Morphology and Function of SEI during Battery Operation

The solid-electrolyte interphase (SEI) on negative electrodes in lithium batteries is one of the main contributors to lithium-ion loss, capacity fade, and limited battery lifetime [1]. Our theory-based continuum models and their experimental validation give insights into the relevant transport and reaction mechanisms for electrons and lithium ions in the SEI.

Standard SEI models focus on capacity fade as a function of time. We developed a series of models to discuss additional dependencies, i.e., SEI morphology [2], potential dependence of SEI growth [3], dependence of SEI growth on cycling conditions [4], and lithium-ion concentration polarization inside SEI [5]. We gain novel insights into transport mechanisms by comparing with dedicated experiments, e.g., battery storage at various state-of-charge [6], differential capacity analysis during cycling [7], and impedance spectroscopy on model electrodes [8].

We highlight the importance of morphology for the function of SEI. It is often reported that SEI is more porous close to the electrolyte than close to the electrode. Our model can predict this dynamic transition in morphology by relating the intrinsic rates of SEI formation reaction and electron transport through the SEI.

On graphite electrodes, Jossen et al. measured the state-of-charge dependence of capacity fade [6]. We show how this reduces the set of potential transport mechanisms responsible for SEI growth [3]. We present the first indirect evidence that neutral radicals carry a negative charge and diffuse through the SEI. Bazant et al. have recently measured and simulated the differential capacity loss during cycling [4,7]. They observe that SEI grows during intercalation dominates. We extend our calendar-life model to describe the observed asymmetry in SEI growth during battery operation. We analyze with 3D micro-structure resolved simulations how the potential dependence of SEI growth results in SEI inhomogeneity throughout the negative electrode.

In this talk, we discuss a physics-based model for impedance spectroscopy of lithium batteries with SEI as porous surface film [5]. Validating our model with experiments of lithium metal electrodes [8], we find large transference numbers for lithium ions. This analysis reveals that lithium-ion transport through the SEI has solid electrolyte character.

This work is supported by the German Research Foundation (DFG) via the research training group SIMET.

Literature

[1] B. Horstmann, F. Single, A. Latz, *Curr. Opin. Electrochem.* **2019**, *13*, 61–69, doi:10.1016/j.coelec.2018.10.013.

[2] P. He, G. Zhang, X. Liao, M. Yan, X. Xu, Q. An, J. Liu, L. Mai, *Adv. Energy Mater.* **2018**, *8*, 1–6, doi:10.1002/aenm.201702463.

[3] F. Single, A. Latz, B. Horstmann, *ChemSusChem* **2018**, *11*, 1950–1955, doi:10.1002/cssc.201800077.

[4] S. Das, P. M. Attia, W. C. Chueh, M. Z. Bazant, *J. Electrochem. Soc.* **2019**, *166*, E107–E118, doi:10.1149/2.0241904jes.

[5] F. Single, B. Horstmann, A. Latz, *J. Phys. Chem. C* **2019**, *123*, 27327–27343, doi:10.1021/acs.jpcc.9b07389.

[6] P. Keil, S. F. Schuster, J. Wilhelm, J. Travi, A. Hauser, R. C. Karl, A. Jossen, *J. Electrochem. Soc.* **2016**, *163*, A1872–A1880, doi:10.1149/2.0411609jes.

[7] P. M. Attia, S. Das, K. Lim, M. Z. Bazant, W. C. Chueh, *J. Electrochem. Soc.* **2019**, *166*, E97–E106, doi:10.1149/2.0231904jes.

[8] F. Wohde, M. Balabajew, B. Roling, *J. Electrochem. Soc.* **2016**, *163*, A714–A721, doi:10.1149/2.0811605jes.