Ga alloy Nanoparticles of Low Size Polydispersity Placed on Highly Ordered Substrates for Supported Liquid Metal Solutions Catalysis

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Many applications of nanomaterials rely on their high specific surface area to generate high performance. In the catalysis field, large distributions of particle sizes and accessibilities on a porous substrate render data modeling and interpretation difficult. Specifically, supported catalytic active liquid metals solutions (SCALMS) catalysts have so far consisted of metal alloy droplets of nonuniform size in inhomogeneous distributions within a meso-/microporous scaffold.[i] The overall catalytic turnovers achieved suggest a high activity, which however has remained non-quantifiable so far given that the number of active sides available and the surface area in contact with the reactants and products are unknown as a consequence. We aim to provide highly ordered model systems of SCALMS catalysts with the ability to tune geometric parameters and quantify turnover as it depends on them. To achieve this, we develop preparative methods capable of controlling the shape of the metal alloy and the porous solid support. In our study, we developed a robust synthesis based on the hot injection method to produce SCALMS nanoparticles, achieving a narrow particle size distribution (PSD). Systematic variation of injection temperature and growth time showed no significant changes in PSD. Spectroscopy and microscopy techniques revealed a 2 nm thick oxide shell around the particles and uniform noble metal distribution within the gallium core, contrary to previous reports.[ii] Using the methylene blue reduction as model reaction, we found that the newly synthesized particles outperform state-of-the-art SCALMS as well as standard Pd/C catalysts. Our methods have allowed us to generate samples presenting Ga-based alloys in spheres of narrow diameter distribution. The stage is set for investigations of catalytic performance upon systematic variation of these geometric parameters.

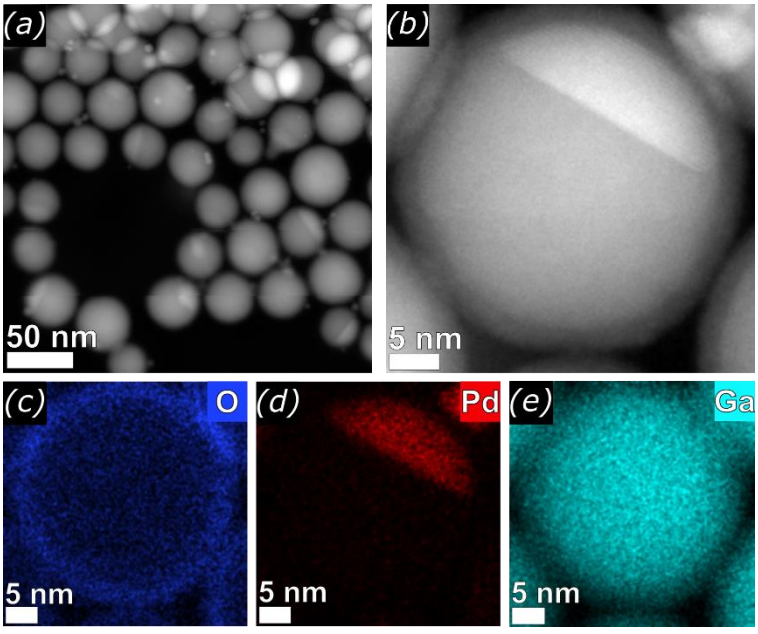

Figure 1: Ga/Pd alloy formation via hot injection method. (a) Dark-Field TEM images of $Ga_{20}Pd$. (b) HAADF-STEM of the phase separated products of Ga/Pd-NPs. (c)-(e) STEM-EDX elemental maps of $Ga_{20}Pd$ (Ga, cyan; Pd, red; O, blue).

i. Taccardi, N. *et al.*, *Nature Chem* , 2017, **9,** 862–867 https://doi.org/10.1038/nchem.2822
ii. S. Carl *et al., J. Phys. Chem. Lett.* 2024, **15,** 17, 4711–4720 https://doi.org/10.1021/acs.jpclett.3c03494