Machine Learning Potentials for Amorphous Solid Electrolytes for All Solid State Batteries.

The topic of solid electrolytes for energy storage systems is both fascinating and crucial from an environmental and sustainability perspective. Liquid electrolytes, commonly made from lithium compounds, face low stability and high cost. Furthermore, lithium extraction is not sustainable for the environment or local communities. Thus, researchers are eager to develop safer and more ecological alternatives. All-solid-state sodium batteries (ASSSBs) are promising candidates for grid-scale energy storage, but there is still much to understand about the electrochemical stability of solid electrolytes with sodium metal. Amorphous sodium phosphorus oxysulfides (aNPOS-SEs) show a high critical current density, but the relationship between their composition, structure, and properties at the atomic level remains unclear.

Computational methods can predict and study the properties of NPS and NPOS solid electrolytes. Classical molecular dynamics is often used to analyze the relationship between composition and properties, but empirical potentials for these systems are not yet available. Although DFT and ab initio molecular dynamics do not require empirical potentials, their high computational cost limits them to small systems.

Machine learning techniques offer a promising solution to develop new reliable potentials for various materials. In this study, we plan to adopt and compare two ML models: the Deep Potential MD approach (a variant of the Behler and Parrinello NN Potentials) proposed by Zhang et al., and the MACE framework developed by Batiata et al. In both models, the total potential energy of the system is assumed to be the sum of atomic energies associated with each atom. These atomic energies depend in a complex and non-linear way on the atomic environment, specifically, on the nature and disposition of surrounding atoms within a cutoff radius. In DeepMD, element-specific Deep Neural Networks (DNN) with several hidden layers and nodes learn the analytical dependence between atomic energy and environment. In contrast, MACE employs Higher Order Equivariant Message Passing Neural Networks.

These models will be trained and tested against energies, forces, and components of the virial tensor computed at the DFT level. In addition, it will be interesting to determine whether these methods can be applied to systems with geometries significantly different from those in the training set.

The developed potentials will be used to obtain a structural description of sodium phosphorus oxysulfides with varying compositions, focusing particularly on the relationship between their structure and their physical and chemical properties (such as ionic conductivity). At the end of the PhD project, I also plan to investigate the Mixed Glass Former Effects (MGFE): examining the effect of mixing anions (O, S) and cations (P, Si, B, Ge) at a constant sodium amount on glass properties.