Soot Primary Particle Size 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
Keywords: soot, primary particle size, effective density, scaling law.
Presenting author email: gkelesidis@ptl.mavt.ethz.ch

Soot impact on health and environment strongly depends on its primary particle size (Rissler *et al.*, 2013). Microscopy can provide primary particle sizes only by tedious analysis of a statistically significant number of images. Alternatively, a scaling law based on particle projected area exponent, D_a, and prefactor, k_a, has been used in tandem with mass-mobility measurements to obtain accurately the primary particle size of zirconia aggregates and agglomerates (Eggersdorfer *et al.*, 2012). However, D_a and k_a of soot aggregates vary between different combustion sources and conditions (Dastanpour *et al.*, 2016). Thus, a better understanding of D_a and k_a dependence on different flame conditions is needed to improve the scaling law predictions for soot primary particle size.

Here, soot growth is investigated by Discrete Element Modeling (DEM) of agglomeration and surface growth (SG) by acetylene pyrolysis. The acetylene concentration was varied during soot growth by agglomeration and SG in order to attain fixed maximum volume fraction, $f_{v,max}$. Higher $f_{v,max}$ are associated with increasing equivalence ratio (Bönig *et al.*, 1990).

Figure 1 shows snapshots of an exemplary soot particle with initial primary particle diameter $d_{p,o}$ = 2 nm growing by agglomeration with SG attaining $f_{v,max}$ of (a) $5{\cdot}10^{-9}$, (b) 10^{-8} and (c) 10^{-7} at residence time, t = 10, 20 and 30 ms. At t = 10 ms, SG is dominant for all three $f_{v,max}$ forming compact soot dimers and keeping the average number of primary particles per aggregate, n_p, below 2. For t = 30 ms, agglomeration gradually takes over and increases n_p. The average primary particle diameter, d_p, grows effectively up to t = 10 ms and levels off after 20 ms. Higher $f_{v,max}$ lead to larger primary particles. Soot aggregates attaining $f_{v,max}$ = 10^{-7} have large d_p and coagulate fast leading to larger n_p and mobility diameter, d_m, than those formed for $f_{v,max}$ = $5{\cdot}10^{-9}$ and 10^{-8} at the same t. Small soot aggregates with d_m up to 30 nm reach asymptotic D_a = 1.13 ± 0.02 and k_a = 1 ± 0.01. The DEM-derived D_a for agglomeration and SG is larger compared to the respective value of 1.07 for sintering (Eggersdorfer *et al.*, 2012). This is because SG increases the aggregate surface (and projected) area by new soot formation, in contrast to sintering where primary particles approach each other fusing into a single sphere.

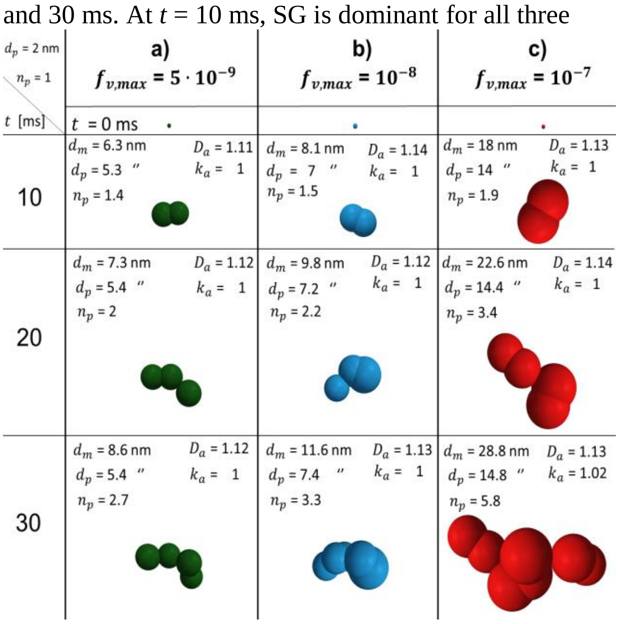

Figure 1. Snapshots of soot particles growing by agglomeration and surface growth (SG) attaining maximum volume fraction, $f_{v,max}$ of (a) $5{\cdot}10^{-9}$, (b) 10^{-8} and (c) 10^{-7}.

As soot aggregates grow larger than 30 nm by agglomeration, D_a decreases and k_a increases logarithmically with d_m. The evolution of D_a and k_a does not change significantly with $f_{v,max}$ and thus correlations describing D_a and k_a as function of d_m are proposed. The scaling law of Eggersdorfer *et al.* (2012) is used in tandem with these relationships to obtain the soot primary particle size from different combustion sources (Rissler *et al.*, 2013; Graves *et al.*, 2015). The accuracy of the scaling law is improved by 15 % when D_a and k_a is varied with d_m. The relationship between n_p, d_m and d_p based on the D_a and k_a derived in this work is compared to agglomeration models (Sorensen, 2011; Eggersdorfer *et al.*, 2012) and mass-mobility measurements of soot aggregates (Rissler *et al.*, 2013).

Bönig, M., Feldermann, C., Jander, H., Luers, B., Rudolph, G., Wagner, H.G.G., (1990) *Proc. Combust Inst.* **23**, 1581-1587.

Dastanpour, R., Rogak, S.N., Graves, B., Olfert, J., Eggersdorfer, M.L., Boies, A.M. (2016) *Aerosol Sci. Technol.* **50**, 101-109.

Eggersdorfer, M.L., Groehn, A.J., Sorensen, C.M., McMurry P.H., Pratsinis, S.E. (2012) *J. Colloid Interface Sci.* **387**, 12-23.

Graves, B., Olfert, J., Patychuk, B., Dastanpour, R., Rogak, S.N. (2015) *Aerosol Sci. Technol.* **49**, 589-598.

Rissler, J., Messing, M.E., Malik, A.I., Nilsson, P.T., Nordin, E.Z., Bohgard, M., Sanati, M., Pagels, J.H. (2013) *Aerosol Sci. Technol.* **47**, 792-805.

Sorensen, C.M. (2011) *Aerosol Sci. Technol.* **45**, 765-779.