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ELECTROCHEMOMECHANICS OF DOPED SOLID ELECTROLYTES AND IMPEDANCE

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Development of next-generation all-solid-state lithium batteries will require a comprehensive understanding solid electrolytes. In particular, the interaction between electrochemical and mechanical behavior has a profound impact on how solid electrolytes perform. The doped garnet ceramic electrolyte $Li_7La_3Zn_2O_{12}$ (LLZO) is a promising candidate for battery applications due to its impressive ionic conductivity [1] and high elastic modulus [2-3]. One of the most crucial issues limiting the deployment of LLZO is that it fails above a critical current [4], at which dendrites nucleate. The mechanism of dendrite formation is still unclear, but is very likely to relate to the mechanical environment state near the metal/solid-electrolyte interface. Another problem with LLZO is that performance-enhancing dopants within the material slowly migrate degrading performance over time, even at relatively low currents. The mechanism of this migration differs substantially from that of multicomponent diffusion in liquids. Our work aims to produce electrochemomechanical dynamic models for doped solid electrolytes to study steady-state operation and impedance, both of which could greatly advance research into the design and characterization of solid electrolytes.

Newman's concentrated solution theory is generalized to relax the electroneutrality constraint and include mechanical effects [5]. Irreversible thermodynamics is used to describe the current in terms of an Onsager-Stefan-Maxwell multicomponent diffusion equation – a thermodynamic consistent formulation useful for multiphysics simulations. A momentum balance connects multiple carriers' diffusion, space charging, and stress to account for the impact of electrochemical/mechanical coupling on solid-electrolyte performance. This framework is applied to study the steady and dynamic response of galvanostatically polarized Li/LLZO/Li cells.

Interfacial impedance has been shown to impact the lifetime of solid electrolytes substantially. We will predict the critical current using interfacial impedance measurements. The impedance of bulk material under different potential biases will be modelled as well. We will investigate how the migration of doped elements, e.g., Al or Ga, could affect both the critical current and the impedance.

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