The voltage and partial pressure dependent defect chemistry of (La,Sr)FeO\(_{3-\delta}\) and its effect on chemical capacitance and oxygen exchange kinetics

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In the search for new high performance solid oxide fuel cells cathodes, understanding the defect chemistry of the materials is of great importance. Not only are defects vital for mass and charge transport, they are also the reaction partners in oxygen exchange reactions, such as the cathodic reduction. In this study, we show how not only the oxygen partial pressure but also the overpotential of polarized electrodes affect defect concentrations and thus the oxygen exchange on polarized (La,Sr)FeO\(_{3-\delta}\) (LSF) model electrodes.

Well defined LSF thin film electrodes were grown by pulsed laser deposition on yttria stabilized zirconia (100) single crystals. Three electrode electrochemical impedance spectroscopy with DC polarization and a variation of oxygen partial pressure were used to determine current overpotential curves and the chemical capacitance of LSF. By investigating the voltage and partial pressure dependence of the chemical capacitance, we could confirm that the oxygen chemical potential alone defines the defect chemical state, regardless whether the chemical potentials were varied via atmospheric oxygen partial pressure or electrode polarization. It is shown, how a detailed defect model can be obtained from measurements of the chemical capacitance.

This detailed understanding of bulk defect chemistry is essential for a proper interpretation of the current-overpotential curves obtained for these LSF electrodes. By simultaneously varying the oxygen partial pressure and the voltage, a fixed defect chemical state could be established in the LSF electrodes despite varying partial pressure. In this manner is became possible to analyze separately how ionic and electronic defects as well as the gas phase affect oxygen reduction and the oxygen evolution reaction on LSF.