Modeling the effect of electrode geometry, and interface properties in solid-state batteries

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Abstract

Electrodes with three-dimensional (3D) architectures offer significant advantages in overcoming capacity and rate performance trade-offs in batteries. These designs can enable higher active material loading while maintaining fixed transport pathways and improving material utilization. Architected electrodes may also help manage the chemical expansion that battery materials experience during charge and discharge cycles. Our models and experiments demonstrate that the benefits of 3D electrode shapes become more pronounced under extreme conditions, such as low temperature operation and high charge/discharge rates. In this talk, we first focus on the influence of electrode shape on the chemo-mechanical response of battery materials, particularly in all-solid-state batteries. We analyze the stresses generated by intercalation-driven volume expansion and show that, while shaped electrodes significantly reduce ion-transport resistance, they also give rise to unfavorable stress concentrations. Overall, we find that fully interdigitated electrode designs strike the best balance between minimizing resistance and managing mechanical stress. In the second part of the talk, we explore the impact of interface patterning on the cycling stability of lithium-metal anodes. To systematically investigate failure mechanisms in solid-state batteries, we introduce a controlled population of defects, in the form of islands or nuclei. In cell fabrication/operation, nonuniformities in current density and mechanical stress can be caused by material defects, limited wetting or contact, void-formation during Li-stripping, and Li-nucleation in anode-free configurations. By introducing defined defects at smaller length scales (≤ 1 μm), we develop model systems to study electro-chemo-mechanical coupling at various stages of metal electrodeposition. This work was supported by Lawrence Livermore National Laboratory LDRD SI 23-SI-002 and LDRD 25-ERD-006. This work was performed under the auspices of the U.S. Department of Energy by Lawrence Livermore National Laboratory under Contract DE-AC52-07NA27344. LLNL release number: LLNL-ABS-2001427.

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