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Synthesis and Investigation of the Electronic Structure of Pd–Ga Intermetallic Compounds as Highly Selective Hydrogenation Catalysts

K. Kovnir[1]*, M. Armbrüster[2], R. Schlögl[2], Yu. Grin[1]

[1] Max-Planck-Institut für Chemische Physik fester Stoffe, Nöthnitzerstr. 40, 01187 Dresden
[2] Dep. Inorganic Chemistry, Fritz Haber Institute, Max Planck Society, Faradayweg 4-6, 14195 Berlin

* Corresponding author: e-mail : kovnir@cpfs.mpg.de, Fax: +49-(0)-351-46464002

Intermetallic compounds consisting of a main group element and a catalytically active transition metal have several advantages over conventional metallic catalysts: due to their well-defined crystal structure, the number of different active-sites is strongly reduced – thus increasing the selectivity. In addition, intermetallic compounds can be obtained as single phase samples avoiding the problems of multiple phases present often observed in conventional catalysts. Furthermore, if covalent bonding is present it can lead to high thermal stability under reaction conditions and reduces the surface segregation – a common problem in conventional bimetallic catalysts.

To prove our concept, the intermetallic compounds PdGa and Pd_3Ga_7 – in both the Pd atoms are isolated by surrounding Ga atoms – were synthesized as single phase samples and characterized as catalysts for the selective hydrogenation of acetylene. As proposed, the investigations revealed that the used intermetallic compounds have considerable higher selectivity as well as enhanced long-term stability compared to commercial Pd/Al_2O_3 and an unsupported silver-rich Pd alloy.[1]

Quantum chemical calculations and bonding analysis by ELI of PdGa reveal strong structuring of the valence region indicating directed (covalent) interactions in PdGa. Each palladium (gallium) atom forms one heteroatomic two-centre bond and participates in six (three) three-centre bonds.

The crystal structure of Pd_3Ga_7 consists of two covalently bonded substructures, one formed merely by gallium atoms, the other built of palladium and gallium atoms.

Despite the covalent character of the chemical bonding, the metallic properties are retained as can be conclude from the metallic-like temperature dependence of the conductivity.

The results obtained reveal the presence of the significant covalent bonding in Pd-Ga intermetallic compounds which explains their enhanced long-term stability and selectivity in the semihydrogenation of acetylene.

References

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