Evaluation of foundational machine learned interatomic potentials for migration barrier predictions

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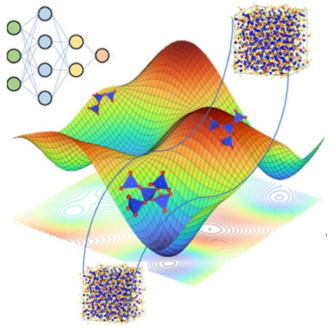
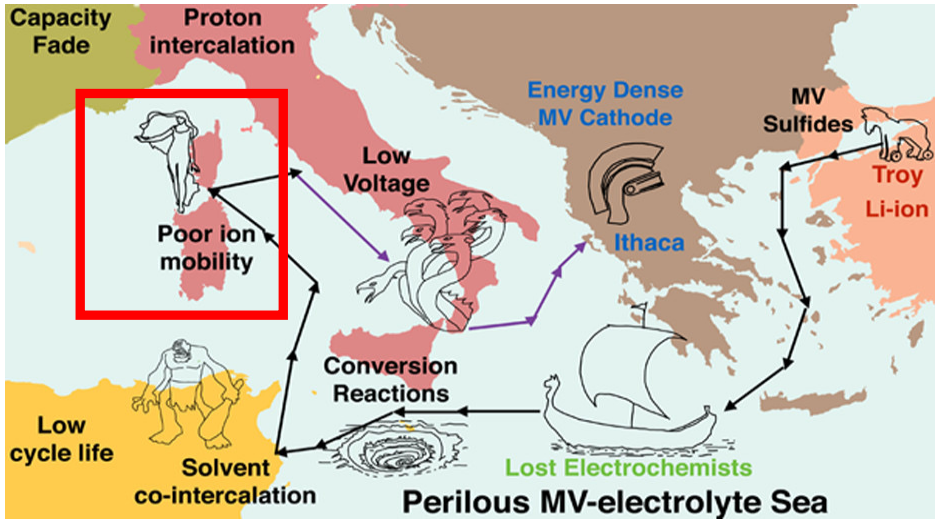
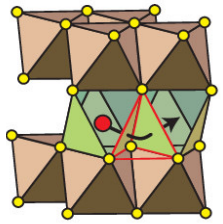
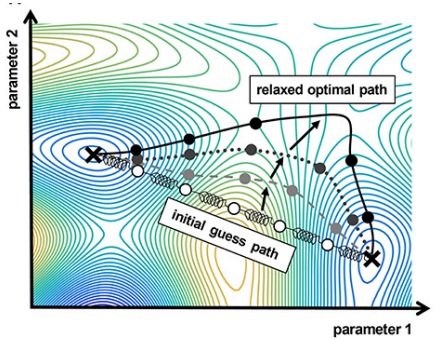
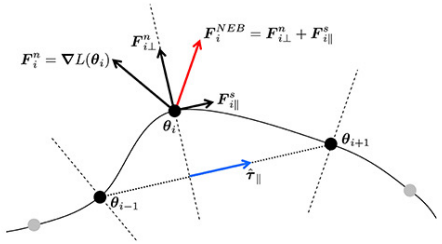


Migration barriers: A central challenge for multivalent cathodes









- Multivalent chemistries (e.g. Mg, Ca-ion etc) offer high volumetric energy density, less toxic and earth earth-abundant
- Ionic Diffusivity (D) within the bulk is a major factor in the rate performance of intercalation electrodes

$$D = D_0 \exp\left(\frac{-Em}{k_B T}\right)$$

D_0: Diffusivity pre-factor
k_B: Boltzmann constant
T: Temperature
E_m: Migration barrier

- Computational technique to predict E_m: ab initio molecular dynamics, nudged elastic band (NEB) with density functional theory (DFT)
- DFT-NEB is computationally expensive and often exhibits convergence difficulties

van der Ven et al. **Ann. Rev. Mater. Res.** 48, 27-55 (2018)
Canepa et al. **Chemical reviews.** 117(5):4287–4341 (2017)

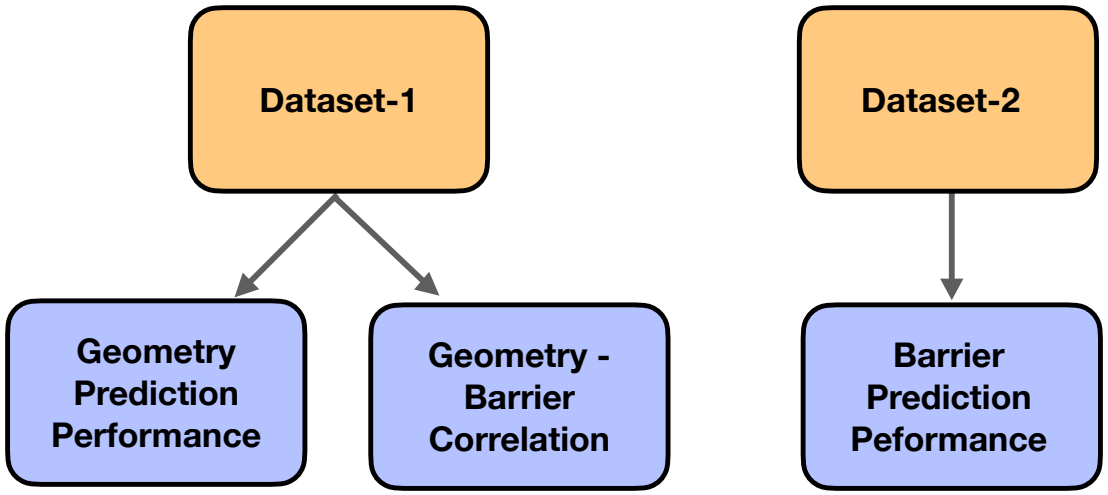
Questions we ask

How accurate are the MLIP-NEB predicted E_m? **(Barrier prediction performance)**

Can MLIP-NEB relaxed image geometries be a better initial guess for subsequent DFT-NEB? **(Geometry prediction performance)**

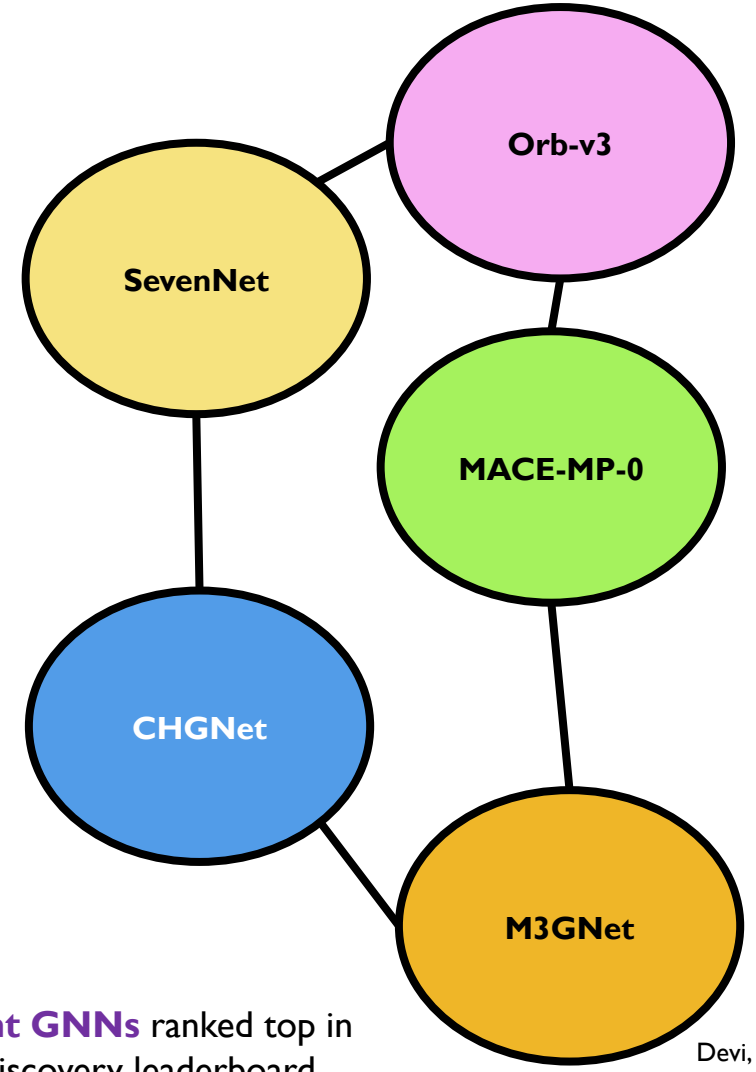
Are geometry and E_m predictions correlated? **(Geometry-barrier correlation)**

Datasets and ground-truth



- **Dataset-1**: 60 DFT-NEB E_m of GGA level accuracy (Includes intermediate relaxed images)
- **Dataset-2** : 574 DFT-NEB E_m of GGA level accuracy compiled from literature
- Both the datasets span over wide variety of battery-relevant chemistries and structures

MLIPs



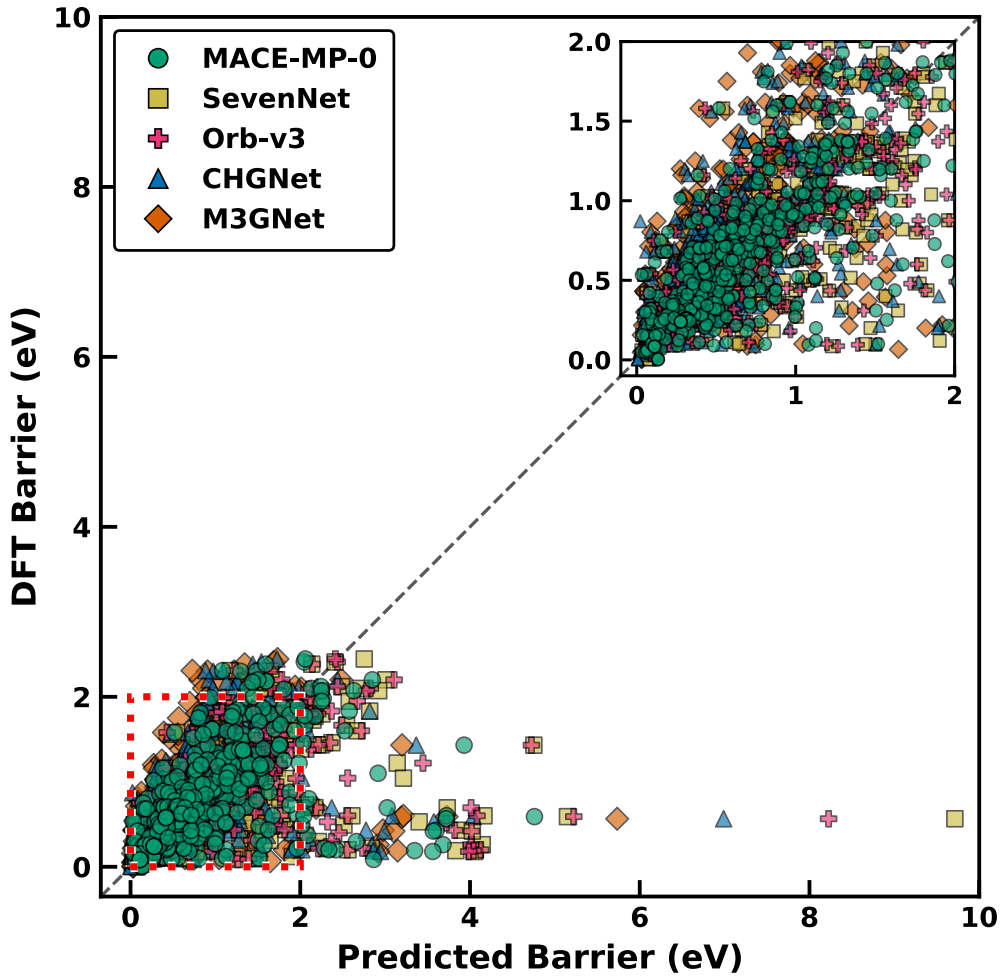
- **Equivariant GNNs** ranked top in matbench discovery leaderboard

NEB parameters

- Image Dependent Pair Potential (IDPP) generated images connected via k = 5 eV/Å^2 spring
- Tangent defined as per Elastic Band (EB) implementation
- Number of images identical to ground-truth
- All calculations run using atomic simulation environment (ASE)

Devi, Reshma et al. **Scientific Data**. 12.1 (2025): 1922

Barrier prediction performance

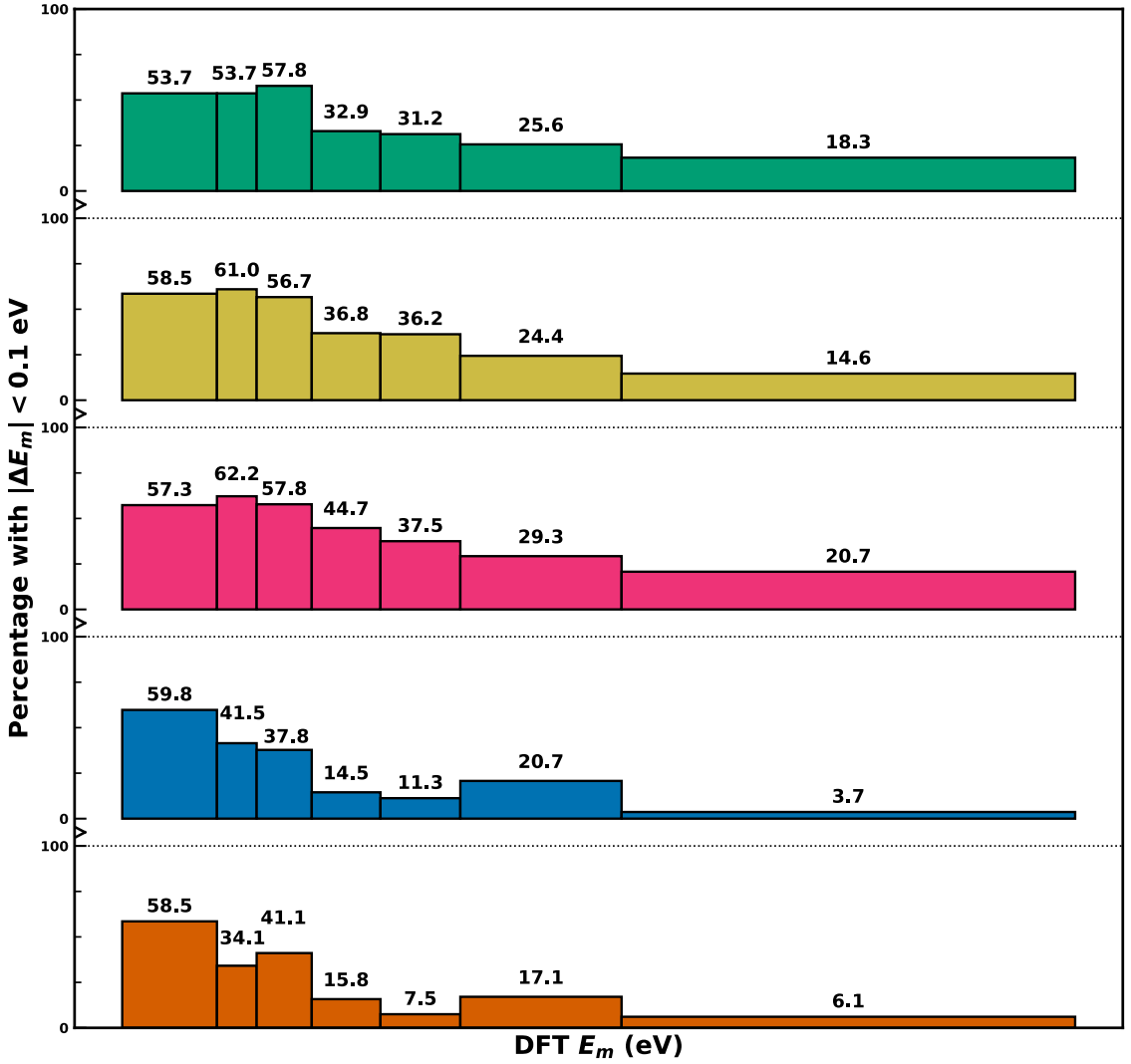


- MACE-MP-0 (M3GNet) shows the lowest (highest) mean absolute error (MAE) of 0.31 eV (0.349 eV) while considering the entire dataset
- MAE excluding the common outliers (> 1eV error) shows the same trend with MACE-MP-0 (M3GNet) exhibiting the lowest (highest) 0.239 eV (0.290 eV)
- Excluding outliers specific to each model:

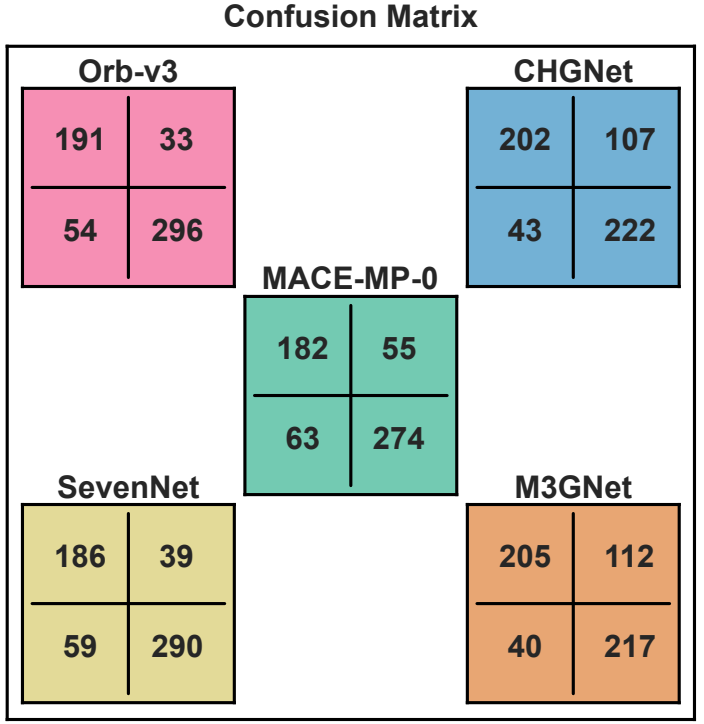
Orb-v3: 0.198 eV
MACE-MP-0: 0.202 eV
SevenNet: 0.203 eV
CHGNet: 0.248 eV
M3GNet: 0.257 eV

- DFT-NEB usually carry a **~60 meV error** in their predictions
- 60 meV change of E_m corresponds to an order of magnitude change in D at 298K
- We define **0.1 eV** as **'acceptable error'**

‘Sweet spot’ in E_m range? Classify good/bad conductors?

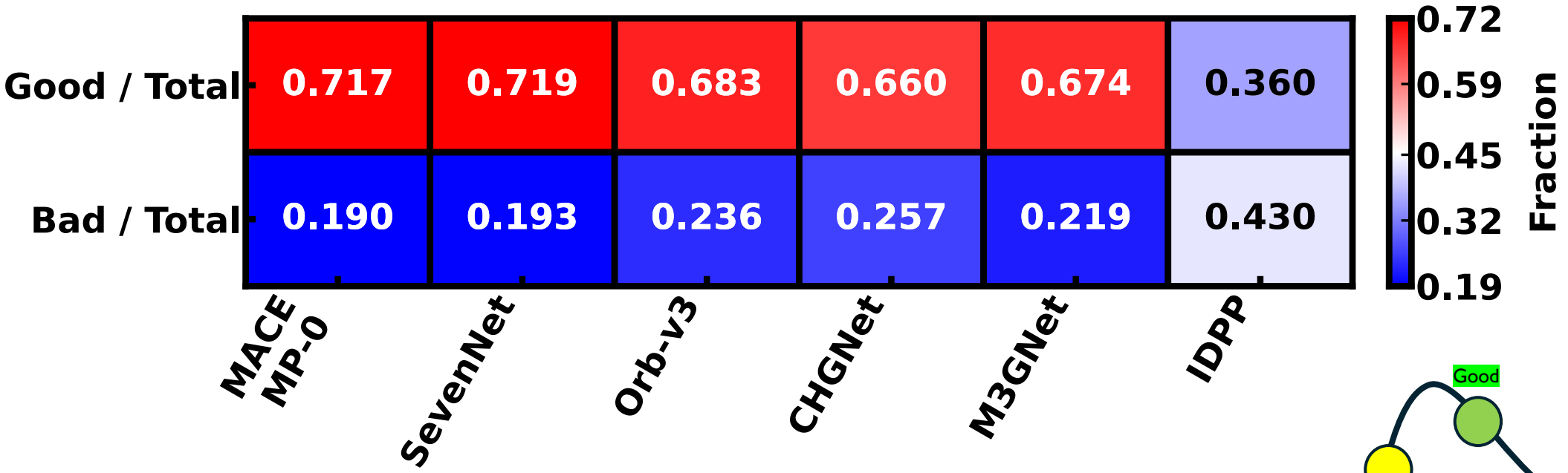


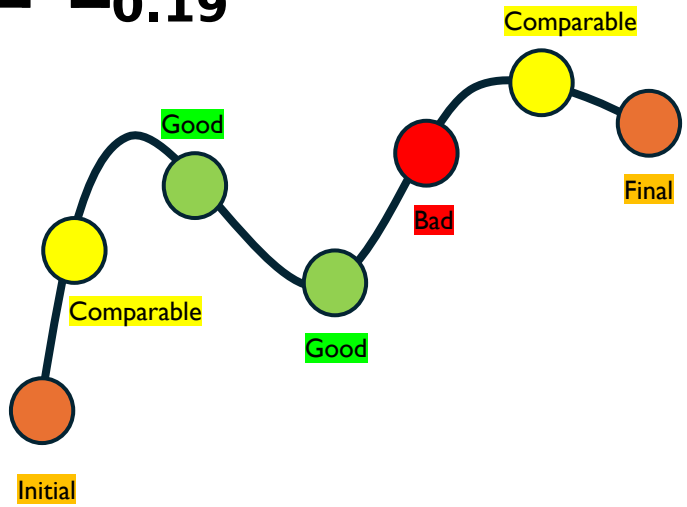
- All models struggle in predictions for systems with high E_m
- Orb-v3 exhibits the slowest degradation
- ‘Simpler’ models show peak performance in a narrow range of low E_m values



- Define < 500 meV as ‘good’ ionic conductor
- Prediction considered as TP (TN) if both the DFT-computed and model-predicted E_m are less than (greater than or equal to) 500 meV
- Orb-v3 reliably classifies 85% of the systems

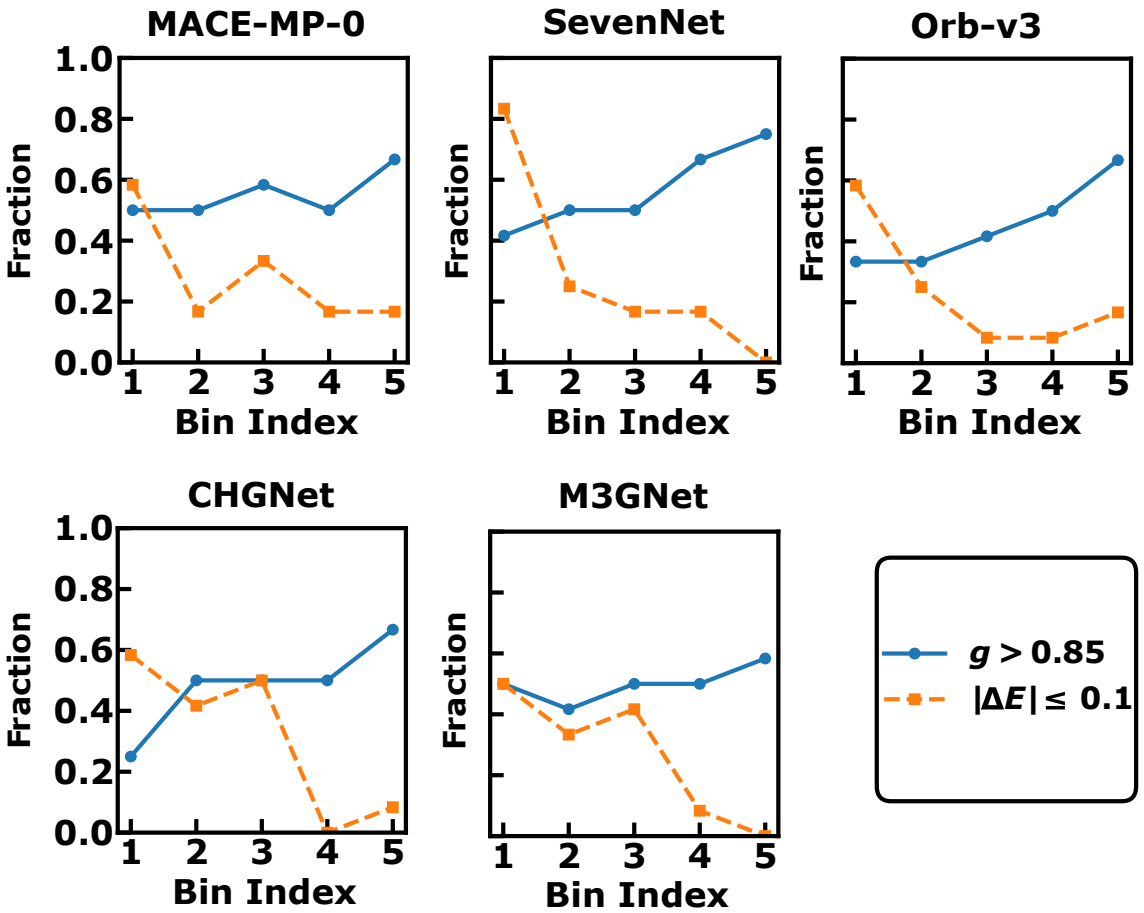
Geometry prediction peformance





- MACE-MP-0 and SevenNet generate better geometries most frequently
- Any MLIP MEP as an initial guess can lead to a good geometry in at least 66% cases
- IDPP and LI show similar performance

Geometry-barrier correlation



- Highest fraction of acceptable E_m predictions at low E_m range
- Highest fraction of 'good' path prediction at high E_m range
- No evident correlation
- The potential energy surfaces are likely 'flat' with variations in local geometries
- Good E_m prediction doesn't necessarily imply good geometry prediction, and vice-versa

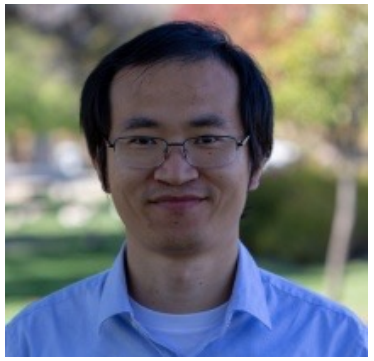
Conclusions

- Foundational MLIPs in their absolute prediction of E_m are not reliable
  - MLIPs perform better if the system can move ions easily
  - Orb-v3 can reliably classify materials, and hence could be used for high-throughput screening
- MLIP-NEB relaxed images as an initial guess can potentially accelerate the subsequent DFT-NEB
- No evident correlation between E_m and geometry prediction accuracy

ML with ~~instead of~~ DFT

Acknowledgments

Dr. Sai Gautam Gopalakrishnan



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Bheemaguli, Achinthya Krishna, Penghao Xiao, and Gopalakrishnan Sai Gautam. "Evaluation of Foundational Machine Learned Interatomic Potentials for Migration Barrier Predictions." arXiv preprint arXiv:2512.03642 (2025).

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