

Chemical Layer Deposition of metal oxide overcoats with targeted porosity by Stoichiometric and Kinetic control

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Abstract

Catalyst overcoating could become an important strategy in catalyst synthesis due to the possibility of tuning surface reactivity, active site stability and general physicochemical properties of the materials.

It has been demonstrated that overcoating via Atomic Layer Deposition (ALD) and subsequent calcination to induce porosity in the overcoat is an effective catalyst stabilization technique [1]. However, this process is known to be expensive and difficult to apply to powders. Using sol-gel chemistry could be an attractive approach but batch deposition approaches often suffer from poor control of the resulting nanomaterial especially in the case of highly reactive oxide precursors such as aluminum alkoxides..

We report a strategy that combines the advantages of ALD and sol-gel chemistry techniques by applying a solution-phase layer-by-layer technique controlled by the stoichiometry and kinetics of precursor deposition. Our procedure allows for the synthesis of overcoats with targeted thickness and porosity by judiciously choosing synthesis parameters such as water/alkoxide ratio, solvent choice or material aging conditions. Our initial study focused on the overcoating of an alumina supported copper catalyst (Fig. 1). The texture of the overcoats was analyzed by N_2 -physisorption and copper accessibility was measured by N_2O titration and the shape of the coating was imaged by STEM-HAADF. Because this layer-by-layer coating method is performed in solution, a vast number of parameters can be controlled compared to gas phase deposition potentially leading to the formation of several novel nano-environments.

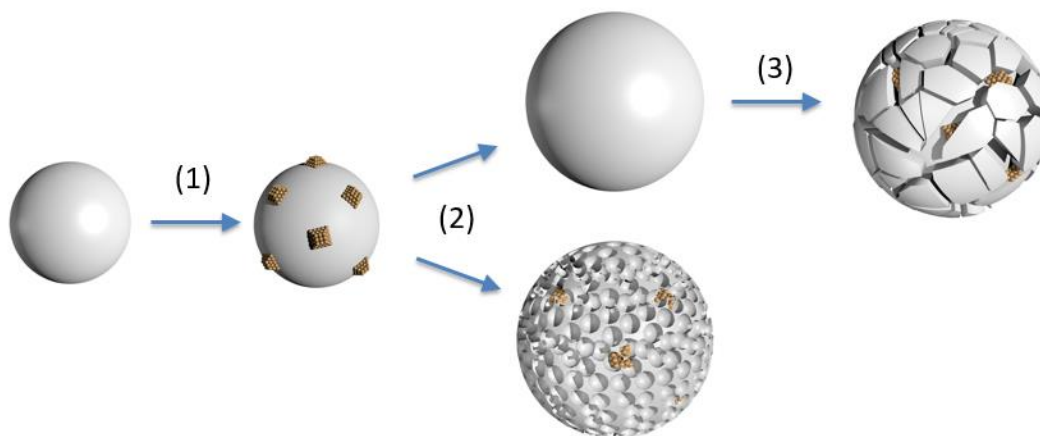


Fig. 1: Steps of catalyst preparation: (1) Impregnation, (2) overcoating, (3) thermal treatment

References

- [1] Brandon J. O'Neill, David H. K. Jackson, Jechan Lee, Christian Canlas, Peter C. Stair, Christopher L. Marshall, Jeffrey W. Elam, Thomas F. Kuech, James A. Dumesic, and George W. Huber, *ACS Catal.* **5**, 2015, 1804–1825
- [2] Rachel A. Caruso and Markus Antonietti, *Chem. Matter.* **13** (2001), 3272-3282