



Abstract ID : 67

The electronic structure of PdAu alloys during selective alkyne hydrogenation

Content

Palladium is an excellent catalyst for hydrogenation reactions due to its high conversion and selectivity. Sub-surface carbon at interstitial sites has been identified as the important phase to achieve the high selectivity.[1] However, high hydrogen partial pressures result in the formation of the β -palladium hydride phase and the catalysts loses its selectivity.[2]

To overcome this issue, we synthesized bimetallic palladium-gold alloys whereas the gold modifies the chemical and electronic properties of the palladium resulting in a higher selectivity compared to pure palladium. The full miscibility of both elements allows a fine adjustment of the desired properties. $\text{Pd}_x\text{Au}_{1-x}$ alloys ranging over the whole range of x were synthesized by co-electrodeposition on various substrates, enabling a precise control of the Pd:Au ratio. The catalytic properties were investigated using the selective hydrogenation of Acetylene towards Ethylene as a model system. It was found that the selectivity of the $\text{Pd}_x\text{Au}_{1-x}$ alloys follows a volcano curve, with the alloy $\text{Pd}_{0.75}\text{Au}_{0.25}$ providing the highest selectivity. To understand this behavior and the influence of Au, the catalyst was investigated at relevant and operando conditions at two NAP-XPS beamlines at BESSY II, namely BEIChem-PGM and CAT@EMIL. NAP-XPS, -HAXPES and -NEXAFS were performed in the pressure range of 1 - 4 mbar at the Pd 3d, Au 4f, C 1s and VB core levels as well as at the Pd L3-edge. To achieve the high pressures needed for the formation of the hydride phase a special reaction cell was used that is capable to resist pressures up to 1 bar and allowing operando NEXAFS investigations. The combination of operando XPS and NEXAFS over the large pressure regime allowed the identification of two relevant phases: interstitial carbon atoms and β -palladium hydride. Both phases were found to be strongly influenced by the presence of Au, with an increasing suppression of these phases with increasing gold content. In addition, also the formation of the hydride phase is strongly influenced by the gold, resulting in a combination of phases that can be linked to the increased selectivity of the alloys.

[1] D. Teschner et al., Science 320 (2008) 86-89.

[2] J.J. Velasco-Vélez et al., Top. Catal. 61 (2018) 2052-2061.

Primary author: Dr ZELLER, Patrick (Fritz-Haber-Institut der Max-Planck-Gesellschaft, Dept. Inorganic Chemistry)

Co-authors: Dr ÖZTUNA, Eylül (Fritz-Haber-Institut der Max-Planck-Gesellschaft, Dept. Inorganic Chemistry); VELASCO-VELEZ, Juan-Jesus (Fritz-Haber-Institut Berlin); KNOP-GERICKE, Axel (MPI for Chemical Energy Conversion); SCHLÖGL, Robert (Fritz-Haber-Institut)

Presenter: Dr ZELLER, Patrick (Fritz-Haber-Institut der Max-Planck-Gesellschaft, Dept. Inorganic Chemistry)

Track Classification: Catalysis