



Universität Stuttgart
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und Energiespeicherung



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Finite Elemente Simulation und experimentelle Validierung der Elektrodenkinetik an einer neuartigen Festoxidzellgeometrie

Masterthesis

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Abstract

Cell geometry plays an important role in the investigation of solid oxide cell (SOC) electrode kinetics. Determination of kinetics from single cell is complex, requiring recording of impedance spectra under load and complex nonlinear least squares (CNLS) fitting with adequate equivalent circuit models. Polarization of symmetrical cells results in operation of electrodes in opposing modes, rendering recorded kinetics not interpretable. AC- and DC analysis of symmetrical cells using reference electrodes can be misleading due to geometrical and electrical asymmetries. A significant increase of joule heat from ionic flow across the thick electrolyte of symmetrical cells at high current densities corrupts recorded kinetics at higher current densities. The pseudo-symmetrical cell geometry has been suggested to allow determination of electrode kinetics to high current densities free of electrode reaction heat (or cooling), electrolyte joule heat as well as free of transport effects. The present thesis verifies these claims through finite element modeling (FEM) and experimental validation. A 2D model, including momentum, mass, heat and charge transport, is developed in COMSOL Multiphysics. The implementation of Butler-Volmer reaction kinetics allows investigation of electrode kinetics through simulated electrochemical impedance spectra (EIS) as well as through current density/overpotential curves (CV). Validation and parameterization of the FEM model is realized with employment of self-made co-sintered LSM/YSZ oxygen electrodes and YSZ electrolytes. Several model parameters are determined experimentally using CNLS fits of recorded EIS spectra. The advantages of the pseudo-symmetrical cell geometry such as negligible impact of electrode reaction heat, joule heat, as well as mass transport on electrode kinetics even at high current densities are confirmed. Comparison between simulation and measurement displays similar tendencies and qualitative agreement. The presence of an electrode process observed in experimental EIS data and still under investigation is proven to account for the non-quantitative agreement between model and measurement. The developed model is employed in numerous studies to investigate impact of diverse parameters (e.g. electrode or electrolyte thickness, conductivities of electronic and ionic phases of the two-phase electrode composite) on electrode kinetics. Further work will include the investigation of the unknown process as well as further development of the FEM including more complex electrochemistry.