ESTIMATION OF EFFECTIVE DENSITY AND FRACTAL-LIKE DIMENSION OF SOOT PARTICLES

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Structure of combustion generated aggregates is important for a number of applications

- Particulate filters (filtration, pressure drop, reactivity of soot cakes)
- Health effects of soot particles (transport, deposition, interaction with tissues)
- Interactions of soot particles with atmospheric constituents
- Performance of carbon black based products
MEASUREMENT METHODS FOR SOOT PARTICLE STRUCTURE

- Tandem mobility-aerodynamic or mobility-mass measurements
  

- Parallel application of SMPS and ELPI and matching the two distributions
  
  Ristimaaki et al (2002), Present work

- Optical methods and microscopy
  
PARTICLE MEASUREMENT SETUP

ELPI: Electric Low Pressure Impactor

SMPS: Scanning Mobility Particle Sizer

LPME: Long Path Multiwavelength Extinction
AERODYNAMIC vs. MOBILITY DIAMETER

(dN/dlnD_p)/N_total

Stage 7
Stage 8
Stage 9
Stage 10
Stage 11
Stage 12

Mobility diameter (nm)

SMPS
ELPI
ELPI-converted to mobility
AERODYNAMIC vs. MOBILITY DIAMETER FOR 3 PASSENGER CAR DIESEL ENGINES

**EURO II**

- 1500 rpm - 2 bar
- 1500 rpm - 5 bar
- 2400 rpm - 6 bar

**EURO III (1)**

- 1500 rpm - 2 bar
- 1500 rpm - 5 bar
- 2400 rpm - 6 bar

**EURO III (2)**

- 1400 rpm - 3 bar
- 1500 rpm - 3.5 bar
- 2500 rpm - 6 bar
DIESEL SOOT FRACTAL AGGREGATES: Definitions

Density and size of primary particles

\[ \rho_0 \approx 2150 \text{ kg/m}^3 \text{ (CV 20\%)} \text{ based on gravimetry vs. LPME} \]

\[ d_0 \approx 32 \text{ nm (CV 20\%)} \text{ based on soot cake permeability and TEM} \]

Number of primary particles per aggregate

\[ N_A = k_g \left( \frac{D_g}{d_0} \right)^{D_f} \]

\[ k_g = \frac{1}{f} \left[ \frac{D_f}{D_f + 2} \right]^{-\frac{D_f}{2}} \]

volume filling factor, Naumman (2003) \( f \approx 1.43 \)

\( D_f \approx 2.4 \text{ on the average} \) Kittelson & McMurry (2002) and others

Geometric diameter

\[ \frac{D_{geo}}{d_0} = \left[ fN_A \right]^\frac{1}{D_f} \]

Diameter of gyration

\[ D_g = \left[ \frac{D_f}{D_f + 2} \right]^{\frac{1}{2}} D_{geo} \]

Mass equivalent diameter

\[ \frac{D_{geo}}{d_0} = \left[ f \left( \frac{D_{mass}}{d_0} \right)^3 \right]^{\frac{1}{D_f}} \]
MOBILITY DIAMETER OF FRACTAL AGGREGATES

\[ D_{me} = h_{KR} D_{geo} = (-0.06483 D_f^2 + 0.6353 D_f - 0.4898) D_{geo} \]

\( h_{KR} \): Kirkwood - Riseman ratio accounting for shielding effects and hydrodynamic interactions

Naumann (2003)

Gyration diameter ~ Mobility diameter

\[ D_g = \left[ \frac{D_f}{D_f + 2} \right]^{\frac{1}{2}} \frac{D_{me}}{h_{KR}} \]

Fractal scaling based on Mobility diameter

\[ N_A = k_g \left[ \frac{D_g}{d_0} \right]^{D_f} = k_m \left[ \frac{D_{me}}{d_0} \right]^{D_f} \]
EFFECTIVE DENSITY OF AGGREGATES

\[ \rho_{\text{eff}} D_{\text{me}}^2 C_c (D_{\text{me}}) = \rho_1 D_{\text{ae}}^2 C_c (D_{\text{ae}}) \]

- \( \rho_{\text{eff}} \): effective density
- \( \rho_1 \): unit density (1 g/cm\(^3\))
- \( D_{\text{me}} \): mobility diameter
- \( D_{\text{ae}} \): aerodynamic diameter
- \( C_c \): Stokes-Cunningham Factor

Basic equations of analysis

\[ \frac{\rho_0}{\rho_1} = \frac{D_{\text{ae}}^2 C_c (D_{\text{ae}}) f h_{KR}^{D_f}}{D_{\text{me}}^{D_f-1} d_0^{3-D_f} C_c (D_{\text{me}})} \]

\[ \frac{\rho_{\text{eff}}}{\rho_0} = \frac{1}{f h_{KR}^{D_f}} \left[ \frac{D_{\text{me}}}{d_0} \right]^{D_f-3} \]
**DI ESEL SOOT AGGREGATE FRACTAL DI MENSIO N**

3 different diesel engines & 1 gen set

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- EURO III (2) 1400 rpm 3 bar
- EURO III(2) 1500 rpm 3.5 bar
- EURO III(2) 2500 rpm 6 bar
- EURO III(1) 1500 rpm 2 bar
- EURO III(1) 1500 rpm 5 bar
- EURO III (1) 2500 rpm 6 bar
- EURO III (1) 2000 rpm 13 bar
- EURO II 1500 rpm 2 bar
- EURO II 1500 rpm 5 bar
- EURO II 2400 rpm 6 bar
- EURO II 2000 rpm 13 bar
- Van Gulijk et al (2003)-5.7 kW

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**fractal dimension**

**mobility diameter (nm)**
DI ESEL SOOT AGGREGATE EFFECTIVE DENSITY
Diesel aggregate size distribution: 5 Engines (1996-2003) with engine displacement 1.9-2.4 litres

Common Rail – 1

\[ \langle \sigma_g \rangle = 1.88 \pm 0.09 \]

Common Rail – 2

\[ \langle \sigma_g \rangle = 1.94 \pm 0.09 \]

Rotary Pump

\[ \langle \sigma_g \rangle = 1.85 \pm 0.04 \]

Common Rail – 3

\[ \langle \sigma_g \rangle = 1.86 \pm 0.15 \]

Pump Unit Injector

\[ \langle \sigma_g \rangle = 1.90 \pm 0.14 \]

Universal Lognormal Shape:

\[ \sigma_g = 1.89 \pm 0.08 \]

cf. Harris & Maricq (2002) 1.7-1.8

Kostoglou & Konstandopoulos (2003)
Steady state shape is determined from the ratio of oxidative fragmentation to coagulation rate

Initial soot aggregate

Contact “necks”

Oxidative fragmentation site

Binary random fragmentation result

Continuous, binary random fragmentation process with size dependent rate:

\[ S_i = Ai^b = Ai^{1/D_f} \]

In the large aggregate limit it can be shown that

\[ \ln \sigma_g = \ln(6) / 2(1+b) \]

For \( \sigma_g = 1.89 \), \( b = 0.42 \) and \( D_f = 2.38 \)

Kostoglou & Konstandopoulos (2003)
CAST SOOT GENERATOR

Provides Reference Soot Size Distributions
SIZE DISTRIBUTIONS FROM CAST

- Dp = 30nm
- Dp = 60nm
- Dp = 91nm
- Dp = 106nm
- Dp = 128nm
- Dp = 143nm
- Dp = 190nm

Number Concentration (#/cm³) vs. Dp (nm)
CAST CALIBRATION WITH SMPS

![Graph showing CAST Dp (nm) vs Measured Dp (nm)]
CAST SOOT AGGREGATE FRACTAL DIMENSION

The graph illustrates the change in fractal dimension with mobility diameter for different soot aggregate sizes. The x-axis represents the mobility diameter (nm) ranging from 0 to 700, and the y-axis represents the fractal dimension ranging from 1.80 to 2.60. Several curves are plotted for different sizes:

- 60 nm (green line)
- 91 nm (pink line)
- 106 nm (red triangle)
- 128 nm (blue line)
- 143 nm (purple line)
- 190 nm (brown line)

The fractal dimension decreases as the mobility diameter increases, with a notable minimum in the range of mobility diameters.
CAST SOOT AGGREGATE EFFECTIVE DENSITY

- 60 nm
- 91 nm
- 106 nm
- 128 nm
- 143 nm
- 190 nm

Effective density (g/cm³)

Mobility diameter (nm)

0.00
0.50
1.00
1.50
2.00
0 100 200 300 400 500 600
OTHER STUDIES

Maricq 2003 ELPI Workshop

Maricq & Xu (in press)
SOOT FRACTAL DIMENSION EVOLUTION IN DIFFUSION FLAME

Fractal dimension of soot from 3 diesel engines and a CAST burner changes non-monotonically with mobility diameter. For diesel soot aggregates $D_f$ decreases sharply from 3 down to $\sim 1.8 - 1.9$ with aggregate size up to about 100 nm. For larger than 100 nm aggregate sizes $D_f$ increases up to $\sim 2.4 - 2.5$. An average $D_f = 2.4$ for the entire aggregate population is consistent with the universal lognormal $\sigma_g$ of 1.89 +/- 0.08 of many diesel size distributions based on population dynamics modelling of random oxidative fragmentation and coagulation. Effective density exhibits a sharp decrease up to aggregate sizes about 200 nm and then a more gradual variation in agreement with the compaction shown by the increase of the $D_f$ up to $\sim 2.4 - 2.5$. 
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