Nanoporous Gold Catalysis

Electronic structure calculations of oxidation reactions on nanoporous gold

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In Short

- Density Functional Theory
- Nanoporous Gold
- Au(321), Au(332)

Bulk gold has been known as an inert material without any specific catalytic activity for almost a century. But then in the 1970s Bond et al. [1] presented small gold particles placed on a SiO2 support that could be used for the hydrogenation of alkenes and alkynes. Since this decade a lot of research has been done on nanostructured gold. This Au-based catalysts can be used for fuel cells, the synthesis of esters or the selective oxidation of alcohols.
The selectivity of gold to partial oxidation products is higher than the selectivity of other metal catalysts, so there is a high interest in this gold based catalysts. A problem of gold nanoparticles as catalyst is, that the efficiency increases if the average particle size is reduced [2], so in most cases the major part of the surface area of the supporting material is not used for the catalytic processes.
In addition to this supported forms of gold catalysts an unsupported form of gold, the nanoporous gold (np-Au), characterized by Zielasek et. al.[3], has recently attracted considerable interest due to its potential use in catalysis. Compared to supported gold nanoparticles the complete entire surface of the material can be possibly usable as a catalytic material. The most prominent example for the use of np-Au as a catalyst is the selective oxidation of methanol.

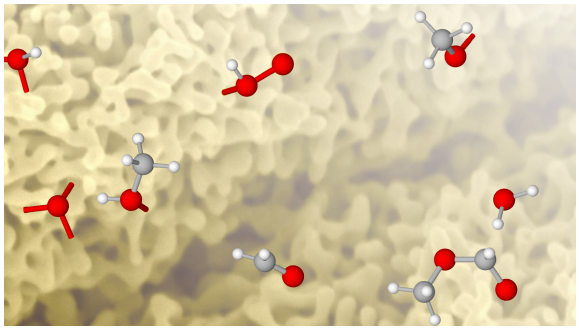

Figure 1: *Possible reactant, intermediate species and reaction products for methanol oxidation on nanoporous gold.*

Although this reactions has been investigated by several groups, the origin of the catalytic activity of np-Au has not been understood completely. The main remaining question that we try to answer is the nature of the active sites of the np-Au. Within DFT (density functional theory) calculations we describe the influence of residual silver atoms in the material and try to explain some possible pathways for the activation of oxygen, the most essential step of most of oxidative coupling reactions.[4]
Another crucial questions addressed by the project is the nature of the active sites of nanoporous gold especially in the context of different alloy compositions. Subsequently, the catalytic behavior of npAu obtained from AuCu alloys will be investigated and compared to AuAg alloys.
For this study, an au(332) surface or a kinked au(321) surface as introduced by Moskaleva [5] represents one of the reactive surfaces of the nanoporous gold. It consists of (111) terraces and zigzag-shaped steps, which may be favourable as possible adsorption positions in partial oxidation reactions. The aim of our work is the elucidation of the detailed mechanisms for total and partial oxidation of methanol and propylene on this model surface. The activation of oxygen is therefore the most elementary step. These catalytic surface reactions are analysed using the exchange-correlation functional PBE implemented in the plane-wave based Vienna ab initio simulation package (VASP) within the supercell approach. In addition AIMD simulations are performed using the program package CP2K.

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https://www.uni-oldenburg.de/tc-kluener

More Information

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Project Partners

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