

Understanding the Silicon Voltage Hysteresis and Relaxation: A Chemo-Mechanical Core-Shell Model

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Silicon anodes are a promising successor to graphite anodes with an increased capacity. One major challenge for their implementation is the understanding and treatment of the silicon voltage hysteresis, observed even during slow cycling and open-circuit storage. This contribution explains the silicon voltage hysteresis and relaxation with a chemo-mechanical core-shell model.

Our study considers the chemo-mechanical interaction between an active silicon core and an inactive shell. The shell can represent the solid-electrolyte interphase (SEI), inactive silicon, or a silicon oxide layer. During cycling, the volume of the silicon anode changes substantially due to (de)lithiation. At the same time, the shell has to accommodate the anode volume changes with purely mechanical deformations. The occurring stresses inside the shell can cause pronounced capacity loss [1]. Moreover, the shell generates stresses acting on the silicon particle and the respective chemo-mechanical potential. Accounting for a visco-elastoplastic shell behavior, our simulation elucidates the observed silicon voltage hysteresis during cycling and open-circuit storage [2]. Furthermore, a recent advancement of our model captures the logarithmic voltage relaxation over weeks [3]. Focusing on the interaction at the core-shell interface, we derived a simple hysteresis model preserving physical information for easy voltage predictions during cycling and open-circuit relaxation.

To conclude, our chemo-mechanical core-shell model explains the silicon voltage hysteresis and long-term relaxation behavior. Thus, our study highlights the impact of interfaces and interphases for silicon anodes, whose understanding is crucial for applying pure silicon anodes.

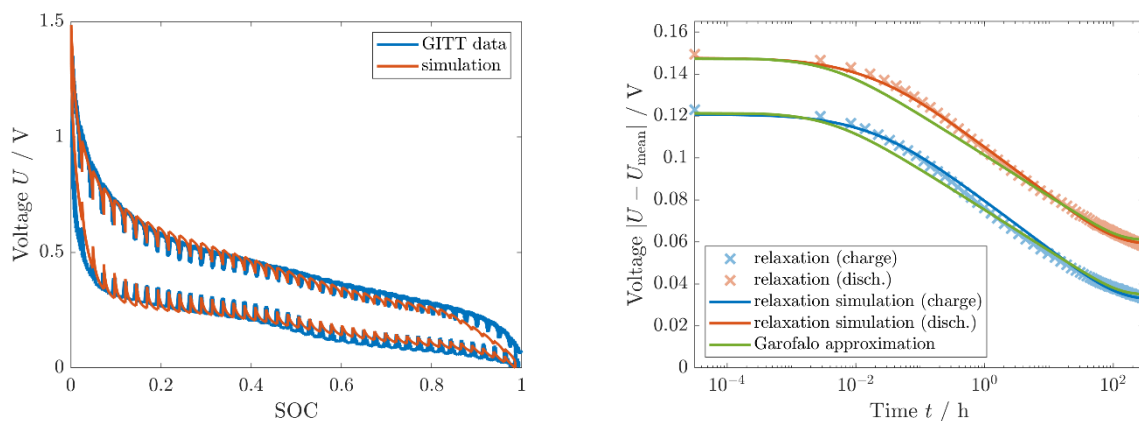


Figure. Simulation of the silicon voltage hysteresis [2] and long-term voltage relaxation [3].

References.

- [1] *Batter. Supercaps* 5 (2022), 2, e202100216, DOI: 10.1002/batt.202100216.
- [2] *Adv. Funct. Mater.* 34 (2024), 7, 2308818, DOI: 10.1002/adfm.202308818.
- [3] *ACS Appl. Mater. Interfaces* 16 (2024), 49, 67609-67619, DOI: 10.1021/acsami.4c12976.