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Modeling soot formation in premixed flames using an Extended Conditional Quadrature Method of Moments

Steffen Salenbauch [a,*], Alberto Cuoci [b], Alessio Frassoldati [b], Chiara Saggese [b], Tiziano Faravelli [b], Christian Hasse [a]

[a] *Chair of Numerical Thermo-Fluid Dynamics, Department of Energy Process Engineering and Chemical Engineering, TU Bergakademie Freiberg, Fuchsmühlenweg 9, Freiberg 09599, Germany*

[b] *Department of Chemistry, Materials and Chemical Engineering, Politecnico di Milano, Piazza Leonardo da Vinci 32, 20133 Milano, Italy*

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ABSTRACT

The scope of this study is the application of the recently developed univariate moment method *Extended Quadrature Method of Moments* (EQMOM) (Yuan et al., 2012) to model soot formation in flames. Furthermore, it is combined with another advanced moment approach, called the *Conditional Quadrature Method of Moments* (CQMOM) (Yuan and Fox, 2011), and this extension leads to a bivariate model.

Retaining the efficiency of a moment method, EQMOM enables the reconstruction of the number density function. CQMOM is a numerically robust multivariate moment method which allows a bivariate soot particle description in terms of particle volume and surface to take into account aggregation. The joint *Extended Conditional Quadrature Method of Moments* (ECQMOM) model combines the advantages of the two methods to arrive at a numerically efficient bivariate moment method which captures both the particle size distribution and the formation of aggregates.

Both the EQMOM and the ECQMOM model are validated against experimental results for premixed burner-stabilized ethylene flames. Thereby, the gas phase is modeled using a modified version of a very detailed, well-established kinetic scheme, which is adapted to be consistent with the moment methods introduced. The results demonstrate the suitability of the applied models to describe both soot precursors and soot evolution in flames. Furthermore, the ability of the moment approaches to represent the statistical soot model accurately is evaluated comparing EQMOM and ECQMOM to other numerical approaches, which are based on the Monte Carlo method, the standard Gaussian Quadrature Method of Moments and the Gaussian-Radau Quadrature Method of Moments, respectively.

1. Introduction

Soot particle formation in combustion has an impact on both combustion efficiency and human health. In terms of legislation, the limitations on soot emissions are tending to become even stricter regarding both volume and number density. In addition, soot plays an important role in radiative heat transfer, where accurate but efficient prediction of the soot evolution is required to precisely model the radiative fluxes in systems like fires [3,4].

Nowadays, most of the detailed phenomenological soot models are based on techniques for solving the population balance equation (PBE), which is a continuity statement written in terms of a number density function (NDF) [5].

Among them, Monte Carlo (MC) based models were developed in order to approximate the PBE-governed soot particle population using an ensemble of stochastic particles [6–12]. Monte Carlo methods are known to yield very accurate results; however, due to their computational expense, their applicability has so far been limited to simple configurations.

Another group of approaches, referred to as Sectional Methods, are based on the separation of the particle size spectrum into a set of size classes [13–22]. While these methods are easy to implement and give detailed information on the particle size distribution, sectional methods are also numerically expensive, especially if the shape of soot particles is described by more than one size property.

The computationally most efficient approach to solving the PBE is given by moment methods. Here, the NDF is not solved directly; instead only a few lower-order moments of the distribution are tracked. As discussed later, this transformation yields unclosed

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* Corresponding author. Fax: +49 3731 394555.
E-mail address: Steffen.Salenbauch@iec.tu-freiberg.de (S. Salenbauch).

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terms in the moment equations. The most widely used moment closure approach is given by the Method of Moments with Interpolative Closure (MOMIC), where unknown moments are interpolated from known ones [23–34].

Another way to achieve closure is given by the Quadrature Method of Moments (QMOM), where the unknown NDF is approximated either by a set of Dirac delta functions or, in newer developments, by kernel density functions [1]. Within recent years, QMOM-based soot models have been applied increasingly [35–40]. Beside univariate approaches, which assume soot particles to be spherical, quasi-multivariate and multivariate approaches have also been developed. Multivariate models offer the possibility to consider aggregation and therefore lead to a more realistic description of the shape of soot particles. However, the univariate moment inversion concept is not easily transferable to multivariate cases [41–43]. Therefore, multivariate moment problems are usually treated using the *Direct Quadrature Method of Moments* (DQMOM) [44]. For instance, Blanquart and Pitsch [39] developed a detailed, trivariate DQMOM-based soot model. The comparison of the results with MC simulations showed that DQMOM yields very accurate results for the PBE. However, DQMOM approaches are numerically challenging, since they require the inversion of a linear system, which may be extremely ill-conditioned [5,45]. Mueller et al. [45,46] therefore suggested a new bivariate approach called *Hybrid Method of Moments* (HMOM), in order to combine the numerical stability of MOMIC with the accuracy of the DQMOM method. The comparison between HMOM, DQMOM and a bivariate MOMIC approach with a MC simulation revealed that the evolution of the soot mass is described adequately using all the moment methods tested [34]. However, with the exception of DQMOM, the moment methods yield deviations from the MC simulations regarding the temporal evolution of the particle number density.

Besides numerical stability and accuracy issues, one of the most severe restrictions of state-of-the-art moment methods is the lack of a resolved NDF. Due to this, source terms in the transport equations cannot be formulated as a continuous function of the particle size. Thus, effects such as the reduced collision efficiency of the smallest particles [47,48] cannot be implemented accurately in standard moment methods with the same precision as in MC.

In order to overcome these limitations, Yuan et al. [1] proposed an *Extended Quadrature Method of Moments* (EQMOM), which enables the shape of the particle size distribution to be reconstructed from a moment set using kernel density functions instead of Dirac delta functions. EQMOM was evaluated for 13 benchmark test cases and further applied to model radiation transport [49], but not yet to model soot formation in flames.

However, EQMOM is a univariate moment method and, therefore, the aggregation of soot particles cannot be accounted for accurately. It is known that even small particles can build aggregates upon collision [33,50–52]. Therefore, aggregation needs to be considered in soot models in order to describe the evolution of soot particle ensembles properly. This implies the application of a bivariate NDF. Yuan and Fox [2] developed a multivariate moment approach called *Conditional Quadrature Method of Moments* (CQMOM) to handle bivariate moment formulations in a numerically robust way. The suitability of CQMOM to describe particle ensembles has already been demonstrated by modeling TiO_2 formation in flames [53,54]. However, in contrast to EQMOM, CQMOM is based on the standard Gaussian-QMOM technique and the NDF is thus not known. A possible modification to standard QMOM techniques such as CQMOM is given by QMOM-Radau, where the Gauss-Radau quadrature interpolation rule is applied to fix a quadrature node at the smallest particle size [55]. However, to the authors' knowledge, QMOM-Radau based approaches have not yet been published for particles or even soot so far.

The scope of this paper is to apply EQMOM and CQMOM to model soot formation in premixed flames. The two models are initially used separately. Then, a combination of the two models, called the *Extended Conditional Quadrature Method of Moments* (ECQMOM) [5], is formulated for sooting flames. To do so, the processes of nucleation, coagulation, condensation and HACA surface growth [12,25,29] are formulated in the context of an EQMOM and a CQMOM approach. In order to obtain a numerically stable moment method, which resolves the soot particle size distribution and captures aggregation, the two approaches are combined to form the ECQMOM method. Special focus is put on the methods' capability to close the moment source terms accurately by comparing the EQMOM-based methods to Gaussian-QMOM models and a MC approach [56]. The gas phase is modeled using a modified version of the extensively validated CRECK mechanism [57–61]. In order to be consistent with the soot model, pyrene (A4-$C_{16}H_{10}$) is set as the largest polycyclic aromatic hydrocarbon (PAH) species in the model, which accounts for all larger ones.

The remainder of this paper is organized as follows: First, the numerical model is introduced and explained in Section 2. The methods of EQMOM, QMOM-Radau, CQMOM and ECQMOM are used to model soot for the first time. Therefore, detailed explanations are given on the application of these methods to the soot model. Next, the kinetic scheme to describe the gas phase is explained. Afterwards, the ability of this scheme to predict with accuracy not only the major species of a fuel-rich flame, but also PAH species is demonstrated in Section 3, where the model is compared to experimental results [62–68]. The kinetic mechanism is then applied to model the gas phase of two sooting reference flames, which serve as validation cases for the moment-based soot models. This involves the simulation results with the univariate EQMOM method being compared to experiments as well as to QMOM, QMOM-Radau and MC results for a premixed burner-stabilized ethylene flame, where aggregation was found to be negligible [69]. A similar comparison to experiments, CQMOM and MC simulations follows for the bivariate ECQMOM soot model introduced. Here, another burner-stabilized ethylene flame, where aggregation is known to be an important effect [70], is chosen as the test flame. Finally, conclusions are drawn in Section 4.

2. Numerical model

2.1. Method of moments

The evolution of the soot NDF $n(t,\mathbf{x};\xi)$ in fuel-rich premixed flames is governed by the PBE:

$$\frac{\partial n(t,\mathbf{x};\xi)}{\partial t} + \frac{\partial \mathbf{u} n(t,\mathbf{x};\xi)}{\partial \mathbf{x}} = \dot{n}(t,\mathbf{x};\xi). \tag{1}$$

In this study, diffusive terms are neglected. For molecular diffusion, this is justified by the high Schmidt number of soot particles, as shown by Bisetti et al. [71]. In addition, thermophoresis is known to have only a minor effect on the transport velocity of soot particles in premixed flames [70,72,73]. Therefore, in this study, soot particles move along the axis with the local gas velocity. The source term $\dot{n}(t,\mathbf{x},\xi)$ accounts for the physical and chemical processes of particle nucleation $\dot{n}_{nuc}$, coagulation $\dot{n}_{coag}$, PAH condensation $\dot{n}_{cond}$ and chemical surface growth $\dot{n}_{sg}$:

$$\dot{n}(t,\mathbf{x};\xi) = \dot{n}_{nuc}(t,\mathbf{x}) + \dot{n}_{coag}(t,\mathbf{x};\xi) + \dot{n}_{cond}(t,\mathbf{x};\xi) + \dot{n}_{sg}(t,\mathbf{x};\xi). \tag{2}$$

In this study, we put the focus on fuel-rich premixed flames. Therefore, soot particle oxidation is not considered, as previous studies have shown that oxidation has a minor effect on the soot evolution in fuel-rich premixed flames [6,24,70,74]. This is confirmed by Xu et al. [70], who experimentally and numerically