Integrated modeling approach decodes solid-state battery microstructures for better performance

January 30 2025, by Amanda Lewis

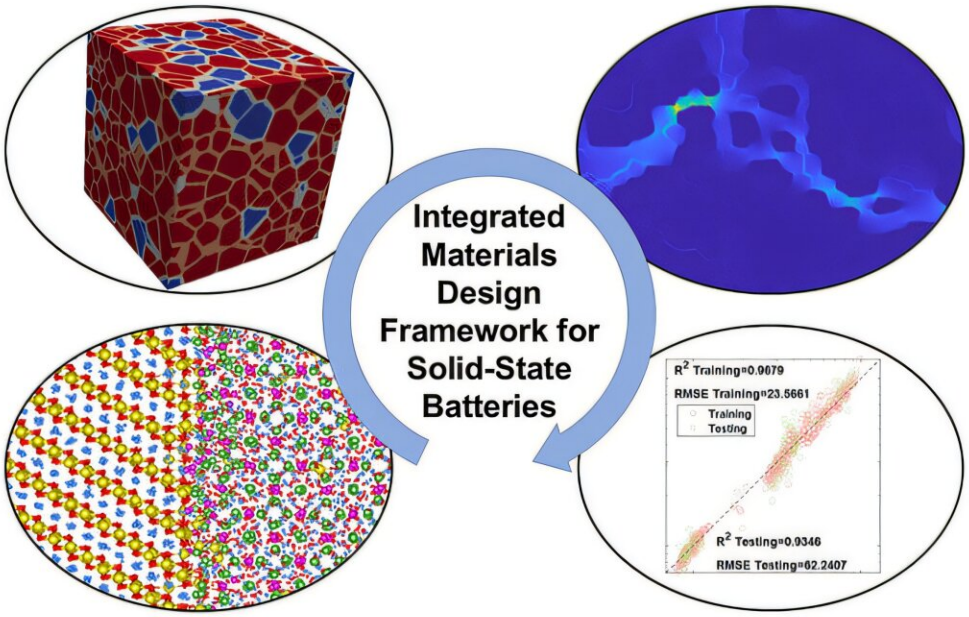

Diagram illustrating the integrated computational framework used to design materials for solid-state batteries. The framework incorporates atomistic simulations of local bulk and interfacial properties, representative multi-phase polycrystalline microstructures, effective property calculations and a machine-learning analysis to correlate microstructure features with effective properties. Credit: Lawrence Livermore National Laboratory

Researchers at Lawrence Livermore National Laboratory (LLNL) have developed a novel, integrated modeling approach to identify and improve key interface and microstructural features in complex materials typically used for advanced batteries. The work helped unravel the relationship between material microstructure and key properties and better predict how those properties affect battery operation, paving the way for more efficient all-solid-state battery design. The research appears in the journal *Energy Storage Materials*.

The team applied their framework to investigate ion transport, an important process for battery function that affects how quickly and efficiently a battery can charge and discharge. The way that ions diffuse through materials is heavily influenced by both the material's intrinsic properties as well as how the material is arranged at the microstructure level.

"Our work introduces a machine learning (ML)-assisted mesoscopic modeling framework to decipher the relationship between microstructural features and ionic transport, representing a cutting-edge approach that combines data-driven techniques with mesoscale modeling," said Longsheng Feng, a postdoc in LLNL's Computational Materials Science Group, Materials Science Division, and the paper's first author.

The work focused on two-phase composites, which are commonly used in solid-state batteries, using $Li_7La_3Zr_2O_{12}$-$LiCoO_2$ as a model system.

"We developed a new method to generate digital representations of the polycrystalline microstructures of two-phase mixtures, combining physics-based and stochastic methods, allowing for efficient, consistent reconstruction of digital microstructures for augmenting microstructural data for training ML models," said Bo Wang, a postdoc and lead co-author of the paper.

The team's new method helped them generate many digital representations of distinct material microstructures with different grain, grain boundary and interface configurations. They then extracted the features of the generated microstructures and employed a ML model to pinpoint specific microstructural features that critically affect effective ionic diffusivity.

"This work builds upon our prior development of a multiscale modeling framework that includes both atomistic modeling and mesoscale simulation capabilities for materials for energy applications," said Brandon Wood, the project's principal investigator.

The team's approach allowed for a comprehensive analysis of very complex microstructural and interface features and their implications for material properties. Their findings confirmed that microstructural feature diversity can significantly impact effective transport properties. Notably, the interface between the two phases played a critical role in determining those properties.

These insights highlight the combined importance of microstructural and interface engineering for improving overall ionic transport properties in composite materials.

"Our established modeling framework can be extended to investigate other critical microstructural and chemical features (e.g., pores, additives and binders), representing the broader impacts and practicality of this approach for materials in energy storage applications and beyond," said Tae Wook Heo, the project's mesoscale modeling lead.

More information: Longsheng Feng et al, Machine-learning-assisted deciphering of microstructural effects on ionic transport in composite materials: A case study of $Li_7La_3Zr_2O_{12}$-$LiCoO_2$, *Energy Storage Materials* (2024). DOI: 10.1016/j.ensm.2024.103776

Provided by Lawrence Livermore National Laboratory