Experimental and Algorithmic Study of Soot Oxidative Fragmentation

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Over the last years, legislation on particulate matter emissions of diesel cars focuses on ultrafine particles (smaller than 100nm) which are claimed to be responsible for climate change and irreversible health effects. It is characteristic that in Euro 5b, a particle number limit was introduced in addition to the already existing particulate mass limit. Particle number and size distribution are unstable and depend on internal processes like coagulation and fragmentation (Friedlander, 2000). While the coagulation kernel has been extensively studied in the literature (e.g. Kostoglou and Konstandopoulos, 2001), there is still lack of understanding about the fragmentation mechanism.

Harris and Maricq (2002) found that soot particle size distributions from various types of diesel engines have a lognormal shape which may be predicted from the solution of the fragmentation-aggregation equation with Brownian coagulation in the continuum regime. Kostoglou and Konstandopoulos (2003) introduced a more-physically based fragmentation mechanism. Accordingly, fragmentation of the soot aggregates is assumed to occur due to surface oxidation of the solid contacts between the primary particles, the so-called “necks”. Necks are smaller than the primary particle size and fragmentation may occur before a significant reduction of the primary particle size.

Herein, we study the behavior of well-characterized soot aggregates generated by CAST in a heated reactor at temperatures up to 850^{o}C. Two different oxidants are used (O2, mixture of O2 & NO2) and different measurement techniques are applied in parallel. For both oxidants, after the burnout of 50 to 70% of soot, a particle number increase and a new particle size distribution are observed. Particles with diameters similar to primary particle diameters appear initially in small concentration, which increases as the temperature increases. A preliminary attempt to understand the fragmentation process impact on the aggregate population dynamics is done by formulating a Monte-Carlo simulation of aggregate fragmentation. The results of the simulation substantiate some of the experimental observations. from the atmosphere (so called air-capture).

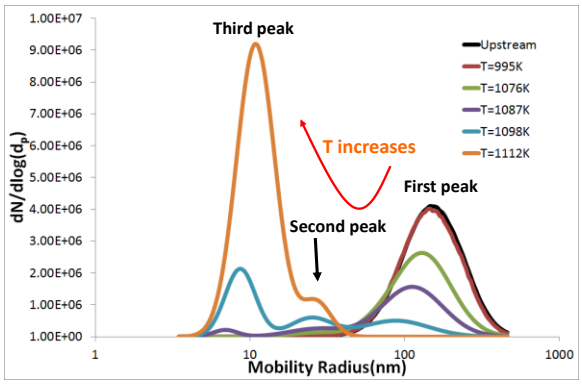

Figure 1. Appearance of oxidative fragmentation modes as temperature is increased.

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