Reactive Molecular Dynamics Frameworks for Describing Transport of Lithium in Solid Electrolyte Interphases

Paolo De Angelis[1, S], Roberta Cappabianca[1], Matteo Fasano[1], Pietro Asinari[1] and Eliodoro Chiavazzo[1, C]

[1]*Politecnico di Torino, Torino, TO, Italy*
eliodoro.chiavazzo@polito.it

In this study, we investigate novel approaches for describing the lithium transport within solid electrolyte interfaces (SEI), that are recognized to play a key role in Li-ion batteries (LIBs), thus aiming at improving the understanding and performance of LIBs - crucial for the green economy transition. First, we focus on the enhancement and reparameterization of the Reactive Force Field (ReaxFF) for more accurate molecular dynamics (MD) simulations, specifically focusing on solid phases of Lithium Fluoride (LiF). Using Python libraries for atomistic simulations, we introduce an automated and interactive protocol for ReaxFF reparameterization. This methodology significantly refines the description of LiF in its solid state, enhancing predictions of solid-phase properties and lithium mobility within the crystal lattice. The reoptimized ReaxFF shows a marked improvement in predicting lithium diffusion, exhibiting a two-order-of-magnitude accuracy at room temperature. Despite these advancements, the study also uncovers challenges in force field parametrization, such as the ReaxFF's sensitivity to training sets and difficulty in interpolating the potential energy surface. This limitation points to the inherent bias in the force field, initially designed for organic systems, and underscores the need for additional corrective energy terms for more accurate modeling of inorganic systems like LiF.

In a second part of the work, we provide future perspective by highlighting the potential of neural network-based force fields (NNFF) as an alternative, possibly offering superior performance but potentially less physical insight. The methodology demonstrated for ReaxFF can be extended to other force fields and potentially incorporate other inorganic compounds in SEI, paving the way for more comprehensive and realistic simulations of LIBs. In conclusion, this research represents a step forward in the accurate simulation of lithium transport phenomena in LIBs, contributing to the development of more efficient and safer battery technologies.