# Linking Particle Design to Cell Performance by Electrochemical Simulations

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Due to their high power-density and rate-capability, rechargeable lithium-ion batteries (LIBs) are a well-established technology with many applications in consumer electronics and electric vehicles. In order to meet the high demands for their use cases in the mobility sector, researchers attempt to push the technology closer to their theoretical limitations. Furthermore, alternative new materials with similar or superior performance are developed which in addition promise improvements in toxicity, safety, sustainability and availability. Especially for the cathode side, this has led to increased investigation and optimization of active material (AM) particles on the micro- and nano-scale. Together with binders and conductive additives, these particles typically form a porous electrode structure that is infiltrated by a liquid electrolyte in the cathode of a LIB. This micro-structure offers a lot of room for structural tuning and optimization. Additionally, AM particles themselves exhibit a complex inner structure comprising grains, grain-boundaries and inner porosity which give rise to complex and anisotropic transport phenomena. Consequently, an additional level of complexity in the optimization process of battery cells is introduced that pushes the development of LIBs. In this regard, modeling tools can reinforce material developments and help in gaining a better understanding of the relevant transport and degradation mechanisms on the various involved length-scales.

From a modeling point of view, many popular transport models like the pseudo-2D-model by Newman and coworkers [1], are well-established on the cell- and micro-scale, but do not incorporate important internal particle phenomena. Consequently, model development is required to establish a link to the particle-scale, which has a strong influence on the final cell performance.

Recent work on the extension of a thermodynamic consistent transport model [2] is presented allowing 3D microstructure-resolved electrochemical simulations. Its finite-volume implementation in the simulation framework BEST [3] allows flexible changes in the model equations for the incorporation and investigation of particle-scale phenomena. Structural information on particles like tomography data can be naturally included in the cubic discretization scheme. We present simulations on 3D particle structures including grains and grain boundaries, micro-porosity as well as internal variations in the material composition. Furthermore, a model with extended electrochemistry incorporates information related to the material's crystal structure and ongoing redox-reactions of the intercalation processes. Using this framework, we are currently investigating the influence of varying grain structures and transport resistances inside cobalt-free high-voltage spinel particles and the interplay with the cathode micro-structure. Results from this work-in-progress will be presented.

[1] Fuller, T.; Doyle, M. et al. J. Electrochem. Soc. 1994, 141 (1). DOI: 10.1149/1.2054684
[2] Latz A.; Zausch J. J. Pow. Sour. 2011, 196 (6), 3296-3302. DOI: 10.1016/j.jpowsour.2010.11.088
[3] BEST - Battery and Electrochemistry Simulation Tool, ITWM. https://itwm.fraunhofer.de/best, (accessed: 2022-02-08).