

Exsolution of Core-Shell Nanoparticles in a Complex Concentrated Perovskite Oxide Thin Film Synthesized in One Step

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Precipitation of metal nanoparticles from a host oxide matrix under a reducing environment, also called “exsolution”, is an in-situ synthesis strategy to obtain well dispersed catalyst nanoparticles and introduce percolating channels on/inside functional oxide thin films [1-3]. Exsolved nanoparticles in thin films are commonly fabricated in multiple steps, first by host oxide film preparation and then by reduction of the host film by heating in a reducing atmosphere such as hydrogen [4-7]. In this work, exsolved nanoparticles were successfully prepared in polycrystalline perovskite complex concentrated oxide thin films in one step during synthesis using pulsed laser deposition (PLD) and without further reduction [8]. This one-step method for exsolved oxide film fabrication can effectively reduce processing time and cost as well as maintain high density of exsolved metal nanoparticles. Additionally, tuning PLD growth parameters can modify morphology and structure of the exsolved phase from metal nanostructures to core-shell nanoparticles. Oxygen vacancies in the host perovskite film induce exsolution, and result in the formation of secondary metal, alloy, or metal-oxide phases. Crystallographic misfit strain between the nanostructures and oxide matrix provides additional driving force for mass transport and the formation of unexpected complex multi-element nanostructures, as well as a percolating channel network [9]. More, grain boundaries between nanocolumnar grains serve as oxygen vacancy sinks, providing reactive sites which localize exsolution and contribute to high exsolution density. The effects of PLD growth parameters on structure and composition of the exsolved oxide films were directly and clearly observed with spherical-aberration-corrected scanning transmission electron microscopy (STEM) imaging with energy dispersive X-ray spectroscopy (EDS) and electron energy-loss spectroscopy (EELS). With this one-step method for exsolved functional oxide thin film fabrication, multiple exsolved phases with various composition and structure can be constructed with advanced functionality for catalysis and/or charge transport [10].

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