Investigation on the Formation and Thermal Stability of Pd-Zn γ-Brass Structures

A. Hamed, G. Canning, A. Dasgupta, R.M. Rioux

The formation of the catalytically relevant and complex intermetallic γ-brass Pd-Zn crystal structure has been examined. Bulk intermetallic compounds are typically formed at high temperatures (dictated by the phase diagram) in an evacuated static environment. We present a simple approach to access bulk and nanoparticle (supported on silica) intermetallic phases using an open system under hydrogen and nitrogen. The approach includes the initial synthesis of Pd/SiO2 by dry impregnation followed by heating in the presence of stoichiometric amounts of metallic Zn in H2.

The method was successful in synthesizing phase-pure γ-brass compounds as bulk materials with exceptional composition control (Pd8Zn44 to Pd10Zn42). The weak thermal stability of bulk γ-brass Pd-Zn structures is suggestive of hydrogen embedded in the lattice. In nanoparticle form, the β phase and line compound, PdZn2, were present at similar synthesis conditions to the bulk γ-brass phase. The mixture of phases potentially indicates inaccessible Pd particles on the silica-supported surface due to interparticle contact or the sensitivity of the intermetallic structures to thermal treatment. The formation and decomposition of the γ-brass phase were investigated by in-situ X-ray absorption near edge spectroscopy (XANES) and in-situ X-ray diffraction (XRD) while considering the influence of synthesis parameters such as temperature, time, and environment. The γ-brass phase eventually decomposes into the thermodynamically stable β phase at temperatures > 500°C, and back to metallic Pd in nanoparticle form with enough thermal exposure. Future efforts will focus on the catalytic reactivity of SiO2-supported nanoparticle form γ-brass Pd-Zn and the catalytic relevance and quantification of hydrogen embedded in the lattice of bulk γ-brass Pd-Zn structures.