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Combustion and Flame 145 (2006) 117–127
Combustion and Flame
www.elsevier.com/locate/combustflame

Numerical simulation and sensitivity analysis of detailed soot particle size distribution in laminar premixed ethylene flames

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Received 29 July 2005; received in revised form 2 November 2005; accepted 23 November 2005

Available online 9 January 2006

Abstract

In this paper, the prediction of a soot model [J. Appel, H. Bockhorn, M. Frenklach, Combust. Flame 121 (2000) 122–136] is compared to a recently published set of highly detailed soot particle size distributions [B. Zhao, Z. Yang, Z. Li, M.V. Johnston, H. Wang, Proc. Combust. Inst. 30 (2005)]. A stochastic approach is used to obtain soot particle size distributions (PSDs). The key features of the measured and simulated particle size distributions are identified and used as a simple way of comparing PSDs. The sensitivity of the soot PSDs to the parameters defining parts of the soot model, such as soot inception, particle and PAH collision efficiency and enhancement, and surface activity is investigated. Incepting soot particle size is found to have a very significant effect on the small-size end of the PSDs, especially the position of the trough for a bimodal soot PSDs. A new model for the decay in the surface activity is proposed in which the activity of the soot particle depends only on the history of that particle and the local temperature in the flame. This is a first attempt to use local flame variables to define the surface aging which has major impact on the prediction of the large-size end of the PSDs. Using these modifications to the soot model it is possible to improve the agreement between some of the points of interest in the simulated and measured PSDs. The paper achieves the task to help advance the soot models to predict soot PSD in addition to soot volume fraction and number density, which has been the focus of the literature.

Keywords: Soot; Particle size distribution; Premixed flames

1. Introduction

Understanding the mechanisms of soot formation is a long-standing challenge in combustion research. Quantitative knowledge of soot formation has been largely derived from three types of work: measurement of soot volume fraction, number density, and particle size distributions (PSD) [1–8]; development of detailed chemical mechanisms for the formation of polycyclic aromatic hydrocarbons [9–16]; and development of soot population dynamics models to describe the evolution of the particle ensemble [5,9,10,13,17–25]. The latest soot models include detailed

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0010-2180/$ – see front matter
doi:10.1016/j.combustflame.2005.11.003

gas-phase reaction kinetics and specifically defined elementary processes leading to soot nucleation, mass and size growth, and oxidation. Advanced and fast numerical techniques are also available to solve the dynamics of soot formation and provide the evolution of soot PSDs [25].

To date most of the modeling work has concentrated on matching the numerical results with mean properties of soot PSDs, including soot volume fraction and total number density. These studies have led to a wide range of basic understanding concerning the chemistry and physics of soot formation, but many questions remain and debate continues. These questions include, for example, the nature and size of soot nuclei, the mechanism of soot inception, the nature and number of chemically active sites on soot surface available for gas–surface reaction, and the sticking probabilities of particle–particle and PAH–particle collision. Over the last decade, progresses have been somewhat limited partly because a further understanding of these issues requires experimental information about soot PSDs beyond their mean properties.

The recent developments of advanced soot measurement techniques, including probe sampling followed by detailed PSD measurements with a scanning mobility particle sizer (SMPS) [3–6], small-angle X-ray scattering [7], and small-angle neutron scattering [8] have provided soot data beyond the mean properties of PSDs. The spatial and temporal evolution of soot PSDs resulting from these experimental developments offers some unique modeling opportunities and challenges. The various issues concerning soot inception and mass growth may now be investigated on the basis of the new and more detailed experimental data and the recent development of advanced numerical techniques for solving the dynamics of soot formation [25–28].

To isolate various issues concerning soot inception and mass growth one is interested in investigating the spatial and temporal evolution of the soot particle size distribution in simple systems, e.g., laminar premixed flames. In a recent study, Zhao et al. [6] presented experimental data on the variations of soot particle size distributions as a function of the maximum flame temperature. These measurements yielded normalized particle size distributions at several heights from the burner for a series of burner-stabilized, laminar, premixed ethylene–oxygen–argon flames at a pressure of 1 bar. The purpose of this paper is to present an attempt at modeling these PSDs and to test the sensitivity of soot PSDs with respect to processes and parameters in the soot model. This information will help to provide insights to further our understanding of the fundamental sooting processes and to refine available models of soot formation.

The base soot model used in the current study is identical to that documented in Appel et al. [13], which has been widely used in recent years. The gas-phase chemistry of this model is largely based on that of Wang and Frenklach [11] for PAH formation in C_2 hydrocarbon flames; the particle inception and growth kinetics and mechanism derive from the early work of Frenklach and Wang [9]. The computational method used to obtain soot PSD is a stochastic one [25–28] which gives the exact solution to the population balance equation describing the dynamics of the particle population. This method may be thought of as a simulation of the inherently random interactions of individual soot particles within a small volume element which is assumed to be representative of its neighborhood as it moves through the flame. It is important to note that the algorithm has a rigorous mathematical basis which does not depend on this heuristic description [29,30].

The paper is organized in the following manner. In Section 2 the experimental data of [6] are discussed in detail and the points for comparison of soot PSDs are outlined. The results obtained from the soot model are then compared to the experimental measurements. In Section 3 we discuss the computational details. In Sections 4 and 5 computational results are compared to the experimental data and numerical sensitivity studies are performed to understand the effect of model parameters on the soot PSDs. Also, inferences are drawn from these analyses and potential improvements are proposed to resolve the model and experimental discrepancies.

2. Experimental data analysis

The ethylene–oxygen–diluent flames in [6], described in Table 1, were chosen for the current study. The experimental details, particle sampling, and measurement techniques have been discussed elsewhere [4–6]. In unburned gas the fuel and oxygen composition was held fixed (24.2 mol% C_2H_4 and 37.9 mol% O_2), but maximum flame temperature differs due to variations of cold gas velocity (series A) or the inert composition (series B). The two sets of flames were designed to provide conditions leading to both unimodal and bimodal PSDs within the measurable range of particle diameters by SMPS.

The PSDs were measured as a function of distance H from the burner surface and usually cover the range of $H = 5$ to 12 mm. In [6] the SMPS mobility diameter was corrected to yield real diameter on the basis of a nanoparticle transport theory developed recently [31,32], which supersedes the Stokes–Cunningham formula of electric mobility. More recent studies [33,34] show that the momentum accom-