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Numerical simulation and parametric sensitivity study of particle size distributions in a burner-stabilised stagnation flame

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ABSTRACT

A detailed population balance model is used to perform a parametric sensitivity study on the computed particle size distributions (PSDs) for a laminar premixed ethylene burner-stabilised stagnation flame. The soot morphology in the post-flame region is studied using computed sintering level distributions, fringe length analysis of the polycyclic aromatic hydrocarbons (PAHs) within the primary soot particles, and TEM-like projections of aggregates. The computed PSDs were sensitive to the minimum particle inception size, the coagulation rate and the inception species concentration. Changes in the particle inception size and the coagulation rate led to an overall shift in the position of the coagulation peak. Only changes in the inception species concentration led to a systematic shift in both the position of the trough between the modes of the bimodal PSD and the coagulation peak at larger diameters. Given the overall model, varying the inception species concentration with each burner-stagnation plate separation was the only means possible to achieve a satisfactory agreement between the experimental and computed PSDs. This study shows that further work is required to better understand the soot precursor chemistry, the inception of soot particles. Additional work may also be needed in the area of experimental mobility sizing for the flame studied here.

1. Introduction

Much progress has been made to understand the chemical and physical processes underlying soot formation. However, details of the individual processes remain an open question [1,2]. In general, a basic understanding of the complex and often competitive processes of soot nucleation and mass/size growth requires detailed kinetic modeling through comparison of model results and experimental data and sensitivity analysis. For this purpose, a range of experimental techniques have been developed to probe the size and mass evolution of soot in flames. Soot volume fraction is most commonly measured using light extinction and scattering [3] and laser light incandescence [4]. However, interpretation of the laser-based experiment requires a fairly precise knowledge about the refractive index of young soot which remains poorly understood. In principle, independent verification of the optical measurements may be made using small-angle neutron scattering [5], small-angle X-ray scattering [6] and thermocouple particle densitometry [7], though very little systematic efforts have been made in that direction. The detailed distribution of particle sizes may be measured by a scanning mobility particle sizer (SMPS) (see, e.g., [8–13]). Though the method is intrusive to the flame, it nonetheless provides more detailed features about the evolution of the particle size distribution (PSD), from very small, incipient particles to aggregates as large as 100 nm in mobility diameter. Ex situ analysis by high-resolution microcopy of particles collected by a rapid insertion technique has been routinely employed (see, e.g., [14–17]). Whilst transmission electron microscopy (TEM) images show the morphology of a soot particle, the higher magnification (see, e.g., [18]) is able to reveal some details of the internal structure of mature soot. Imaging incipient soot a few nanometers in size by TEM is challenging [10], because of potential sample damage by the vacuum environment and the high-energy electron beam of TEM, as shown recently by Schenk et al. [17]. Recent advances in Helium ion microscopy (HIM) [17,19] offer a

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low-energy, softer and high-contrast solution to incipient soot imaging. The chemical composition of soot may be analysed using laser microprobe spectroscopy [14,20] and photoionisation aerosol mass spectroscopy [21].

A large portion of the experimental data that are available with regards to the time evolution of soot PSD has been gathered in premixed flames [8–10,13,21–24]. A sample probe is typically placed across the flame and PSDs would be measured by a SMPS. This sampling technique is inherently intrusive and perturbs the flame [8,25]. For this reason, a premixed burner-stabilised stagnation flame (BSSF) configuration was introduced where the sample probe is integrated into a water-cooled stagnation plate as a flow stagnation surface for which pseudo-one-dimensional numerical solution of the flame problem becomes feasible [26]. The probe in that setup may be treated as the flame boundary condition; thus, eliminating the problem in earlier setups in which the probe effects on fluid mechanics and reaction kinetics cannot be quantified easily. The setup, along with the numerical modeling method for such flames, removes the need to carry out arbitrary "time or spatial distance shifting" as it was customarily employed in comparison of experimental data and modelling result.

Beyond the probe effect, measurements by SMPS coupled with BSSF faces other complications when such measurements are used for testing models. Like any other techniques for probing soot in flames, the interpretation of the data is not always straightforward. Mobility measurements yields essentially the collision diameter of the particles. Thus two factors can impact a proper comparison between model and experiment. The first factor lies in the unknown morphology of the particles undergoing mass and size growth. The second factor is related to the internal structure of the particles and thus the mass density of the particle. HIM measurements have shown that particles < 10 nm in size can exhibit odd, non-spherical shapes [17,19]. Considering the experimental issues just discussed, it would be beneficial to carry out a study in which the model and experimental uncertainties are considered sequentially. This paper will be a step in that direction.

The soot PSDs for an ethylene BSSF were modelled by Lindstedt and Waldheim [27] using a surface-volume description of particles and a sectional method. To counteract the excessive depletion of the small particles in their model, they introduced a collision efficiency for coagulation which varies between 1 for particle diameters greater than 5 nm and about 0.01 for pyrene. However, there are still some unresolved problems. For example, the model is unable to predict particle diameters across all burner-stagnation plate separations. It remains unclear whether the discrepancy is caused by the particle morphological assumptions of the model in the simulation or by other factors.

There are also many detailed models of soot formation and solution methods. For example, Kraft and co-workers [28–34] employ a detailed population balance model which is solved using a stochastic method. The model describes particles as aggregates composed of primary particles which are in turn composed of individual polycyclic aromatic hydrocarbons (PAHs), thus containing information on particle size, morphology, and the internal structure of nanoparticles. This particular modelling approach has been successfully applied to the analysis of a variety of nanoparticles. It was used to simulate the PSDs of soot particles in laminar premixed flames [22,35,36] and to uncover the various factors that govern the shape of PSDs and their time evolution [22,35]. Specifically, Singh et al. [35] performed a sensitivity analysis of the PSDs to various kinetic parameters in the hydrogen-abstraction–carbon addition (HACA) mechanism [37]. A unique feature of the model is that it resolves the size and connectivity of the primary particles in an aggregate; therefore, TEM-like projections of aggregates could be produced to visualise the temporal evolution of the fractal dimension in different flames [36]. Similar investigations of soot PSDs, morphology and composition have been performed in the context of engines [38]. The stochastic approach was also used to follow the morphology of aerosols in Titan's atmosphere [39] and to study the sintering of titania [40], silica [41] and silicon [42]. One has to be aware, though, that the increasing complexity of the model comes at a cost of including a large number of parameters, some of which were calibrated against experiments, while others remain poorly known.

The **purpose of this paper** is to investigate whether the model can describe the evolution of soot PSDs observed in the BSSF of [26]. The flame chemistry and structure was computed using a pseudo-one-dimensional stagnation flow flame code with appropriate boundary conditions. The particle dynamics were solved using a detailed population balance model. A thorough parametric sensitivity study is carried out here to understand how the various submodels and model parameters impact the various PSD features quantitatively and to shed light on the mobility measurement, especially concerning the particle morphology and its effect on the interpretation of the mobility diameter.

The paper is organised as follows: Section 2 introduces the gas-phase chemical mechanism, the governing equations and boundary conditions for the burner-stagnation flame configuration, and key aspects of the detailed population balance model. Sections 3.1 and 3.2 present the temperature and species profiles, and the PSDs. Section 3.3 presents the parametric sensitivity study of the computed PSDs. Section 3.4 shows various aspects of the soot morphology calculated from the detailed population balance model. Lastly, implications on the experimental measurements are discussed in Section 4.

2. Computational method

The computational method consists of two parts. In the first part, temperature and species profiles are computed using a modified version [26] of Oppdif [43,44], including calculation of the source terms by the Method of Moment with Interpolative Closure (MOMIC) using the code published by Revzan et al. [45]. A gas-phase chemical mechanism, and species thermodynamic and transport properties are supplied as input. The transport equations of the moments of the PSD are solved to approximately account for the production and consumption of key gas-phase species due to inception, surface growth, oxidation and condensation processes. A total of six moments, including the zeroth moment, were solved using MOMIC to close the moment transport equations. In the second part, a detailed population balance model is applied as a post-processing step where the computed temperature and species profiles from Oppdif are supplied as input. This two-step methodology has been applied to the studies of a number of laminar premixed flames [46–48] and ideal reactor simulations [49].

Oppdif simulations were performed using an unburned-gas composition (molar basis) of 16.3% ethylene, 23.7% oxygen and 60% argon (an equivalence ratio of 2.07), a cold-gas velocity of 8 cm/s (STP), a gas temperature of 473 K at the burner surface and at atmospheric pressure. The method for determining the gas temperature at the burner surface has been discussed in [26]. Windward differencing was used and multi-component transport and thermal diffusion were considered. About 200 grid points were found to be sufficient for convergence. The energy equation was solved with both gas and particle radiation.

The detailed population balance model requires the computed profiles from Oppdif to be expressed in terms of the residence time of a Lagrangian particle travelling from the burner to the stagnation plate. The combined axial convective velocity and thermophoretic velocity were used to perform the conversion as per Abid et al. [26].