

SEI Growth on Graphite and Silicon Electrodes

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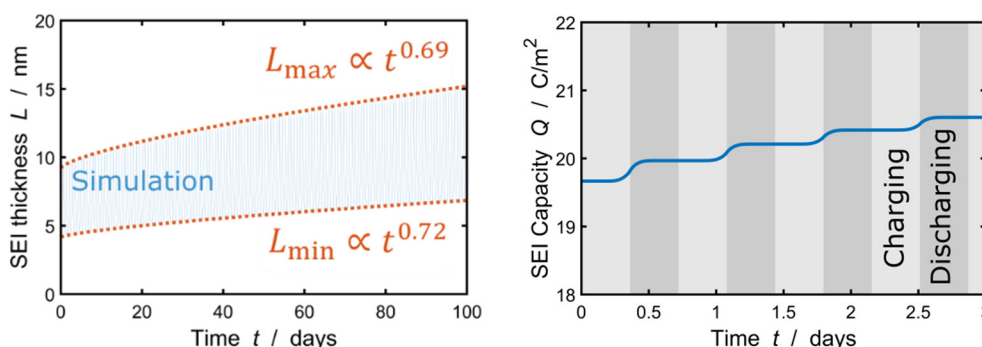
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Electrolyte reduction products form the solid-electrolyte inter-phase (SEI) on negative electrodes of lithium-ion batteries. Even though this process practically stabilizes the electrode-electrolyte interface, it results in continued capacity-fade limiting lifetime and safety [1]. Most SEI models focus on capacity fade as a function of time. By modelling additional properties and dependencies, our recent continuum theories give new insights into the growth and deformation of SEI structure and its function [2,3].

We give arguments that the diffusion of neutral radicals limits the growth of SEI thickness [4], as it predicts the observed potential dependence of SEI growth [5]. 3D micro-structure resolved simulations show that this potential dependence results in SEI inhomogeneity throughout the negative electrode. We extend this calendar-life model to describe the observed asymmetry in SEI growth during battery operation [6] and validate it with recent experiments of differential capacity loss during cycling [7].

Silicon as material for negative electrodes offers high theoretical energy densities. Unfortunately, it exhibits a large volume change during lithium intercalation. This exerts stress on the SEI, leads to continued SEI fracture, and constitutes a major source for capacity fade of silicon anodes. We present a combined mechanical and electrochemical theory of silicon electrodes which captures SEI growth, elastic deformation, plastic flow, and cracking during battery cycling. In this way, we study the evolution of SEI structure during cycling of silicon electrodes.

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References

- [1] B. Horstmann, F. Single, and A. Latz, *Curr. Opin. Electrochem.* **13**, 61 (2019).
- [2] F. Single, B. Horstmann, and A. Latz, *Phys. Chem. Chem. Phys.* **18**, 17810 (2016).
- [3] F. Single, B. Horstmann, and A. Latz, *J. Electrochem. Soc.* **164**, (2017).
- [4] F. Single, A. Latz, and B. Horstmann, *ChemSusChem* **11**, 1950 (2018).
- [5] P. Keil *et al.*, *J. Electrochem. Soc.* **163**, A1872 (2016).
- [6] L. Kolzenberg, A. Latz, and B. Horstmann, *ChemSusChem* **13**, 3901 (2020).
- [7] P. M. Attia *et al.*, *J. Electrochem. Soc.* **166**, E97 (2019).