From Particle-Scale Information to Cell Performance

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Due to their high energy density, Lithium-Ion batteries (LIBs) continue to play an important role in applications like electric vehicles and mobile devices. As this technology is already well-established the cells are approaching their theoretical limits in terms of energy density. One strategy to overcome these limitations is the development of high voltage cathode materials. However, typical issues of these materials are the stability and cycling performance, which have yet to be solved. The electrodes typically consist of active material particles, conductive additives and binding agents which form a complex microstructure which is soaked with the liquid electrolyte. The properties of the active material particles (e.g. mechanical strength, chemical stability and ionic conductivity) play a major role for the performance of the electrode and battery, respectively. As these properties are largely determined by chemical composition and inner particle structure, an understanding of electrochemical processes on the micro- and nanometre scale is important for material design. Relevant phenomena which need to be considered are micro-porosity, anisotropic Lithium diffusion and grain structures. Some of these properties can be tailored during material synthesis. To name some examples, tunable features include attuned crystal structures like single-crystalline particles, core-shell structures or particles with a full internal composition-gradient. The combination of modelling with improved analytical techniques provides a tool box for optimization on particle level.

From a modelling point of view, many popular electrochemical battery models, like the widely known pseudo-2D-model developed by Newman and coworkers[1], are not capable of accurately capturing phenomena on the particle scale. This introduces the need for the extension of existing and well-established models, such that the interplay of particle and cell performance can be investigated with this crucial information incorporated. Furthermore, due to the difficulty of experimental in-situ investigations of the interior behaviour of particles embedded in electrode structures, such extended models can provide new insights into electrochemical battery processes.

In our work, we tackle this issue by extending a thermodynamic consistent transport theory[2] allowing 3D microstructure-resolved electrochemical simulations where the particle-scale information can be included. Implementation in the simulation framework BEST[3] allows flexible changes in the model equations as well as utilization of structural information on particles like tomography data. Latest results of this ongoing effort will be shown.

***Keywords**:* Li-ion batteries, continuum modelling, micro-structure resolved simulation.

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