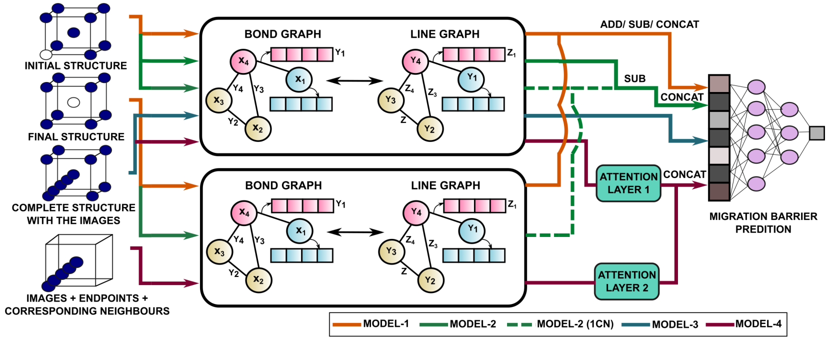

Leveraging transfer learning to predict migration barriers in battery materials

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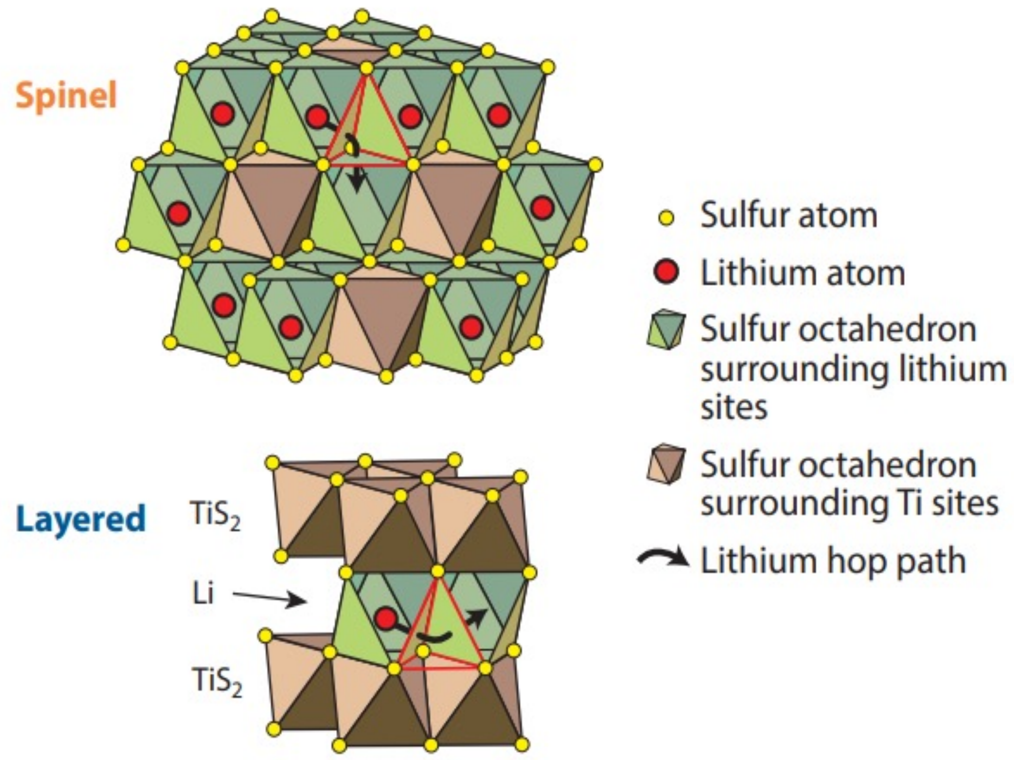
Migration barriers govern rate performance in batteries



How to accurately and swiftly predict migration barriers? Use machine learning?

Intercalation electrodes: ionic diffusivity (D) within the bulk a major factor in rate performance

$$D = D_o \exp\left(-\frac{E_m}{k_B T}\right)$$

D_o : Diffusivity pre-factor (carrier concentration, correlations, etc.)
k_B: Boltzmann constant
T: Temperature
E_m: Migration barrier

Migration barrier: dominant factor determining diffusivity

Experimental measurements of E_m: variable-temperature impedance spectroscopy, variable temperature nuclear magnetic resonance, etc.

Computational predictions of E_m: ab initio molecular dynamics, nudged elastic band (NEB) with density functional theory (DFT)

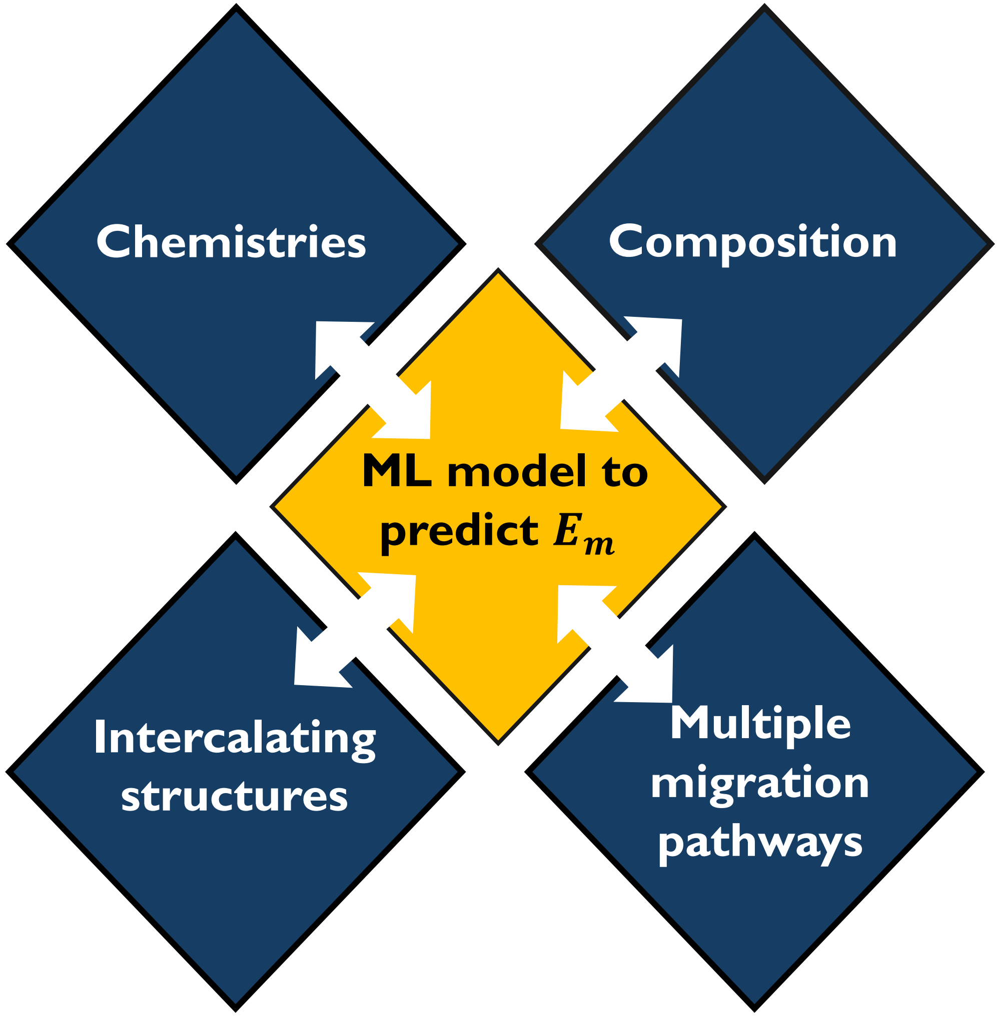
van der Ven et al. **Ann. Rev. Mater. Res.** *48*, 27-55 (2018)

What we need for fast and accurate E_m predictions?

How accurate are first-principles calculations in predicting E_m?

Is there a dataset of E_m values belonging to a diverse set of structures, chemistries, compositions, and migration pathways?

If there is a dataset, how do we construct a generalizable model that accurately predicts E_m?



Accuracy

On average accuracy is in the 140-180 meV range

MAE of SCAN (140 meV): lowest

MAE of GGA: 178 meV

Qualitative trends: reliable with GGA

SCAN: computational cost+convergence

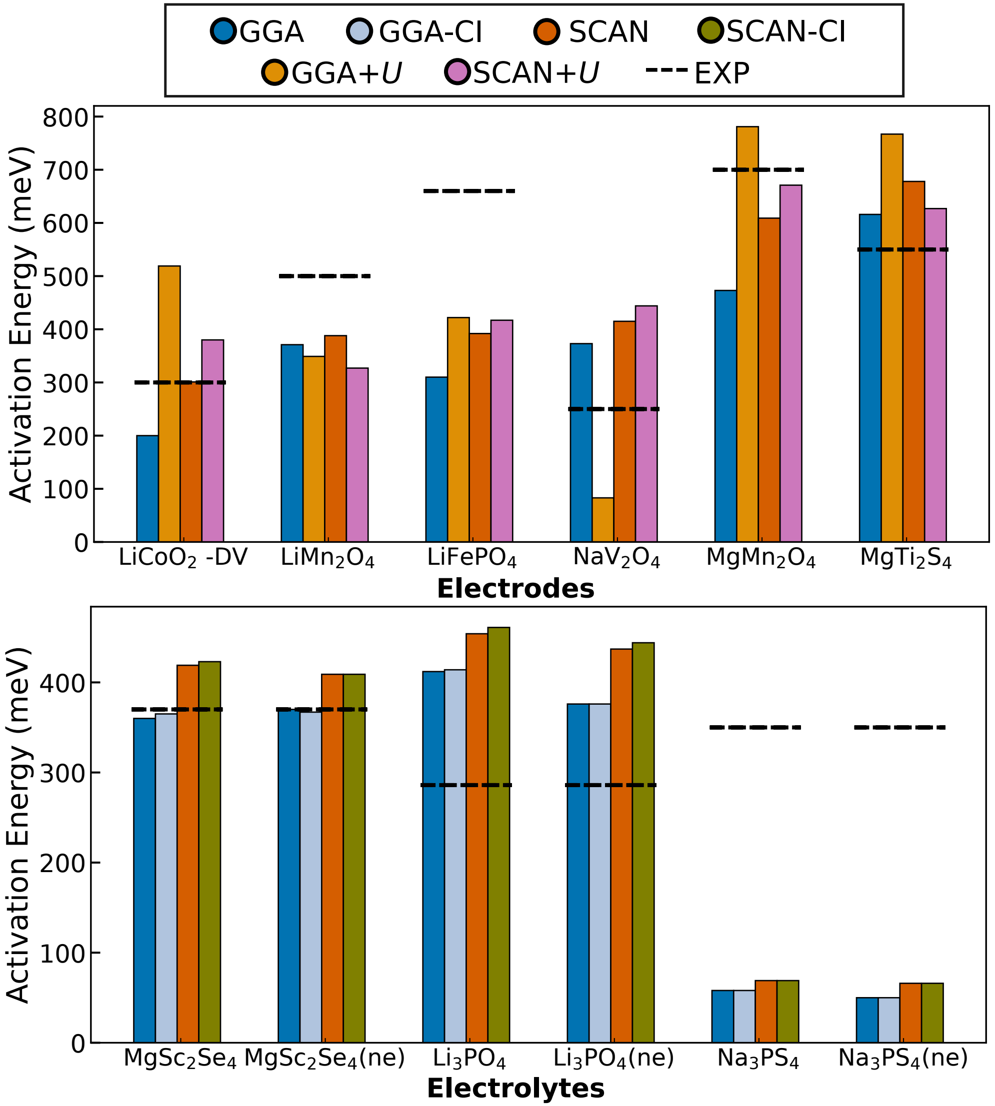
60 meV in E_m: ~1 order of magnitude D at 300 K for micron-sized particles

125 meV in E_m: same D at 300 K, one order change in particle size

SCAN: strongly constrained and appropriately normed;
GGA: Generalized gradient approximation
U: Hubbard *U* correction

CI: Climbing image; EXP: Experimental

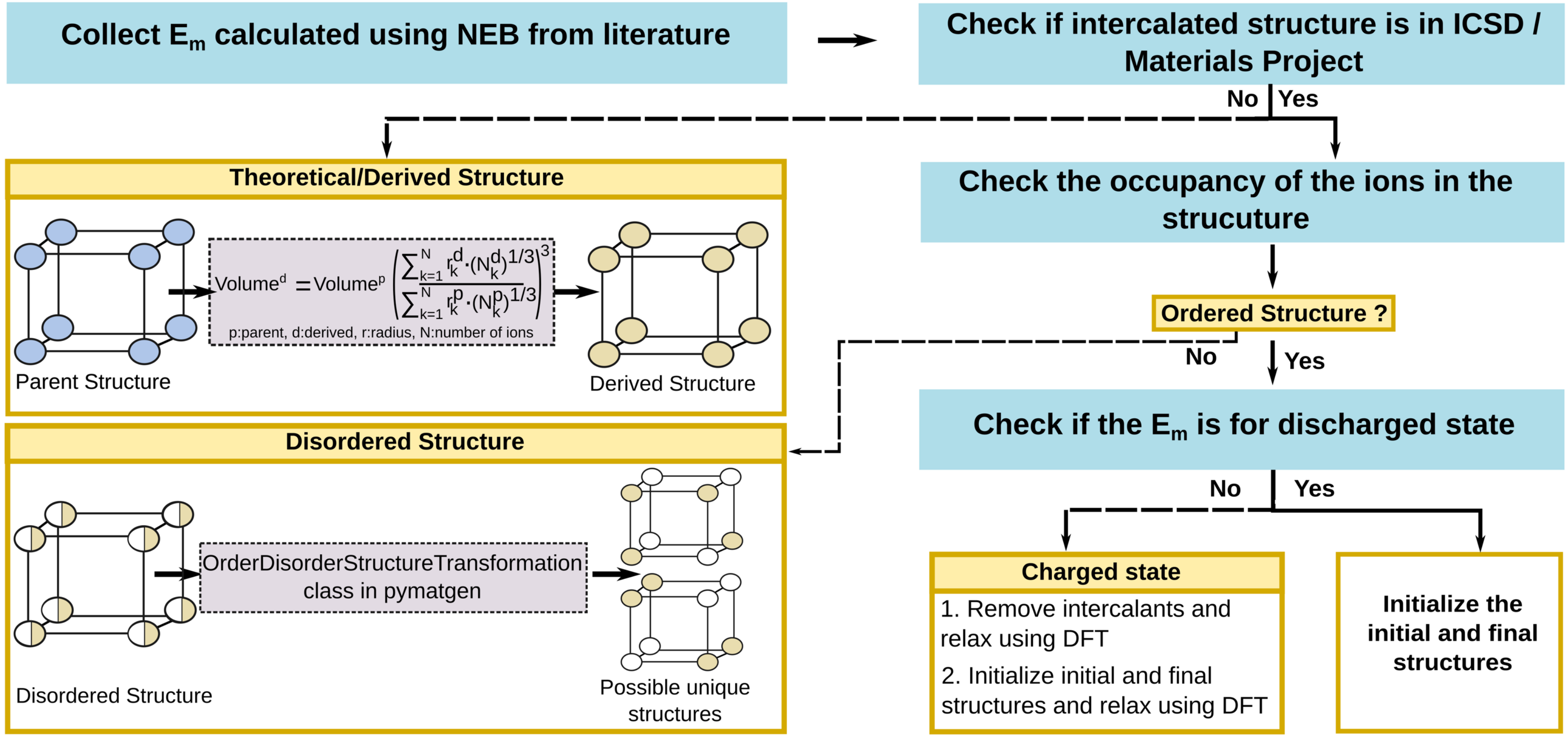
ne: Background charge; DV: divacancy



Devi et al., **npj Comput. Mater.** *8*, 160 (2022)

Dataset

Workflow to generate a curated dataset of E_m



Consider papers that reported unambiguously the methods and the structure used

Generalized gradient approximation or its Hubbard *U* preferred as the functional

Well labelled images of E_m and/or the minimum energy pathway (MEP)

Statistics

619 datapoints spanning 58 space groups across 7 crystal systems

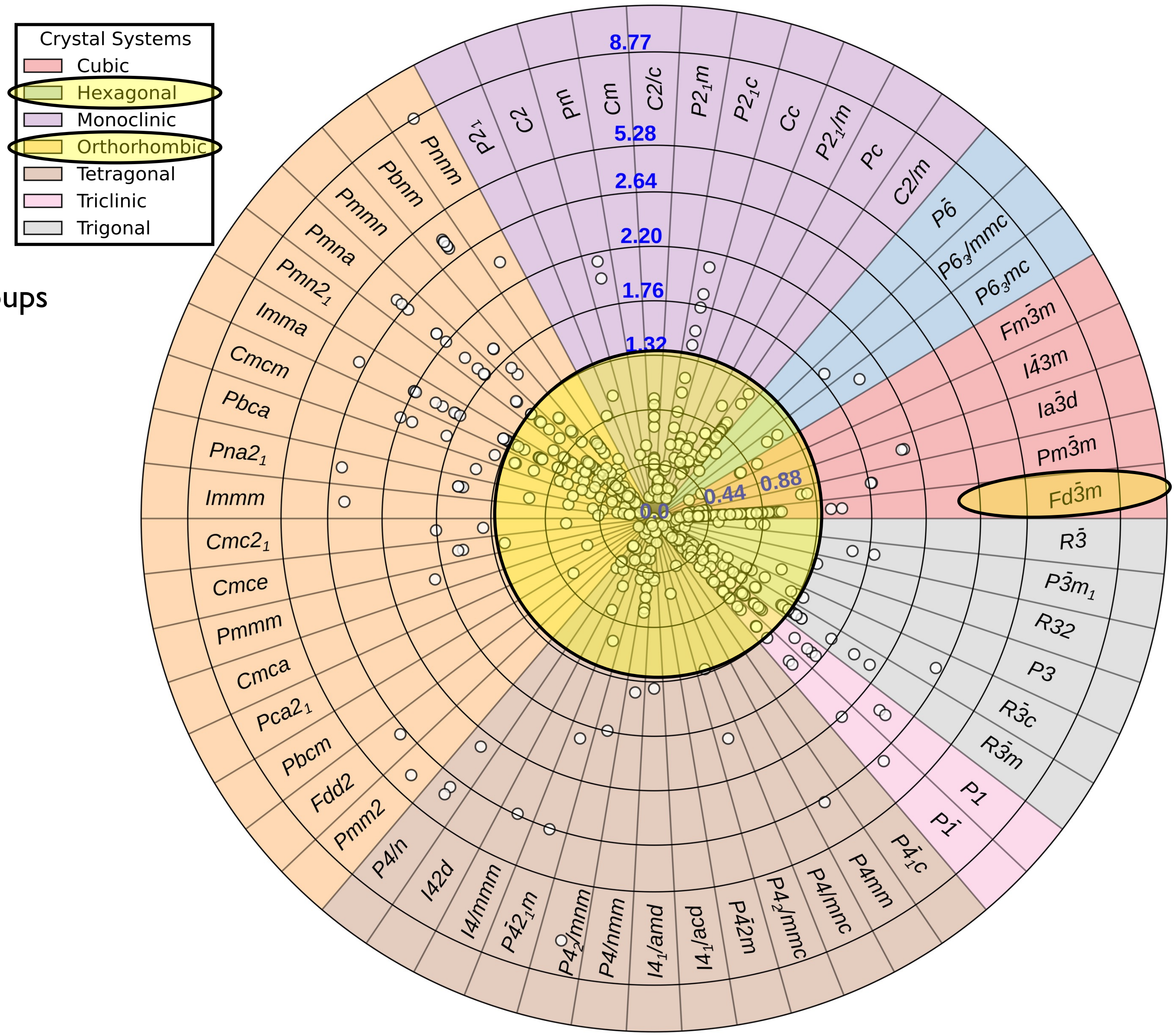
Range: 0.03 to 8.77 eV

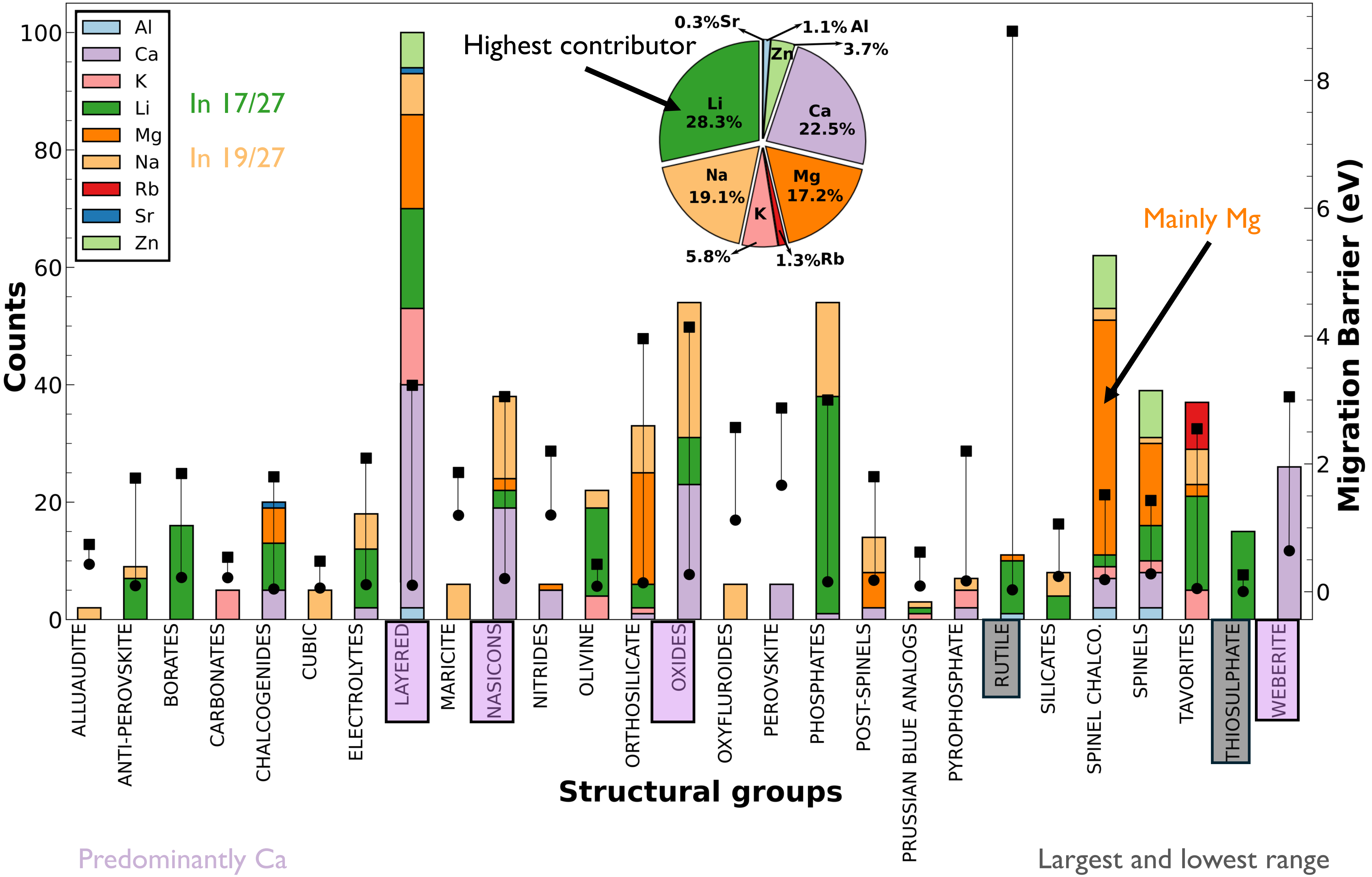
528 electrodes
91 electrolytes

Cubic spinels: 94 entries

Orthorhombic (206): highest
Hexagonal (6): lowest

73.4%: E_m < 1 eV
19.4%: 1 eV < E_m < 2 eV
7.2%: E_m > 2 eV



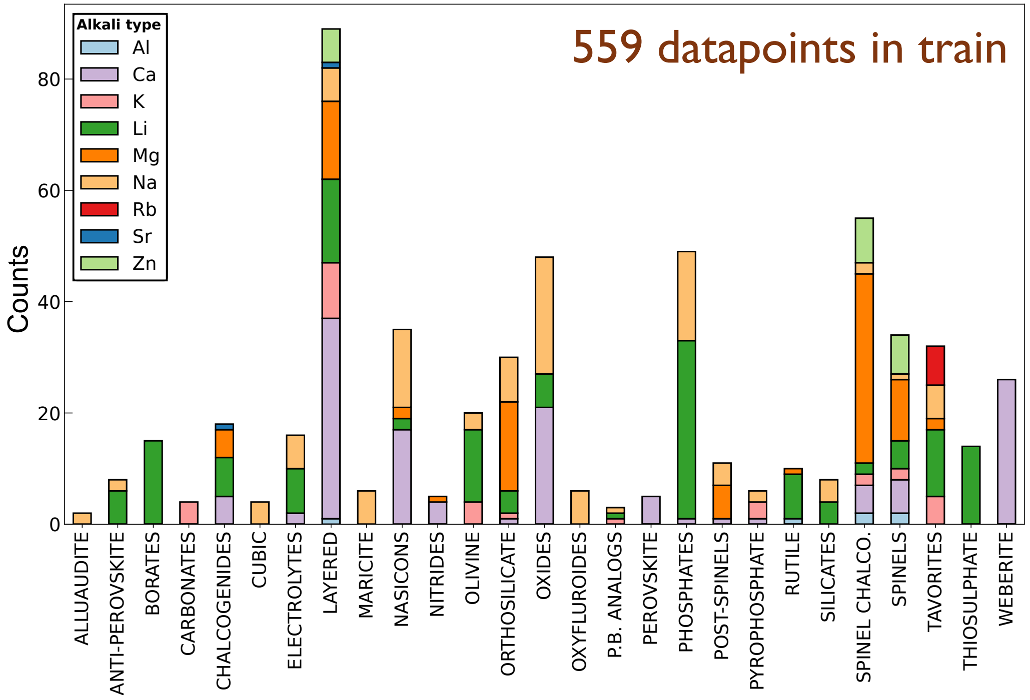


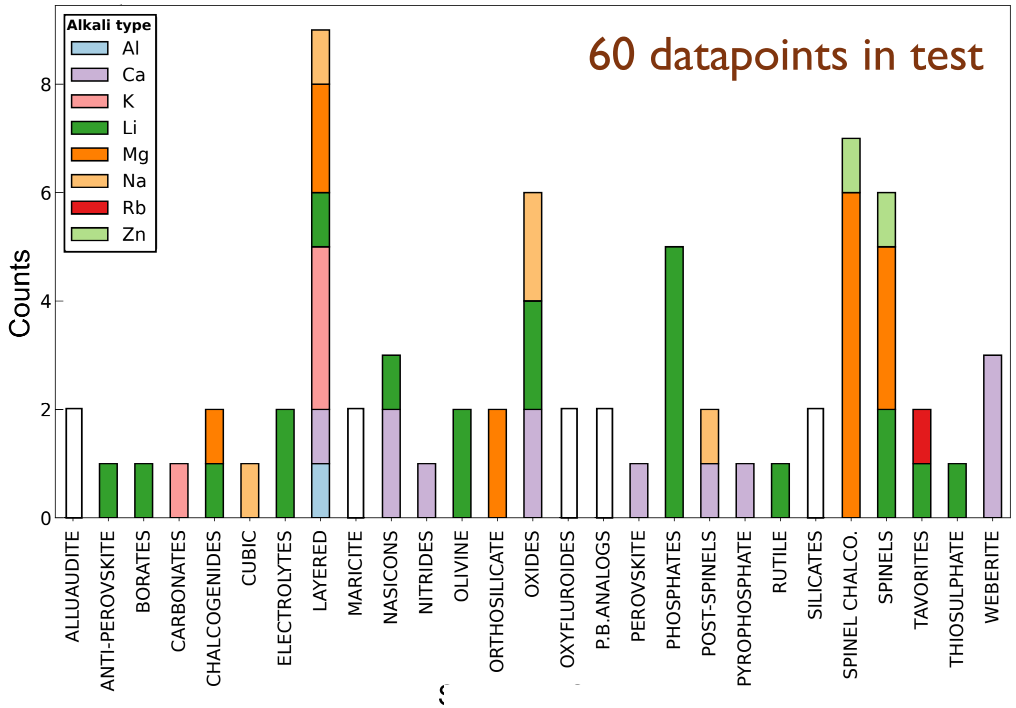
Predominantly Ca

Largest and lowest range

Generalizable model

Careful split of dataset

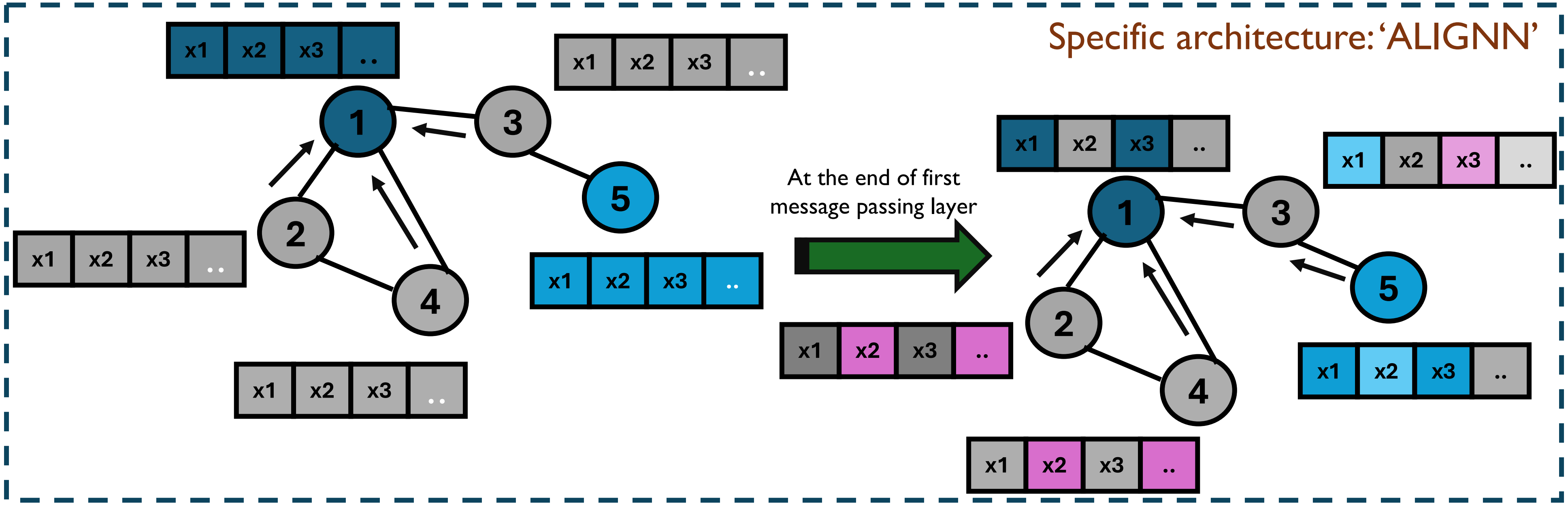


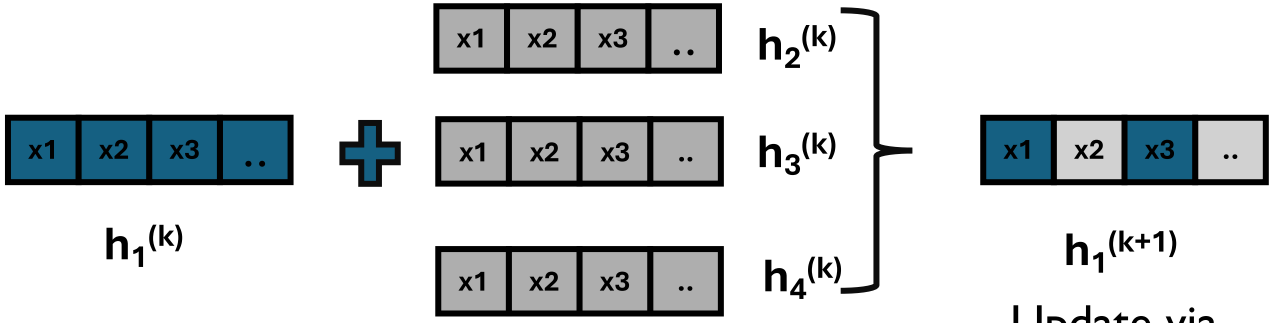


← Structural groups →

- Similar distribution in train and test
- Prevent unfair penalization: 1 test datapoint for groups contributing 1-2%
- Random sampling within each structural group

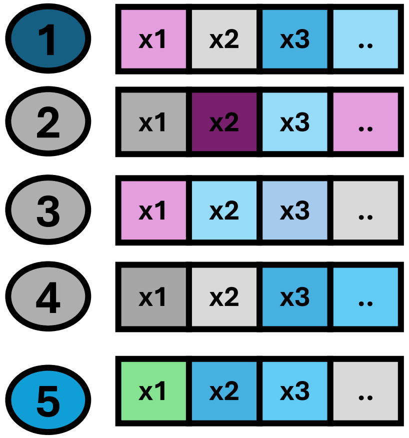
Base model architecture: graph neural networks





Aggregate

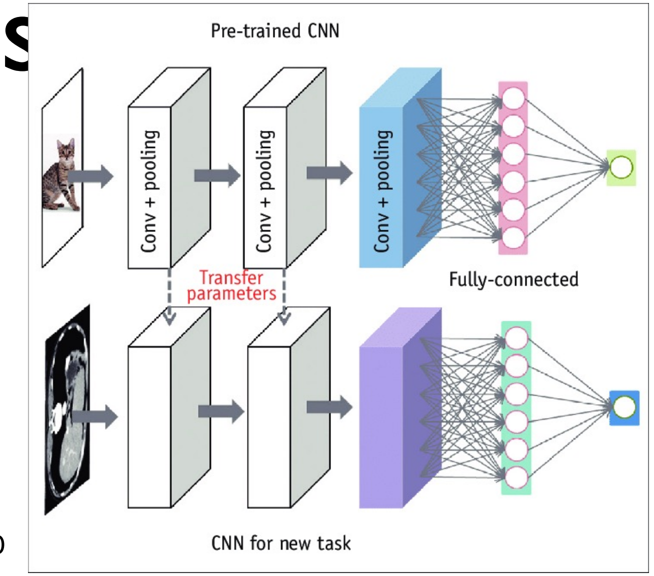
Update via activation



After multiple message passing layers

ALIGNN: Choudhary and DeCost, **npj Comput. Mater.** 7, 185 (2021)

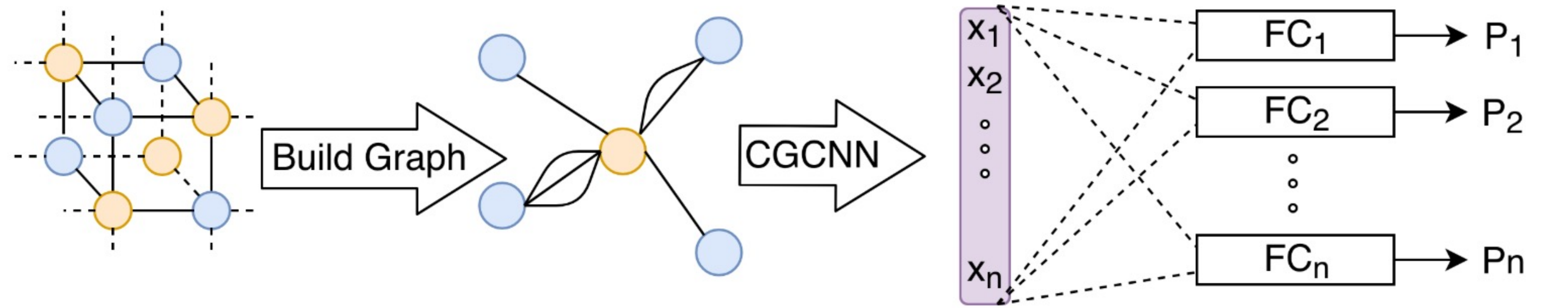
Transfer learning and 'MPT' models



Pre-train (PT) on a large dataset and fine-tune (FT) on a target, smaller dataset

Do et al., **Korean J. Radiol.** 2020

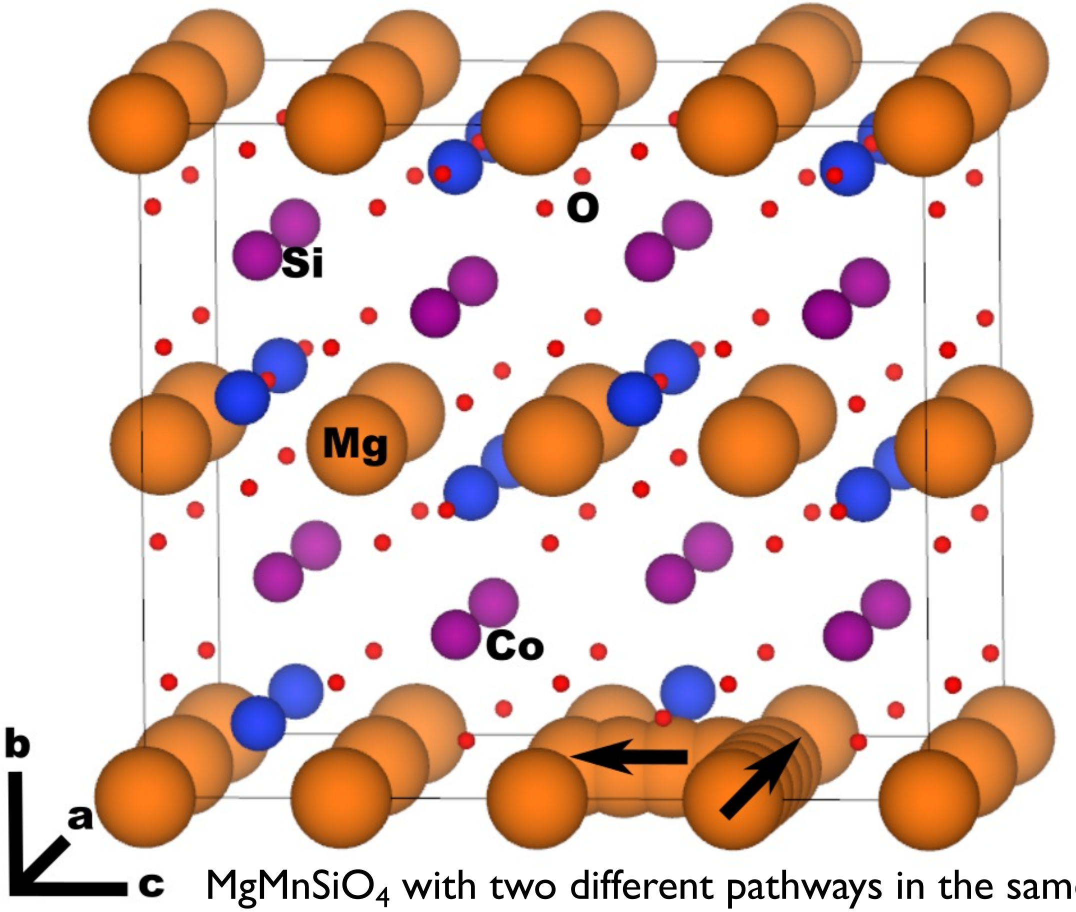
Multi-property pre-trained (MPT) models: PT on seven bulk properties simultaneously



Devi et al., **npj Comput. Mater.** *10*, 300 (2024)

Sanyal et al., **arXiv** 1811.05660 (2018)

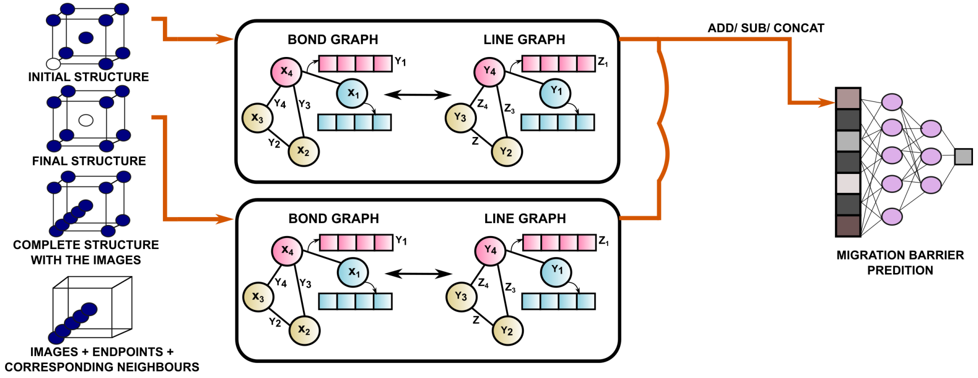
Distinguish multiple paths in a structure: use modified model architectures



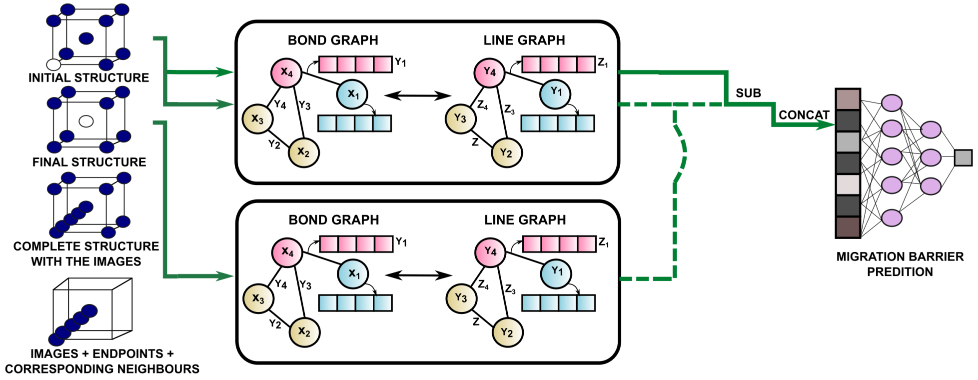
$MgMnSiO_4$ with two different pathways in the same structure

Distinguish multiple paths in a structure: use modified model architectures

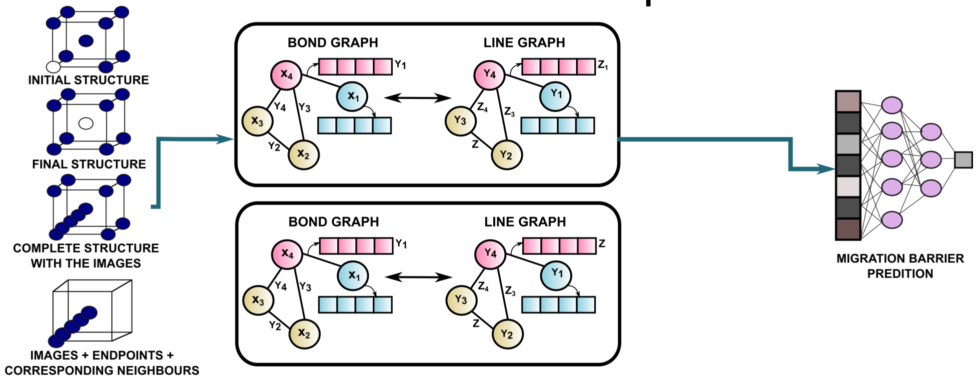
Model 1: Take initial and final as input



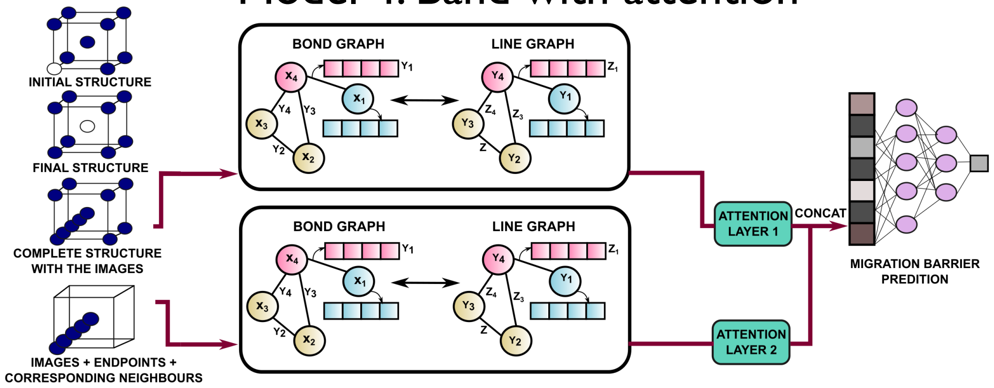
Model 2: Take initial and delta as input



Model 3: Band as input



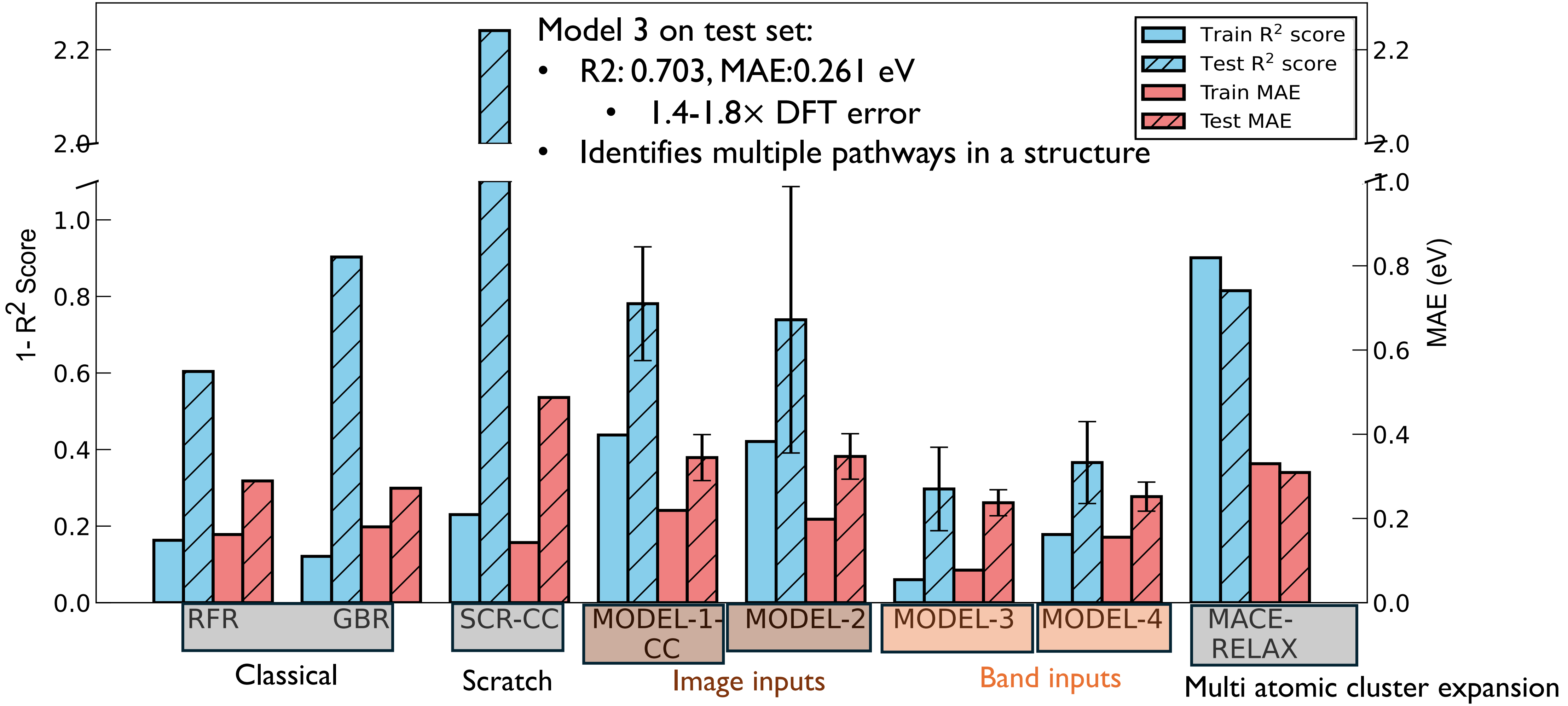
Model 4: Band with attention



Model-3: best performing model

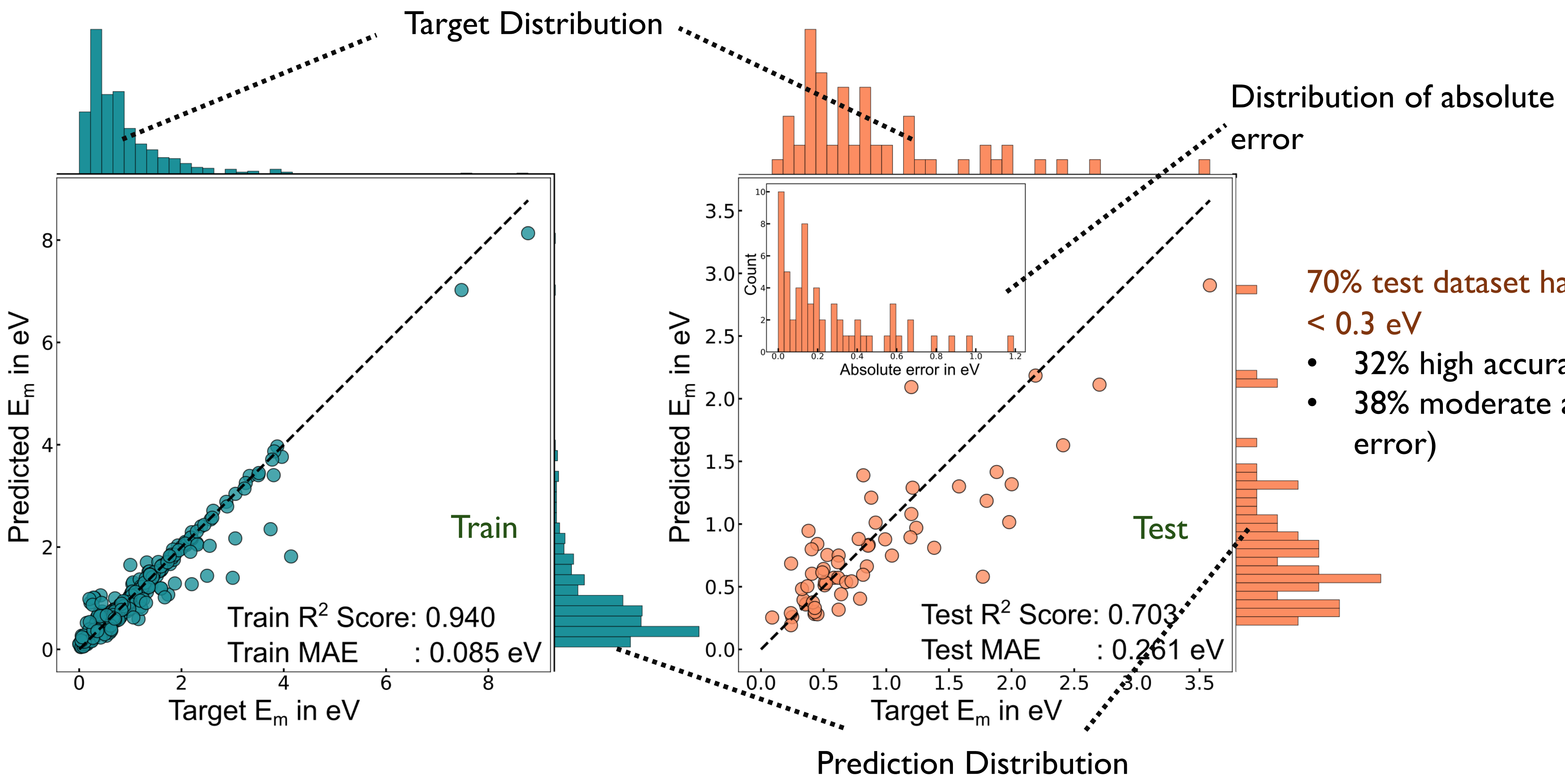
Model 3 on test set:

- R2: 0.703, MAE:0.261 eV
    - 1.4-1.8× DFT error
- Identifies multiple pathways in a structure



RFR: Random forest regressor; GBR: Gradient boosted regressor; SCR: Scratch; CC: Concatenate

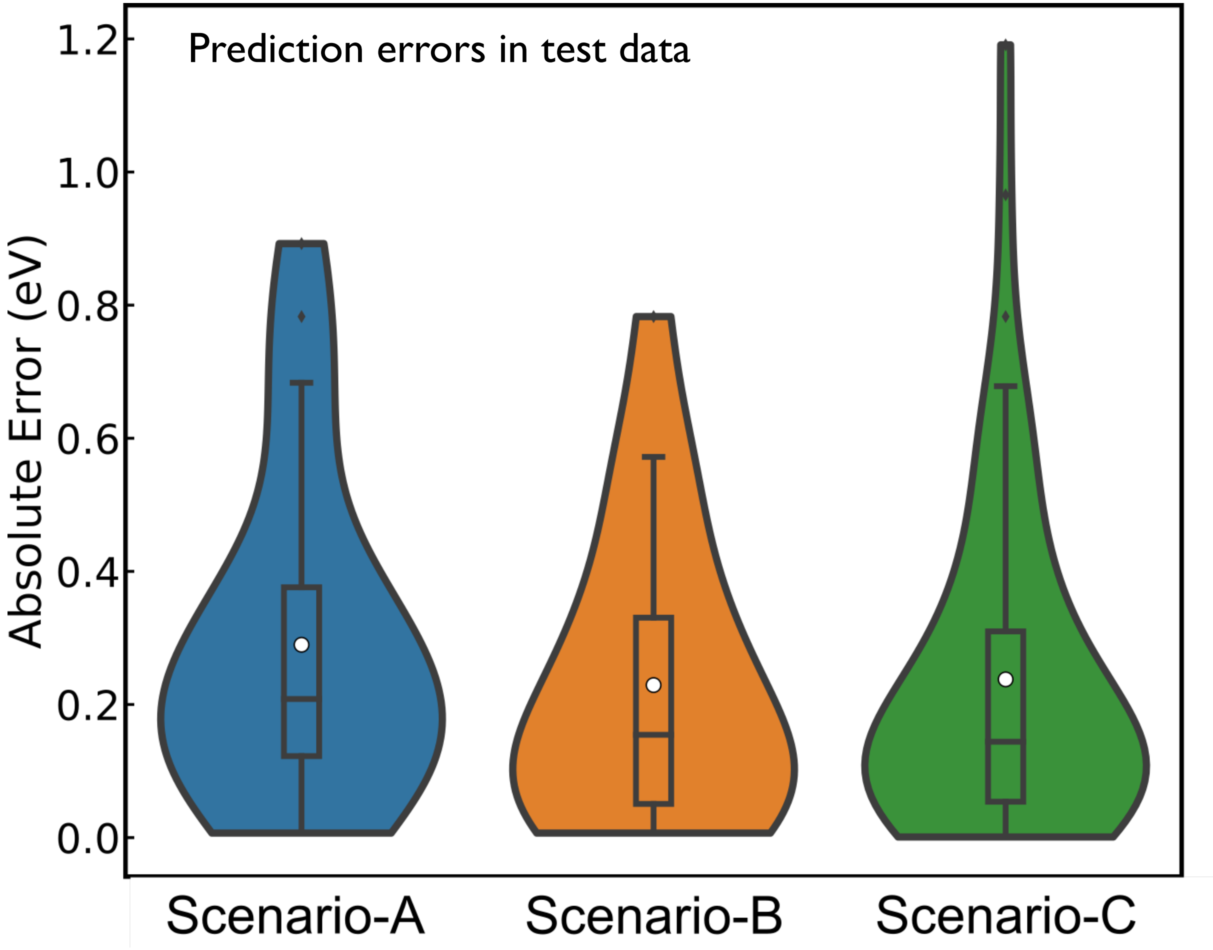
Model-3 performance



70% test dataset has prediction errors < 0.3 eV

- 32% high accuracy (< 0.1 eV error)
- 38% moderate accuracy (0.1-0.3 eV error)

How does Model-3 generalize?



Generalization across migration pathways in a structure

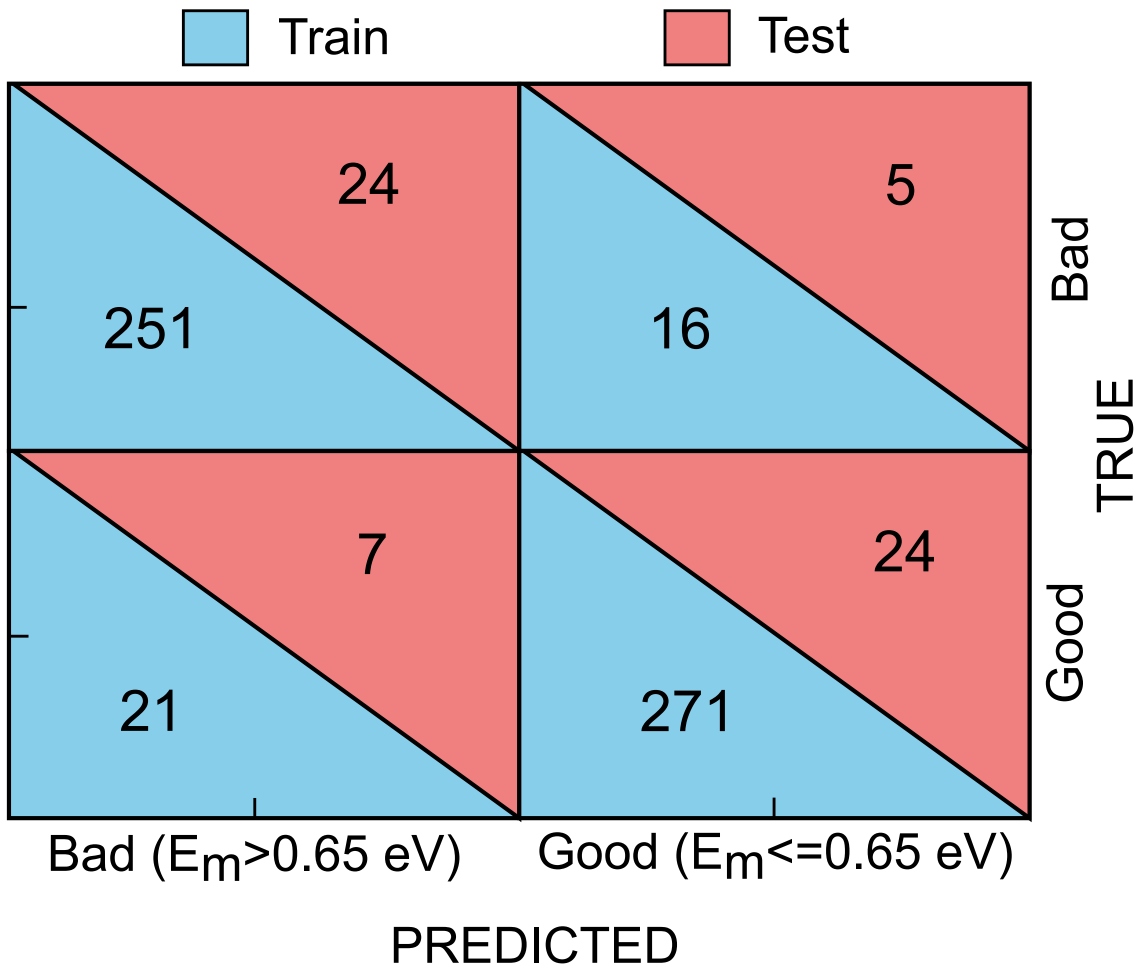
Generalization across intercalant composition (charged vs. discharged)

Generalization across chemistries (different intercalant/transition metal/anion, same structure)

Lower mean/median errors for Model-3 in scenarios B and C: better generalization across composition and chemistry compared to migration pathways!

- Can be used as a screening tool

Model-3 as a classifier



Threshold: 0.65 eV

- Nano-sized cathode
- C/2 rate, 300 K

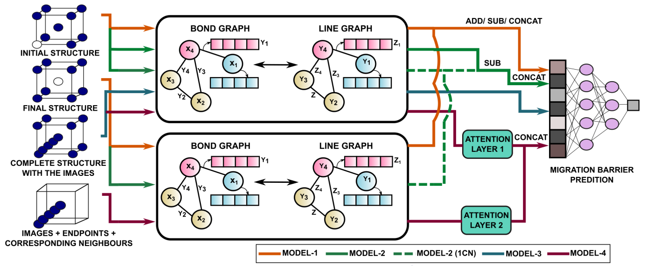
Accuracy: 80%

Precision:
82.8% (good conductors)
77.4% (bad conductors)

Model-3 can be good classifier!

- Especially for identifying good conductors

Conclusions



- Predicting E_m: critical for discovery of new materials with 'high' intercalant mobilities
- Accuracy: DFT gets qualitative trends right, reasonably close to experiments
- Dataset: curated 619 datapoints of calculated E_m
- Generalizable model to predict E_m
  - Leverage transfer learning, modified graph networks
  - Model-3 classifies multiple migration pathways, generalizes across chemistries and compositions

"A literature-derived dataset of migration barriers for quantifying ionic transport in battery materials", R. Devi, A. Balasubramanian, K.T. Butler, and G. Sai Gautam, *Scientific Data* 12, 1922 (2025).

"Leveraging transfer learning for accurate estimation of ionic migration barriers in battery materials ", R. Devi, K.T. Butler, and G. Sai Gautam, *arXiv*, 2508.06436 (2025). *[Coming soon on npj Comput. Mater.]*

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