}$$

$$\xi_{stat} \in A, A \in \{\text{stable,radical}\}, \tag{4}$$

$$\xi_{typ} \in B, B \in \{\text{largePAH,cluster,agglomerate}\}. \tag{5}$$

As demonstrated in Eqs. (4) and (5), the two coordinates ξ_{stat} and ξ_{typ} are of discrete nature and this aspect is utilized in the following. Using the concept of conditional density functions, the joint NDF statement $f(\underline{\xi};\underline{x},t)$ can be written as (time and space dependencies are dropped in the remainder for brevity)

$$f(\underline{\xi}) = f_{H/C}(\xi_{H/C}|\xi_{nc},\xi_{stat},\xi_{typ})\cdot f_{nc}(\xi_{nc}|\xi_{stat},\xi_{typ})\cdot n(\xi_{stat},\xi_{typ}), \tag{6}$$

where f_{nc} represents the distribution of ξ_{nc} at a fixed value of (ξ_{stat},ξ_{typ}) and $f_{H/C}$ the distribution of $\xi_{H/C}$ given a certain state of $(\xi_{nc},\xi_{stat},\xi_{typ})$. Due to the discrete nature of ξ_{stat} and ξ_{typ} and their particular domain of definition, the joint bivariate distribution $n(\xi_{stat},\xi_{typ})$ can only assume 6 different values, one for each combination (u,v) of the respective domain parameters $\xi_{stat} = \xi_{stat,u}$, $\xi_{typ} = \xi_{typ,v}$. Thus, the problem is fully described by 6 bivariate NDFs $\Pi_{u,v}(\xi_{nc},\xi_{H/C})$,

$$\Pi_{u,v}(\xi_{nc},\xi_{H/C}) = f^{u,v}_{H/C}(\xi_{H/C}|\xi_{nc})\cdot f^{u,v}_{nc}(\xi_{nc})\cdot n_{u,v}, \tag{7}$$

with a marginal NDF $f^{u,v}_{nc}(\xi_{nc})$ depending only on ξ_{nc}, a conditional NDF $f^{u,v}_{H/C}(\xi_{H/C}|\xi_{nc})$ and the number density $n_{u,v}$ for each specific combination of (u,v).

Two important aspects need to be pointed out here. First, the steps presented above are without loss of generality meaning that no approximations are made. The formulation however allows the soot model to cast one joint quadvariate NDF $f(\underline{\xi})$ to 6 bivariate NDF statements $\Pi_{u,v}$. This is numerically preferable for the moment-based approach CQMOM used in this work. Second, although the overall NDF has been split into 6 bivariate distributions, these NDFs are not independent from each other but they are strongly coupled by various