Element-Specific Neighbor Lists for Periodic Graph Neural Networks

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Abstract

In recent years, periodic message-passing graph neural networks (MPGNN) for materials discovery have accelerated atomistic simulations in complex systems. They usually use the atomic decomposition of total properties to truncate the interatomic correlations to a local environment. One key component of MPGNNs is the neighbor list, which defines the interactions between atoms in a material. However, traditional fixed cutoff for neighbor lists consider each element similarly, which leads to a huge neighbor list because of the inclusion of redundant neighbors, which becomes a bottleneck as working on huge crystal graphs is computationally expensive. Furthermore, if we look deeper into the periodic table, the elements interact because of their electronic configuration and size. If we consider a hydrogen atom and a transition metal (TM), the bonding and the interaction with their local environment will be different. Hence, in this study, we propose a pairwise element-specific neighbor list that dynamically adjusts the neighbor list size based on the type and coordination environment of each atom in the material. By this way we can adjust the range of interaction with respect to the elements, which will reduce the computational cost and will help in efficient learning.

1. Introduction

Recently, machine learning techniques have become prevalent tools in the materials science community for atomistic simulation and modeling, enabling expedited simulations with accuracy comparable to ab initio methods utilizing materials databases. Conventional machine learning techniques have been used for numerous property prediction tasks, including formation energy, band gap, etc. Periodic message-passing graph neural networks (MPGNN) for materials discovery have achieved great success in accelerating atomistic simulations in complicated systems in recent years.[1, 2, 3, 4] GNN excels in representing the topological structures of materials through graphs. Now, GNN has attained exceptionally high accuracy in predicting material properties for handling very complex chemical systems and also running simulations like molecular dynamics for big crystal systems (containing a few hundred atoms or even thousands), which is impossible by first-principle calculations.

2. Related Works

Many work has been done with fixed cutoff neighbor search. [5, 6, 7]. Some methods, such as quantized bounding volume hierarchies for neighbor search and solid angle nearest neighbor search, are used with classical potential to simulate the molecular dynamics of particles.[8, 9] Boris et. al. 2023 also consider species-dependent cutoffs for neighborhood search. In chemically diverse systems such as bio-molecules, there is significant variation in the distribution of distances between atoms and their neighbors depending on chemical species, and this has been used to reduce the number of neighbor pairs while preserving relevant interactions. [10] Norwood and Bhowmik (2023) used geometry-aware MACE with attention and layer-specific universal cutoff for long-range interaction. [11]

3. Experimental Setup

3.1 Data:

The data used in this work is a cathode materials database for Na-polyanion cathodes. We only consider sodium alluadites [Na_2TMSiO_4 and $Na_{2.56}TM_{1.72}(SO_4)_3$; where TM: Fe, Mn, Co, Ni]. The data contains sampled ground state structures of the sodium alludites with different concentrations of sodium and ab initio molecular dynamics snapshots. [12, 13] All the structures in the data are the supercell structures. This data set is good for our experiment as it has a very diverse local environment and many chemical species. The distribution of energy and maximum force are shown in Fig. 1.

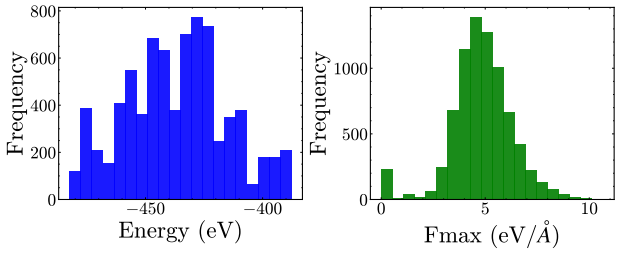

Fig. 1: The distribution of total energy (left) and maximum force (right) in the dataset.

3.2 Model and approach

We have considered MACE architecture for this experiment, which is proven to be one of the most popular MPGNN architectures among the community for machine-learned atomistic simulations. [14] As a global experimental setup, we consider 3 layers

of mace with 32 channels and 3-body interactions up to spherical harmonics l=2. A global cutoff of 5 Å is used, but the edge indices are further calculated with respect to the covalent radius of the elements. The model is trained for 500 epochs. The pairwise cutoff is given in the expression 1, and the element-specific cutoff is given in expression 2 below.

$$r_c^{ij} = r_i + r_j + buffer \tag{1}$$

$$r_c^{i} = \alpha r_i + buffer \tag{2}$$

Where, r_c^{ij} is the pairwise cutoff for elements i and j having covalent radii r_i and r_j respectively. r_c^i is the cutoff radius for element i, α is the multiplication factor, and *buffer* is for smooth cutoff interactions.

In order to investigate the effect of species-dependent neighbor lists, we changed the amount of buffer in eq. 1 from 1.0 Å to 3.0 Å for our first three experiments reported in Table. 1

4. Results

Table 1: The table showing validation RMSE errors in energy (meV/atom) and forces (meV/ Å and ≈ t/epoch is in sec.

	Avg. NL	ESNL	E_{RMSE}	E_{RMSE}	t/epoch
Exp. 1	6.0	+	5.9	175.3	30
Exp. 2	16.8	+	4.8	125.3	54
Exp. 3	31.2	+	**4.1**	**114.3**	100
Exp. 4	38.9		4.2	122.3	140

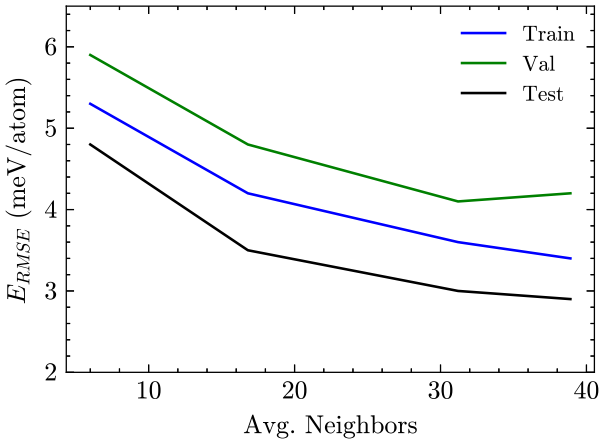

Fig. 2: Trend of train, validation and test RMSE in energy with respect to average number of neighbors.

The training summary is reported in the table: 1. We can clearly see the improvement of model accuracy depends on the neighbor list, i.e., on the complexity of the local environment. But when we used the species-based neighbor list, we considered the way elements interact with their local environment. Hence, we only consider necessary neighbors with respect to the chemical species. We found that for an average neighbors of 31.2, the force and energy root mean square error (RMSE) is the least. But for a fixed cutoff, including a larger number of neighbors for the crystal graph is not improving any accuracy for the same setting of the experiment, but the computational cost increases with a huge input graph because of a larger average number of neighbors. Fig. 2 and fig. 3 show the trend of train, validation, and test errors for energy and force, respectively, with respect to the average number of neighbors.

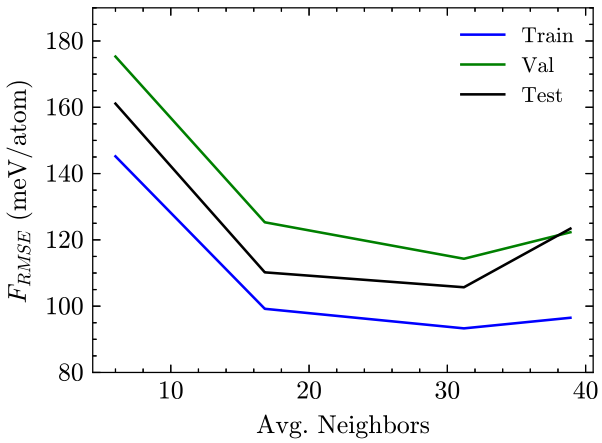

Fig. 3: Trend of train, validation and test RMSE in force with respect to average number of neighbors.

5. Conclusion

In this study, we consider a very simple expression for a species-dependent neighbor list for periodic MPGNN. Hence, the approach can consider a chemistry-informed neighbor list without any redundant neighbors. The training errors agree with our intuition that unnecessary neighbors are nothing but redundant information for training, hence not helping in the improvement of model accuracy. This redundant neighbor also results in large crystal graphs, which means we need more computation time for training. However, we can consider more advanced neighbor searches, like solid angle-based searches or Voronoi-based searches, to build a more efficient neighbor list, which can be explored in future work, and also other hyperparameters and model complexity can be addressed along with all this chemistry-informed neighbor list search.

Acknowledgments

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