

Screening Energetically Stable Structures in Solid-State Ionics Applications

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Probabilistic materials screening holds significant potential for rational design of innovative energy storage devices provided that a screening methodology relies on efficient simulation methods augmented by advanced mathematical techniques. Here, we focus on the computational design of solid electrolytes with particular attention paid to garnet-type electrolyte $\text{Li}_7\text{La}_3\text{Zr}_2\text{O}_{12}$ (LLZO), which is of great interest for all-solid-state Li-ion batteries due to improved energy density and safety over traditional liquid electrolytes [1]. Its performance is stabilized at room temperature by aliovalent substitution of Li^+ by $\text{Al}^{3+}/\text{Ga}^{3+}$ or Zr^{4+} by Ta^{5+} [2-4]. The high computational cost required for the selection of adequate structures prior to large-scale atomistic simulations can be reduced by using efficient optimization techniques for screening energetically stable structures from a pool of LLZO garnets with Li/Zr vacancies and Al/Ga/Ta substitutions. With this purpose, we propose a new algorithm, Search for Optimal Structure (SOS), that converts a high-dimensional optimization problem into a reduced discrete optimization model with the pre-computed component of a score function for fast calculations (see workflow in Figure 1). The SOS algorithm has been implemented in Julia and is intended for efficient search of configuration space using customizable classic energy models and meta-heuristic optimization strategies. SOS combines speed and accuracy while remaining lightweight and easy to integrate into existing workflows. It scales well to large and compositionally complex systems, supports a wide range of energy models—from simple empirical rules to high-fidelity surrogates—and is compatible with any meta-heuristic solver. By not relying on DFT, SOS offers a fast and adaptable framework that can be extended to diverse materials and applications.

Comparison of our approach with the methods of similar scope demonstrates a significant improvement in performance (up to several orders of magnitude faster), accuracy (at least twice more accurate over the random search algorithm, see Figure 2) and flexibility.

References

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Figures

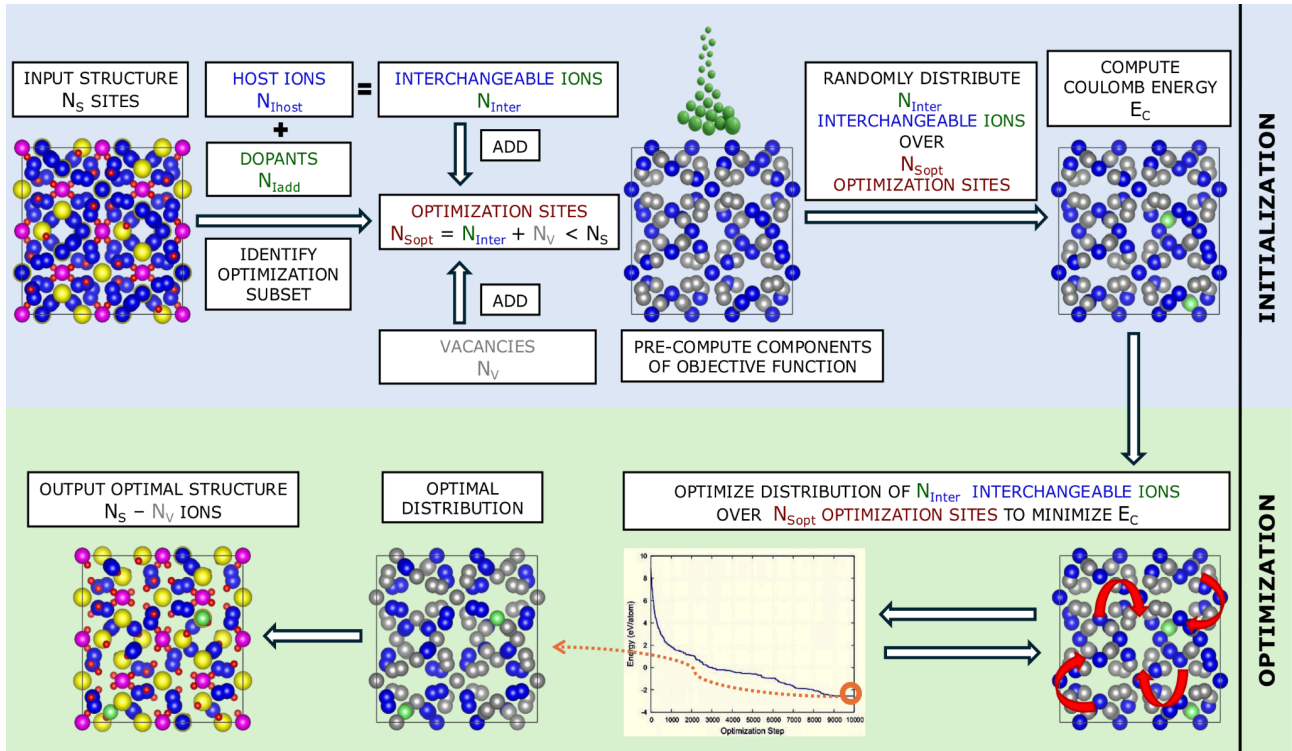


Figure 1. The SOS algorithm workflow. The first row (blue) illustrates the initialization steps, including the identification of optimization sites, pre-computation of interaction energy components, and generation of an initial random distribution of interchangeable ions. The second row (green) depicts the optimization stage, where the distribution of ions is optimized and the final atomic structure is generated.

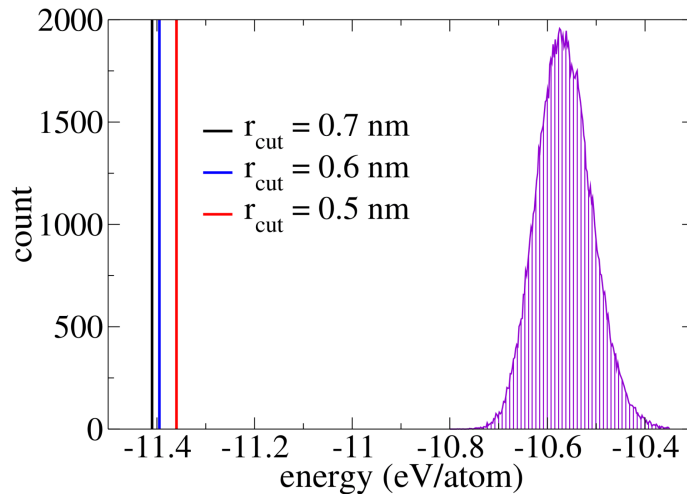


Figure 2. $3 \times 3 \times 3$ Ga^{3+} -doped LLZO supercell (5076 atoms) with 54 Ga^{3+} dopants. Energy histogram computed with the Buckingham force field from random structure generation (10^5 structures). Vertical lines indicate the lowest-energy structures identified in three independent SOS runs with interaction energy cutoff radius set to 0.7, 0.6, and 0.5 nm.