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Brownian dynamics simulation of soot primary particle aggregation in laminar ethylene diffusion flames

Ya-fei Wang, Qun-xing Huang *, Fei Wang, Yong Chi, Jian-hua Yan

State Key Laboratory of Clean Energy Utilization, Zhejiang University, Hangzhou, 310027, China

HIGHLIGHTS

- A more realistic simulation of soot primary particle aggregation is proposed.
- This simulation approach can dynamically display soot morphology evolution.
- Fractal properties of simulated soot aggregates agree with the experimental data.

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ABSTRACT

The aggregation of soot primary particles along the centerline of laminar ethylene diffusion flames is simulated using the Brownian dynamics method incorporating the cluster–cluster aggregation model. Unlike previous studies, this simulation approach takes more key parameters into account, such as the temperature, dynamic viscosity and velocity of the flame gas, the primary particle diameter distribution and the soot volume fraction. Given that the change in the O_2 concentration would greatly affect these simulation parameters, three aggregation simulations for flames at different O_2 concentrations are implemented to check the applicability of this simulation approach for different sets of simulation parameters. These simulations dynamically display the soot morphology evolution from isolated mature primary particles to fractal-like aggregates along the flame centerline, and simulation results indicate that the fractal properties of simulated soot aggregates are almost identical, yielding a fractal dimension of 1.70-1.74 and a fractal prefactor of 2.13-2.23, and the mean number of primary particles per simulated soot particle is in the range of 1-83. Moreover, as the O_2 concentration increases from 21% to 40%, the simulated height of the soot aggregation region along the flame centerline decreases from 25 mm to 10 mm, and the simulated time of soot aggregation reduces from 18.5 ms to 8.8 ms. On the whole, these simulation results agree well with the experimental data.

1. Introduction

A detailed knowledge of soot aggregate structures is the foundation for the theoretical calculation of various basic properties of soot aggregates, such as the optical cross-sections [1], the heat transfer [2] and the surface area per unit volume [3]. Transmission electron microscopy (TEM) studies have shown that soot aggregates consist of spherical mature primary particles and have fractal structures [4–6]. Moreover, Forrest and Witten [7] suggested the following statistical

* Corresponding author.
E-mail address: hqx@zju.edu.cn (Q.-x. Huang).

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scaling law to describe the fractal structures of soot aggregates:

$$N = k_f \left(\frac{2R_g}{\overline{d}_p} \right)^{D_f}, \tag{1}$$

where N is the number of primary particles in the aggregate, $\overline{d}_p$ is the mean diameter of the primary particles, R_g is the radius of gyration of the aggregate, k_f is the fractal prefactor, and D_f is the fractal dimension. However, the direct experimental determination of soot aggregate structures is difficult and requires either the extensive data reduction of stereopair soot images or the alternative evaluation from projected soot images [8]. As a result, the computer simulation of soot aggregation, by which soot aggregate structures can be obtained easily, has attracted increasing attention from researchers.

Over the past decades, the Brownian dynamics (BD) method has been widely used to simulate the soot aggregate formation [9–11]. This method generally begins the aggregation simulation by dispersing primary particles uniformly in a cube, and then primary particles are allowed to move in random directions, collide and form aggregates. Based on whether aggregation in the simulation occurs between isolated primary particles and clusters or between clusters, these aggregation simulations can be divided into two models: particle–cluster aggregation (PCA) and cluster–cluster aggregation (CCA). Using the BD method incorporating the CCA model, Mountain and Mulholland [9] created monodisperse aggregates consisting of 10–700 uniform primary particles, and their fractal properties were $1.7 < D_f < 1.9$ and $k_f = 1.58$. In addition, Watanabe and Tanaka [10] performed the BD simulation of the aggregation of submicron particles in static gas. It was shown that the D_f of aggregates generated with the CCA model and the PCA model were 1.2 and 1.6, respectively. However, the fractal properties of aggregates generated by these implementations of the BD method were not well consistent with the experimental data of $1.7 < D_f < 1.9$ and $2.1 < k_f < 2.4$ [8,12–14], and the soot volume fraction in these BD simulations was much higher than the experimental value. In general, the existing implementations of the BD simulation method have been oversimplified, and the Langevin equation used to describe the translational and rotational motion of Brownian particles and the momentum exchange during collision are not solved simultaneously. For example, Li et al. [11] conducted a three-dimensional on-lattice CCA simulation to dynamically display the aggregation process, where primary particles and clusters are only allowed to move translationally in six directions (up, down, left, right, front and back) with a constant displacement at each step. To reconstruct fractal-like aggregates with the prescribed D_f and k_f, Thouy and Jullien [15] modified the standard CCA and proposed the tunable dimension cluster–cluster aggregation (TDCCA), based on a hierarchical scheme of aggregation of equal-sized clusters at each step. Subsequently, Filippov et al. [2] developed a more general TDCCA algorithm, which allows clusters with equal number of primary particles to combine, so that the primary particle number in simulated aggregates is not necessarily a power of 2. These TDCCA algorithms can generate monodisperse aggregates with a wide range of D_f (1.3–2.3) and k_f (1.3–2.3). Although the fractal structures created with the TDCCA model can be qualitatively similar to real soot aggregates, the simulation process does not comply with the randomness of the Brownian motion of primary particles and clusters. At each step of the simulation, the newly-added primary particle or cluster is repeatedly positioned relative to the fixed aggregate based on the hierarchical scheme until Eq. (1) gets satisfied for the prescribed D_f and k_f. In other words, the implementation process of the TDCCA model cannot represent the actual aggregation process of soot primary particles.

The above literature review has shown that the existing soot aggregation simulations often fail to meet the following two requirements simultaneously: reconstructing soot aggregates with the fractal properties consistent with the experimental data and conforming to the actual soot aggregation process. Therefore, implementing the soot aggregation simulation of meeting the two requirements becomes the aim of this paper. In this study, the BD method was still adopted to describe the translation and rotation of primary particles and aggregates. Unlike the previous studies, the simulation approach presented here took more key parameters into account, such as the temperature, dynamic viscosity and velocity of the flame gas, the primary particle diameter distribution and the soot volume fraction. These simulation parameters were experimentally obtained from the centerline of laminar ethylene diffusion flames, and the simulation dynamically displayed the aggregation process of soot primary particles along the flame centerline. The comparison of the simulated soot aggregates with those captured experimentally was carried out to evaluate the simulation results. Because the change in the O_2 concentration would greatly affect simulation parameters, soot aggregation simulations for flames at different O_2 concentrations were performed to check the applicability of this simulation approach for different sets of simulation parameters.

2. Simulation method

2.1. Motion equation of isolated primary particles and aggregates

Brownian dynamics can be used to describe the motion of tiny particles. It is a simplification of Langevin dynamics and corresponds to the limit where no average acceleration occurs [16]. For an isolated primary particle with the mass m and the translational velocity $\mathbf{v}$ immersed in the flame gas, its translational motion can be expressed by the Langevin equation:

$$m\frac{\mathrm{d}\mathbf{v}}{\mathrm{d}t} = \mathbf{F}_d + \mathbf{F}_g + \mathbf{F}_b, \tag{2}$$

where $\mathbf{F}_d$, $\mathbf{F}_g$ and $\mathbf{F}_b$ are the drag force, the gravitational force and the Brownian force, respectively. In terms of the primary particle diameter d_p, the translational velocity of the flame gas $\mathbf{V}$ and the gravitational acceleration $\mathbf{g}$, $\mathbf{F}_d$ and $\mathbf{F}_g$ can be written