Discovering oxide solid electrolytes through topology-constrained crystal structure prediction

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Oxide superionic conductors have garnered significant attention due to their high stability compared to other material groups. However, the number of known oxide superionic conductors in existing databases remains limited. Interestingly, most known oxide solid electrolytes exhibit a unique structural characteristic: their backbone framework follows a corner-sharing topology. Building on this insight, Jun et al. identified several new oxide superionic conductors by exploring Li-containing oxide materials with corner-sharing frameworks in the Materials Project database [1]. However, the discovered candidates exhibit lower ionic conductivity than other known oxide superionic conductors. This underscores the need to discover new oxide superionic conductors that are not yet present in existing databases.

To identify materials in unexplored spaces, crystal structure prediction (CSP), which generate atomic structures for given compositions, are essential. Among these techniques, the genetic algorithm is widely used for structural optimization, typically involving energy evaluations and structural refinements via DFT calculations. Previously, we developed a method that combines GA with machine-learning potentials (MLPs), achieving orders-of-magnitude faster predictions with significantly higher accuracy compared to conventional approaches such as USPEX [2]. We confirmed the reliability of this method for ternary materials containing up to 50 atoms per unit cell. However, oxide solid electrolytes are typically quaternary compounds with unit cells containing ~100 atoms, making their prediction infeasible using conventional CSP methods.

In this study, we developed a CSP algorithm that leverages the inherent corner-sharing framework of solid oxide electrolytes. We validated the method by applying it to known oxide structures and found that it successfully identifies quaternary corner-sharing crystal structures with up to 160 atoms per unit cell. Using this approach, we explored common-element compositions of oxide solid superionic conductors and identified several promising candidates with novel structural framework. Additionally, we uncovered a design principle for oxide superionic conductors and proposed a full quaternary compositional space that may yield high-performance materials.

References

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