

In situ studies on the chemical activity of complex oxides

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The complex oxides are candidate materials for a wealth of electrochemical devices, but key problems remain in understanding the role of defects on the reactions that take place. In this presentation, we discuss two examples in which we employ *in situ* synchrotron X-rays to probe defect behavior as the oxide undergoes chemical reaction.

In the first example, we study SrIrO₃ as a model catalyst for the oxygen evolution reaction (OER). We start from the initial single crystalline state and find that the surface amorphizes at the beginning of the OER. This is due to direct participation of surface oxygen in the reaction, leading to its gradual removal from the oxygen sublattice. Interestingly, once formed, the thickness of the amorphous layer remains stable with time. We will describe a general model for the reaction and discuss the importance of the surface layer in terms of catalyst activity / stability and design.

In the second example, we investigate SrCoO₃ epitaxial films and their behavior during oxidation and reduction. When reduced, the original perovskite phase transforms into brownmillerite, with a stoichiometry of SrCoO_{2.5}; in an oxygen environment, this can be fully reoxidized, reforming the SrCoO₃ phase. We first discuss the kinetics of these processes and the nucleation and growth of the different phases. We then describe the results of coherent X-ray studies, using X-ray photon correlation spectroscopy (XPCS) to investigate the dynamics of the phase transitions. The advantages of this relatively new technique, which will become widely available with fourth generation synchrotrons, for the study of phase transitions will be described in detail. We find that dynamics of the transition for SrCoO₃ grown on SrTiO₃ (001) versus those grown on (LaAlO₃)_{0.3}(Sr₂TaAlO₆)_{0.7} (001) differ substantially due to the differing strain states.

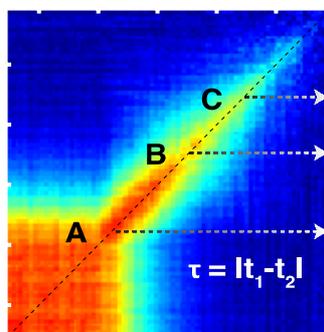


Fig 1. Two-time correlation map of the SrCoO_x film during oxidation.