Lithium Localization by Anions in Argyrodite Solid Electrolytes from Machine-Learning-Based Simulations

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The introduction of density functional theory (DFT) has improved the study of material properties. This has enabled significant breakthroughs in solid electrolytes, which have emerged as promising candidates for next-generation energy storage systems. However, DFT faces limitations due to the extremely high computational costs required for large atomic cells and long simulation times. In current study, we introduce AI-based simulations based on neural network potentials (NNPs), extending capabilities of DFT to explore the effect of anions on lithium diffusion in Li argyrodite (Li_6PS_5X, X=Cl and Br)[1]. Our investigation categorizes lithium frameworks into two distinct cages, demonstrating that sulfur ions in these cage centers bind the surrounding lithium ions. As a result, we propose a strategy to enhance lithium ion conductivity by minimizing the occupation of sulfur ions in cage centers. This finding critically refines our understanding of solid electrolytes, connecting theoretical insights with practical applications. The incorporation of NNPs not only improves the prediction of material properties but also deepens our comprehension of the fundamental mechanisms. The results represent a pivotal advancement in solid-electrolyte research, merging large-scale computational techniques with in-depth material analysis and providing a notable perspective to the field.

References

1. Hyun-Jae Lee, Hyeonjung Kim, Sungyoung Ji, Kyuri Choi, Ho Choi, Woosang Lim, Byungju Lee. Advanced Energy Materials, **2024**, 14, 2402396.