A Fracture Mechanics-Based Model of Lithium Dendrite Growth in Partially Filled Cracks of Solid Electrolytes

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Abstract

Solid-state batteries represent a transformative advancement in energy storage, offering enhanced safety and higher energy density compared to traditional liquid-electrolyte Li-ion batteries. These benefits arise from the solid electrolyte's ability to suppress flammability, enable the use of lithium-metal anodes, and increase battery cell energy density. However, lithium dendrite penetration remains a significant challenge in solid electrolytes, often causing short circuits and battery failure. Internal defects, such as cracks and grain boundaries, are known to trigger lithium dendrite formation in solid electrolytes. Understanding the interplay between lithium transport and internal defects is therefore critical to suppress dendrite formation and prolong battery life.

While significant progress has been made in understanding dendrite propagation within filled cracks, recent studies reveal that dendrites can initiate and drive crack propagation even before the crack is completely filled. Here, we develop a computational model coupling solid electrolyte deformation, stress-driven lithium diffusion along the dendrite-electrolyte interface, and stress-dependent Butler-Volmer kinetics of Li deposition into the crack. Using this model, we examine how interfacial diffusivity, interfacial resistance, and applied current density influence dendrite-driven crack growth. Our results show that dendrite thickening, rather than lengthening, promotes early fracture, particularly under conditions of low interfacial diffusivity and high current density.

These findings provide valuable insights into the mechanical and electrochemical interactions within solid-state batteries, offering critical guidance for designing more robust solid electrolytes and creating safer, longer-lasting energy storage systems.

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