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Detailed particle nucleation modeling in a sooting ethylene flame using a Conditional Quadrature Method of Moments (CQMOM)

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Abstract

In this work, a detailed kinetic mechanism for soot formation and evolution (D'Anna *et al.* 2010) is implemented in a method of moments framework using the concept of 'Conditional Quadrature Method of Moments' (CQMOM) (Yuan and Fox, 2011). Particle nucleation and growth pathways are described in detail tracking moments of the number density function (NDF) for large PAHs, soot clusters and agglomerates in both stable and radical forms. Individual entities of these groups are characterized by both their size and their H/C ratio, the latter providing information on the chemical structure and the reactivity of the particles. This leads to a multivariate population balance statement. The CQMOM implementation of the soot model is then applied to predict particle formation in a lightly sooting burner-stabilized premixed ethylene flame with an equivalence ratio of $\phi = 2.1$. Several experimental data sets for this target flame are available in the literature and they are used in this study for comparison with the simulation results. The suitability of the soot model to predict particle matter under such conditions is shown, as both the onset of soot formation by nucleation and the subsequent growth are captured. Modeling results offer further insights into the relevance of agglomeration and dehydrogenation in this lightly sooting flame.

Keywords: Soot modeling; Multivariate population balance equation; Premixed flames; Method of moments; QBMM

1. Introduction

Understanding particle nucleation is still one of the major challenges regarding soot formation

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[1–4]. Recent experimental studies have offered a detailed insight into the relevant steps leading to the formation of soot particles [1–2,5]. As a consequence, soot models have been further developed describing both the transition between the gaseous and solid phase and the evolution of the growth and oxidation of particles in detail [6–8].

One of these detailed kinetic soot models has been developed by D'Anna and coworkers and validation was conducted both in premixed and diffusion flames for various fuels [6,9–10]. It considers all important physical and chemical processes, such as soot nucleation both by the dimerization of polycyclic aromatic hydrocarbons (PAHs) [11] and by the molecular growth of PAHs involving both acetylene and aromatic addition [12]. Furthermore, the model captures not only the evolution of the size (the number of C atoms, respectively) of soot clusters and agglomerates but also of large PAH molecules in both radical and stable form. Particles are known to undergo dehydrogenation, which is a key process influencing the chemical structure and therefore the reactivity of soot [13]. This process is also described in the model tracking the H/C ratio of each entity. Altogether, this leads to a multivariate number density function (NDF) and different numerical approaches are available to solve the corresponding population balance equation (PBE).

One possibility is to use a multi-sectional method; this has also been done for the model described above [9]. Another possible approach is given by moment methods, such as MOMIC [14], HMOM [15] or quadrature-based moment methods (QBMM) [16]. Moment methods are known to be well-suited and established for soot simulation in laminar and turbulent flames [17–19]. Recently, the quadrature-based moment methods (QBMMs) are increasingly being applied in the field of particle formation [20–22].

Among the different QBMM approaches available, a suitable choice for multivariate distributions is given by the Conditional Quadrature Method of Moments (CQMOM) [23], which has already been applied successfully to model soot formation in flames [24]. It is within the scope of this study to use the detailed soot model presented above in a *method of moments* context based on CQMOM. The model is applied to simulate a burner-stabilized, premixed $C_2H_4/N_2/O_2$ flame with an equivalence ratio of $\phi = 2.1$. Specifically, the flame investigated is a fundamental one in the field of soot research, being a target flame for both the 'International Workshop on Laser-Induced Incandescence (LII)' [25] and the 'International Sooting Flame Workshop' (ISF) [4]. The lightly sooting character of this flame challenges soot modeling as both the nucleation and the growth mechanism are important under such conditions and the onset of soot and the formation of the first agglomerates needs to be effectively described to predict the total concentration of particle matter. To the authors' knowledge, detailed modeling results for this flame have not yet been published in the archival literature.

The remainder of this paper is structured as follows. First, the physical and chemical reactions considered in the detailed soot model are explained. Then, the implementation of the soot model in a QBMM framework is presented, explaining the derivation of CQMOM-based formulations of the source terms. Afterwards, the new implementation of the soot model is applied to simulate the formation of soot in the premixed flame described above. Experimental data from different groups are available [26–30] and thus, the modeling results are validated against measurement data. Finally, using the combined information of both the modeling and experimental results, the soot characteristics of the flame investigated are discussed, with special focus on the transition from molecules to small particles, the agglomeration and the evolution of the H/C ratio.

2. Soot model

Hydrocarbon oxidation and pyrolysis are described by a detailed kinetic mechanism able to model several premixed flames at atmospheric pressure [6,12]. Fuel-rich conditions promote molecular growth and hydrogen loss. Acetylene (C_2H_2) and methane are the most abundant gaseous species; benzene is the first product of the molecular growth process. PAH formation is modeled by the HACA and the resonantly stabilized free radical (RSFR) mechanism [6] and molecular growth is described up to pyrene (A_4). The kinetic gas-phase mechanism consists of 460 reactions involving 120 species. All the compounds with molecular masses larger than A_4 are not treated as individual species, but considered as lumped species whose evolution is described by a PBE statement. Their reactions are described on the basis of similarity with gas-phase reactions involving PAHs. The molecular growth for the lumped species is initiated by an H atom loss through the H-abstraction reaction by H or OH or the spontaneous formation of radicals. Termination reactions of aromatic radicals are also considered. The formation of pericondensed-aromatic hydrocarbons (PCAHs) is modeled by the addition of C_2H_2 whereas the formation of incompletely-condensed aromatics (aromatic-aliphatic-linked hydrocarbons, AALH) is modeled both by an H-atom substitutions by a pericondensed structure and by the addition of aromatic radicals to non-aromatic double bonds.

Both PCAHs and AALHs can undergo unlimited growth, forming extremely large molecules. Moreover, AALH can also undergo dehydrogenation reactions, migrating to pericondensed