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Brief Communication

Distribution of the number of primary particles of soot aggregates in a nonpremixed laminar flame

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1. Introduction

Investigations on the morphology of soot aggregates generated in hydrocarbon/air combustion systems are of considerable importance to our understanding of the physical and chemical processes of soot inception, growth, and oxidation. Nonintrusive methods for soot diagnostics, such as LII (laser-induced incandescence) [1] and laser light-scattering techniques [2], have been developed. These techniques require a reliable independently obtained description of soot morphology for calibration and/or estimation of optical properties of soot. It has been generally accepted that soot aggregates consist of polydispersed carbon nanoparticles. The mean primary particle diameter, $\bar{d}_p$, and the distribution of the number of primary particles per aggregate, N, are two of the key variables to characterize the soot aggregates.

The diameters of the primary particles that constitute soot aggregates in general cover a range of 10–50 nm and follow a normal distribution [3]. At a given location in a laminar diffusion flame, the mean primary particle diameter, $\bar{d}_p$, can be treated as nearly constant [3–5]. The number of primary particles per aggregate, N, however, covers a wide range, from 1 to several hundred. A widely accepted viewpoint is that the distribution of N follows a lognormal distribution (normalized) that can be described by two parameters, N_g and σ_{2g} [3],

$$\mathrm{PDF}(N) = \exp\left[-\left(\frac{\ln(N)-\ln(N_g)}{2^{0.5}\ln(\sigma_{2g})}\right)^2\right] \times \left(N \cdot \ln(\sigma_{2g}) \cdot (2\pi)^{0.5}\right)^{-1}, \qquad (1)$$

where PDF(N) is the probability distribution function of N, N_g the geometric mean of N, and σ_{2g} the geometric standard deviation. Several results for N_g, σ_{2g}, and the arithmetic mean of N, $\bar{N}$, have been reported [3,6] for different flame conditions. To our best knowledge, however, experimental data on the probability distribution of N, from which N_g and σ_{2g} are derived, for soot aggregates sampled within a laminar diffusion flame environment have not been published. The objective of the present investigation is to report such experimental data and to gain a better understanding of the distribution of N of soot aggregates thermophoretically sampled from a laminar ethylene/air diffusion flame by analyzing thousands of aggregates in TEM (transmission electron microscopy) images.

2. Method and results

The experiment was carried out in an atmospheric-pressure, axisymmetric-coflow laminar ethylene/air diffusion flame. The burner for generating the flame has been previously described in detail [7,8]. It was similar to those used by Santoro et al. [9], Dobbins

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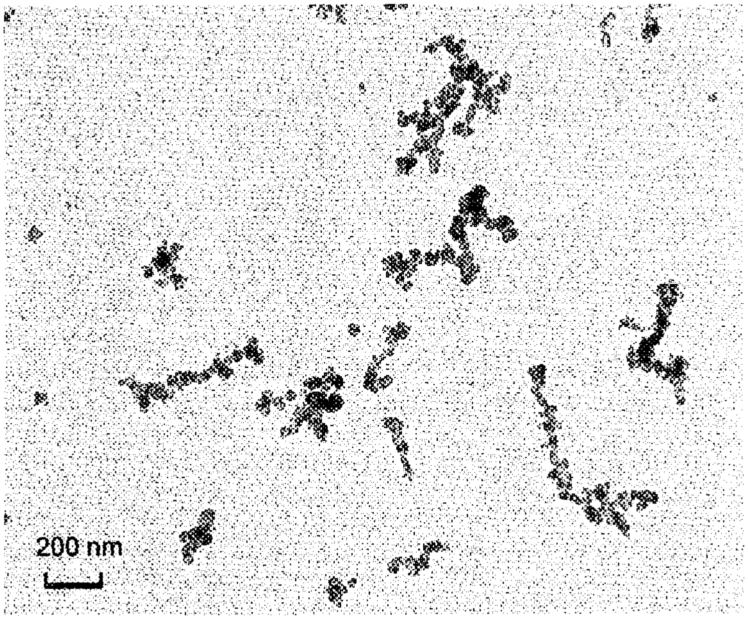

Fig. 1. A typical TEM image of soot aggregates sampled at the centerline of the flame at at height 42 mm above the exit of the fuel tube.

and Megaridis [5] and Köylü et al. [6]. The fuel-flow rate was 3.23 cm^3/s, and the air-flow rate was 4733 cm^3/s at the standard condition. These conditions resulted in a visible flame height of about 67 mm. The thermophoretic sampling (TS) instrumentation of the present experiments was similar to that used by Dobbins and Megaridis [5]. The sampling probe consists of a carbon-coated circular copper TEM grid (Electron Microscopy Sciences, Catalog # LC200-Cu, diameter 3 mm) and a stainless steel grid holder. The probe was rapidly inserted into the flame and then rapidly withdrawn after an exposure time of 25 ms. This procedure was controlled by a double-acting pneumatic cylinder system. The time interval has been confirmed by high-speed digital video. The sampling location was at a height of 42 mm over the exit of the fuel tube and at the centerline of the flame. At this flame height, the maximum soot volume fraction in the radial direction appears on the flame centerline.

After the sampling procedure, the TEM grids were examined with a Philips CM20 transmission electron microscope. The microscope is equipped with an UltraScan 1000 CCD camera (Gatan Inc.) which can directly generate digital images for analysis. The images were subsequently analyzed using an image-processing software package (Media Cybernetics, Image Pro Plus). Each image file has a calibrated length scale that can be directly used by the image processing software. Fig. 1 shows a typical TEM image of soot aggregates sampled in this study.

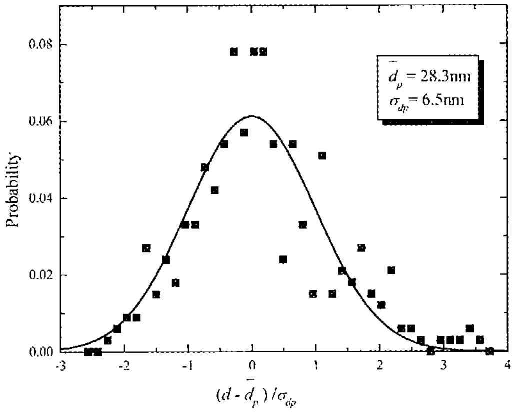

Fig. 2. Probability distribution of the primary particle diameters and the normal distribution fitting curve.

Primary particle diameters d_p were measured manually with high-magnification TEM images by detecting the apparent circular outlines of the particles near the edges of the aggregates. In total, 385 primary particles were measured. The diameters of the particles cover a range of 13–50 nm, with an arithmetic mean diameter $\bar{d}_p = 29.2$ nm and a standard deviation of $\sigma_{dp} = 7.1$ nm. When a normal distribution was fitted to the measured diameters, the resulting normal distribution parameters were $\bar{d}_p = 28.3$ nm with $\sigma_{dp} = 6.5$ nm, as shown in Fig. 2. These results are quite close to those reported by Dobbins and Megaridis [5] and Köylü et al. [6] at a comparable relative height in a similar diffusion flame.

The number of primary particles per aggregate (N) was obtained from low-magnification TEM images, based on the projected area of each aggregate and an empirical equation [10],

$$N = k_a (A_i/A_p)^{\alpha}, \tag{2}$$

where A_i is the projected area of the aggregate, $A_p = \pi \bar{d}_p^2/4$ the cross-sectional area of a single primary particle with a diameter of $\bar{d}_p$, k_a the projected area constant, and α the projected area exponent. The mean primary particle diameter $\bar{d}_p = 29$ nm and constants $k_a = 1.10$ and $\alpha = 1.08$ [11] were employed in this paper. In total, 3488 aggregates within 62 TEM images were analyzed and the statistical distribution of N was obtained. N was found to be in the range 1–1610. A lognormal distribution model was applied to fit the statistical data. Considering that the lower limit of N in the statistical results is 1 instead of 0, Eq. (1) was multiplied by a proportionality coefficient to ensure that the integral of the scaled PDF(N) from 1 to infinity equals 1, to be consistent with the normalization of the statistical results. A similar correction was conducted in the fitting when a lower cutoff of $N > 5$ was employed (see below).

It was found that the distribution of N is poorly fit by a lognormal model over the range from 1 to infinity, as shown in Fig. 3 (dash-dotted line and unfilled circles). However, if a lower limit of $N > 5$ was used as a cutoff to remove data less than or equal to 5, a lognormal distribution model would represent the data reasonably well with $N_g = 23.2$ and $\sigma_{2g} = 4.15$, also shown in Fig. 3 (dashed line). We obtained the arithmetic mean of N, $\bar{N} = 43.8$, with a standard deviation of $\sigma_N = 74.7$ without the lower limit, and $\bar{N} = 54.8$ with $\sigma_N = 80.6$ with the threshold of $N > 5$. The results for $\bar{N}$ are much smaller than reported by Köylü et al. [6] at a comparable height in a similar flame using similar techniques (TS/TEM).

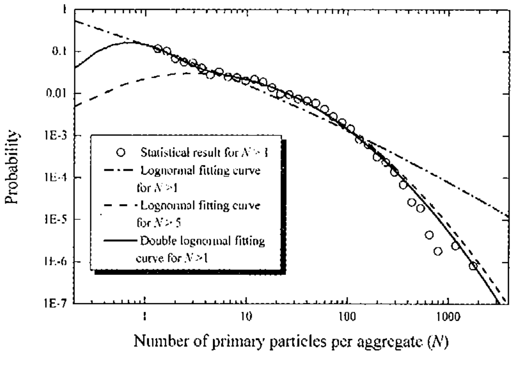

Fig. 3. Probability distribution of the number of primary particles per aggregate, the lognormal fitting curves with or without a lower cutoff limit, and the double lognormal fitting curve.

A double lognormal model,

$$\begin{aligned}\mathrm{PDF}(N) = {} & a \cdot \exp\left[-\left(\frac{\ln(N) - \ln(N_{g_1})}{2^{0.5}\ln(\sigma_{2g_1})}\right)^2\right] \\ & \times \left(N \cdot \ln(\sigma_{2g_1}) \cdot (2\pi)^{0.5}\right)^{-1} \\ & + (1-a) \cdot \exp\left[-\left(\frac{\ln(N) - \ln(N_{g_2})}{2^{0.5}\ln(\sigma_{2g_2})}\right)^2\right] \\ & \times \left(N \cdot \ln(\sigma_{2g_2}) \cdot (2\pi)^{0.5}\right)^{-1},\end{aligned} \tag{3}$$

was also applied to fit the distribution of N without the lower cutoff. In Eq. (3), a is the weight factor, N_{g_1} the first geometric mean N, σ_{2g_1} the first geometric standard deviation, N_{g_2} the second geometric mean N, and σ_{2g_2} the second geometric standard deviation. A proportional coefficient was also applied in fitting Eq. (3) to the data for the reason mentioned earlier. The resulting fit represent the statistical data very well with $a = 0.21$, $N_{g_1} = 1.13$, $\sigma_{2g_1} = 2.06$, $N_{g_2} = 22.6$, and $\sigma_{2g_2} = 3.91$, in the entire range of N as shown in Fig. 3 (solid line and unfilled circles).

From the above results, it seems very difficult for the lognormal distribution model to describe the N distribution throughout, based on N calculated from the statistical equation, i.e., Eq. (2). On the other hand, the double lognormal model can represent the statistical results of the aggregate size distribution more precisely. Consequently, it might lead to more accurate estimate of the optical properties of soot aggregates based on the RDG/PFA theory.

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