Microstructure-Resolved Impedance Simulations: Investigation of the Binder and Conductive Agents-Network in Lithium-Ion Batteries

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Lithium-Ion batteries currently perform better than most other battery systems due to their outstanding energy and power density. At high current densities the transport of Li-ions in the electrolyte is crucial for the battery performance. To reduce transport limitations, the optimization of electrode designs is important. The impedance of the porous electrode is commonly investigated as a characteristic performance indicator. However, the performance of the electrode is often affected by local structural inhomogeneities due to compression in the calendering process or an unfavorable binder and/or carbon black distribution. For example, harsh drying conditions were found to cause binder migration to the electrode surface and consequently reduce the rate capability [1]. In this contribution we apply 3D microstructure-resolved simulations [2] to Li-ion battery positive electrodes, which allows us to study in detail the effect of local structural inhomogeneities on the electrode 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 cells [3] 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 microstructure generator [4]. 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 technique [5]. With the help of our simulations we are able to extract the contribution of the carbon black and binder network to the electrochemical performance of these cathodes. Additionally, different drying conditions are mimicked through different models for the spatial distribution of binder and carbon black. Through these variations the effect on the electrode impedance and cell performance is investigated.

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