Comparison of two different methods for the determination of the fractal dimension of soot aggregates: TEM measurement and serial analysis of the aerodynamic and electrical mobility diameters

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INTRODUCTION

Morphology of particles generated during hydrocarbon or biomass combustion is fundamental as data for characterizing the optical and aerodynamic behaviour of these particles. The fractal nature of soot particles is well known since the works of Jullien and Botet (1987). Nevertheless, the determination of the fractal morphology of these aggregates is based on direct analysis of transmission electronic microscopy (TEM) micrograph which represents a long and tiresome work.

In order to determine the fractal morphology, we propose in this work to use the method based on serial analysis of electrical mobility and aerodynamic diameters of soot aggregates. This method has been recently used by Van Gulijk *et al.* (2004), and seems to bring morphological information systematically higher than the TEM analysis. We will present the theoretical approach, the experimental setup used and the results obtained for aggregates generated during the combustion of acetylene (C_2H_2), toluene (C_7H_8) and PolyMethyl Methacrylate (PMMA, $C_5H_8O_2$). These results will be compared to TEM results, and discrepancies will be analysed and explained.

THEORETICAL APPROACH

We propose here to determine the fractal dimension of soot particles by comparing the aerodynamic and electrical mobility diameters. The key feature of this approach is the link introduced by Rogak and Flagan (1990) between the electrical mobility diameter D_m and the gyration diameter D_g of aggregates. The β ratio between these two diameters is defined as:

$$D_g = \beta . D_m \qquad (1)$$

Introducing this definition in the well-known fractal relation we obtain:

$$N_p \propto \left(\frac{D_g}{D_{pp}}\right)^{D_f} \propto \left(\frac{D_m}{D_{pp}}\right)^{D_f} \qquad (2)$$

N_p : number of primary particles in the aggregate ; D_{pp} : primary particle diameter ; D_f : fractal dimension.

On the other hand, in order to link the electrical mobility to the aerodynamic diameter, we compare the relaxation times of the aggregate and of the equivalent aerodynamic sphere:

$$m_{agg} . B_{agg} = m_{sph} . B_{sph} \qquad (3)$$

The aggregate mass m_{agg} and mobility B_{agg} are defined as:

$$m_{agg} = \frac{\pi}{6} . N_p . \rho_{pp} . D_{pp}^3 \quad ; \quad B_{agg} = \frac{C_c(D_m)}{3.\pi.\mu_g.D_m} \qquad (4)$$

and for the sphere:

$$m_{sph} = \frac{\pi}{6} . \rho_0 . D_a^3 \quad ; \quad B_{sph} = \frac{C_c(D_a)}{3.\pi.\mu_g.D_a} \qquad (5)$$

ρ_{pp} is the density of the primary soot particles, $\rho_0 = 1 g/cm^3$, μ_g is the gas viscosity, $C_c(D)$ the Cunningham correction factor and D_a the aerodynamic diameter. Then we can link the electrical mobility diameter to the aerodynamic diameter:

$$\frac{\pi}{6} . N_p . \rho_{pp} . D_{pp}^3 . \frac{C_c(D_m)}{3\pi \mu_g D_m} = \frac{\pi}{6} . \rho_0 . D_a^3 \frac{C_c(D_a)}{3\pi \mu_g D_a} \qquad (6)$$

Furthermore we introduce the relation between the number of primary particles N_p to the electrical mobility diameters D_m and we simplify the slip factor $C_c(D)$ as a function of $D^{1-\alpha}$. Finally we can establish the following relation between D_a and D_m (Van Gulijk *et al.* 2004) :

$$\frac{D_a}{D_{pp}} \propto \left(\frac{D_m}{D_{pp}}\right)^{\frac{(D_f - \alpha)}{(3-\alpha)}} \qquad (7)$$

According to this relation we have determined the fractal dimension of soot aggregates by comparing the electrical mobility and the aerodynamic diameters.

EXPERIMENTAL DEVICE

Soot particles are generated in a furnace of approximately 1 m^3. The furnace is surmounted by a hood having a 114 mm diameter vertical ventilation duct. Three different fuels have been used: a gas (acetylene), a liquid (toluene) and a solid (PMMA). Particle sampling is done with an isokinetic probe and a two-stages heated dilution device (FPS 4000, *Dekati*). A specific soot mobility diameter is selected by a Differential Mobility Analyser (DMA 3936, *TSI*). Then the aerodynamic diameter is measured with an Electrical Low Pressure Impactor (ELPI, *Dekati*). The morphology of soot particles is also determined by sampling on TEM grids and a micrograph analysis program has been developed based on previous works (Köylü *et al.*, 1995).

RESULTS AND DISCUSSION

The experimental evolution of the logarithm of the aerodynamic diameter as a function of the logarithm of the electrical mobility diameter is detailed on Figure 1.

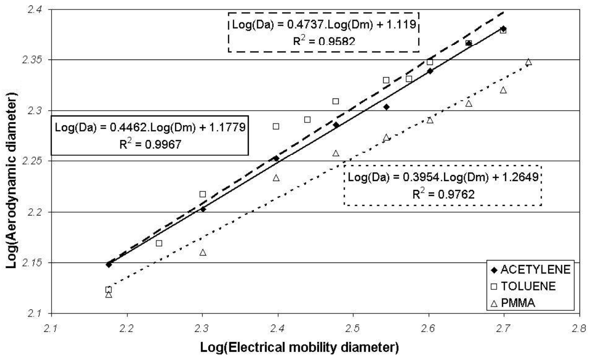

Figure 1: Experimental evolution of aerodynamic diameter as a function of electrical mobility diameter

From the slopes of the linear regressions in Figure 1 and eq. (7) the fractal dimensions are obtained. These values are compared to the ones obtained from TEM analyses in Table 1.

Table 1: Comparison of fractal dimension determined by TEM analyses, uncertainty in brackets, and DMA-ELPI method, discrepancy with TEM in brackets.

Fuel	$D_{f TEM}$	$D_{f DMA\text{-}ELPI}$
Acetylene (C_2H_2)	1.93 (5%)	2.19 (17%)
Toluene (C_7H_8)	1.81 (5%)	2.36 (19%)
PMMA ($C_5H_8O_2$)	1.72 (7%)	2.15 (19%)

A systematic overestimation (20%) of fractal dimension is observed for the DMA-ELPI method. To explain this discrepancy we discuss the hypothesis of linear relation between these two diameters. As Rogak & Flagan (1990) have done theoretically we present in Figure 2 the evolution of the D_g/D_m ratio as a function of the electrical mobility diameter and as a function of the number of primary particles in the aggregates. It appears that this ratio is constant only for electrical mobility diameters above 250 nm, which approximately corresponds to 40 primary particles. Then we have only compared the fractal relations on electrical mobility diameter and gyration diameter for particle diameters bigger than 250 nm.

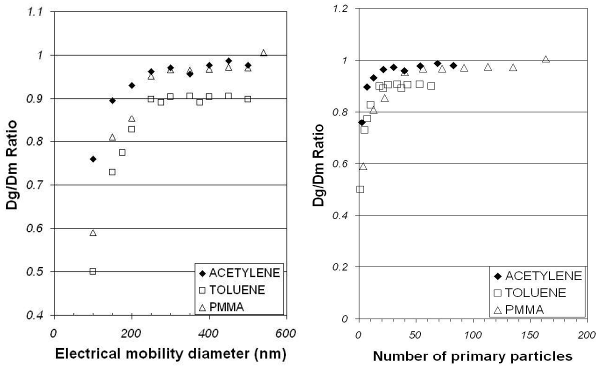

Figure 2: Evolution of $\beta = D_g/D_m$ ratio as a function of electrical mobility diameter and number of particles

The results are given in Table 2 and show a very good agreement between both methods.

Table 2: Fractal dimensions determined by TEM, uncertainty in brackets, and DMA-ELPI methods considering only D_m over 250 nm, discrepancy with TEM in brackets.

Fuel	$D_{f TEM}$	$D_{f DMA\text{-}ELPI}$
Acetylene (C_2H_2)	1.93 (5%)	1.99 (3%)
Toluene (C_7H_8)	1.81 (5%)	1.82 (1%)
PMMA ($C_5H_8O_2$)	1.72 (7%)	1.79 (4%)

CONCLUSIONS

Two methods for the determination of fractal dimension have been used and compared in this study. With the assumption of a constant β ratio on the whole size range the DMA-ELPI method shows a systematic over-estimation compared to the TEM analysis. In contrary when only the aggregates above 250 nm are considered, a good agreement is found. The conclusion is that DMA-ELPI and TEM methods lead to similar fractal dimension when only the β constant size range is considered (here N_p>40 and D_m>250 nm).

Keywords: *Soot aggregates, fractal dimension.*

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