The importance of passive materials in thick Li-ion battery electrodes

Tobias Knorr [a,b], Simon Hein[a,b], Benedikt Prifling[c], Matthias Neumann[c], Volker Schmidt[c], Timo Danner [a,b], Arnulf Latz[a,b,d]

[a] *Helmholtz Institute Ulm for Electrochemical Energy Storage (HIU), Ulm, Germany*
[b] *German Aerospace Center (DLR), Institute of Engineering Thermodynamics, Stuttgart, Germany*
[c] *Ulm University, Institute of Stochastics, Ulm, Germany*
[d] *Ulm University, Institute of Electrochemistry, Ulm, Germany*
E-mail: tobias.knorr@dlr.de

Due to their outstanding energy and power density, Li-ion batteries are widely used in portable electronic devices and electric vehicles. The porous composite of a Li-ion battery electrode generally consists of active material (particle diameter ~ 10-20 µm), conductive carbon (particle diameter ~ 100 nm) and polymeric binder. The so-called carbon binder domain (CBD), a microporous phase formed from carbon black and the binder, is distributed in the macro-pores of the electrode and both, provides better mechanical stability as well as electronic contact. However, at the same time the CBD increases the tortuosity of Li-ion transport pathways in the electrolyte. At high current densities, this increases mass transport limitations and reduces the performance of the battery cell[1]. It has already been shown that the production process has a significant effect on the morphology and spatial distribution of the CBD. For instance, harsh drying conditions can cause binder migration to the electrode surface, which amplifies transport limitations and performance losses[2].

In our contribution we present results of pore scale simulations in Li-ion batteries[3], which explicitly consider the morphology and spatial distribution of the CBD. In these simulations we study 3D realizations of NMC cathodes created by a 3D stochastic microstructure generator[4] with varying density, particle size, thickness and CBD content.

In a first step we determine effective conductivities of the virtual samples which provides insight on limiting processes during operation of the battery cell. Moreover, this is an important input to volume-averaged models of the Newman-type. In a second step, we simulate the electrochemical performance of the virtual electrodes with an extended version of the "Battery and Electrochemistry Simulation Tool" (BEST) considering both the transport of electrons in the conductive carbon network as well as Li-ion transport in the pore space of the CBD. In our simulations we see that insufficient contact and high electronic resistance reduces the capacity at low CBD content for typical cathode densities around 3 g/cm³. Interestingly electronic conductivity is not limiting the electrode capacity at high electrode densities, which opens up new strategies for the electrode design of for high-energy density batteries.

Acknowledgement
This work has been funded by the 'Bundesministerium für Bildung und Forschung' within the project HiStructures under the reference numbers 03XP0243E and 03XP0243D.

References:

1. Hein, S. *et al.* Influence of conductive additives and binder on the impedance of lithium-ion battery electrodes: effect of morphology. *J. Electrochem. Soc.* **167,** 013546 (2020).
2. Jaiser, S. *et al.* Investigation of film solidification and binder migration during drying of Li-Ion battery anodes. *J. Power Sources* **318,** 210–219 (2016).
3. Danner, T. *et al.* Thick electrodes for Li-ion batteries: A model based analysis. *J. Power Sources* **334,** 191–201 (2016).
4. Westhoff, D., Manke, I. & Schmidt, V. Generation of virtual lithium-ion battery electrode microstructures based on spatial stochastic modeling. *Comput. Mater. Sci.* **151,** 53–64 (2018).