Applying Transfer Learning to dGNN Models for Defect Formation Energy Predictions

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Abstract

We aim to enhance Crystal Graph Convolutional Neural Network (CGCNN) models for predicting defect formation energies in crystal structures. Our modifications focus on improving accuracy, reducing overfitting, and enhancing interpretability. These advancements support efficient material property predictions, and reduce the need for Density Functional Theorem calculations.

Previous Work

CGCNN excels at predicting properties for pristine crystals. Adapting it to defect structures requires adjustments due to limited defect data. We employ transfer learning, leveraging the pristine crystal database to enhance defect energy predictions.

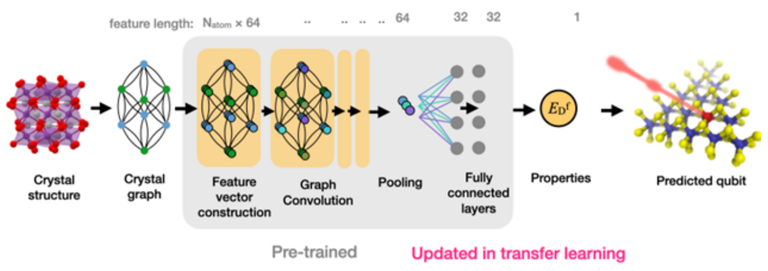

Figure 1: A crystal graph-based approach initially trained on pristine structures. Transfer learning refines the model for defect formation energies.

Model	MAE
> From scratch	0.382
Transfer learning (embedding + conv all frozen, 1 fc free)	0.41
Transfer learning + 1CNN	0.45
Transfer learning + 1 fc frozen + 1 additional fc	No Improv.
> Transfer learning + 1 fc free + 1 additional fc + 1CNN	0.378
Transfer learning + embedding free + fine-tuning at end	0.26
Transfer learning + embedding free	0.30
Transfer learning + embedding free + 1 additional fc	0.32
Fine-tuning +Adam optimizer + 1 fc	0.240
> Fine-tuning +Adam optimizer + 1 CNN (matching)	0.243
Fine-tuning (everything free)	0.249
Fine-tuning + adding 1 fc	0.277

Figure 2: Previous work did not improve results with transfer learning. Best outcomes were with fully connected layers and adaptive moment estimation. (Red text = fine-tuning)

dGNN Model

The defect Graph Neural Network (dGNN), enhances prediction accuracy of defect formation energies. The dGNN incorporates elements from the original CGCNN model, optimized for defect-specific predictions.

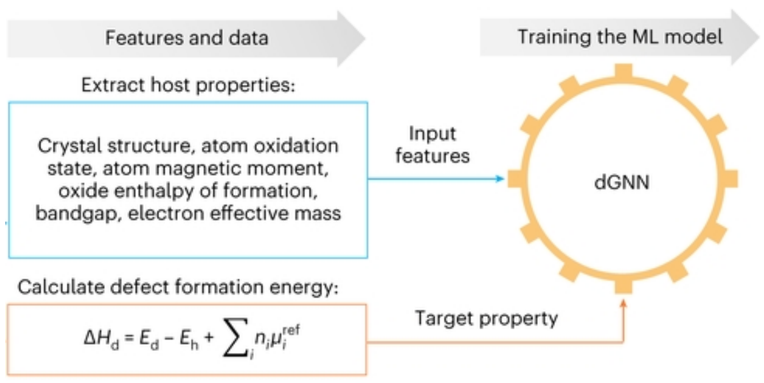

Figure 3: Workflow of the dGNN model, embedding specific features into the ML model to predict defect formation energy. It utilizes features from both the pristine crystal structure and specific crystal defects.

Key differences between the original CGCNN model and the newer dGNN model include:

- Pooling operation (node level)
- Vector features (oxidation states)
- Radial basis set expansion
- Larger database (≈ 1500 vs. ≈ 800)

Potential Improvements

Advised Experts:
Experts specialize in various crystal structures, combining insights for precise defect energy predictions.

Transfer Learning:
Pre-train on a large crystal structure dataset for pristine crystal formation energy and fine-tune on a smaller defect formation energy dataset for enhanced accuracy.

Cross Validation Functions:
Cross-validation reduces the high variance that results from the noise present in small datasets, improving model reliability.

Pooling Functions:
Different pooling functions can be experimented with. The dGNN uses a defect specific pooling function with node-level pooling that singles out the defect site. Other pooling functions that can be applied are dropout-style pooling and max pooling.

dGNN Implementation:
Using the same improvements used in the dGNN model for predicting defect formation energies such as improved dataset size, vector features, and gaussian filters.

Results and Conclusions

Cross-validation has improved CGCNN models' performance in predicting defect formation energies by increasing robustness and reducing overfitting. Transfer Learning did not see improved results while using dGNN, but average pooling yields the best results for pretraining the model on pristine crystal formation energy.

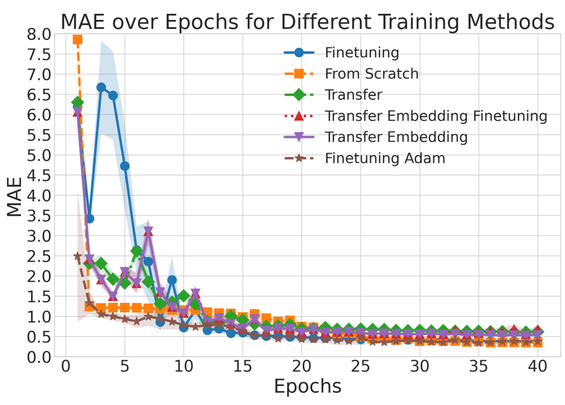

Figure 4: MAE of the test set at each training epoch for various models. This uses the k-fold cross-validation for each model. This test was done on the CGCNN, and the best performing model included an adaptive moment estimation model coupled with transfer learning.

Cross-Validation Technique	Mean MAE	Std MAE	Min MAE	Max MAE
K-Fold	0.935	1.165	0.287	8.440
Monte Carlo	1.126	1.356	0.291	9.035
Leave-One-Out	0.729	0.638	0.002	3.325
Leave-Two-Out	0.597	0.421	0.037	2.424
Bootstrapping	1.147	1.341	0.350	9.508

Figure 5: Comparison of cross-validation methods on a single model. The most effective method is the Leave-Two-Out cross-validation method as it achieves the least amount of variance. Only a small percentage of Leave-Two-Out and Leave-One-Out CV were performed due to large computing times.

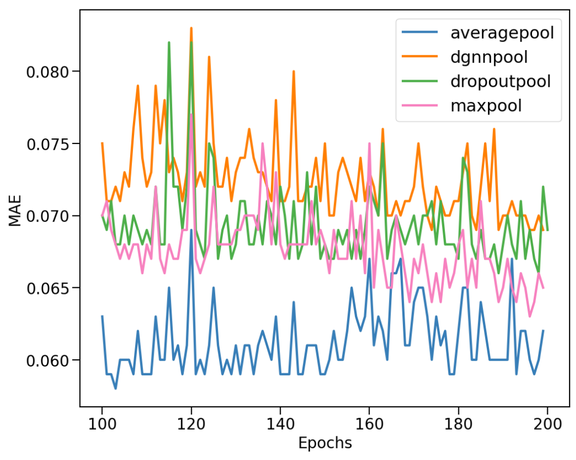

Figure 6: A comparison of different pooling functions used for pretraining pristine crystal formation energy while using dGNN. The best results yielded from using the average pooling function, and fine-tuning using the dGNN pooling.

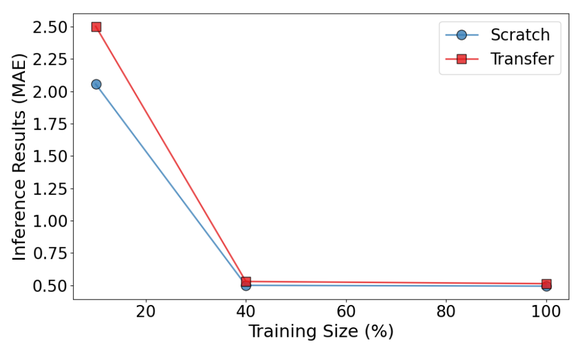

Figure 7: Comparison of using transfer learning and using a model with no transfer learning to predict defect formation energies with different levels of data availability. Inference results are plotted for 3 different percentages of data availability (10, 40, 100) and the from scratch model consistently performs better.

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Acknowledgments

We express our gratitude to the University of North Texas for essential resources and support. Additionally, we thank the National Science Foundation for generous funding (Award #2244259, Beyond Language: Training to Create and Share Vector Embeddings) and invaluable assistance.

References

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