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Mobility size and mass of nascent soot particles in a benchmark premixed ethylene flame

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ABSTRACT

The burner stabilized stagnation flame technique coupled with micro-orifice probe sampling and mobility sizing has evolved into a useful tool for examining the evolution of the particle size distribution of nascent soot in laminar premixed flames. Several key aspects of this technique are examined through a multi-university collaborative study that involves both experimental measurement and computational modeling. Key issues examined include (a) data reproducibility and facility effects using four burners of different sizes and makers over three different facilities, (b) the mobility diameter and particle mass relationship, and (c) the degree to which the finite orifice flow rate affects the validity of the boundary condition in a pseudo one dimensional stagnation flow flame formulation. The results indicate that different burners across facilities yield nearly identical results after special attention is paid to a range of experimental details, including a proper selection of the sample dilution ratio and quantification of the experimental flame boundary conditions. The mobility size and mass relationship probed by tandem mass and mobility measurement shows that nascent soot with mobility diameter as small as 15 nm can deviate drastically from the spherical shape. Various non-spherical morphology models using a mass density value of 1.5 g/cm^3 can reconcile this discrepancy in nascent soot mass. Lastly, two-dimensional axisymmetric simulations of the experimental flame with and without the sample orifice flow reveal several problems of the pseudo one-dimensional stagnation flow flame approximation. The impact of the orifice flow on the flame and soot sampled, although small, is not negligible. Specific suggestions are provided as to how to treat the non-ideality of the experimental setup in experiment and model comparisons.

1. Introduction

Advances in experimental probing of sooting flames have contributed to refined theoretical and modeling studies of soot formation [1,2]. A case in point is the use of a scanning mobility particle sizer (SMPS) and the burner-stabilized stagnation (BSS) flame sampling technique [3–6] to follow the evolution of the particle size distribution function (PSDF) of nascent soot formed in premixed flames. The BSS flame was introduced to address inherent flame perturbations occurring during probe sampling. In this technique, a stagnation surface of well-defined temperature is combined with the sampling probe. This setup enables comparisons between experimental observations and soot modeling results in a less ambiguous manner, in that the flame boundary conditions are defined and the probe itself serves as the boundary condition downstream of the flame. Using this technique, soot PSDFs have been studied in a series of flames burning a range of fuels [3–5,7]. The results have been used in exploring chemical and physical processes of soot formation in detailed models [6,8–11]. Among other things, the BSS flame configuration offers the advantage that the flow field of the flame may be treated

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more directly such that experimental and modeled PSDFs can be compared directly without having to carry out an artificial shift of any experimental or computed profiles [3].

Despite the advantages just mentioned, the accuracy of the BSS flame method coupled with probe sampling and SMPS analysis still can be impacted by several factors. These include a possible burner effect due to differences in burner size/material and how the porous plug is cooled. This can lead to variations in the heat loss to the burner and thus the maximum flame temperature, the flame position, and the preheat zone temperature gradient. Other factors include difficulties in quantifying sample dilution and its calibration during mobility measurement, how the flame gas sample is diluted and transmitted to the mobility sizer and, to an extent, the mobility sizer and its setting. The suitability to directly compare the experiment and pseudo one-dimensional (1-D) simulation also can be questionable for an otherwise 2-dimensional (2-D) flame. The far-side boundary condition applied thus far [3,8,10] uses zero convective velocity, whereas the actual experiment continuously extracts a small flow through the sampling orifice along the centerline of the flame.

Recent efforts have suggested that experimental observations of fundamental flame properties are the most useful when the data of overlapping experiments from several laboratories may be crosschecked to yield an assessment of the random and systematic errors (see, e.g., [12,13]). In this spirit and to assess the accuracy of the BSS flame/SMPS technique, we report here a coordinated, collaborative effort with the goal to evaluate the uncertainty of the BSS flame technique and its sampling method on the same benchmark flame (Flame C3 of Abid et al. [3]) using facilities in three laboratories. The assessment includes, among others, three burners that differ in size and design, two SMPS component models, and two independent calibration procedures. As a tertiary objective, we also aim to make improvements to data reported by Abid et al. [3] on the same benchmark flame.

In addition to the above objectives, we report the results of direct numerical simulation of the BSS flame in a 2-D axisymmetric configuration. The flow rate at the stagnation surface is finite during soot sampling and the 2-D simulation can be used to assess this effect on the 1-D assumptions currently taken in the flame and soot models. Lastly, we note that the interpretation of the mobility size is another open question [10]. Mobility size can deviate from the true size even for a sphere [14–16] but the full extent of deviation for nascent soot has only recently been realized.

Helium ion microscopy (HIM) techniques and other related studies of nascent soot [17–19] have shed new light on the morphology nascent soot. In agreement with theoretical predictions [20,21], nascent soot particles are hardly spherical [17,18]. A separate diagnostic may prove to be necessary to unravel the relationship between mobility size and particle mass due to the uncertainty in the mass density of the particle material [22], structural intricacies and compositional complexity [23,24]. This directly impacts detailed modeling because the primary size parameter that is modeled is mass and not the particle diameter. Here, we use the centrifugal particle mass analyzer (CPMA) [25,26] to examine this relationship. In the CPMA, the balance between the electrostatic force and the so-called centrifugal force allows for particle mass to be classified independently without any knowledge about particle shape and morphology. We note that the mass–mobility relationship has been studied for larger, mature soot [27–29] but this relationship is unavailable for nascent flame soot during its size/mass growth.

In summary, mobility measurements of a benchmark flame are carried out on four different burners across three laboratories (Stanford, Shanghai Jiao Tong and Tsinghua). The mobility diameter of nascent soot is evaluated by measuring the particle mass in tandem (UC Riverside and Stanford). Lastly, DNS modeling of the experimental flame was carried out at University of Duisburg-Essen to provide a better understanding of the 2-D effects on flame modeling and to yield suggestions about how the underlying BSS flame and PSDFs of nascent soot are best modeled using the pseudo 1-D approximation.

2. Experimental

Similar mobility measurement techniques were employed across laboratories at Stanford, Tsinghua and Shanghai Jiao Tong to observe detailed sooting behavior. Key burner and experimental parameters are summarized in Table 1. Briefly, burner-stabilized flames were stabilized on respective burners at atmospheric pressure with an unburned composition of 16.3% (mol) ethylene and 23.7% (mol) oxygen in argon (Flame C3 of Abid et al. [3,30]). The unburned gas has an equivalence ratio, ϕ, of 2.07 and a cold velocity of 8 cm/s (298 K and 1 atm).

At Stanford, two burners of different diameters (5.0 and 7.6 cm) were used to evaluate any possible burner size dependency. Unless otherwise indicated, the Stanford results are reported for measurements made with the 5.0 cm burner. The outer body of both burners is brass. The burners are water-cooled from an inner concentric channel within the burner body. The Shanghai Jiao Tong burner is a duplicate of the Stanford 5.0 cm burner. Tsinghua, on the other hand, uses a McKenna burner with a bronze porous plug 6.0 cm in diameter. Water-cooling in the McKenna burner occurs in small tubes embedded within the porous plug. In addition to the difference in the porous plug material and thickness (see Table 1), there are other differences between the McKenna burner and the Stanford burners. Among them, the Stanford burners can have the porous plug plate replaced, whereas the McKenna burner has the porous plug plate permanently fixed into the burner housing. All flames were isolated from the ambient air by a shroud of nitrogen at a linear velocity of 25 cm/s (298 K and 1 atm) through a concentric porous ring. The gas flows were all metered using critical orifices calibrated independently in each laboratory. The uncertainty in the flow rate is estimated to be 0.5%, mostly due to room temperature fluctuation that would impact the nozzle flow.

Temperature was measured by fine wire thermocouple coated with a Y/BeO mixture to prevent catalysis on the surface. Sizes of the coated thermocouple beads and wires are listed in Table 1. Radiation correction is carried out according to the procedure of Shaddix [31]. The gas properties were estimated by solving for flame structure and

Table 1
Key parameters of the experimental apparatus and models of ultrafine condensation particle counter (UCPC).

Facility	Burner/porous plug						Sample orifice		Thermocouple		UCPC
	Source	Body material	Plug material	Plug thickness (cm)	Pore size (μm)	Diameter (cm)	Diameter (μm)	Length (cm)	Wire diameter (μm)	Bead size (μm)	
Stanford	In-house	Brass	Bronze	1.3	10	5.0 and 7.6	127	30.5	130[a]	300[a]	3025
Shanghai Jiao Tong	In-house	Brass	Bronze	1.3	10	5.0	130	30.5	150[a]	380[a]	3776
Tsinghua	McKenna	SS	Bronze	1.5	70–130	6.0	160	30.5	135[a]	320[a]	3776

[a] Coated.