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Coupled Surface Growth: A Phase Transformation Perspective

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Abstract

The mechanical failure of solid electrolytes (SE) driven by lithium dendrite growth remains a critical barrier to Solid-State Battery (SSB) development for which modeling of the stress-state of the system is essential. Current modeling approaches rely on bulk growth or consider the growing dendrite as rigid and neglect the specific surface-growth kinematics of lithium plating [1, 2, 3, 4]. We address this by proposing a novel formulation that treats surface growth as a phase transformation in which one of the phases is preserved. We distinguish between a growing phase (anode) and a conducting phase (SE) through which material is supplied to the former, where the interface represents a material boundary for the conductor but a non-material boundary for the accreting solid. A fictitious displacement mapping handles the evolution of the conducting phase to accommodate the surface growth of the growing phase, thus introducing the initial, the reference and the current configurations. We use standard tools of continuum thermodynamics to derive the balance laws and jump conditions which have a non-conventional form due to the different nature of the interface for each phase. We identify the driving force that includes the elastic energy density of the growing phase with the chemical potential of the diffusing material and the mechanical stress and strain tensors. Notably, it does not involve jumps in these quantities but their actual values at the interface,

unlike in the classical phase-transformation models [5]. Analysis of a one-dimensional boundary value problem highlights the intrinsic dependence of accretion on the system's chemo-mechanical state. We demonstrate that compressive stress acts as a regulatory stabilizer, modifying the thermodynamic driving force to enable steady-state solutions in otherwise unstable regimes. This theory offers a physically consistent foundation for predicting stress evolution and interface stability in SSBs with lithium metal anodes and provides a rigorous treatment of surface growth coupled with multiphysics of the battery.

Keywords:

Surface growth Phase transformation Driving force Multiphysics coupling

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