

Fatigue fracture predictions in composite solid-state battery electrode microstructures using the phase field approach

Adam Boyce¹

¹ University College Dublin, Ireland. adam.boyce@ucd.ie

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Abstract

Solid-state batteries (SSBs) offer a step-change in performance (up to 2x) over incumbent liquid-electrolyte lithium-ion technologies and are widely regarded as a key enabler for next-generation electric vehicles and consumer electronics [1]. A typical SSB comprises a lithium-metal anode, a ceramic solid electrolyte, and a composite cathode consisting of electrochemically active particles, solid electrolyte, and conductive additives. Despite rapid progress, widespread adoption of SSBs remains limited by severe chemomechanical degradation mechanisms. At the anode, lithium metal is prone to mossy dendrite formation, leading to electrolyte fracture and short-circuiting, while at the cathode, repeated electrochemical cycling induces cracking and interfacial delamination that progressively isolates active material. In this work, we focus on fatigue-driven delamination in composite electrode microstructures. Delamination at particle-electrolyte interfaces is increasingly recognised as a dominant degradation mechanism in SSBs, directly contributing to impedance growth and capacity fade. However, most existing modelling approaches treat fracture as a monotonic process and do not capture the progressive nature of damage accumulation under repeated cycling [2]. To address this gap, we adopt a phase-field fatigue fracture framework capable of describing sub-critical crack growth under cyclic chemomechanical loading. The formulation couples lithium transport, large-strain deformation, and a fatigue-degraded fracture toughness, enabling the gradual evolution of interfacial cracks over many charge-discharge cycles. Microstructurally resolved finite-element models are constructed for both layered oxide cathodes (e.g. NMC-based composites), capturing particle morphology and

interface properties. Simulations demonstrate the fatigue-driven delamination at particle–electrolyte interfaces over 20-30 cycles, consistent with experimentally observed early-life degradation in solid-state cells. The framework predicts spatially heterogeneous damage evolution, highlighting the role of local stress concentrations, particle size, and cycling amplitude. Model predictions are compared against experimental observations from electrochemical cycling and post-mortem characterisation reported in the literature, showing good qualitative agreement in both damage morphology and degradation trends. The proposed approach provides a physics-based tool for predicting fatigue-induced failure in composite SSB electrodes and offers design guidelines for improving interfacial stability through microstructural optimisation, materials selection, and cycling protocol design.

Keywords:

Chemomechanics Phase-field Fatigue Solid-state battery Delamination

References:

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