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Particle size distribution of nascent soot in lightly and heavily sooting premixed ethylene flames

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ABSTRACT

The evolution of the nascent soot particle size distribution function (PSDF) was determined by mobility sizing for two series of atmospheric pressure premixed ethylene flames in the burner stabilized stagnation flame configuration. The first series of flames has an equivalence ratio of 1.8, corresponding to conditions just above the sooting limit. The second series has an equivalence ratio of 2.5 and is quite heavy in soot production. In each series, six flames were tested in which the cold gas velocity is varied to obtain flame temperatures ranging from 1559 to 1941 K. The temperature profiles were carefully determined and the comparison to pseudo-one dimensional simulations was satisfactory. It was found that the evolution of the PSDFs with respect to flame stoichiometry, temperature and growth time is consistent with the understanding of kinetic competition during soot formation. Finite rate kinetic limitations are observed at lower temperatures and thermodynamic reversibility occurs at higher temperatures. The observed PSDF features are highly sensitive to competition among the various processes of soot formation, from nucleation to coagulation and gas–surface reactions. The PSDFs are mostly bimodal with both nucleation and coagulation mode particles present. The evolution of the PSDF indicates a strong contribution to the mass of coagulation-mode by the nucleation-mode particles. The measured PSDFs offer comprehensive, canonical data sets useful for testing models of soot formation.

1. Introduction

Combustion generated particulates are significant sources of anthropogenic aerosols that affect human health [1–8] and the global climate [9–18]. For example, ultrafine soot particles can penetrate deeply into the human respiratory tract and be transported into the blood stream [19]. Soot is also thought to increase Arctic warming [18] by depositing on polar ice, enhancing sunlight absorption and thereby intensifying polar ice melting [17]. Over the last two decades, knowledge about fundamental processes and mechanisms of soot formation has grown tremendously [20–30]. However, there remain uncertainties in the details of many processes. This is especially true for kinetic rates of competing processes of soot nucleation and growth. As such, detailed experimental observations continue to be essential for testing fundamental models of soot formation [29].

A recent approach has been to couple experimental observations of the detailed particle size distribution function (PSDF) with soot model predictions to improve the quantitative understanding of fundamental sooting processes [31–47]. The PSDF of nascent soot from premixed ethylene flames has been heavily studied and the detailed features have been used to extract useful kinetic information about soot growth. Shape and other properties of the PSDF were shown to be related to rates of competing processes ranging from nucleation rate to surface growth mechanism and rates [41,45].

Bimodal PSDFs have been consistently observed in previous studies of relatively lightly sooting premixed ethylene flames ($\Phi = 1.92$) [34] and ($\Phi = 2.07$) [31,32,37–39,47]. In these premixed flames, small soot particles just a few nanometers in size form a nucleation tail that appear to exist at all stages of growth. The persistence of the tail leads to a bimodal PSDF even in the later stage of soot growth. It is known that the shape of soot PSDF is dependent on many competing kinetic processes, which in turn, is a strong function of flame temperature [34,37,38]. At low temperatures, the rate of soot nucleation and growth appears to be kinetically limited, and the nucleation tail tends to be pronounced. Towards higher temperatures, soot nucleation and growth is hindered by reversibility in the nucleation and

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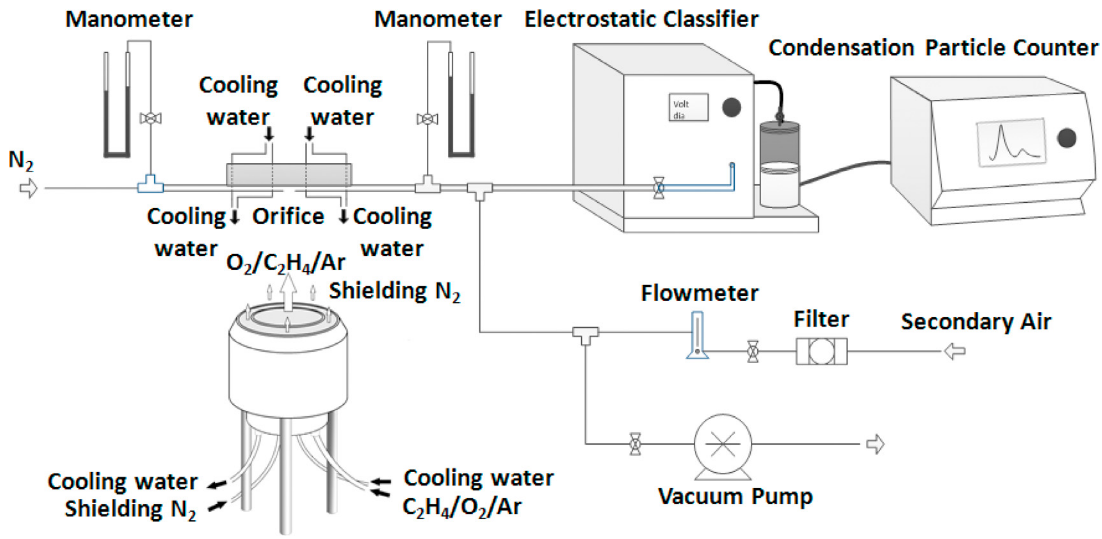

Fig. 1. Schematic of experimental setup.

Table 1
Summary of the two series of ethylene flames studied.

Flame	Cold gas velocity,[a] v_0 (cm/s)	Maximum temperature,[b] T_m (K)	Sheath velocity,[a] (cm/s)	Burner surface temperature,[b] T_0 (K)	Stagnation surface temperature,[b] T_s (K)	Largest separation, $H_{p,\max}$ (cm)
K-Series: 15.0% (mol) C_2H_4-25.0% (mol) O_2–Ar ($\Phi = 1.8$)						
K1	3.5	1680 ± 75	13.3	443 ± 20	387 ± 10	1.4
K2	5.0	1790 ± 84	15.5	545 ± 30	378 ± 10	1.4
K3	6.0	1826 ± 87	17.7	562 ± 30	385 ± 10	1.4
K4	7.0	1882 ± 92	19.9	561 ± 20	389 ± 10	1.4
K5	7.5	1902 ± 93	19.9	621 ± 20	383 ± 10	1.4
K6	8.5	1941 ± 96	24.3	603 ± 30	386 ± 10	1.4
L-Series: 13.6% (mol) C_2H_4-16.4% (mol) O_2–Ar ($\Phi = 2.5$)						
L1	4.5	1559 ± 61	13.3	383 ± 20	396 ± 10	1.5
L2	5.5	1622 ± 66	15.5	371 ± 20	393 ± 10	1.4
L3	6.5	1654 ± 66	17.7	399 ± 20	393 ± 10	1.2
L4	7.5	1713 ± 72	19.9	503 ± 20	397 ± 10	1.0
L5	11	1787 ± 76	26.6	445 ± 20	393 ± 10	0.8
L6	15	1893 ± 85	31.0	323 ± 20	394 ± 10	0.8

[a] STP condition (298 K and 1 atm).
[b] Measured for the largest separation of the burner to stagnation surface, $H_{p,\max} = 1.4$ cm for the K-series of flame and $H_{p,\max} = 1.5$, 1.4, 1.2, 1.0, 0.8 and 0.8 cm for Flames L1 through L6.

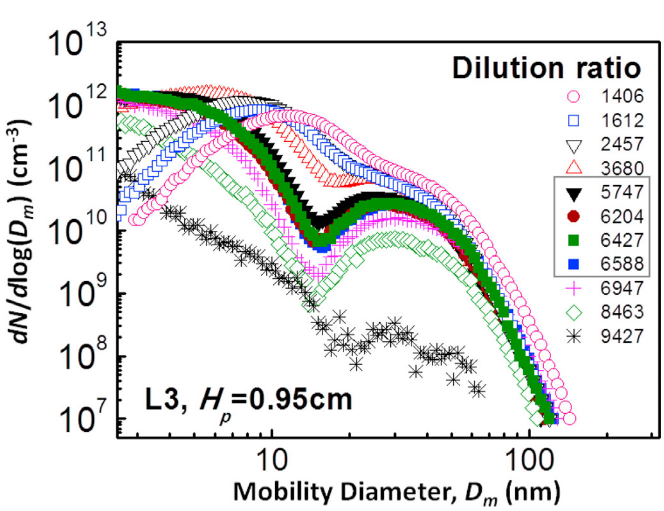

Fig. 2. Effect of the dilution ratio on PSDFs measured for Flame L3 ($\Phi = 2.5$) with $H_p = 0.95$ cm. The solid symbols indicate the range of the appropriate dilution ratio over which the measured PSDFs are found to be independent of the dilution ratio.

growth processes. As flame temperature increases, the nucleation tail diminishes in both the number and particle size, leading to a shift of the trough of the bimodal PSDF to smaller particle size. Unfortunately, the aforementioned phenomena are observed almost exclusively in lightly sooting flames. It is difficult to generalize any of the observations made in lightly sooting flames for heavier soot flames, as the flame stoichiometry should have a strong impact on the soot nucleation and growth process [19,48].

The evolution of PSDF in a fairly heavily sooting ethylene flame ($\Phi = 2.5$) was reported previously [35,36]. The observation shows that the nucleation tail is replaced by a log-normal distribution, as if nucleation stops at some point of particle growth. It was speculated that the absence of persistent nucleation is caused by the depletion of PAHs because they are scavenged by soot particles. Unfortunately, the experiment data reported in [35,36] may be contaminated by experimental artifacts primarily because of particle losses in the sample probe. The primary objective of that series of study was, in fact, not to obtain the size distribution, but it was on the particle chemical composition using photo-ionization mass spectrometry. For that reason, particle sampling was optimized for composition measurement only.

In this paper, we carry out a systematic experimental study for nascent soot PSDFs by comparing a series of premixed ethylene flames close to the sooting limit ($\Phi = 1.8$) to a second series of heavier sooting flames at $\Phi = 2.5$, each over a range of maximum flame temperatures. The experimental data enable the examination of the impact of equivalence ratio and flame temperature on soot nucleation and growth in a manner more comprehensive than previously possible. The measurements also provide useful data over the range of