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Energy Conversion in Solid Oxide Cells: Gaining In-Depth Understanding through Model Experiments

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Solid oxide cells comprise a large class of electrochemical systems with multiple applications in energy conversion (solid oxide fuel and electrolysis cells), heterogeneous catalysis, gas analysis (lambda sensors), and others. The research questions associated with these cells and their materials range from the optimization of commercialized systems to the basic scientific understanding of electrochemical elementary processes. A sound knowledge of the relevant elementary parameters and current pathways is thereby of great importance both for the application case and for purely academic understanding. Identifying these pathways and disentangling elementary parameters such as ionic and electronic conductivities, reaction resistances, interfacial and chemical capacitances is thus the aim of many research works in this field of science. Well-defined model systems are a powerful tool to achieve this goal, as they offer the opportunity to exactly knowing the geometry of the studied electrode and to modify it in a controlled manner. This is the basis for drawing sound conclusions about current pathways and relevant elementary parameters. Moreover, this type of samples is an ideal platform for analytical measurements, as their electrochemically active interfaces are easily accessible by surface sensitive techniques. In this contribution, a couple of examples are presented, where model samples based on thin films are employed to extract elementary properties of functional materials used in solid oxide electrochemical devices:

i) The formation and electrochemical switching behaviour of exsolution catalysts was studied by a combination of electrochemical techniques and *in-situ* surface analytical measurements. These experiments

allowed extensive conclusions to be drawn about both the switching mechanism and the reaction mechanism of H_2 oxidation/ H_2O splitting on metal-decorated perovskite electrodes.

ii) To study the effect of exsolutions on CO_2 electrolysis, *in-situ* near ambient pressure XPS was performed on exsolution decorated perovskite-type electrodes. In this case the polarization induced formation of different carbon species – a carbonate and graphitic carbon – was observed, with the carbonate being regarded as an active intermediate in the CO_2 reduction mechanism.

iii) Finally, it will be shown how the mechanistic understanding obtained on model-type thin film systems can be used to significantly improve the performance of real 3D porous Ni/Ce_{0.9}Gd_{0.1}O_{1.95- δ} (GDC) electrodes in a knowledge-driven manner. By fitting the measured electrode impedance to the correct analytic transmission line circuit, separation and quantification of the individual contributions to the electrode polarisation resistance is possible, and comparison with model studies yields very good quantitative agreement. Moreover, we can demonstrate that fully oxidic GDC electrode functional layers can even outperform state-of-the-art Ni/GDC electrodes in H₂/H₂O atmosphere and exhibit also excellent kinetics in CO/CO₂.



Alexander K. Opitz is the head of the research group Electrochemical Energy Conversion at TU Wien(Vienna, Austria). There he also received his PhD in 2011. He was a visiting scientist at MIT in 2017 and returned to a tenure track position at TU Wien, where he is now an Assistant Professor in the research division of Technical Electrochemistry. He is doing research in the fields of Solid State Ionics (electrode kinetics, current pathways, and electrochemically active zones of solid state electrochemical systems), heterogeneous catalysis (in-situ spectroscopic and analytic studies on the surface chemistry and catalytic activity of electrodes), and materials chemistry (synthesis and characterization of novel, alternative materials for solid oxide cells).