

Microstructure-Resolved Impedance Simulations for the Characterization of Li-Ion Battery Electrodes

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Li-Ion batteries are commonly used in portable electronic devices and state-of-the-art electric vehicles due to their outstanding energy and power density. At high current densities, e.g. during fast charging, the transport of Li-ions in the electrolyte is decisive for the performance of the battery cell and optimized electrode designs are required to reduce mass transport limitations. In this respect the impedance of the porous electrode is a characteristic performance indicator and is commonly evaluated integrally with the help of equivalent circuit models. However, often the performance of the electrode is affected by local structural inhomogeneities due to compression in the calendaring process or an unfavorable binder and/or carbon black distribution. For instance, it was found that harsh drying conditions cause binder migration to the electrode surface and consequently reduce the rate capability¹. In this contribution we present simulated impedance spectra of Li-ion battery positive electrodes based on 3D microstructure-resolved simulations² which allows us to study in detail the effect of local structural inhomogeneities on the electrode impedance and, thus, performance.

NMC electrodes with different thickness and density were prepared and characterized electrochemically by galvanostatic cycling and electrochemical impedance spectroscopy. Impedance spectra were recorded on symmetrical cells³ which are especially advantageous for the characterization of electrode transport properties. Reconstructions of the electrodes were created with the help of synchrotron tomography and a 3D stochastic structure generator. The resulting microstructures are then input to microstructure-resolved electrochemical simulations. Impedance spectra of the symmetrical cells and half-cells with Li counter electrode were simulated with a potential step and current relaxation technique⁴. With the help of our simulations we are able to extract the contribution of the carbon black and binder network to the overall pore transport resistance by comparing our simulations to the experimental data. Additionally, we use different models for the spatial distribution of binder and carbon black to mimic different drying conditions and investigate the effect on the electrode impedance and cell performance.

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