Nascent Soot Formation by Agglomeration and Surface Growth

G.A. Kelesidis, E. Goudeli and S.E. Pratsinis

Department of Mechanical and Process Engineering, ETH Zürich, CH-8092 Zürich, Switzerland
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Presenting author email: gkelesidis@ptl.mavt.ethz.ch

Soot can be classified in two categories based on its formation stage, namely nascent (particles of 1-10 nm mobility diameter) and mature soot (fractal-like aggregates with mobility diameters between 10 and 100 nm). Nascent soot particles have been observed in a wide range of combustion sources, from laminar diffusion and premixed flames to diesel engines (Dobbins, 2007). There are major concerns about the adverse effects of nascent soot particles on public health due to their small size and high specific surface area (Pedata *et al.*, 2015). Thus, a better understanding of nascent soot formation is required for identification of process parameters that determine soot particle size and morphology.

Here, nascent soot growth is investigated by Discrete Element Modeling (DEM) of agglomeration and surface growth (SG) by acetylene pyrolysis. The model is validated with theoretical expressions for pure agglomeration (Goudeli *et al.*, 2015) as well as with SG with and without coagulation at full coalescence. The evolution of nascent soot structure is benchmarked against previous numerical studies (Morgan *et al.*, 2007).

Nascent soot growth by agglomeration with or without acetylene pyrolysis is compared to that with full coalescence. Neglecting the non-spherical or fractal-like nature of soot underestimates its mobility diameter and polydispersity up to 40 %. The DEM-obtained size distributions of soot growing by agglomeration with SG are in good agreement with microscopic (Schenk *et al.*, 2013) and mass-mobility measurements (Camacho *et al.*, 2015) in a standard burner-stabilized stagnation ethylene flame.

The evolution of nascent soot structure from spheres to aggregates is quantified by mass fractal dimension, D_f, and mass-mobility exponent, D_{fm}. Figure 1 shows the evolution of DEM-derived mass fractal dimension, D_f (line) of nascent soot particles growing by agglomeration and SG at the flame conditions of Schenk *et al.* (2013) as a function of mobility diameter, d_m. The evolution of D_f is compared to that obtained from microscopic measurements of Schenk *et al.* (2013; symbols, insets). The images and D_f of Schenk *et al.* (2013; insets and symbols) are similar to representative DEM-obtained blue images and D_f of soot particles of the same d_m in Figure 1.

Soot particles are quite spherical for $d_m < 6$ nm where SG is dominant, consistent with Schenk *et al.* (2013). When SG ends ($d_m \approx 10$ nm) and agglomeration starts to prevail, D_f starts to decrease as more open and rather elongated structures are formed by DEM (insets for d_m = 14, 18 and 22 nm). New soot layers are formed on the primary particle surface by acetylene surface reaction and bury the original primary particle boundaries, making the aggregates more spherical. Both DEM- and microscopy-obtained aggregates are much more compact than ballistic-grown agglomerates in the free molecular regime (Ball and Jullien, 1984).

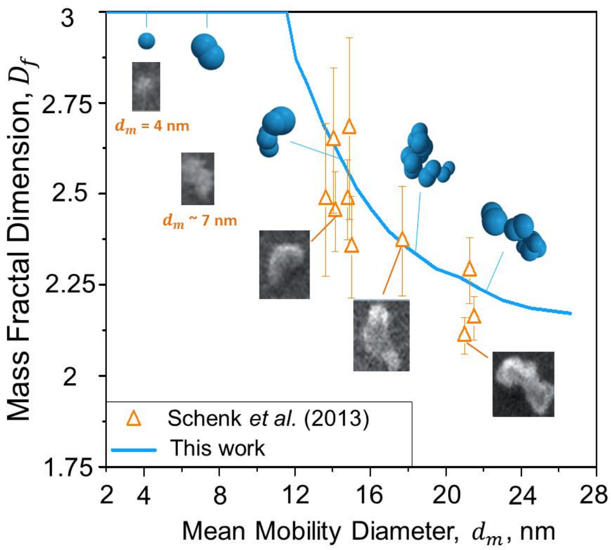

Figure 1. The evolution of the DEM-derived D_f (line) of soot particles growing by agglomeration with surface growth (SG) is in excellent agreement with microscopic measurements of Schenk *et al.* (2013; triangles, insets).

The DEM-derived D_{fm} is also in excellent agreement with mass-mobility measurements (Camacho *et al.*, 2015), pointing out the capacity of the present model to accurately capture soot dynamics by agglomeration and surface growth (SG).

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