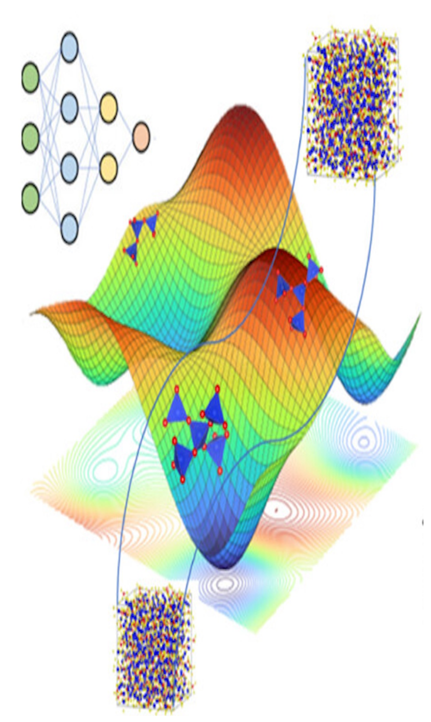
Evaluation of Foundational Machine Learned Interatomic Potentials for Migration Barrier Predictions

Achinthya Krishna Bheemaguli[1], Penghao Xiao[2], Sai Gautam Gopalakrishnan[1]

[1]Department of Materials Engineering, Indian Institute of Science, Bengaluru, 560012, Karnataka, India

[2]Department of Physics and Atmospheric Science, Dalhousie University, Halifax, Canada

INTRODUCTION



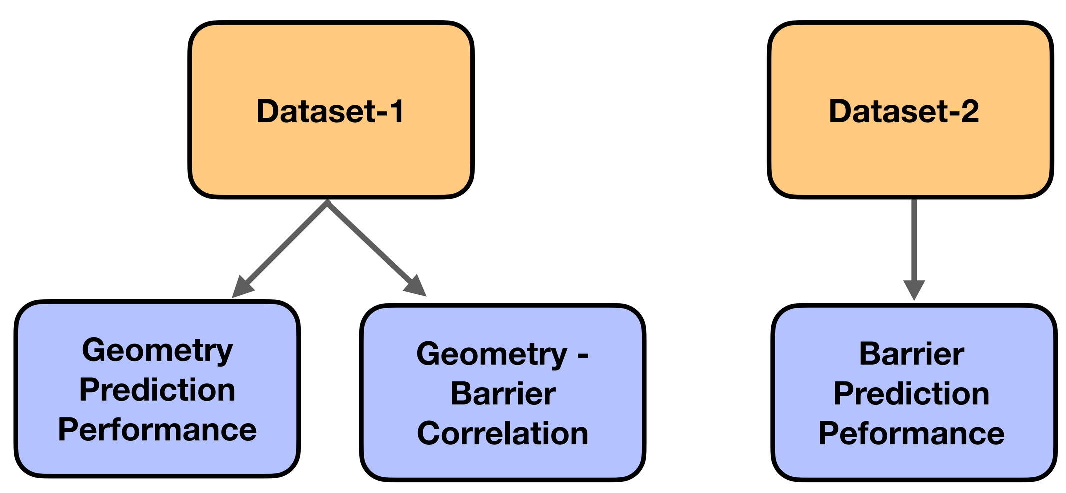
- Ionic diffusivity is exponentially proportional to ion migration barrier, E_m
- Density functional theory (**DFT**) based nudged elastic band (**NEB**)[1] calculations for E_m prediction are **computationally expensive**
- Can graph neural network (GNN) based machine learned interatomic potentials (**MLIPs**)[2] achieve quick and accurate E_m predictions, when integrated with NEB?
- Can **MLIPs** provide a superior initial guess for the minimum energy path (MEP) than linear interpolation (**LI**)?

METHODS

MLIP	Training data	Key features
MACE-MP-0	MPTrj dataset	E(3)-equivariant GNN that captures many-body interactions
SevenNet-MF-ompa	MPTrj, OMat24, sAlex	Equivariant GNN incorporating multifidelity learning with efficient parallelization
Orb-v3	MPTrj, OMat24, Alex	Roto-equivariance inducing regularized GNN with analytical energy gradients (conservative forces) and (effectively) infinite neighbors
CHGNet	MPTrj dataset	GNN including magnetic moment inputs, thus incorporating information on atomic charges
M3GNet	MPTrj dataset	Includes three-body interactions within its GNN

[3]

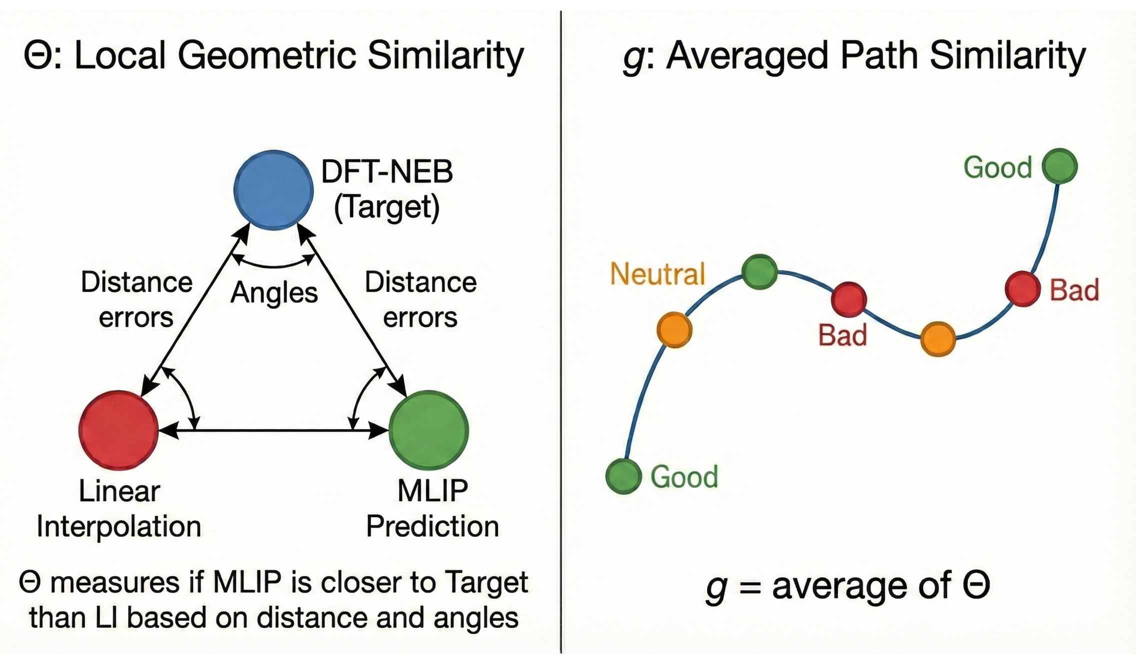
Workflow



- All MLIP-NEB[4] calculations done with atomic simulation environment (ASE[5])
- NEB Settings:
  - **7 images** for Dataset-1, **3 images** for Dataset-2
  - Initial MEP generated using **image dependent pair potential (IDPP)** technique[6]
  - Tangent and spring force defined as per **elastic band (EB)** method with full spring force
  - Spring constant of 5 eV/ Å^2, forces converged within |0.05| eV/Å

Geometry Metric

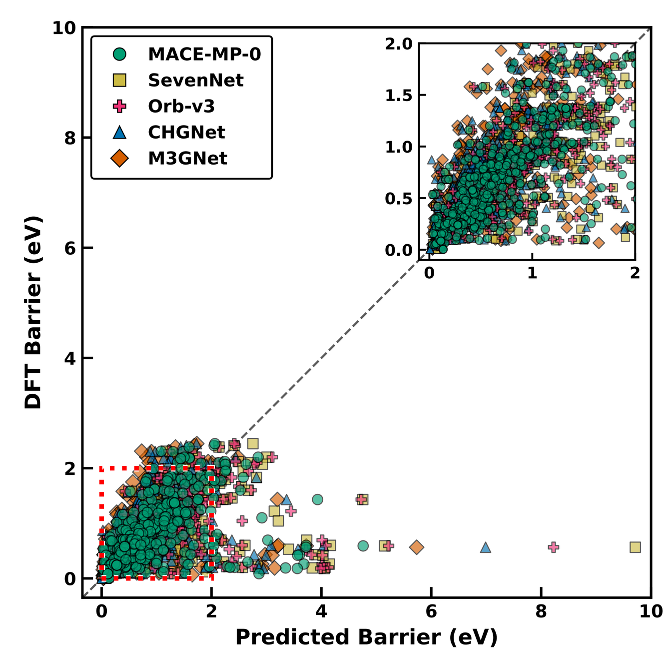
- Use θ to quantitatively assess the errors in local geometry predictions
- Identify the nearest neighbors of the migrating ion
- Calculate the absolute errors in pairwise distances
- Calculate the absolute errors in solid angles
- Calculate δ, defined as maximum value among the differences in the mean and maximum errors of distances and angles between the MLIP-NEB and LI structures
- Classify structure as 'Good (1)', 'Bad (-1)', or 'Comparable (0)' based on the value of δ



- For a given system containing i intermediate images, $g = \frac{\Sigma_i \theta}{\Sigma_i i}$

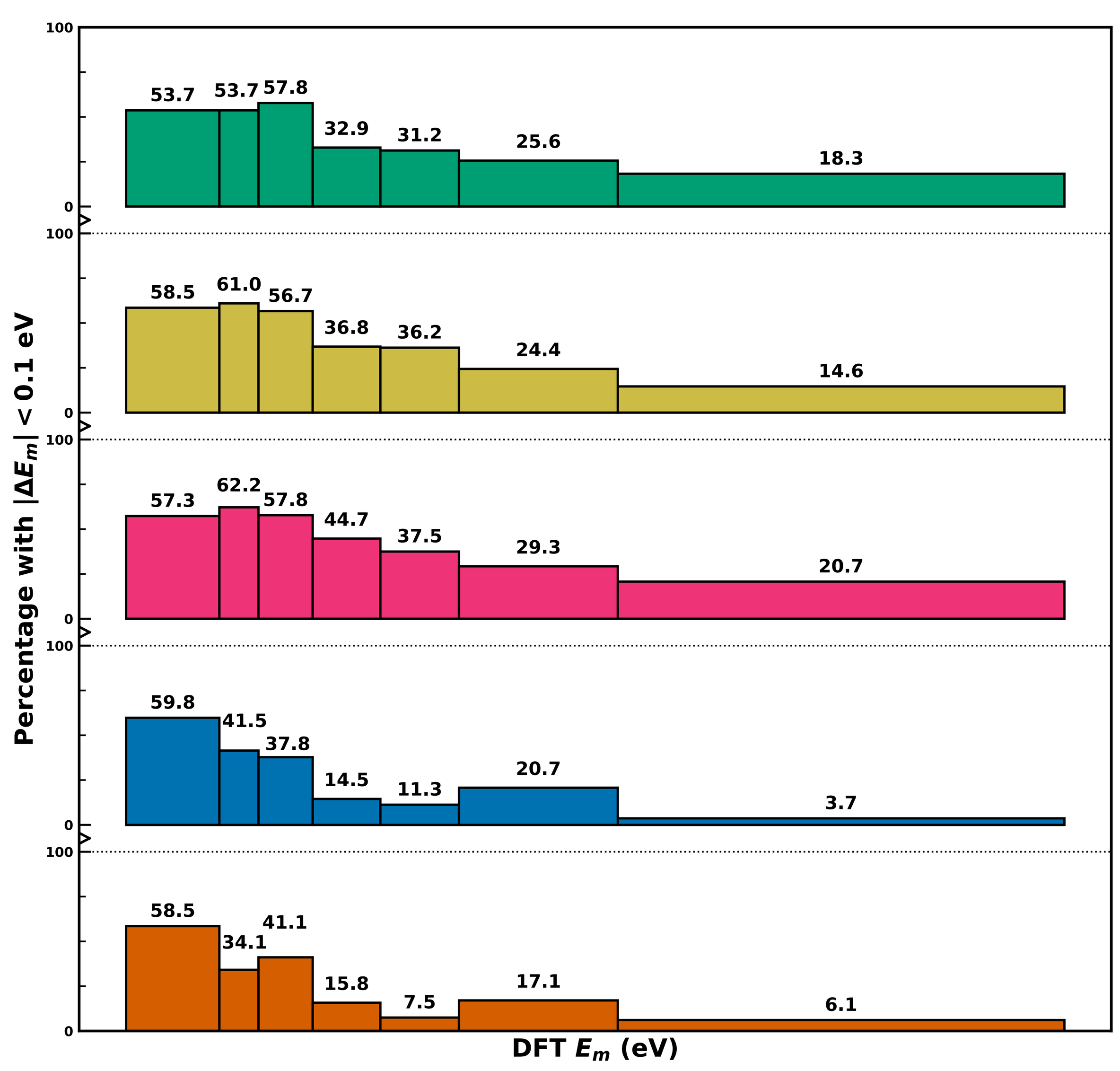
RESULTS

Barrier Prediction Performance



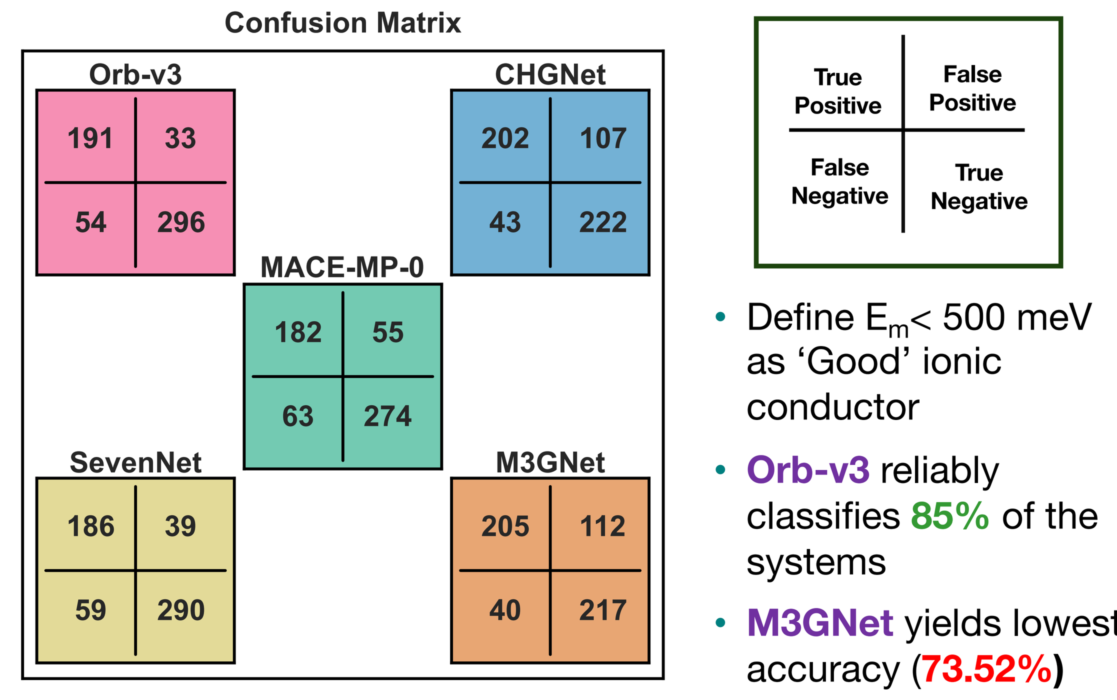
- **M3GNet** and **CHGNet** tend to underestimate the barrier heights
- Mean absolute errors (MAE) in E_m, excluding specific outliers:
  - **Orb-v3:** 0.198
  - **MACE-MP-0:** 0.202
  - **SevenNet:** 0.203
  - **CHGNet:** 0.248
  - **M3GNet:** 0.257

Predictions Over Different Barrier Ranges



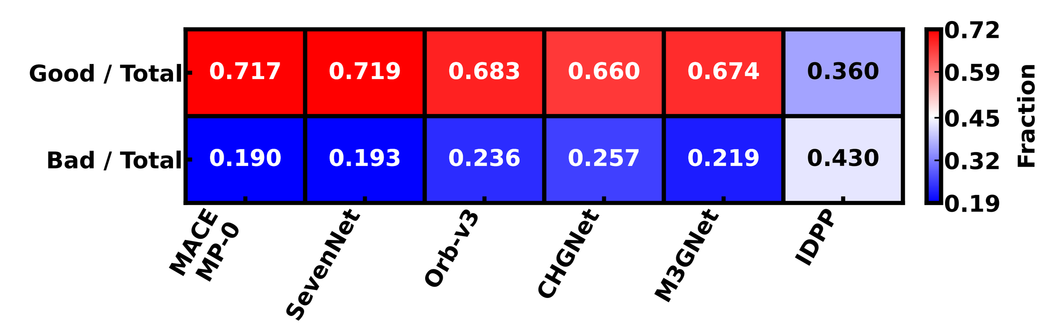
- Each bin from left (low E_m) to right (high E_m) contains equal data points
- All models struggle with high E_m predictions
- **Orb-v3** exhibits the slowest degradation
- Simpler models achieve their best performance over a narrow range of E_m

Barrier Classification Performance



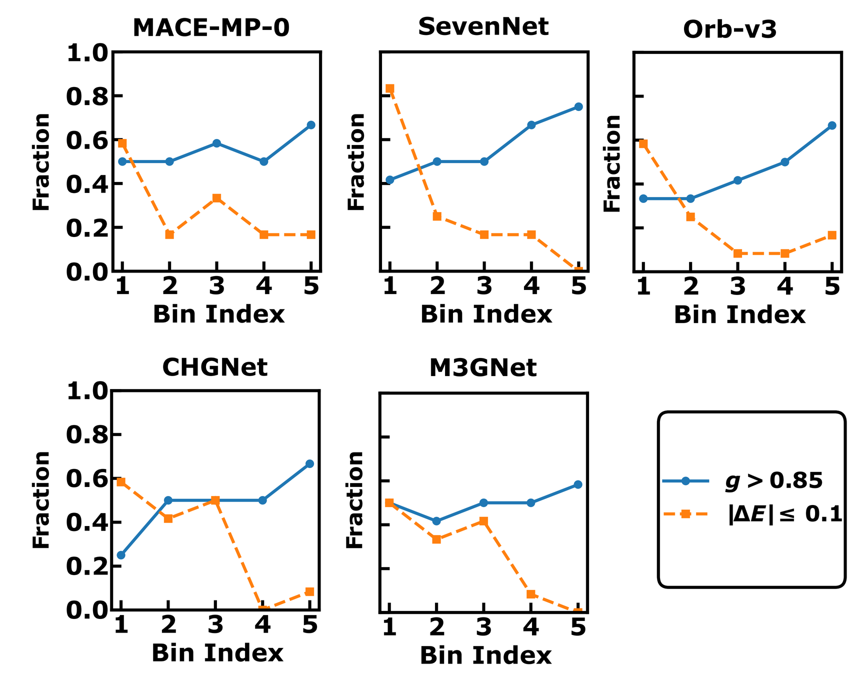
- Define E_m< 500 meV as 'Good' ionic conductor
- **Orb-v3** reliably classifies 85% of the systems
- **M3GNet** yields lowest accuracy (**73.52%**)

Geometry Prediction Performance



- All MLIPs give good geometries in **>66%** cases
- **MACE-MP-0**, and **SevenNet** exhibit best performance

Geometry-Barrier Correlation



- Low (high) bin index contains low (high) E_m systems
- We observe no evident correlation
- Good E_m prediction doesn't imply good geometry prediction, and vice versa
- Might be associated with 'flatness' of potential energy surface

CONCLUSIONS

- **MLIPs (Orb-v3)** can be used for **high-throughput screening** of ionic conductors
- **MACE-MP-0** and **Orb-v3** exhibit the lowest MAE across the entire dataset and datapoints that are not outliers, respectively
- Using MLIP-relaxed NEB images as an **initial guess** can indeed reduce the computational expense of subsequent DFT-NEB

ACKNOWLEDGMENTS

- ACENET, Digital Research Alliance of Canada
- Supercomputer Education and Research Centre, Indian Institute of Science
- Jülich Supercomputing Centre, Germany

REFERENCES

1. Henkelman et al., The Journal of chemical physics,113(22):9901–9904, 2000.
2. Choi et al., JACS Au, 5(4):1499–1518, 2025.
3. Devi et al., Scientific Data, 2025.
4. Riebesell et al., Nature Machine Intelligence, 7(6):836–847, 2025.
5. Hjorth et al., Journal of Physics: Condensed Matter, 29(27):273002, 2017.
6. Smidstrup et al., The Journal of chemical physics, 140(21), 2014.

Bheemaguli, Xiao, and Sai Gautam, "Evaluation of Foundational Machine Learned Interatomic Potentials for Migration Barrier Predictions (under review), arXiv 2512.03642 (2025).

International Meeting of the Battery Research Society (IMBRS 25), Bangalore, India

Achinthya K Bheemaguli
PhD Student
Advisor: Prof. Sai Gautam Gopalakrishnan
Department of Materials Engineering
Indian Institute of Science
Email: achinthyab@iisc.ac.in