Combining experiment and theory to design the next generation of metal oxide energy storage materials

Montaha Anjass,[a,b,c] Timo Jacob[c] and Carsten Streb[a,b]*

[a] Institute of Inorganic Chemistry I, Ulm University, Albert-Einstein-Allee 11, 89081 Ulm, Germany
[b] Helmholtz Institute Ulm for Electrochemical Energy Storage (HIU), Helmholtzstraße 11, 89081 Ulm, Germany
[c] Institute of Electrochemistry, Ulm University, Albert-Einstein-Allee 47, 89081 Ulm, Germany

Introduction

The global transition from traditional fossil-fuel-based systems to carbon neutral sustainable energy schemes requires fundamentally new concepts for energy conversion and storage. One of the most promising materials classes in this field are molecular metal oxides, so-called polyoxometalates (POMs). POMs have revolutionized energy materials research as they can bridge the gap between molecular designer materials and technologically important solid-state metal oxides.[1,2] This has led to ground-breaking studies demonstrating their high performance in challenging energy applications including water electrolysis into H2 and O2[3] and as unique electrode materials in Lithium and Post-Lithium ion batteries. Here I combined experimental and theoretical methods to explore the fundamentals of electronic storage and electron transfer in molecular vanadium oxides or polyoxovanadates (POVs).[4] To further study this extraordinary behaviour, I used high-level theoretical computations (density functional theory, DFT) to analyze the electronic structure and predict the redox-processes of \( \text{FeV}_{12} \). To further study this extraordinary behaviour, I used high-level theoretical computations (density functional theory, DFT) to analyze the electronic structure and predict the redox-processes of \( \text{FeV}_{12} \).

Structure and Formation of Polyoxometalates

Polyoxometalate formation processes - self-assembly

Electrochemical Tuning via Heterometal Functionalization

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Electron paramagnetic resonance spectroscopy (EPR) and Density functional theory computations

Experimental spectroscopic (EPR) data suggested that the first two electrons transferred to \( \text{FeV}_{12} \) are stored on the iron, leading to the reduction of Fe\(^{III}\) to Fe\(^{II}\). This is unusual as it leads to a low-valent metal site (Fe) in a high-valent POV framework (containing V\(^{IV}\)).

Outlook

Demonstrate the power of combining theory and experiment as an advanced and efficient way of predicting molecular materials properties for energy storage and conversion.