Reaction and Ion Transport at Solid-state battery Electrode-electrolyte Interface from Machine Learning Molecular Dynamics

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Atomistic-level understanding of the chemical reactions forming the solid-electrolyte interphase (SEI) in solid-state lithium batteries has remained challenging, primarily due to the limited resolution in experimental techniques and the insufficient accuracy in large-scale simulations. In this work, we combine on-the-fly active learning based on Gaussian Process regression (FLARE) with local equivariant neural network interatomic potentials (Allegro) to construct a machine-learning force field (MLFF) to perform large-scale long-time explicit reactive simulation of a complete symmetric battery cell with ab initio accuracy. The MLFF is validated with experimental values of mechanical properties of bulk lithium and diffusion coefficient of solid electrolyte. For the symmetric battery, we observe prominent fast reactions at the interface and characterize the dominant reaction products along with their evolution time scales, using unsupervised learning techniques based on atomic geometry descriptors. Our simulation reveals the kinetics and the passivation involved in the chemical reaction responsible for the SEI formation. The methods in this study are promising for acceleration analysis of atomistic mechanisms in complicated heterogeneous systems and provide design insights for the development of solid-state batteries.