Structural and Catalytic Investigation of Palladium-Gallium Intermetallic Compounds

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Introduction

Palladium in catalysis

Acetylene hydrogenation to ethylene (C₂H₂ + H₂ → C₂H₄) is a common method to remove traces of acetylene in the ethylene feed for the production of polyethylene 1,2. Typical supported Pd catalysts show high activity but only limited selectivity and limited stability 3,4.

Increase of selectivity

Elimination of hydride formation 2,3, Active site isolation 4,5, Modification of electronic structure and thermodynamic properties 6,7,8.

Palladium intermetallic compounds

Pd-Ga intermetallic compounds are particular interesting as potential catalysts because of the isolation of Pd atoms in the structure.

Goal

Determine thermal stability in different gas atmospheres with in situ XRD, in situ XAS and thermal analysis, CO chemisorption and investigation of selectivity and reactivity for catalytic hydrogenation of acetylene.

In situ XRD

Determination of the structural stability in helium and hydrogen using in situ XRD. Only thermal lattice expansion and no phase transition was observable. Increasing particle size lead to decreasing line width.

Thermal analysis of PdGa and Pd3Ga7 in hydrogen: The slight mass loss for both samples are due to the reduction of a surface oxide layer and the exothermic DSC peaks are induced by sintering effects.

CO Chemisorption

Investigation of the surface stability: hydrogen treatment at elevated temperature with following CO chemisorption to detect surface decomposition and Pd segregation.

In situ EXAFS

In situ XAS in hydrogen and during acetylene hydrogenation is used for determination of the thermal stability and the detection of a possible hydrogen inclusion. In contrast to in situ XRD which provides structures and lattice parameters of crystalline phases EXAFS gives information about single atomic distances in crystalline and non-crystalline phases.

Results

Bulk characterisation of PdGa + Pd3Ga7.

High thermal stability under different atmospheres. Annealing of the ballmilling samples lead to sintering.

Catalytic studies of PdGa + Pd3Ga7: Preliminary results

The Pd-Ga alloys show activity for hydrogenation reactions. The selectivity for the hydrogenation of acetylene to ethylene is higher compared to the commercial catalyst Pd on Al₂O₃.

CO chemisorption

No surface decomposition and Pd segregation detectable.

Outlook

• Further preparation of high surface area samples.
• Quantitative catalytic studies.
• Surface investigation with XPS, IR and ISS.

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Literature