Interface and Interphase Modelling Approaches Within OPINCHARGE

Lukas Köbbing,[a,b] Manuel Lopez,[d] Diego del Olmo,[d] Beñat Larrarte-Lizarralde,[d] Elixabete Ayerbe,[d] Arnulf Latz,[a,b,c] and Birger Horstmann[a,b,c]

[a] *German Aerospace Center (DLR), Institute of Engineering Thermodynamics, Wilhelm-Runge-Straße 10, 89081 Ulm, Germany*
[b] *Helmholtz Institute Ulm (HIU), Helmholtzstraße 11, 89081 Ulm, Germany*
[c] *Ulm University, Faculty of Natural Sciences, Albert-Einstein-Allee 47, 89081 Ulm, Germany*
[d] *CIDETEC, Basque Research and Technology Alliance (BRTA), Paseo Miramón 196, Donostia/San Sebastián 20014, Guipuzcoa, Spain*

The OPINCHARGE modelling activities aim to generate an improved understanding of interfaces and interphases inside lithium-ion batteries. The DLR investigates the chemo-mechanical interaction between next-generation silicon anodes and the covering SEI layer. It was identified that the mechanical deformation of the shell in a core-shell setup causes the experimentally observed voltage hysteresis and slow voltage relaxation of silicon nano-anodes [1, 2]. Based on the chemo-mechanical knowledge, the DLR derived a simplified and easy-to-use voltage hysteresis model that preserves the physical information at the interface and captures voltage relaxation processes [2]. Recently, the study was generalized from a spherical symmetric setup to elliptical nanowires to identify the effect of asymmetric geometries. Therein, local changes in the surface-to-volume ratio of the anode lead to heterogeneous stress and lithium concentration distributions [3].

CIDETEC complements with research on the growth of SEI layer during formation process based on kinetic Monte Carlo (kMC) model interactions among electrode and electrolyte particles. Current improvements focus on the combination of kMC with a macroscale model, Single Particle Model (SPM), to be able to capture the voltage evolution during the entire SEI formation process. Further, to streamline feature extraction in experimental cells to parametrize and validate the models, CIDETEC is also creating Distribution of Relaxation Times (DRT) model based on ML to create an automatized methodology to analyze Electrochemical Impedance Spectroscopy (EIS) tests.

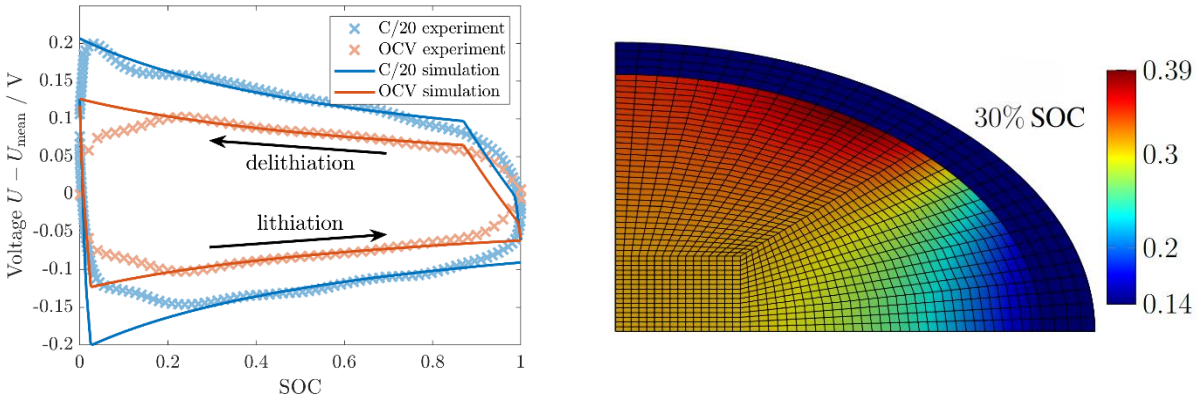

Figure. Voltage hysteresis of spherical silicon-SEI system in reduced model [2] and lithium concentration distribution in elliptical nanowire covered by SEI [3].

References.

[1] *Adv. Funct. Mater.* 34 (2024), 7, 2308818, DOI: 10.1002/adfm.202308818.
[2] *ACS Appl. Mater. Interfaces* 16 (2024), 49, 67609-67619, DOI: 10.1021/acsami.4c12976.
[3] *Batter. Supercaps* (2025), e202400604, DOI: 10.1002/batt.202400604.