Continued SEI Growth and its Impact on the Silicon Potential Hysteresis

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

The solid-electrolyte interphase (SEI) essentially impacts the performance and lifetime of lithium-ion batteries. Although SEI research has been ongoing for many years, various central aspects of this thin passivation layer are still not conclusively clarified. Therefore, we examine the growth mechanisms and mechanical behavior of the interphase in detail. Improving the SEI will lead to longer-lasting and, consequently, more environmentally-friendly batteries.

The main reason for capacity fade during open-circuit storage in state-of-the-art lithium-ion batteries is the continued growth of the SEI. However, there is still an ongoing debate in the literature about the relevant transport process leading to the long-term growth of the SEI. Therefore, we investigate the characteristics of electron diffusion and solvent diffusion as the most widely used mechanisms [1]. We focus on the experimentally observed dependence of the capacity loss depending on the state-of-charge (SOC) and the typical square-root behavior in time for open-circuit storage. Our simulation shows that the electron diffusion mechanism can explain both the SOC dependence and the time behavior with the same parameters. In contrast, solvent diffusion can reproduce only the correct SOC dependence or the time dependence. A parameter study reveals no intermediate regime between the reaction-limited and the diffusion-limited regimes for solvent diffusion. Furthermore, self-discharge leads to an interplay between the SOC and time dependence. Taking this into account, our simulation can reasonably reproduce experiments observing large capacity fades.

For future applications like aviation, a next generation of lithium-ion batteries with a higher storage capacity is necessary. From the anode side, silicon is the most promising candidate due to its high theoretical capacity. However, silicon anodes show a significant volume expansion the SEI has to withstand. Therefore, large strains and plastic flow will rapidly arise inside the SEI [2]. Further, silicon possesses an open-circuit potential hysteresis, which is problematic for a precise estimation of the state-of-charge. This voltage hysteresis has been explained with plastic models for silicon thin films and large particles but not for amorphous silicon nanoparticles. We demonstrate that our chemo-mechanical model of a silicon nanoparticle and SEI can reproduce the open-circuit potential hysteresis observed in experiments [3].



1.5 CITT data Simulation C/20 Simulation GITT 0.5 0 0 0 0.2 0.4 0.6 0.8 1

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

Figure 2: Simulation of the potential hysteresis in silicon nanoparticles [3]

- 1. Köbbing, L.; Latz, A.; Horstmann, B. J. Power Sources 2023, DOI: 10.1016/j.jpowsour.2023.232651.
- 2. Kolzenberg, L.; Latz, A.; Horstmann, B. Batter. Supercaps 2022, 5, DOI: 10.1002/batt.202100216.
- 3. Köbbing, L.; Latz, A.; Horstmann, B. 2023 (in preparation).