Investigating disorder effect in Li_6PS_5Cl via machine-learning potential

Solid-state electrolytes with argyrodite structures, have attracted considerable attention due to their superior safety and higher ionic conductivity than other solid electrolytes. Although experimental efforts have been made to enhance conductivity by disorder engineering, the underlying diffusion mechanism is not yet fully understood. Moreover, existing theoretical analyses based on *ab initio* molecular dynamics (MD) simulations have limitations in addressing various types of disorder at room temperature. In this study, we directly investigate Li-ion diffusion in Li_6PS_5Cl at 300 K using large-scale, long-term MD simulations empowered by machine-learning potentials (MLPs). The computed Li-ion conductivity, activation energies, and equilibrium site occupancies align well with experimental observations. Notably, Li-ion conductivity peaks when Cl ions occupy 25% of the 4c sites. In addition, Li-ion diffusion shows non-Arrhenius behavior, leading to different activation energies at high temperatures. These phenomena are explained by the interplay between inter- and intra-cage jumps. By elucidation of the key factors affecting Li-ion diffusion in Li_6PS_5Cl, this work paves the way for optimizing ionic conductivity in the argyrodite family. This study was published in ACS Applied Materials & Interfaces 2024, **16**, 46442.