

Synthesis and Characterisation of Aerosol Atomic Clusters: A link between Atmospheric Sciences and Nanotechnology

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Atomic clusters are aggregates comprised of a few to several hundred atoms, arranged in ways that can range from highly-ordered to amorphous structures. Depending on the number of atoms they contain, they can exhibit properties that differ substantially from their larger particle or bulk material counterparts, making them particularly interesting for a number of applications, including, e.g., medicine, catalysis, and sensing. At the same time, atomic clusters are the bridge between gas and particle phase in the atmosphere of the Earth, warranting full understanding of their formation and behaviour to explain the conditions under nucleation events take place in different environments.

Early studies on atomic clusters have employed synthesis techniques under high vacuum conditions, including laser ablation, gas condensation, and chemical vapor deposition, followed by ion-mobility, mass, and/or photoelectron spectrometry to determine their properties. In high vacuum systems, the clusters can have extremely high purity and be spatially separated with atomic precision, enabling examination of their electronic structures that define their unique properties and consequently their potential applications.

Developments in aerosol technology over the past decade have demonstrated that atomic clusters can be produced by atmospheric-pressure methods employing flames (Fujiwara and Pratsinis, 2017), glowing wires (Peineke et al., 2009), or spark discharges (Maisser et al., 2015). These activities have gone hand-in-hand with advancements in aerosol instrumentation involving high-flow Differential Mobility Analysers and Mass Spectrometers, employed individually, in parallel, or in tandem (Hogan and Fernandez de la Mora, 2009).

In contrast to high vacuum methods, the composition of clusters produced by atmospheric-pressure aerosol-based techniques strongly depends on the purity of the carrier gas, and particularly trace amounts of oxygen, nitrogen, and water, as has been recently demonstrated by Maisser et al. (2021) for spark ablation. These observations were recently confirmed by advanced computational chemistry methods (cf. Figure 1; Bhowmick et al. 2022), shedding light into their stability and potential use. Despite the impurities in the carrier,

however, a big fraction of these clusters is in the pure form, whereas those that are combined with other atoms coming from the carrier gas could be post-treated depending on the desired application.

In the framework of this talk I will provide an overview of atmospheric-pressure methods for synthesizing aerosol atomic clusters, and highlight the unique applications that these can have in sensing and catalysis. In doing so, the talk will also cover recent advancements in aerosol instrumentation that have enabled atomic cluster characterisation, highlighting the existing challenges and the opportunities they can offer in aerosol-based nanotechnology but also for atmospheric nucleation studies.

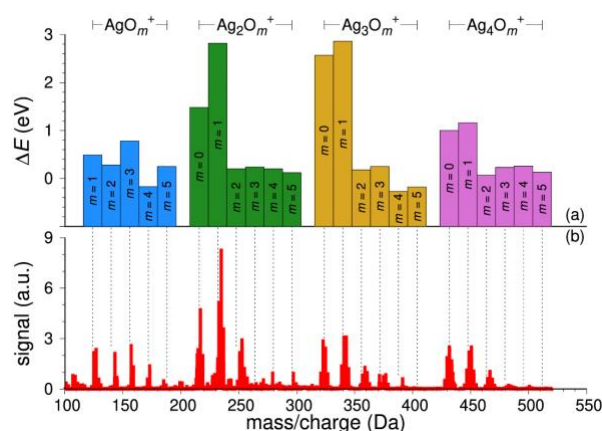


Figure 1. Stabilization energies, ΔE , of the Ag_nO_m⁺ clusters ($n = 1-4$ and $m = 0-5$) calculated by the CCSD(T) method (a), and mass spectra of atomic silver clusters produced by atmospheric-pressure spark ablation. Adopted by Bhowmick et al. (2022).

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