

Measurement of nanoparticles true volume and density in the aerosol phase

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Harnessing the high surface-to-volume ratio of aerosol nanoparticles is essential for improved photo-and-thermal catalysis, nanotube synthesis, and medicine. However, adverse effects on health and environment due to fossil fuel combustion, brake, and tyre wear are still to be assessed and mitigated. All these phenomena are linked to fundamental nanoparticle properties, but in the aerosol phase current knowledge is limited to the measurement of equivalent metrics. The mass is commonly measured using particle mass analysers, relying on knowledge of the particle charging state, from which particle volume can be inferred using a predefined bulk density. Online measurements of volume have been attempted based on charging [1]. Semi-online and offline techniques for volume measurement also exist such as tapered element oscillating microbalance and microscopy retrieval. However, most of these approaches rely on a prescribed density, which is commonly retrieved from aerosol-microscopy techniques [2] or pycnometry. These methods have high uncertainty and very low throughput, but the error propagation inherent to using inadequate densities has dramatic consequences, especially in material synthesis, metrology, and models, e.g., optical and global climate models.

Therefore, it is crucial to develop techniques that can measure the true particle volume and infer the material density of nanoparticles with unknown properties, e.g., alloys, oxidised metals, matured soot, or coated particles. In this work, we propose two prototype techniques aiming at measuring the mass-to-charge change between bare particles and fully-encapsulated particles (Δm) using a centrifugal particle mass analyser (CPMA), along with a final size measurement, i.e., electrical mobility or aerodynamic, of the combined droplets (d_{mf}, d_{aef}). Knowing a pair ($\Delta m, d_{*f}$) allows the true volume of the particles inside the droplets to be calculated. The density is then recovered from the mass and true volume.

The first technique uses a growth apparatus to condense oil on the surface of the particles. To achieve full encapsulation in a spherical droplet, a moderately high temperature is needed within the saturator to evaporate the oil and large temperature difference is required to reach the radial growth mode of the droplets. The minimum T_{sat} and ΔT_{min} are then set with the previous constraints, whereas ΔT_{max} is both constrained by the minimum energy required for unwanted homogeneous nucleation and the higher limit of detection of the instruments. Validated with PSL nanoparticles of multiple sizes, the technique is applied to silver agglomerates and is able to recover the true volume and density, as well as several important properties such as the agglomerates porosity and packing factor.

The second technique employs electrostatically-enhanced coagulation of oppositely charged sample nanoparticles and oil droplets. The collision-coagulation takes place inside a dedicated large volume mixing chamber. The resulting combined droplets are measured with a CPMA and an aerodynamic aerosol classifier (AAC), the latter allowing for charge-independent size measurements. Through this combination the volume of silver agglomerates has been recovered and we show that the volume error using an equivalent sphere approximation reaches up to 60 %.

Our results will analyse an extended set of samples and compare the suitability of each technique.

[1] Su *et al.*, *Aerosol Sci. Technol.*, **2019**, 53, 12, 1453–1467.

[2] Park *et al.*, *J. Nanopart. Res.*, **2004**, 6, 267–272.