Multiple Reaction Paths for CO Oxidation on a 2D SnO\textsubscript{x} Nano-Oxide on the Pt(110) Surface: Intrinsic Reactivity and Spillover

An interface stabilized SnO\textsubscript{x}/Pt(110) nano-oxide characterized by a c(2 \times 4) surface reconstruction is prepared and characterized by low-energy electron diffraction (LEED), synchrotron radiation photoemission spectroscopy (SRPES), and scanning tunneling microscopy (STM). Based on the experimental data, atomic models for the nano-oxide are proposed and then validated by comparing the experimental results with the outcome of first-principle calculations. The reactivity of the nano-oxide toward CO is investigated, obtaining that the c(2 \times 4) reconstruction efficiently oxidizes CO to CO\textsubscript{2}. The SnO\textsubscript{x} nano-oxide on the Pt(110) surface can act as a reservoir for oxygen that can diffuse on the adjacent Pt areas where it oxidizes CO. This spillover effect endows the SnO\textsubscript{x}/Pt(110) system with enhanced tolerance to CO poisoning.

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Contributors: Zheng, J., Busch, M., Artiglia, L., Skála, T., Rossmeisl, J., Agnoli, S.
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A DFT Structural Investigation of New Bimetallic PtSnₓ Surface Alloys Formed on the Pt(110) Surface and Their Interaction with Carbon Monoxide

Two surface alloys with p(3 x 1) and p(6 x 1) periodicity have been identified after the deposition of metallic Sn on the (1 x 2)-Pt(110) surface. These two structures have been characterized by low energy electron diffraction (LEED), scanning tunneling microscopy (STM), and photoemission spectroscopy. Based on the experimental results and density functional theory (DFT) calculations, we propose atomic models for these surface alloys, which both consist of a highly corrugated row structure with a very similar surface motif. CO temperature-programmed desorption (TPD) experiments indicate that CO desorbs from the PtSnₓ surfaces at about 415-425 K compared to 495 K on the clean Pt(110). The energetics and geometry of the CO chemisorption sites have been studied by DFT calculations, obtaining an adsorption energy of 0.7-0.86 eV on p(3 x 1) and 0.9-1.05 eV on p(6 x 1). Overall our theoretical and experimental results indicate that the introduction of Sn strongly reduces the CO adsorption energy on the (110) oriented PtSnₓ surfaces.
Beyond the top of the volcano? - A unified approach to electrocatalytic oxygen reduction and oxygen evolution

We study the oxygen reduction (ORR) and the oxygen evolution reaction (OER) and based on previous obtained mechanistic insight we provide a unified general analysis of the two reactions simultaneously. The analysis shows that control over at least two independent binding energies is required to obtain a reversible perfect catalyst for both ORR and OER. Often only the reactivity of the surface is changed by changing from one material to another and all binding energies scale with the reactivity. We investigate the limitation in efficiency imposed by these linear scaling relations. This analysis gives rise to a double volcano for ORR and OER, with a region in between, forbidden by the scaling relations. The reversible perfect catalyst for both ORR and OER would fall into this "forbidden region". Previously, we have found that hydrogen acceptor functionality on oxide surfaces can improve the catalytic performance for OER beyond the limitations originating from the scaling relations. We use this concept to search for promising combinations of binding sites and hydrogen donor/acceptor sites available in transition metal doped graphene, which can act as a catalyst for ORR and OER. We find that MnN4-site embedded in graphene by itself or combined with a COOH is a promising combination for a great combined ORR/OER catalyst.

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Targeted design of α-MnO₂ based catalysts for oxygen reduction

The paper focuses on theoretical and experimental aspects of an oxide surface optimization for oxygen reduction reaction (ORR). Various doped α-MnO₂ based electrocatalysts were prepared by microwave-assisted hydrothermal synthesis and electrochemically characterized to validate density functional theory (DFT) based predictions of the oxidation state and local structure effects on the catalytic activity of α-MnO₂ catalysts in ORR. Both theory and experiments conclude that the highest activity in ORR is to be expected in the case of clustered Mn³⁺ sites which yield activity comparable with that of the polycrystalline Pt. These active sites have to be formed under in-operando conditions and their formation is hindered in doped alpha-MnO₂ catalysts. The activation of Mn³⁺ or Mn⁴⁺ based active sites leads to a shift in selectivity of the ORR process towards 2 electron formation of hydrogen peroxide.

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Enhancing Activity for the Oxygen Evolution Reaction: The Beneficial Interaction of Gold with Manganese and Cobalt Oxides

Electrochemical production of hydrogen, facilitated in electrolyzers, holds great promise for energy storage and solar fuel production. A bottleneck in the process is the catalysis of the oxygen evolution reaction, involving the transfer of four electrons. The challenge is that the binding energies of all reaction intermediates cannot be optimized individually. However, experimental investigations have shown that drastic improvements can be realized for manganese and cobalt-based oxides if gold is added to the surface or used as substrate. We propose an explanation for these enhancements based on a hydrogen acceptor concept. This concept comprises a stabilization of an $\cdot$OOH intermediate, which effectively lowers the potential needed for breaking bonds to the surface. On this basis, we investigate the interactions between the oxides and gold by using DFT calculations. The results suggest that the oxygen evolution reaction overpotential decreases by 100–300 mV for manganese oxides and 100 mV for cobalt oxides.

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