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

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The production of Li-Ion battery electrodes is a highly interconnected process and many parameters determine the functionality of the final battery cell. Therefore, characterization techniques are very important to monitor the quality of the electrodes and to analyze deviations in electrode performance. The impedance of the porous electrode is a characteristic performance indicator, relatively easy to measure, and the corresponding spectra provide a comprehensive overview of characteristic timescales of different processes. For a detailed analysis impedance spectra are 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 calendering 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 capability[1]. In this contribution we interpret impedance spectra of Li-ion battery positive electrodes with the help of 3D microstructure-resolved simulations[2]. This 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 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 structure generator[4]. The resulting microstructures are then input to microstructure-resolved electrochemical simulations. 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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