Bunsentagung 2006
25.-27.05.06
Erlangen / Deutschland

Active-Site Isolated Pd-Ga Intermetallic Compounds for the Selective Hydrogenation of Acetylene

J. Osswald[1], K. Kovnir[1,2], M. Armbrüster[1,2], R.E. Jentoft[1], R. Giedigkeit[2], T. Ressler[1], Y. Grin[2], R. Schlögl[1]

[1]*Abteilung Anorganische Chemie, Fritz-Haber-Institut der MPG, Faradayweg 4-6, 14195 Berlin*
[2]*Max-Planck-Institut für Chemische Physik fester Stoffe, Nöthnitzerstr. 40, 01187 Dresden*

Selective hydrogenation of acetylene ($C_2H_2 + H_2 \rightarrow C_2H_4$) is an important industrial process to remove traces of acetylene in the ethylene feed for the production of polyethylene. A typical hydrogenation catalyst is palladium dispersed on metal oxides which shows high activity but only limited selectivity and long-term stability. The limited selectivity of Pd catalysts in acetylene hydrogenation can be attributed to the presence of ensembles of active sites and, hence, to the number of possible adsorption configurations of acetylene. Active-site isolation leads to increased Pd-Pd distances on the catalyst surface and to only weakly π-bounded acetylene on top of one Pd atom which results in the superior catalytic properties. Well-defined Pd-Ga intermetallic compounds were selected to verify the concept of active-site isolation. PdGa and Pd_3Ga_7 are potential catalysts for the selective acetylene hydrogenation because of the increased Pd-Pd interatomic distances and decreased Pd-Pd coordination numbers in their structures.

Thermal and structural investigations of PdGa and Pd_3Ga_7 were performed by TG/DSC, in situ XRD, and in situ EXAFS (Pd K-edge) in inert and reactive gas atmosphere to determine the stability of the isolated Pd atoms under hydrogenation conditions. The catalytic performance of Pd-Ga intermetallic compounds was tested by acetylene hydrogenation in the presence of ethylene excess. Considerably higher selectivity and long-term stability were found compared to a commercial supported Pd catalyst and an unsupported silver-rich Pd alloy.

The results presented show that structurally well-defined intermetallic compounds possess a high potential to investigate the validity of structural concepts like active-site isolation in heterogeneous catalysis. New catalyst systems found thereby may exhibit superior catalytic properties such as improved selectivity and long-term stability, and may be promising candidates for future industrial applications.