Soot Primary Particle Size by Agglomeration and Surface Growth

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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, \(k_0\), 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 \(k_0\) of soot aggregates vary between different combustion sources and conditions (Dastanpour et al., 2016). Thus, a better understanding of \(D_a\) and \(k_0\) 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, \(f_{v,\text{max}}\). Higher \(f_{v,\text{max}}\) are associated with increasing equivalence ratio (Bönig et al., 1990).

Figure 1 shows snapshots of an exemplary soot particle with initial primary particle diameter \(d_{p,0} = 2\) nm growing by agglomeration with SG attaining \(f_{v,\text{max}}\) of (a) \(5 \times 10^{-9}\), (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 \(f_{v,\text{max}}\) 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 \(f_{v,\text{max}}\), lead to larger primary particles. Soot aggregates attaining \(f_{v,\text{max}} = 10^{-7}\) have large \(d_p\) and coagulate fast leading to larger \(n_p\) and mobility diameter, \(d_m\), than those formed for \(f_{v,\text{max}} = 5 \times 10^{-9}\) and \(8 \times 10^{-8}\) at the same \(t\). Small soot aggregates with \(d_{p,0}\) up to 30 nm reach asymptotic \(D_a = 1.13 \pm 0.02\) and \(k_0 = 1 \pm 0.01\). 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.

As soot aggregates grow larger than 30 nm by agglomeration, \(D_a\) decreases and \(k_0\) increases logarithmically with \(d_{p,0}\). The evolution of \(D_a\) and \(k_0\) does not change significantly with \(f_{v,\text{max}}\) and thus correlations describing \(D_a\) and \(k_0\) as function of \(d_{p,0}\) 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 \(k_0\) is varied with \(d_{p,0}\). The relationship between \(n_p, d_{p,0}\) and \(d_p\) based on the \(D_a\) and \(k_0\) 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).


