Understanding instabilities in lithium based batteries – dual-layer SEI and isolated lithium formation

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Lithium metal batteries with liquid electrolytes have regained attention as candidates for next-generation high-energy-density batteries [1]. However, the low cycle life is an obstacle to commercialization. The performance and durability of lithium metal batteries are largely influenced by the operating conditions and often find their underlying cause in the nano- and microscale physical processes at the interfaces of the negative electrode. The continuous growth of solid electrolyte interphase (SEI) consumes lithium and electrolyte. Additionally, stripping is not completely reversible, and parts of lithium remain isolated from the current collector [2]. This so-called isolated lithium remains attached to the anode only via an insulating SEI.

We develop thermodynamic consistent mesoscale models to explain observed instabilities on the negative electrode and investigate its dependence on operating conditions. For the SEI growth under storage conditions [3], we present a surface growth model [4] combined with a diffusion-based SEI growth mechanism [5,6]. We observe a universal instability [7] that can explain the emergence of a dual-layer SEI with a compact inner layer and a porous outer layer. For lithium stripping, we present a generalized phase-field model of the dissolution of a single lithium whisker covered by SEI [8]. We find that the instability caused by the interaction between lithium metal and SEI leads to the formation of isolated lithium.

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