Data-driven exploration of halide spinels for high performance ionic conductors

Panyalak Detrattanawichai[1], Aron Walsh[1]
[1]Department of Materials, Imperial College London, Exhibition Road, London SW7 2AZ, UK

a.walsh@imperial.ac.uk

Halide spinels (AB_2X_4, where X = Cl^-, Br^-, I^-) represent a promising class of materials for high performance solid-state battery electrolytes, yet their compositional space remains largely underexplored.[1] Here, we present a high-throughput screening (HTS) framework that integrates materials databases, chemical heuristic rules, and machine learning to identify synthesisable candidates with high ionic conductivity. Our workflow explores tens of thousands of potential compositions across the first 83 elements of the periodic table. We apply sequential computational filters based on structural feasibility, thermodynamic stability, and electronic properties to reduce the search space. We also employ a statistical machine learning model based on experimentally known and unknown compositions to identify predicted synthesisable compounds.[2] A key challenge when studying ion diffusion properties of halide spinels is how to account for the strong influence of cation disordering within the crystal structure. To address this question, we identify the preferred degree of disorder by calculating the equilibrium cation inversion parameter (x_{eq})[3] and generating representative disordered structures using the special quasirandom structure (SQS)[4] method. By employing the machine learning interatomic potential MACE-MH-1 foundation model,[4] we accelerate molecular dynamics simulations by an order of magnitude, enabling the efficient calculation of diffusion coefficients. Overall, we show a robust and scalable data-driven workflow for the study of disordered functional materials, which will accelerate the discovery of high-performance candidates for next-generation solid-state batteries.

References

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Figures

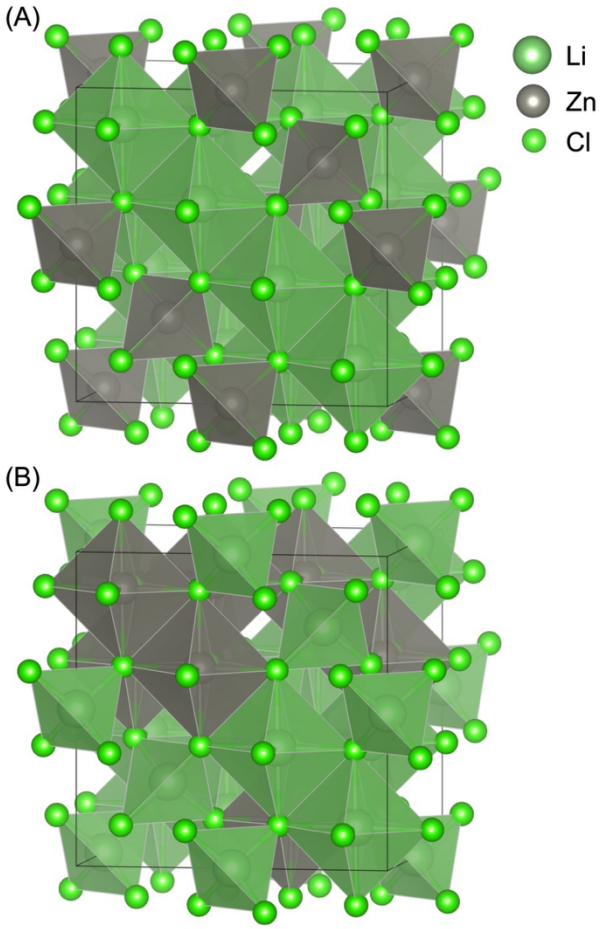

Figure 1. Crystal structures of $ZnLi_2Cl_4$ in (A) normal and (B) inverse configurations where all Zn and half Li atoms exchange their positions.