Phase-field fracture predictions for composite solid-state battery cathode microstructures

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Abstract

The solid-state lithium-ion battery (SSB) is an emerging technology with the potential to provide significant improvements in energy storage capacity relative to the ubiquitous liquid-based (LIB) equivalent. Mass adoption of SSB technology will have enormous benefit on myriad industries, i.e. automotive and aerospace, and is critical to mitigate climate change via reduction of CO2 emissions (1). SSBs normally comprise a multi-material composite cathode, a high-capacity lithium-metal anode, and a ceramic electrolyte separator. The limiting lithium-dendrite problem at the anode has received significant attention and remains a key challenge. Comparatively, the composite cathode has received limited focus despite its equally restrictive issues, specifically the understanding and mitigation of fracture-based degradation mechanisms during cycling – this is the focus of the present study.
An SSB cathode is a heterogeneous structure with transition metal oxide ceramic active particles, ionically conductive oxide or sulphide ceramic electrolyte and a polymer binder containing carbon particulate for electrical conductivity. Poor interfacial contact and active particle volume changes during charge transfer reactions lead to interfacial fracture of the active material-electrolyte interface. This reduces battery power, capacity, and ultimately leads to complete failure.

Predictions of microstructural fracture in a solid-state battery (SSB) or lithium-ion battery (LIB) using the phase field approach are usually made for a single active particle (2) or within a group of particles across an electrode subvolume (3). The present study takes a significant step forward by capturing degradation in the multi-material composite structure of an image-based SSB electrode microstructure obtained through a combination of focused ion beam and scanning electron microscopy. This study simultaneously predicts the fracture of active particles, the solid-electrolyte, and the particle-electrolyte interface using a coupled chemo-mechanical model of a realistic composite electrode structure. This framework offers a deep mechanistic understanding of heterogeneous SSB microstructural degradation. Furthermore it serves as a valuable tool for SSB electrode design to mitigate fracture-induced degradation through careful selection of particle, electrolyte and conductive additive distributions.

References

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