Understanding isolated lithium formation in lithium metal batteries with liquid electrolytes

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Lithium metal batteries (LMBs) with liquid electrolytes are promising candidates for next-generation high-energy-density batteries, currently limited by their cyclability [1]. A main cause for the low cycle life of LMBs is the non-reversible stripping of lithium during discharge [2]. Lithium gets isolated from the current collector and trapped in the insulating remaining solid-electrolyte interphase (SEI) shell [2,3]. However, a fundamental understanding of this process and the stripping dynamics remain elusive. We performed a combined theoretical and experimental study to understand isolated lithium formation during stripping [4]. We derive a thermodynamically consistent model of lithium dissolution underneath the SEI to predict the stripping dynamics of lithium during dissolution. We probe our predictions by resolving the structures after stripping with cryogenic transmission electron microscopy (TEM). We find that locally preferred stripping occurs due to the interaction with lithium and the SEI, which leads to isolated lithium formation. The cryo TEM results reveal that these local effects are particularly pronounced at kinks of lithium whiskers. Heterogeneous SEI, heterogeneous stress fields, or the geometric shape of the deposits can cause these local effects. Further, the amount of isolated lithium formation depends on the operating conditions, where higher stripping current densities lead to less isolated lithium. We conclude that in order to fully mitigate isolated lithium, a planar lithium morphology and a homogeneous SEI must be achieved.

Literature:

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