Real-time Characterization of Fractal-like Aerosols

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Aerosol particles are formed by natural and man-made processes, often resulting in fractal-like particle structures. These particles can be held together by either physical forces (van der Waals or electrostatic) resulting in agglomerates or chemical forces (sinter, ionic or covalent) resulting in aggregates. The transport and optical properties of these structures are determined mainly by the composition, number, diameter, and geometric arrangement of their constituent primary particles (Meakin, 1988). These properties may also affect the health impact of these particles as the potential toxicity of inhaled nanoparticles correlates better to their surface area than mass. Thus, real-time characterization of primary particle sizes and specific surface area of gas-borne nanoparticles is necessary for continuous monitoring of aerosol manufacturing and airborne pollutant particle concentrations.

Mostly, ex situ methods have been used to characterize such structures in terms of primary particle diameter and specific surface area by counting microscopic images, nitrogen adsorption, X-ray diffraction, and light scattering.

Here, zirconia (ZrO_2) nanoparticles are generated by scalable spray combustion and their mobility diameter and mass are obtained by differential mobility analyzer (DMA) and aerosol particle mass (APM) analyzer measurements (Eggersdorfer et al. 2012a). Using these data and a newly-developed power law between mobility diameter and primary particle diameter (Eggersdorfer et al., 2012b), the structure of fractal-like particles is obtained in almost real-time (mass-mobility exponent, prefactor and average number and surface area mean diameter of primary particles), for the first time to our knowledge. The so-determined primary particle diameter, d_{va}, is in good agreement with those measured by both nitrogen adsorption (squares) and microscopic analysis (diamonds, Fig. 1). The extent of aggregation is determined by the convergence of the measured d_{va} to that determined using a power law for agglomerates or aggregates. That way the effect of flame spray process parameters (e.g. precursor solution and oxygen flow rates as well as zirconium concentration) on fractal-like particle structure characteristics is investigated in detail during particle synthesis. The primary particle diameter varied between 5 and 25 nm depending on process conditions. Longer particle residence times at high temperatures and high precursor concentrations resulted in larger primary particles with increased degree of aggregation (Fig. 1). When the primary particle growth is coagulation-limited, i.e. the particles have sufficient time between collisions to fully coalesce, agglomerates can form downstream in the reactor at lower temperatures. In contrast, aggregates form when particles encounter collisions before sintering ceased (sintering-limited). Predominantly agglomerates and aggregates of nanoparticles were formed at low and high particle concentration, respectively.

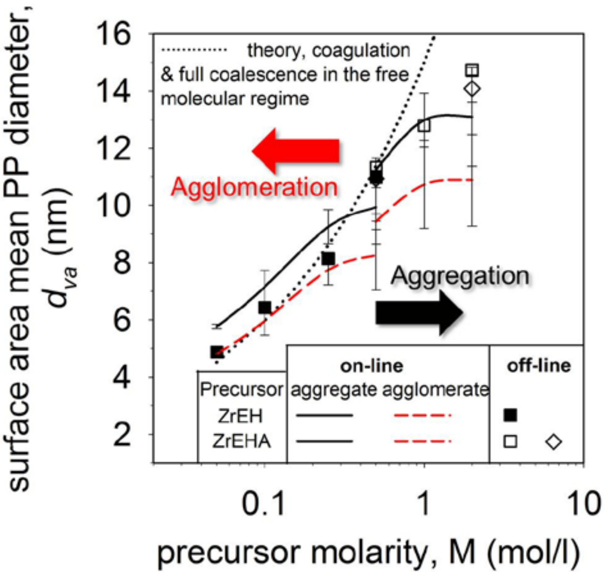

Figure 1. The effect of Zr precursor concentration on surface area mean primary particle diameter, d_{va}, of ZrO_2 particles made by flame spray pyrolysis measured **on-line** by DMA-APM measurements and **off-line** by nitrogen adsorption (squares) and transmission electron microscopy (diamonds). There is a transition from coagulation-limited growth (agglomeration) to sintering-limited growth(aggregation) with increasing precursor concentration.

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