

Modelling of lithium whisker growth

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Lithium metal batteries are promising candidates for next-generation batteries due to their high specific energy [1]. However, lithium metal anodes show poor cycling stability in commercially available carbonate-based electrolytes. The cycling inefficiencies can be traced back to lithium whisker growth. This high surface-area lithium deposition type leads to accelerated solid electrolyte interphase (SEI) formation and the formation of dead lithium during stripping [2,3]. We investigate the emergence of lithium whiskers in the early stages of electroplating. After the nucleation phase, lithium is deposited underneath the SEI. The SEI stretches with the growing nucleus and may break eventually. We model lithium whisker growth as the extrusion of lithium through cracks in the SEI. In the expected stress conditions, lithium experiences significant creep deformation, which provides a material transport mechanism toward the whisker root. By modeling the flow of lithium as a Herschel-Bulkley fluid, we predict whisker growth rates consistent with experimental observations. The whisker diameter is determined by the crack size of the SEI. Engineering the SEI properties can mitigate whisker emergence and is the most promising approach to achieving safe lithium metal batteries.

[1] Horstmann, B., Werres, M. et al. Strategies towards enabling lithium metal in batteries: Interphases and electrodes. *Energy Environ. Sci.* 14, 5289–5314 (2021)

[2] von Kolzenberg, L., Werres, M., Tetzloff, J. & Horstmann, B. Transition between growth of dense and porous films: theory of dual-layer SEI. *Phys. Chem. Chem. Phys.* 24, 18469–18476 (2022)

[3] Werres, M. et al. Origin of Heterogeneous Stripping of Lithium in Liquid Electrolytes. *ACS Nano* 17, 10218–10228 (2023)