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Simulation and analysis of the soot particle size distribution in a turbulent nonpremixed flame

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ABSTRACT

A modeling framework based on Direct Simulation Monte Carlo (DSMC) is employed to simulate the evolution of the soot particle size distribution in turbulent sooting flames. The stochastic reactor describes the evolution of soot in fluid parcels following Lagrangian trajectories in a turbulent flow field. The trajectories are sampled from a Direct Numerical Simulation (DNS) of a *n*-heptane turbulent nonpremixed flame. The DSMC method is validated against experimentally measured size distributions in laminar premixed flames and found to reproduce quantitatively the experimental results, including the appearance of the second mode at large aggregate sizes and the presence of a trough at mobility diameters in the range 3–8 nm. The model is then applied to the simulation of soot formation and growth in simplified configurations featuring a constant concentration of soot precursors and the evolution of the size distribution in time is found to depend on the intensity of the nucleation rate. Higher nucleation rates lead to a higher peak in number density and to the size distribution attaining its second mode sooner. The ensemble-averaged PSDF in the turbulent flame is computed from individual samples of the PSDF from large sets of Lagrangian trajectories. This statistical measure is equivalent to time-averaged, scanning mobility particle size (SMPS) measurements in turbulent flames. Although individual trajectories display strong bimodality as in laminar flames, the ensemble-average PSDF possesses only one mode and a long, broad tail, which implies significant polydispersity induced by turbulence. Our results agree very well with SMPS measurements available in the literature. Conditioning on key features of the trajectory, such as mixture fraction or radial locations does not reduce the scatter in the size distributions and the ensemble-averaged PSDF remains broad. The results highlight and explain the important role of turbulence in broadening the size distribution of particles in turbulent sooting flames.

1. Introduction

Soot is formed as result of fuel pyrolysis or combustion under rich conditions. Soot aggregates consist of elementary spherical particles, which are referred to as *primary particles*. The primary particle size falls in the range $\mathcal{O}$(1–10 nm) [1] and is affected by the residence time in the flame environment and by the fuel [2].

Since primary particles are arranged in complex, three-dimensional structures, a definition of a characteristic length of a soot aggregate is not straightforward. It is usually defined as a function of the number and size of the primary particles describing soot as fractal aggregates [3], or as a characteristic length depending on the measurement technique [4], such as the mobility diameter [5]. The characteristic size of a typical soot aggregate spans from a few nanometers for nucleated particles composed of a single primary particle up to $\mathcal{O}$(1–10 μm) due to the hundreds or thousands of primary particles in carbon black [6].

The size of soot aggregates is crucial in determining the risk posed to the population and the environment [7]. In particular, ultrafine particles below 100 nm are very hazardous, since they can penetrate the human cell membrane easily [8]. Moreover, these particles are difficult to filter out of the exhaust, since they escape the post-combustion cleaning devices, contributing to air pollution [9]. For these reasons, regulations on soot emissions

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distinguish the particulate matter emissions thresholds by considering the particulate size [10,11]. As the regulations are gradually tightened, the need to characterize the soot particle size distribution function (PSDF) and its evolution in flames is critical.

Several intrusive experimental techniques allow for the determination of the PSDF, such as probe-based particle mobility measurements [5,12,13] or grid-based sampling followed by TEM image postprocessing [14]. The sampling process induces flame perturbations [15] and results may suffer from particle loss and growth due to coagulation in the extraction probe and sampling lines. In addition, the measurements are time-averaged over several minutes, although fast-scanning technology has been developed recently, providing size distribution information with a time resolution of 1 s [16]. In this context, numerical simulations are a very useful tool due to the time and space resolution they provide. Two possible modeling approaches are widely employed for the purpose of simulating soot size distributions. The sectional method [17,18], based on a direct discretization of the population balance equation [19], and the Monte Carlo method [20], which reproduces the evolution of a population of particles via stochastic methods.

Numerical [18,21,22] and experimental measurements [1,5,12] show that the PSDF in laminar flames evolves from unimodal, at locations close to the burner, to bimodal further downstream. Bimodality is defined here to indicate that the PSDF has two different modes, appearing as two distinct peaks in size space. The first mode is located at small particle sizes. The peak of second mode is at larger sizes and is due to growth processes, such as PAH condensation, acetylene-driven surface growth (HACA [23]), and coagulation of existing soot particles. Bimodality originates from the persistent nucleation in the presence of large particles [5].

In real devices, combustion occurs in the turbulent regime. Turbulence imposes chaotic changes to the thermochemical properties of the mixture, hence the time average of the PSDF at a given location can be very different from its instantaneous realization. This is not the case in laminar flows, where the flows are steady. It is expected that the PSDF in a turbulent flow flame differs from that observed in laminar flames. The knowledge of PSDF in turbulent flames is limited, reflecting the challenge of performing detailed measurements in turbulent flames, and the high computational cost of executing DNS of turbulent sooting flames. Chowdhury et al. [24] have recently performed scanning mobility particle size (SMPS) measurements on probe-sampled soot collected on the centerline of a turbulent sooting flame of ethylene. In contrast with known trends in laminar flames, the authors found that the PSDF does not show a second mode. Instead, it appears as a polydisperse distribution with only one evident peak, at small particle size, and a marked tail towards larger sizes, spanning over many orders of magnitudes. These findings agree with mobility measurements of alumina powder in turbulent flows [25]. In the present study, statistics of the PSDF of soot particles in a turbulent nonpremixed flame for which data from a Direct Numerical Simulation are available [26], are computed using a Direct Simulation Monte Carlo (DSMC) approach [20], coupled with a detailed soot model. We aim to explain the mentioned difference between findings in laminar and turbulent flames.

The paper is organized as follows. Firstly, the physical models describing soot formation and growth, and the DSMC algorithm for the evolution of the soot population are briefly introduced. A test case is discussed in order to facilitate the understanding of the relevant physical processes. Then, a comparison with a set of experimental measurements in laminar premixed flames is performed. Finally, results obtained from the target turbulent flame at selected locations are presented.

2. Soot physical models and statistical description

A soot particle is described as a fractal aggregate with volume V and surface S, consisting of n_p primary particles with diameter D_p [27]:

$$D_p = 6VS^{-1} \quad \text{and} \quad n_p = \frac{1}{36\pi} V^{-2} S^3. \tag{1}$$

Soot nucleation [28,29], acetylene-driven surface growth (HACA [23]), condensation of PAH species over the particle surface [28,29], oxidation due to OH and O_2 [30,31], and particle coagulation are included in the model. The rates of these processes depend on the thermochemical state of the background gaseous mixture as well as the volume and surface of the soot particle. The details of the processes are available in Refs. [28,29] and references therein, and only a short description is given here.

Nucleation of a soot particle occurs when two dimers collide [32], forming a spherical particle of diameter $D_0 = 0.94$ nm. A dimer is a molecular cluster composed of two Polycyclic Aromatic Hydrocarbon (PAH) molecules. Dimers condense on the surface of soot particles (condensation) or collide with each other forming a new soot particle (nucleation). The dimers concentration is assumed to be in steady state, so that their rate of formation is equal to the sum of the soot nucleation and condensation rates [28]. In this work, only naphthalene dimers are considered for soot nucleation, with a sticking coefficient equal to 0.002 [28,33,34]. This modeling choice is not necessary, but is adopted here for consistency with the modeling approach used in the DNS [26].

Soot oxidation results in soot particles loosing mass. The oxidation model and rates employed in the present work are discussed in detail in Ref. [28]. A particle is removed if its volume becomes smaller than the volume of nucleated particles. Thus, the oxidation model accounts for complete particle depletion. Oxidation induced fragmentation [35] is not included.

Here, the term *coagulation* refers loosely to the formation of a new particle as a consequence of the collision of two particles. The coagulation model reflects experimental observations on soot particles and aggregates at various stages of soot growth. Small particles in laminar flames shortly after nucleation behave as liquid-like [36]. On the contrary, large particles stick to each other to form larger aggregates. These hypotheses lie at the basis of the modeling approach for the outcome of a coagulation event as explained in Ref. [28]. The outcome depends on the volume of the colliding particles, which are distinguished in small and large. The threshold that differentiates small and large particles is set at a critical size D_t. In our work, we consider $D_t = 1$ nm. This value yields results that are consistent with the HMOM approach used in the DNS of sooting turbulent nonpremixed flame in Ref. [26]. Two small colliding particles coalesce into a spherical particle, two large particles stick in a chain, while a small particle splashes on a large one. The volume is always conserved, while the surface of the resulting aggregate is computed following the functional expressions given in Ref. [28]

It should be noted that there is little experimental evidence supporting this modeling approach conclusively, although this coagulation model has been applied extensively to several laminar flames [28,29] and shown to produce volume fractions, number densities, and particle sizes that are in good agreement with experimental data.

The collision diameter of the aggregate is computed from the volume, the surface, and the fractal dimension as [37,38]:

$$D_c = \frac{6}{(36\pi)^{1/D_f}} V^{1-2/D_f} S^{3/D_f-1}. \tag{2}$$

The value of the fractal dimension is $D_f = 1.8$ [28]. A mobility diameter [39] D_m may be computed adopting a model that relates