Synthetic Architecture of Integrated Nanocatalysts with Controlled Spatial Distribution of Metal Nanoparticles

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Synthetic Architecture of Integrated Nanocatalysts with Controlled Spatial Distribution of Metal Nanoparticles

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ABSTRACT: Integrated nanocatalysts (INCs) with high compositional and structural complexities are recognized as a new class of heterogeneous catalysts exhibiting more advantageous features than the conventional ones. In this work, we have developed a general synthetic protocol for the design and fabrication of INCs by controllable integration of hollow or non-hollow Cu_2O, noble metal nanoparticles (MNPs, M = Au, Pd, and Pt) and mesoporous silica ($m$$SiO_2$) into a single and well-defined matrix. The synthetic protocol was based on stepwise fabrication manner involving sol-gel process to coat mesoporous silica, Ostwald ripening process to generate void space, and galvanic replacement process to deposit ultrafine catalytic MNPs. As a demonstration, in total, our method gives rise to 14 different kinds of INCs with two to four components, such as Cu_2O@M, Au@Cu_2O@M, Au@$h$$Cu_2O$@M, and Au@$h$$Cu_2O$@M@$m$$SiO_2$, *etc*. Interestingly, INCs with the various spatial distributions of MNPs on $m$$SiO_2$ were constructed by using Cu_2O as a sacrificial template under deliberately controlled pH condition. For instance, location regulation of MNPs was achieved on the external surface of $m$$SiO_2$ wall or inside the $m$$SiO_2$ cavity due to the different redox couple of metal precursors and different dissolution rate of Cu_2O during the galvanic replacement process. The workability of the designed INCs was also examined with 4-nitrophenol reduction in the liquid phase and the enhanced catalytic activity was found with catalysts having more exposed MNPs on the external silica surface.