Understanding the Silicon Voltage Hysteresis by considering the impact of the Solid-Electrolyte Interphase (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

Long-range Battery Electric Vehicles (BEVs) and electric planes require the development of next-generation lithium-ion batteries (LIBs) with increased storage capacity. Silicon stands out as the most promising next candidate for the anode, primarily due to its remarkably high theoretical capacity. Furthermore, silicon is an abundant, cheap, and widely spread material.

A major challenge for the implementation is detrimental heat generation during fast-charging due to the significant voltage hysteresis of silicon anodes. The hysteresis leads to an unclear state-of-charge (SOC) to voltage relation impeding precise SOC estimation.

The voltage hysteresis behavior of silicon anodes was rationalized before by three approaches: phase transformations, plastic flow of silicon, and slow diffusion. They explain the voltage hysteresis of crystalline silicon, thin films, and large anode particles. Nevertheless, these approaches are not able to explain the hysteresis observed for silicon anodes consisting of amorphous nanoparticles, the commercially relevant materials.

Our 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 layer that grows on negative electrode particles due to electrolyte decomposition [1]. For silicon particles, volume change leads to significant strains and plastic deformation occurring within the SEI [2]. We demonstrate that our chemo-mechanical model agrees with the Plett model and reproduces the observed open-circuit voltage hysteresis in experiments [3]. Furthermore, our visco-elastoplastic SEI model reproduces the voltage difference between slow cycling and the relaxed open-circuit voltage.

To summarize, we for the first time explain the silicon hysteresis with a visco-elastoplastic particle-SEI model and discuss options to mitigate the related heat generation. This detailed understanding can improve battery management systems.

1. Köbbing, L.; Latz, A.; Horstmann, B. *J. Power Sources* **2023**, 561, 232651.
2. Kolzenberg, L.; Latz, A.; Horstmann, B. *Batter. Supercaps* **2022**, 5, e202100216.
3. Köbbing, L.; Latz, A.; Horstmann, B. *Adv. Funct. Mater*. **2024**, 34, 2308818.