Modelling of Lithium Droplet Formation During Lithium Dissolution

Martin Werres[1,2] , Arnulf Latz[1,2,3] , Birger Horstmann[1,2,3]

[1]German Aero Space Center (DLR), Stuttgart, Germany
[2] Helmholtz Institute Ulm (HIU), Ulm, Germany
[3] Ulm University, Ulm, Germany

Lithium metal anode batteries have regained a lot of research attention as they are promising candidates for next-generation energy storage systems. However, the poor Coulombic efficiency limits the battery's cyclability. The main causes for low efficiencies are the irreversible loss of active lithium due to the formation of a solid electrolyte interface (SEI) and the formation of unreactive "dead" metallic lithium which is disconnected from the current collector [1]. Experiments show that during the dissolution of lithium whiskers, which grow as irregular structures during plating, the tip cannot be dissolved [2]. We developed a generalized phase-field model of the dissolution process to gain insights into the underlying mechanisms of "dead" lithium formation and capacity fade.

Our model describes the dissolution of a single lithium whisker in a liquid electrolyte based on lithium surface tension and the interaction between lithium and the interphase, utilizing the framework of non-equilibrium thermodynamics [3]. We study under which circumstances instabilities occur and dead lithium forms. Our model predicts the nucleation of an instability for low stripping current densities, induced by interactions with the SEI. This leads to an incomplete dissolution and the formation of an electrically disconnected metallic lithium droplet, in agreement with optical microscope observations of lithium dissolution [2].

References

[1] Fang, C. et al., Nature **572**, 511-515 (2019)
[2] Steiger, J. et al., J. Power Sources **261**, 112-119 (2014)
[3] Horstmann, B. et al., J. Phys. Chem. Lett. **4**, 4217-4222 (2013)