

Spatially resolved modeling and simulation of degradation in Lithium ion batteries

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SEI growth and Lithium plating are two of the most important aging and degradation mechanisms in Lithium ion batteries. In both cases the development of a mesoscopic structure is initiated on atomistic scale via electrochemical and chemical reactions at the interface of anode and electrolyte and continues to grow into a nanoscale structure. The growth is mediated by a complex interplay of reactions and transport mechanisms. The evolved structures change decisively the local nanoscale electrochemical environment at the interface and have influence on the macroscopic behavior and functionality of the battery. A thorough understanding of these multiscale processes in batteries requires going beyond a pure atomistic description on one side and a pure macroscopic porous electrode modeling on the other side. The challenge for a theoretical description on the intermediate scale from nanometer to micrometers scale is to capture and predict local spatial structures, which are formed by and influence the interplay of transport and electrochemistry on the nanoscale. In our contribution, new theoretical models for SEI growth and 3D structure resolved simulations of Lithium plating and their influence on the local electrochemical environment are presented.

Our structure resolved model for SEI growth [1,2] reveals new universal features of the growth of multilayered SEI films due to the coherent interplay of transport and reaction mechanisms. We demonstrate how the evolved internal structure of the SEI determines the global homogeneity of the growth behavior of the SEI. By comparing with experiments [3] we show that calendar aging due to SEI growth can neither be explained by electronic or solvent transport within the SEI nor by a tunneling mechanism. The only transport limiting mechanism consistently describing the SOC dependent growth pattern is neutral Lithium diffusion due to interstitial transport.

The model for Lithium plating [4] considers the spatially resolved interplay of reversible Lithium film formation and dissolution, electrochemical intercalation from the electrolyte and chemical intercalation from the already plated film. We demonstrate that depending on the local intercalation rate the global transport behavior of Lithium and local current distribution can be considerably influenced by the already deposited Lithium leading in some cases leading to a striking reversal of intercalation currents. Experimentally known influences of lithium stripping/plating on the current voltage curves are reproduced in the simulations and can be correlated with local plating behaviors.

References:

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