Elucidating the Silicon Voltage Hysteresis by Mechanical Coupling of Anode Particles and the SEI

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

Silicon promises to be a superior next-generation anode material. However, a major challenge of silicon anodes is the significant voltage hysteresis reducing efficiency and leading to detrimental heat generation. Additionally, the hysteresis hinders precise state-of-charge estimation. Our recent research identifies the chemo-mechanical coupling of silicon and the Solid-Electrolyte Interphase (SEI) as the reason for the substantial voltage hysteresis. The SEI is a thin passivating film that grows on negative electrode particles due to electrolyte decomposition [1]. For silicon particles, volume changes lead to significant strains and plastic deformation within the SEI [2]. As anode particle and SEI are mechanically coupled, the stress generated inside the SEI impacts the stress inside the anode, affecting its potential. Our chemo-mechanical model reproduces the observed open-circuit voltage hysteresis [3]. Furthermore, our visco-elastoplastic SEI model reproduces the voltage difference between slow cycling and the relaxed voltage. This detailed physical understanding can improve the performance of silicon anodes.

[1] L. Köbbing et al. J. Power Sources 2023, DOI: 10.1016/j.jpowsour.2023.232651.
[2] L. Kolzenberg et al. Batter. Supercaps 2022, DOI: 10.1002/batt.202100216.
[3] L. Köbbing et al. Adv. Funct. Mater. 2024, DOI: 10.1002/adfm.202308818.