## Modeling of the Solid-Electrolyte Interphase: Transport Mechanisms and Mechanics

Lukas Köbbing<sup>1,2</sup>, Arnulf Latz<sup>1,2,3</sup>, Birger Horstmann<sup>1,2,3</sup> <sup>1</sup>German Aerospace Center (DLR), Wilhelm-Runge-Straße 10, 89081 Ulm, Germany. <sup>2</sup>Helmholtz Institute Ulm (HIU), Helmholtzstraße 11, 89081 Ulm, Germany. <sup>3</sup>Ulm University, Albert-Einstein-Allee 47, 89081 Ulm, Germany. lukas.koebbing@dlr.de

The solid-electrolyte interphase (SEI) considerably affects the performance and lifetime of lithium-ion batteries. Although the SEI has been investigated for many years, various central aspects of this thin passivation layer are still ambiguous due to its generic complexity. Therefore, we thoroughly investigate the growth mechanisms and the mechanical behavior of the SEI.

The long-term growth of the SEI is the main reason which determines the shelf-life of state-of-the-art lithium-ion batteries. Nonetheless, the relevant transport mechanism responsible for the continued growth of the SEI is still highly debated in the literature. Thus, we carefully compare the two mostly considered mechanisms, namely electron diffusion and solvent diffusion [1]. We investigate whether the two mechanisms can describe the observed state-of-charge (SOC) dependence and the typical square-root behavior in time for capacity loss during open-circuit battery storage. We demonstrate that the electron diffusion mechanism reproduces both the SOC dependence as well as the time dependence. Contrarily, we show that solvent diffusion can describe either the SOC dependence or the typical behavior in time. We demonstrate that there is no intermediate regime between the reaction-limited and the transport-limited regimes, which reproduces both dependencies at the same time. In addition, we examine the interdependence of the SOC and square-root time behavior of SEI growth. Due to self-discharge, the combination of both aspects leads to deviations from the typical square-root behavior in time. Our simulations of capacity fade account for this effect and reasonably match capacity fade experiments.

Another substantial aspect of lithium-ion battery research is the transition to next-generation anode materials. As a promising candidate with a high theoretical capacity, silicon receives a lot of research interest. Two major drawbacks of this material are the massive volume expansion and the observed open-circuit voltage hysteresis. Similar to graphite, the SEI covers silicon anodes and has to withstand the expansion and shrinkage of the anode during lithiation and delithiation [2]. We check whether the plasticity of the silicon particle will contribute substantially to the voltage hysteresis. Further, we examine the impact of stress and plastic deformations of the SEI on the volume expansion and the voltage hysteresis. We demonstrate that the mechanics of both silicon anode and SEI will determine the observed voltage hysteresis.



Figure 1: Scheme of electron diffusion and solvent diffusion [1]



- 1. Köbbing, L.; Latz, A.; Horstmann, B. Preprint arXiv 2022, DOI: 10.48550/arXiv.2209.10854
- 2. Kolzenberg, L.; Latz, A.; Horstmann, B. Batter. Supercaps 2022, 5. DOI: 10.1002/batt.202100216