

AIM-ing for Catalyst Synthesis with Single-Atom Precision

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Abstract

A grand challenge in the field of heterogeneous catalysis is to identify and fully characterize sites that are competent for catalysis of desired chemical transformations, ideally with high selectivity, high activity, and high stability. We have found that automated AIM (ALD-like chemistry in MOFs, where ALD is atomic layer deposition) can be used for chemically clean vapor-phase installation of uniform arrays of identically structured, few-atom catalysts on the reactive nodes of suitably chosen metal-organic framework (MOF) materials. This methodology, along with a solution-phase analogue termed SIM, can yield metal-oxygen, metal-sulfur, or metal(0) clusters of predetermined size, shape, and chemical composition.

This lecture will sketch the approach to synthesis and characterization of MOF-supported arrays of well-defined clusters, and then illustrate, via one or two brief case studies, their application as catalysts for desirable, but challenging gas- and condensed-phase chemical transformations. Together with input from computational modeling, these kinds of experiments can be used to address fundamental questions in contemporary Catalysis Science that require atomically precise knowledge of the siting and composition of pre-catalysts, activated catalysts, co-catalysts, reactants and products, i.e. questions of chemical selectivity, chemical confinement, and modulation of activity via control over catalyst metal-atom nuclearity, and general questions regarding emergent complexity in catalytic systems.