Solid-state batteries (SSBs) are promising for future battery generations, offering inflammable solid Li-ion conductors and high energy density with Li metal anodes. Cubic garnet-type Li7La3Zr2O12 (LLZO) solid electrolytes show excellent ionic conductivity and stability with high-voltage cathodes. However, LLZO reacts with Li metal, forming an amorphous solid electrolyte interphase (SEI) with high impedance that limits battery performance.

This project studies the impact of dopants in LLZO on the electronic structure and stability of the Li/LLZO interface. We use first-principles calculations, machine learning interatomic potentials, and mechanical simulations to develop a computational framework for characterizing the SEI and its evolution over time in pure and doped LLZO. We investigate Li-ion migration in complex Li/LLZO systems with grain boundaries and interphases. Our analysis includes realistic interface structures to determine the origin of impedance and instability. By providing insights into the mechanical behavior of the Li/LLZO interface, this work can contribute to the development of next-generation solid-state batteries with improved performance and reliability.