Chemo-Mechanical SEI Model for Silicon Electrodes

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One challenge in the development of silicon electrodes is their large volume expansion. This leads to mechanical stress on the SEI and accelerates battery ageing [1]. In addition, their commercial use is limited by the voltage hysteresis. This hysteresis leads to lower efficiency, harmful heat generation and makes it difficult to estimate the state of charge. Our presentation shows how the two effects are related and how mechanical effects consistently explain the observed silicon voltage hysteresis.

We have developed a chemo-mechanical model that takes into account the interaction between active silicon and a surrounding inactive phase in a core-shell geometry. The shell can be regarded as the solid-electrolyte interphase (SEI). The silicon volume expansion induces visco-elastoplastic flow of the shell, which in turn exerts a constant stress on the silicon core. Thus, our model predicts the experimentally observed hysteresis of silicon stress during cycling and after short-term relaxation [2]. Furthermore, our mechanical model allows the description of long-term logarithmic stress relaxation over weeks [3].

In conclusion, we explain the silicon voltage hysteresis and long-term relaxation with a visco-elasto-plastic core-shell model. We gratefully acknowledge funding by the European Union's Horizon Europe within the research initiative Battery 2030+ via the OPINCHARGE project under the grant agreement number 101104032.

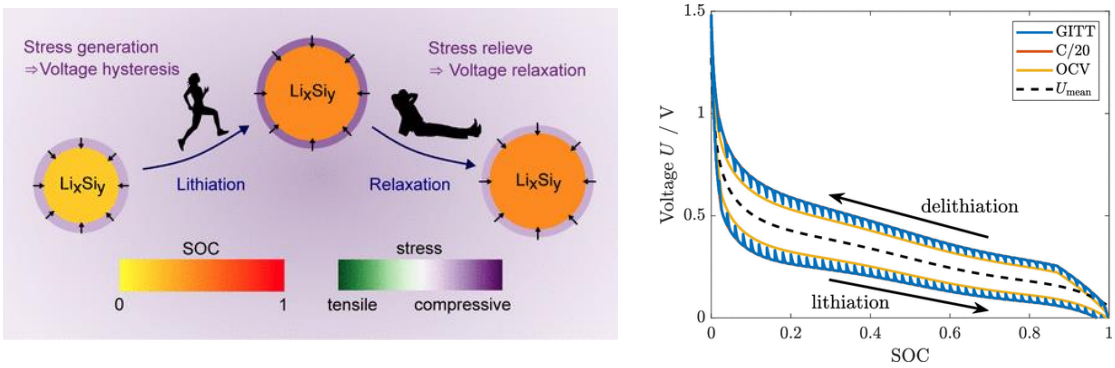

Figure 1: a) Scheme of chemo-electrochemical core-shell model (b) Voltage hysteresis during cycling and after relaxation predicted by the hysteresis model. Reprinted from Ref. [3].

References:

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[2] L. Köbbing, A. Latz, B. Horstmann, Adv. Funct. Mater. **2024**, 34, 2308818
[3] L. Köbbing, Y. Kuhn, B. Horstmann, ACS Appl. Mater. Interfaces **2024**, 16, 67609-67619