Characterization and Optimization of Lithium-Ion Battery Materials via Particle-Scale Measurements and Simulations

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A precise characterization and optimization of electrochemically active materials (AM) plays an important role in further advancing lithium-ion battery technology. These active materials are typically polycrystalline particles with a diameter of several microns. The properties of AM particles are a major factor in the resulting energy density, rate capability and cycling stability of a battery cell. Therefore, quantification and prediction of material properties is imperative for battery development. However, on electrode scale AMs are mixed with conductive additives and binders forming a complex porous electrode microstructure filled with a liquid electrolyte. This poses several challenges for material characterization. On the one hand, most conventional methods rely on measurements of composite electrodes with many design variables. This introduces uncertainties in the characterization process. For example, the intrinsic AM conductivity is masked by the conductive network that is formed together with the conductive additives. On the other hand, polycrystalline AM particles also inherit a complex inner microstructure themselves, which has a direct influence on its electrochemical performance. A better understanding of the inner particle structure and its related transport processes is required to identify and eliminate bottlenecks for mass and charge transport. These aspects motivate the development of characterization and simulation techniques on the particle scale.

For our investigation, we apply a complementary approach by combining simulations and experiments on the particle scale. We developed an optimized single-particle measurement (SPM) setup allowing electrochemical measurements on single AM particles. This includes rate tests, electrochemical impedance spectroscopy (EIS), and galvanostatic intermittent titration technique (GITT). Thereby, information on electrochemical properties of AMs can be acquired without any influence of an electrode microstructure. To complement these measurements, we extended a previously developed transport model, cf. [1,2]. The derivation of the model relies on a physics-based continuum modelling approach, which only uses physically interpretable parameters as input. By combining experiments and simulations, we investigate and quantify how different particle structures as well as surrounding conditions of the experiment influence the accuracy of the SPM measurement setup. Furthermore, different particle properties can be linked to interior dynamics and, consequently, to particle performance via simulations with varying 3D particle microstructures.

References:

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