Micromechanics of lithium dendrite growth in solid electrolytes

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Abstract

Lithium-ion batteries with solid electrolytes are transforming electric vehicles by enabling faster charging, extended driving range, and enhanced safety compared to conventional batteries with liquid electrolytes. However, solid electrolytes are prone to cracking near the anode during operation. Lithium deposition into these cracks during charging can lead to dendritic structures that propagate across the electrolyte, potentially causing short circuit. Despite extensive efforts to understand and mitigate lithium dendrite growth in solid electrolytes, the influence of size effects, that is, the increased strength of lithium at dendritic length scales, has been completely overlooked. Recent experiments on compressed lithium layers between ceramic substrates have quantified these size effects, which can now be incorporated into multiphysics models of dendrite growth.

This study develops a comprehensive model that integrates the transport of lithium ions within the solid electrolyte, the deposition of ions into cracks, the viscoplastic flow of lithium within the cracks, the pressure exerted on crack walls, and the fracture mechanics of the electrolyte. Size effects are included by accounting for strain rate gradients in the energetics of lithium.

A parametric analysis is conducted to determine how the ratio of dendrite thickness to viscoplastic length scale influences crack propagation rates and dendrite growth dynamics. These findings provide a basis for future operando experiments, where advanced imaging techniques can monitor the evolving morphology and crystallography of dendrites in real time.

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