Materials Science & Engineering Doctoral Defense

Copper-based Nanostructures and Atomically Dispersed Metal Species for Heterogeneous Catalysis

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Abstract

Copper-based nanostructures and atomically dispersed metal species have been widely used in heterogeneous catalysis for energy and environmental applications. Nanostructures with desirable physicochemical properties play an important role in controlling the performance heterogeneous catalysts. In this dissertation, systematic research has been carried out on Cu-based and atomically dispersed catalysts with an emphasis on developing new synthesis routes to produce novel catalysts and exploring the synthesis-structure-performance relationships. Hollow nanostructures of copper oxides with high-surface area exhibit notable activity and stability towards low-temperature CO oxidation. These tubular nanostructures can be produced utilizing either an electrochemical corrosion process or by the Kirkendall effect. It is found that the presence of appropriate Cu2O-CuO interfaces governs the observed activity of low-temperature CO oxidation. Therefore, optimizing and stabilizing Cu2O-CuO interfacial sites are highly advantageous for low-temperature CO oxidation reaction on Cu-based catalysts.

Copper-based catalysts effectively reform methanol into hydrogen with low selectivity toward CO. During the steam reforming of methanol (SRM) reaction, however, thermal sintering of Cu species usually occurs at reaction temperatures. It is expected that an epitaxial relationship between CuO nanoparticles (NPs) and ZnO nanowires (NWs) should stabilize the mobile Cu species. Compared to non-epitaxial CuO/ZnO catalyst, the epitaxially grown CuO/ZnO catalysts demonstrated significant sintering resistance during SRM reaction.

Water-gas shift (WGS) is an important reaction for producing hydrogen. It is hypothesized that atomically dispersed Ce species, which strongly interact with high-surface-area γ-Al2O3 can be used as anchoring sites for active Cu species during the WGS reaction. Synthesis protocols have been developed to deposit both atomically dispersed Ce and Cu species on high-surface-area Al2O3 support. These catalysts demonstrated their enhanced stability during WGS reaction.

Functional CeOx nanoclusters supported atomically dispersed Pt catalysts are successfully synthesized via strong electrostatic adsorption. The as-prepared catalyst demonstrates remarkable activity in CO oxidation following reductive activation, which surpasses that of previously reported CeO2-supported Pt catalysts. The activation and regeneration processes of the Pt/CeOx/SiO2 catalysts have been investigated.

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