The microstructure of soot aggregates produced by the Combustion Aerosol Standard (CAST) generator

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Introduction

In the present work we characterize the microstructure of soot nanoparticles produced by a well-known soot generator, the Combustion Aerosol Standard (CAST), which produces soot aggregates with selected size distributions in a repeatable manner, in a range of mobility diameters from 30 to 200 nm. To our knowledge these are the first systematic characterization results for the structure of CAST aggregates and they should be useful in future research where the effect of soot aggregate structure on the phenomena under investigation needs to be studied.

Nanoparticle Characterization

In order to achieve the unbiased sampling of the original aggregate structure, a thermophoretic sampler was designed and evaluated. The evaluation was based on digital image processing and analysis of electron microscopy images of the CAST aggregates. We reconstructed the size distribution of the particles through projected area aggregate analysis and it was found to be in good agreement with the results obtained with a Scanning Mobility Particle Sizer (SMPS), validating thus the thermophoretic sampler. Subsequently for each of the pre-set soot size distributions (with nominal mean diameter $D$) of the CAST we evaluated the actual mobility diameter $D_{me}$, the size of primary soot particles, the average number of primary particles per aggregate, $<N>$, the fractal dimension, $D_f$, the fractal prefactor, $k_f$, the filling factor, $f$, and the overlap coefficient, $C_{ov}$. The fractal dimension and fractal prefactor of the CAST soot aggregates are given in Table 1.

Table 1. Morphological properties of CAST soot aggregates

<table>
<thead>
<tr>
<th>Nominal, $D$ (nm)</th>
<th>$D_{me}$ (nm)</th>
<th>$D_f$</th>
<th>$k_f$</th>
</tr>
</thead>
<tbody>
<tr>
<td>190</td>
<td>210</td>
<td>2.25</td>
<td>3.95</td>
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<tr>
<td>143</td>
<td>156</td>
<td>2.27</td>
<td>3.69</td>
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<tr>
<td>128</td>
<td>138</td>
<td>2.14</td>
<td>4.28</td>
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<tr>
<td>106</td>
<td>121</td>
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<tr>
<td>91</td>
<td>104</td>
<td>2.03</td>
<td>5.25</td>
</tr>
<tr>
<td>60</td>
<td>78</td>
<td>2.15</td>
<td>4.97</td>
</tr>
</tbody>
</table>

The $D_f$ results were compared with those obtained with an independent method based on combined inertial (ELPI) and mobility (SMPS) size measurements [1] and are shown to be in good agreement (Figure 1), while the $k_f$ values are in agreement with those known for soot aggregates in the literature. To facilitate use of these results in computational studies involving the transport and deposition of soot aggregates a computational engine has been developed (“Digital CAST”, d-CAST), allowing the generation of soot aggregates with the same microstructural properties as those found in this work (Figure 2).
Figure 1. Fractal Dimension against Mobility Diameter obtained with two independent methods

Figure 2. Digital-CAST (d-CAST) generated aggregates with same properties as actual CAST aggregates.

Conclusions
Soot aggregates produced at different operating points of the CAST burner have been characterized in terms of their structure and a database of $D_f$, $k_p$, $C_{ov}$, $N_p$, $d_0$ has been established. The morphological results obtained by image analysis were found to be in good agreement with an independent method based on parallel SMPS-ELPI measurements and a previously developed theory that allows the calculation of $D_f$ from such measurements. The d-CAST software provided faithful aggregate particle morphologies that were used for ab initio simulations in a variety of applications, including filtration and deposition in realistic geometries.

References
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Zurich, June 26th – 29th, 2011
A Day in the Life of the Aerosol & Particle Technology Laboratory
Motivation

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
CAST: Combustion Aerosol Standard

Provides Reference Soot Size Distributions but not enough quantitative data on their morphology exist
CAST Soot size distributions

Nominal CAST size

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

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Nominal vs. Measured (SMPS) Size

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Methods to Measure Aggregate Morphology

- 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
  - Sorensen and co-workers (1995-2004), di Stasio et al. (1999-2004), Brazil et al. (1999), Wentzel et al. (2003), Present work

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Example: Diesel Soot Characterization

Combined SMPS-ELPI Measurements

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Re=72

\n
\varepsilon \approx 3.25 \times 10^5 \text{K/m}

V_{th} \approx 1.65 \times 10^{-2} \text{m/sec}

\tau \approx 24 \text{msec}

Thermophoretic Sampler

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Image Analysis

Aggregate Projected Area, \( A_p, d_{pa} \)

Projected area diameter

Mobility diameter

Aggregate Size (nm)

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From Projections to 3-D Structures

Aggregate Projected Area, $A_p$, $d_{pa}$

From 2-D to 3-D

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

$N_A = k_a \left( \frac{A_a}{< A_p >} \right)^a$

Method as in:

Oh and Sorensen (1997)
Brazil et al. (1999),
Wentzel et al. (2003)
Park et al. (2003)
and others

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Scaling Analysis

$y = 1.9398x + 0.2861$
$R^2 = 0.9459$

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CAST Primary vs. Aggregate particle size

\[ y = 0.0838x^{1.1845} \]

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CAST Nanoparticles Characterization

<table>
<thead>
<tr>
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<th>$C_{ov}$</th>
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<td>4.97</td>
<td>0.44</td>
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</tbody>
</table>

\[
D_g = d_0 \left[ \frac{N_A}{k_g} \right]^{\frac{1}{D_f}}
\]

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Comparison with Other Method

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Comparison with Theory

Gyroscopic Diameter from Image Analysis vs Computation from Mobility Diameter

\[ D_g = d_0 \left[ \frac{N_A}{k_g} \right]^{\frac{1}{D_f}} \]

\[ D_g = \frac{d_{me}}{(-0.06483D_f^2 + 0.6353D_f - 0.4898) \left( \frac{D_f}{D_f + 2} \right)^{1/2}} \]

Wentzel et al. (2003)
Konstandopoulos (2003)

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Digital Combustion Aerosol Standard: d-CAST

\[ d_{\text{me}} = 78 \text{ nm} \]

\[ d_{\text{me}} = 104 \text{ nm} \]

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Digital Combustion Aerosol Standard: d-CAST

\[ d_{me} = 121 \text{ nm} \]

\[ d_{me} = 138 \text{ nm} \]

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Digital Combustion Aerosol Standard: d-CAST

\[ d_{me} = 156 \text{ nm} \]

\[ d_{me} = 210 \text{ nm} \]

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Employ Actual Soot Aggregates in Simulations

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Example: Soot Accumulation on Interdigitated Sensor

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Example: Ab Initio Computation of Filtration Efficiency

See Poster by N. Vlachos, L. Chasapidis and A. G. Konstandopoulos
Conclusions

- Soot aggregates produced at different operating points of the CAST burner have been characterized in terms of their structure and a database of $D_f$, $k_g$, $C_{ov}$, $N_A$, $d_0$ has been established.

- The morphological results obtained by image analysis are shown to be in good agreement with an independent method based on parallel SMPS-ELPI measurements and a previously developed theory.

- A Digital CAST (d-CAST) has been programmed to provide faithful aggregate particle morphologies for ab initio simulations in a variety of applications.

- Current work focuses on extending the size specific CAST characterization in terms of chemical reactivity.

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Acknowledgements

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