# Topological Analysis of Machine-Learned Interatomic Potential for Solid Electrolytes Discovery

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Developing next-generation energy storage technologies is vital for widespread renewable energy adoption, improved transportation, grid stability, and greenhouse gas emission reduction. In this pursuit, the quest for solid electrolytes offers promising avenues for safer, more stable batteries with higher energy density [1]. Ionic conductivity prediction for materials often relies on resource-intensive ab initio computational methods such as the Nudged Elastic Band (NEB) and Molecular Dynamics (MD) [2, 3, 4]. By leveraging the theoretical understanding of ionic motion in solids, one may extract ionic mobility information from the interatomic potential (IAP) [5], which still relies on computationally demanding Density Functional Theory (DFT) calculations.

Machine learning (ML) may offer a solution to computational limitations in material science. By analyzing large datasets, efficient ML models can be created, reducing computational costs for predicting material properties. Given that labeled ionic conductivity data for solids is scarce, one may employ semi-supervised learning, as has been done in [6] with hierarchical clustering on various structure descriptor representations. Alternatively, ML, and especially Graph Neural Networks (GNN), may be employed to bypass the expensive DFT calculations and efficiently predict IAP [7]. Trained on an extensive dataset, like structure relaxation trajectories from the Materials Project database [8], such efficient ML IAP models may be utilized in NEB and MD calculations in place of computationally demanding DFT. An issue with this approach, however, is that both NEB and MD may necessitate extrapolating the ML IAP significantly beyond the original training domain, which could result in high errors. Avoiding these errors would require utilizing active learning [9], albeit at the expense of sacrificing some of the improvement in the computational demand.

In this work, we propose an alternative approach where we study the topology of the ML IAP by only perturbing a single atom at a time within a structure. By doing so, we minimize the need for extensive extrapolation of the model beyond its training domain, thus facilitating more accurate predictions with reduced error. With this approach, we build physically motivated heuristic descriptors and evaluate their performance on the available labeled computational [2] and experimental [6] data. Our choice of ML IAP is the M3GNet model [7] trained on the Materials Project data [8], although the proposed method could be used with any IAP.

Within our method, each structure is considered as having a single mobile ion per unit cell and a frozen framework formed by the remaining atoms. A PES scan is performed by placing the mobile ion at a set of locations on a regular 3d grid and evaluating the potential energy for the resulting structure. The grid spans over the entire unit cell with axes parallel to the lattice vectors. An example result of such a scan is shown in Fig. 1, top. Assuming connectivity of nearby grid nodes, each location is then marked with the minimal energy required for the mobile ion to get there from its equilibrium position, thus producing the minimal energy level map, as depicted in Fig. 1, bottom.

The obtained minimal energy level maps can be used to extract valuable topological information about the system. When constructed for a 2x2x2 supercell with the mobile ion equilibrium positioned at its center, the lowest level map value at the supercell's surface should indicate the lowest barrier for ion percolating into an adjacent cell (Minimal Percolation Energy, MPE). While the frozen framework approximation should result in the overestimated MPE, we assume that the introduced error does not significantly change the relative ordering of different materials with respect to the obtained values. Another useful value that can be extracted from the minimal energy level map is the fractional volume accessible to the ion under a given energy threshold. We denote this value as the Free Volume (FV) and assume that higher values of FV should be associated with higher ion mobility. We test this assumption on the extensive ab initio MD dataset [2], and observe good correspondence between the FV and the values of the diffusion coefficients extracted from ab initio MD simulations at $T = 1000$ K, as shown in Fig. 2. Additionally, we perform a similar validation on experimental data collected from various sources by [6]. Figure 3 shows the relation between the FV and experimental conductivity at room temperature. While the correlation is less pronounced than in the simulated dataset, we can still observe that higher FV values tend to correspond to higher average conductivity, and vice versa. These findings highlight the promising potential of our proposed topological descriptors in identifying new solid electrolytes.

In this study, we have introduced a novel approach by proposing topological features extracted from the IAP, leveraging the speed advantages of ML IAP. The proposed features demonstrate reasonable performance in predicting conductivity when evaluated against existing data. Looking ahead, we anticipate conducting ab initio validation of structures identified by our proposed features from the Materials Project dataset, ultimately contributing to the development of more efficient solid electrolytes for advanced energy storage applications.

## References


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## Figures

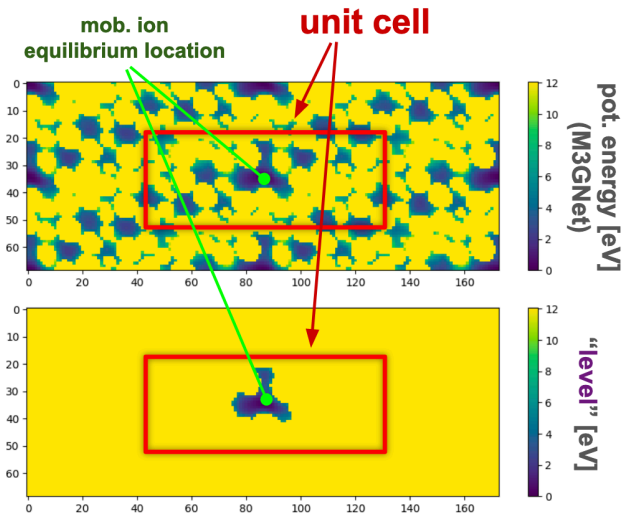


Figure 1: Potential energy surface for $Li_5TiN_3$ (mp-686129), as predicted by M3GNet [7] (top), and corresponding minimal energy level map (bottom).

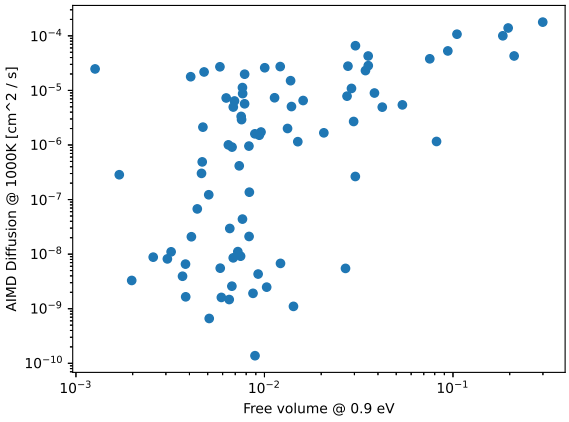


Figure 2: Free volume with 0.9 eV threshold versus AIMD diffusion coefficient at 1000K [2]

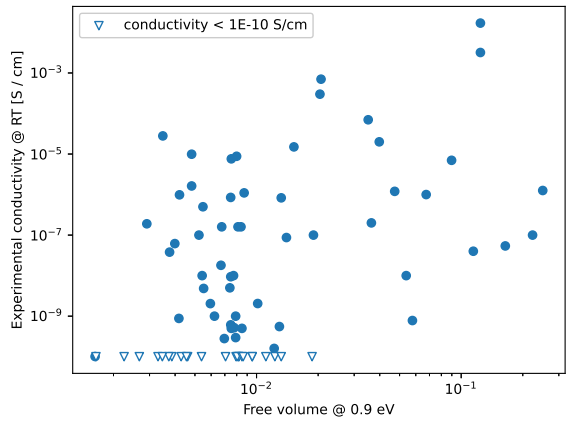


Figure 3: Free volume with 0.9 eV threshold versus experimental conductivity at room temperature [6]