Engineering complexity in metal nanoparticles: Defects and interfaces

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Meeting competing demands for performance and sustainability in applications such as catalysis requires the design and synthesis of increasingly complex nanostructured materials with finely tuned and multifunctional surfaces and interfaces. This added complexity, in turn, provides more opportunity for the introduction of structural and compositional heterogeneity during nanoparticle synthesis. As a result, it is necessary to continually develop creative synthetic approaches for minimizing heterogeneity and/or for tuning architectural parameters such as composition, defects, and interfaces in a controlled and deliberate manner to accurately correlate these features with the performance of the material.

This talk will describe approaches developed in our lab for differentially tuning the rates of competing chemical processes to manage complexity during nanomaterials synthesis in order to generate materials with tailored defects and interfaces. In the first approach, visible light illumination is used as an orthogonal parameter to the chemical composition of the reaction solution to modulate the relative rates of reductive and oxidative chemical processes during materials synthesis. This approach enables the formation of hybrid nanostructures composed of a plasmonic metal and a more reactive metal, with both core-shell and core-satellite architectures. It also provides a means for reconfiguring monometallic Ag nanoparticles to tune defect structure while maintaining the same overall surface facet structure and surface-binding molecules, to separate the influence of these parameters in studies of materials performance and reactivity. The second example is an integrated electrochemical approach that enables the synthesis of shaped nanoparticles directly on electrode surfaces. While nanoparticles synthesized in colloidal solution can be processed and cast onto electrodes, doing so adds an additional fabrication step—increasing time and cost—and faces challenges in controlling particle dispersion on the surface. Our approach links metal nanoparticle synthesis with real-time monitoring of chemical changes in the reaction solution using a combination of colloidal particle synthesis, electrochemical particle synthesis, and electrochemical measurements. This provides a pathway for the directed adaptation of the extensive library of existing shaped colloidal nanoparticle syntheses to growth on a surface—something that remains a non-trivial challenge.