Behaviour of Lithium During Stripping – Isolated Lithium Formation

Martin Werres[a,b †], Yaobin Xu [c], Hao Jia[d], Chongmin Wang[c], Wu Xu[d], Arnulf Latz[a,b,c], Birger Horstmann[a,b,c‡]

[a] Institute of Engineering Thermodynamics, German Aerospace Center (DLR), Wilhelm-Runge-Str.10, Ulm, Germany
[b] Helmholtz Institute Ulm (HIU), Helmholtz Straße 11, Ulm, Germany
[c] Environmental Molecular Science Lab, Pacific Northwest National Laboratory, Richland, WA, 99354, USA
[d] Energy and Environment Directorate, Pacific Northwest National Laboratory, Richland, WA, 99354, USA
[e] Institute of Electrochemistry, Ulm University, Helmholtz Straße 16, Ulm, Germany
[†] martin.werres@dlr.de, [‡] birger.horstmann@dlr.de

Lithium metal (Li) batteries are promising candidates for next-generation high-energy density batteries, yet they suffer from low cycle life [1]. It is commonly observed that Li gets isolated from the current collector during cycling, diminishing the available capacity [2]. This isolated lithium is trapped in an insulating solid-electrolyte interphase (SEI) shell [2,3]. However, a fundamental understanding of how Li gets isolated is still lacking. Here, we present a combined experimental and theoretical study to uncover the origin of isolated Li formation [4]. We derive a thermodynamic consistent model for stripping of a single Li whisker, accounting for the interaction between Li and SEI. We find that this interaction leads to locally preferred stripping and isolated Li formation upon further stripping. Cryo transmission electron microscopy investigations of Li whiskers during stripping reveal that these local effects are pronounced at kinks and the tip of Li whiskers. Sources for heterogeneity can be locally varying geometry or heterogeneous SEI. Further, simulations reveal that higher stripping current densities lead to less isolated Li formation. This can be understood in terms of the auto-inhibitory behaviour of the stripping process, where the instability is suppressed above a critical current [5]. We conclude that isolated Li can only be fully avoided when Li-deposition is planar, and the SEI is homogeneous.

References

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