

Vorstellungsvortrag zum Habilitationsvorhaben

Manganese Oxides as Biomimetic Catalysts for Water Oxidation: Atomic Structure and Mechanistic Implications

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Water oxidation (splitting of water molecules into gaseous O_2 , protons and electrons) takes place in green plants and cyanobacteria as part of biological photosynthesis and has shaped the atmosphere of our planet by producing oxygen. Importantly, water oxidation is also pivotal for the sustainable production of non-fossil fuels as water is the optimal source of electrons to reduce protons or CO_2 and convert them into storable fuels (H_2 or carbohydrates). One of the main requirements for the successful practical use of this process is the development of efficient noble-metal-free catalysts that can facilitate this highly energy demanding reaction. In biological systems, water oxidation is catalyzed by a Mn_4CaO_5 center where the metal ions are connected by bridging oxygens and form a small oxide-like cluster. This renders inorganic Mn oxides very attractive water oxidation catalysts for utilization in renewable fuel synthesis.

I will talk about the structural requirements for catalytic activity in synthetic manganese oxides. As most of the catalytically active Mn oxides are non-crystalline materials, the efficient method for their structural characterization is X-ray absorption spectroscopy (XAS). I will present XAS results for Mn oxides synthesized via different synthetic routes and will identify the structural motifs essential for their catalytic activity. A special focus will be given to Mn oxides as electrocatalysts for the anodic water oxidation in an electrolysis cell, one of the most straightforward ways to convert the (solar) electrical energy into H_2 as a storable fuel. Furthermore, the combined results from XAS and electrochemistry provide information on the reaction mechanism of water oxidation catalyzed by inorganic oxides and imply that amorphous Mn oxides are non-classical heterogeneous catalysts. Their activity is not restricted to the surface and is determined by the dynamic interactions of several Mn ions. The structure allows for reversible transition between Mn(III) and Mn(IV) as well as bridging-type changes during the catalytic cycle. Based on the obtained results, I will discuss possible strategies for development of catalysts with optimized activity.

