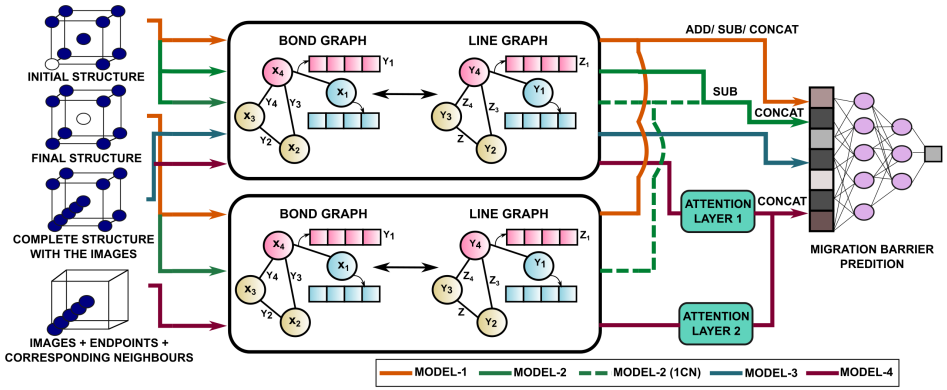

Optimal use of transfer learning for predicting ionic migration barriers in solids

[1]Reshma Devi, [1]Avaneesh Balasubramanian, [2]Keith T. Butler, [1]Sai Gautam Gopalakrishnan

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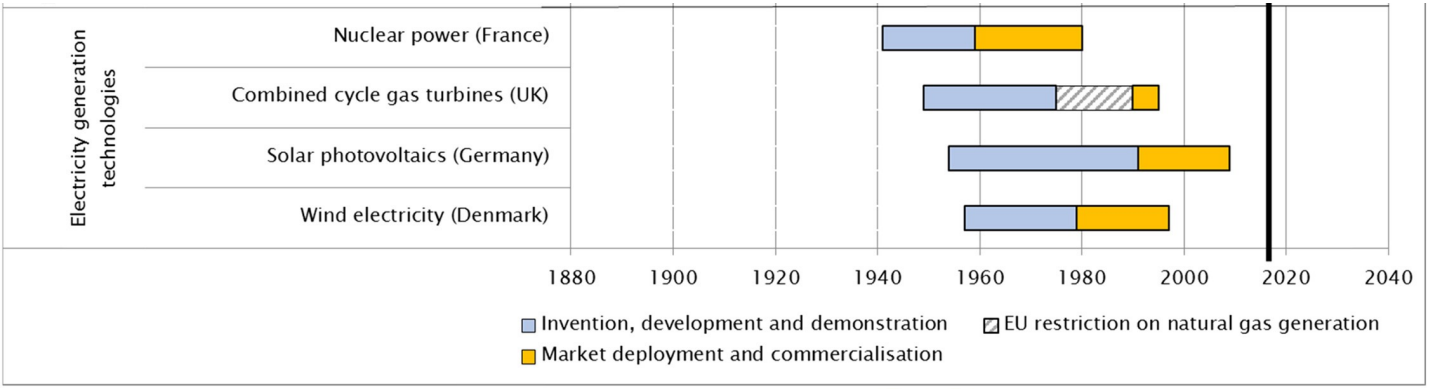
Institut für Nachhaltige Wasserstoffwirtschaft (INW-1), Forschungszentrum Jülich
Sep 15, 2025

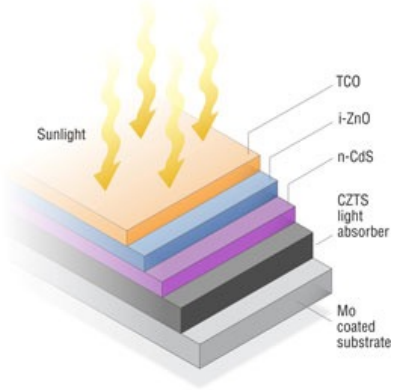
We work broadly on energy materials



Technological innovation cycle → slow in energy sector

Need rapid identification of novel materials for improving performance

Design better electrodes and solid electrolytes



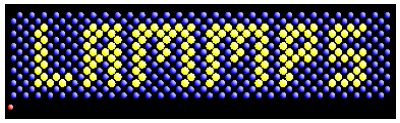
Develop better light-absorbing semiconductors

Identify better materials for H_2 generation and sensing

Identify novel materials for applications

Understand underlying materials phenomena better

Make theory better/build models



Acknowledgments

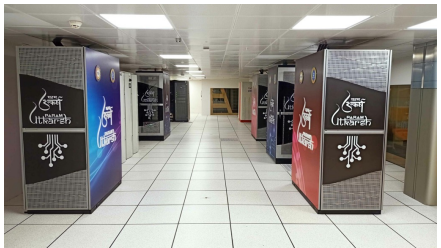
Group picture in May 2025

Dr. Keith Butler

Reshma

Avaneesh

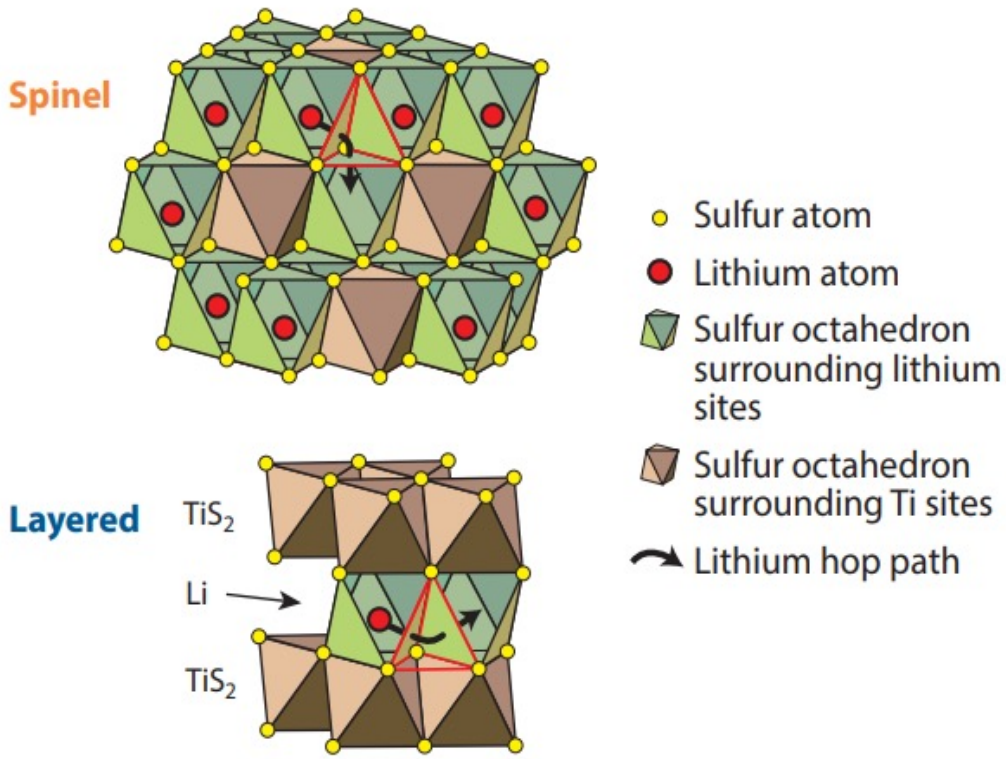
SERC (IISc)



Param Utkarsh (CDAC)

Archer (UK)

Migration barriers govern rate performance in batteries



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

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

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

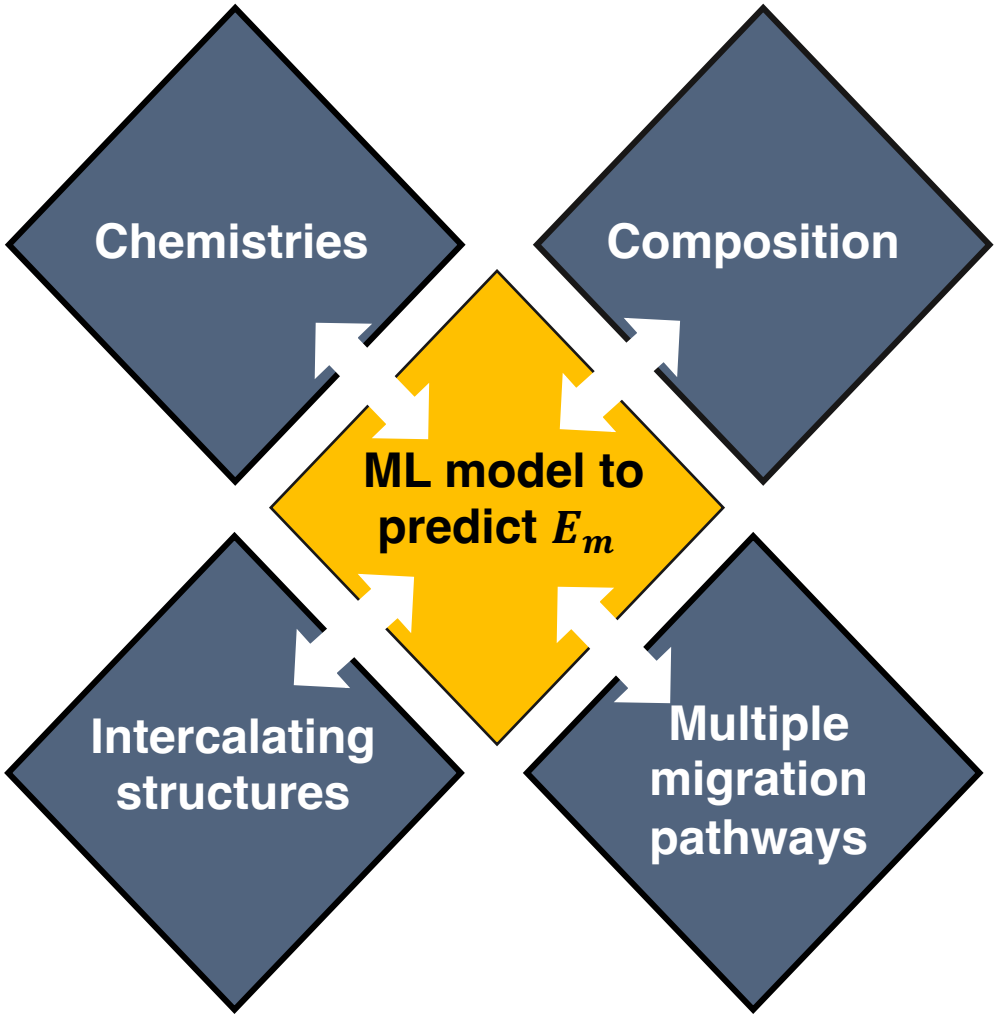
Migration barrier: dominant factor determining diffusivity

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)

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

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?

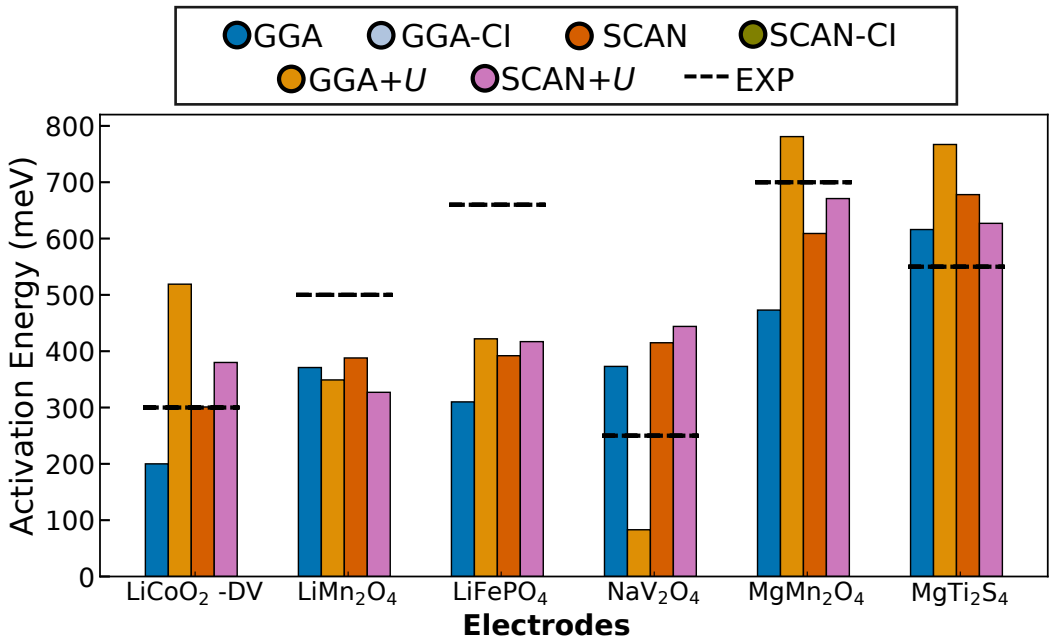
Is there a dataset of E_m values belonging to a diverse set of structures, chemistries, compositions, and migration pathways?

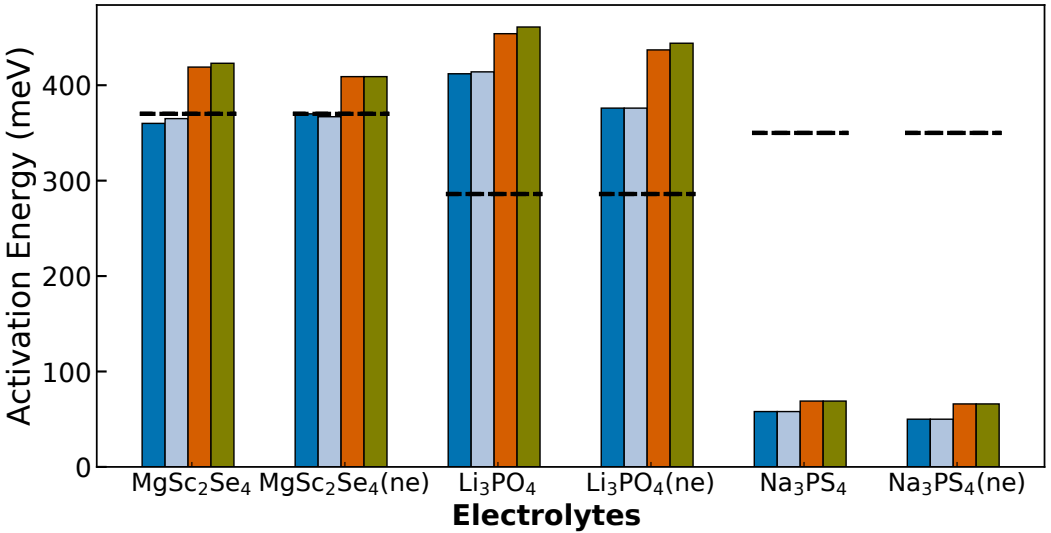
If there is a dataset, how do we construct a generalizable model that accurately predicts E_m?

- Using transfer learning

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

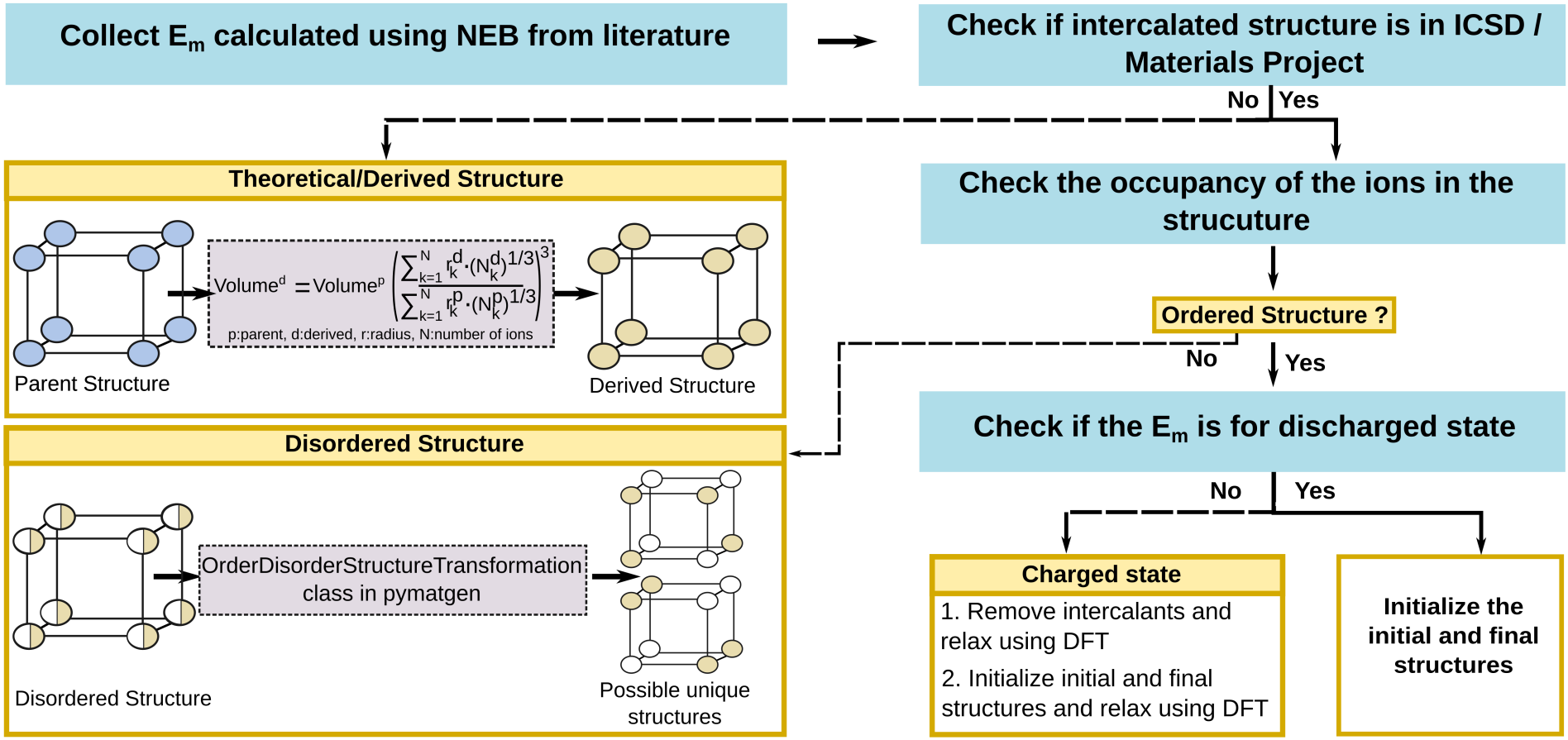
CI: Climbing image; EXP: Experimental

ne: Background charge; DV: divacancy

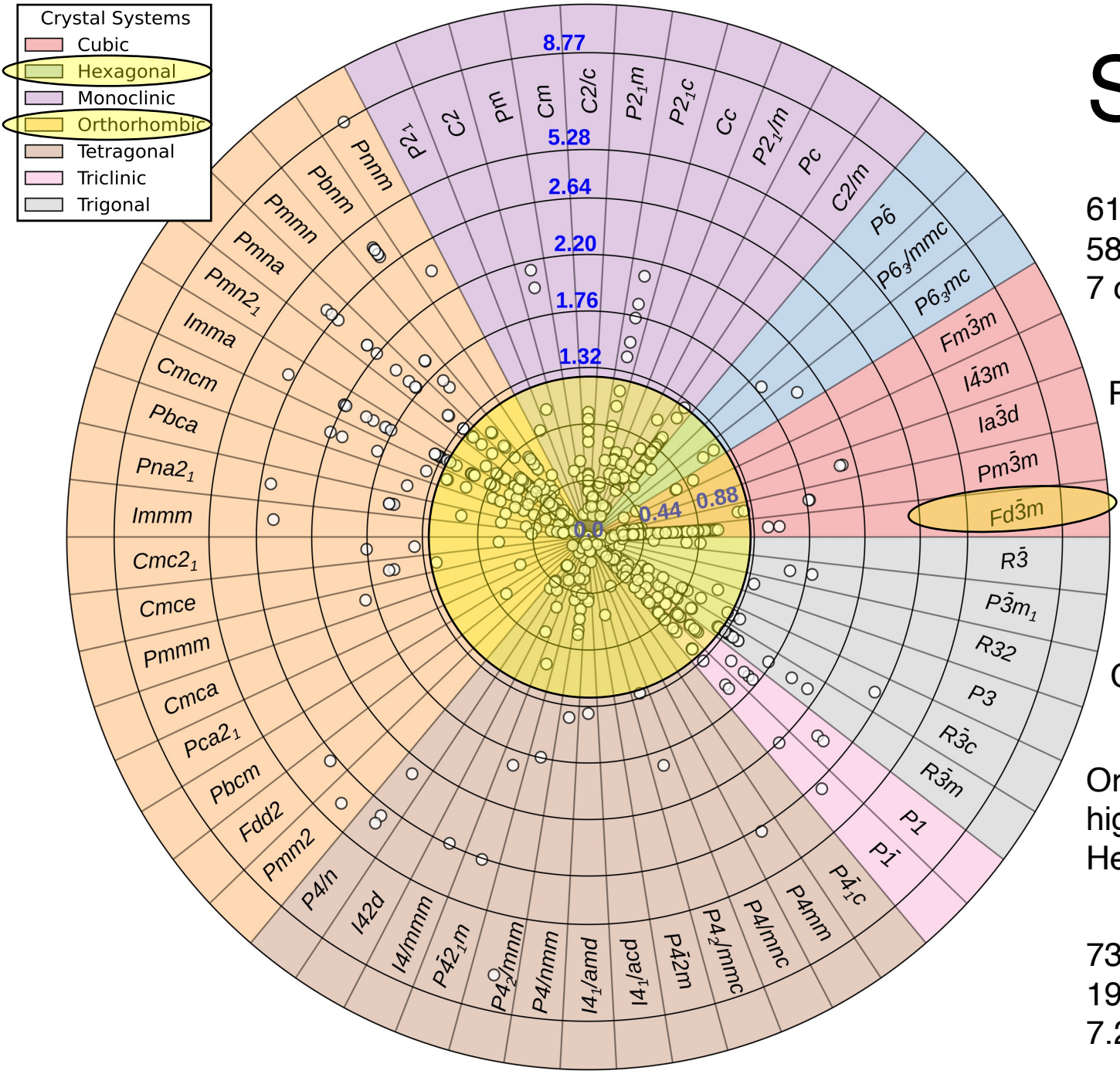
Devi et al., **npj Comput. Mater.** *8*, 160 (2022)

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

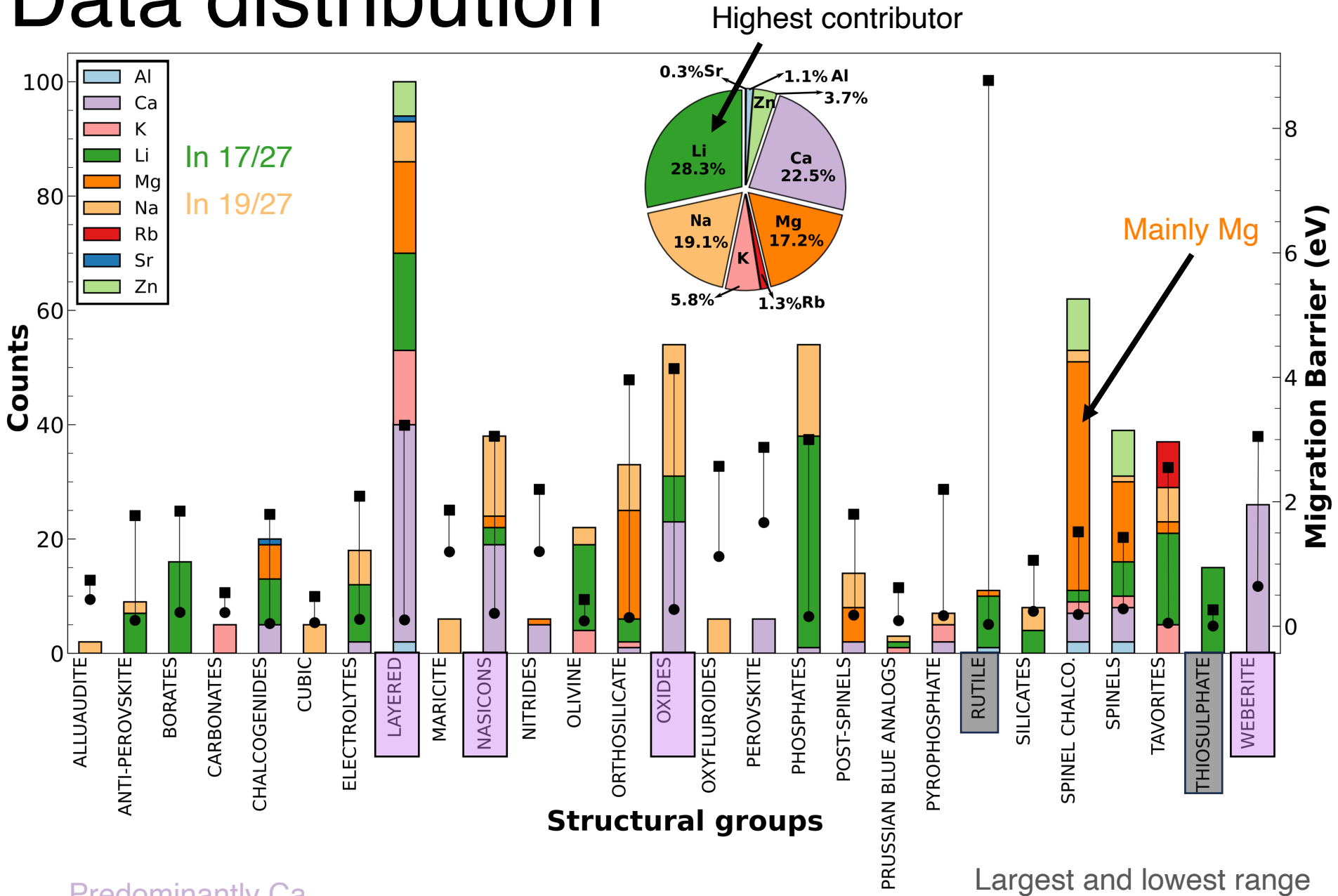
528 electrodes
91 electrolytes

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

Data distribution

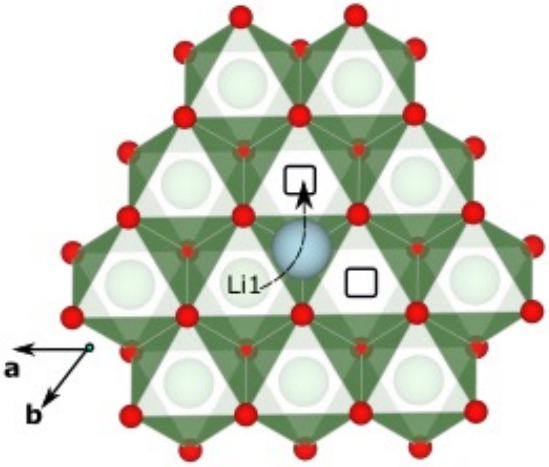


How to create generalizable models?
Transfer learning

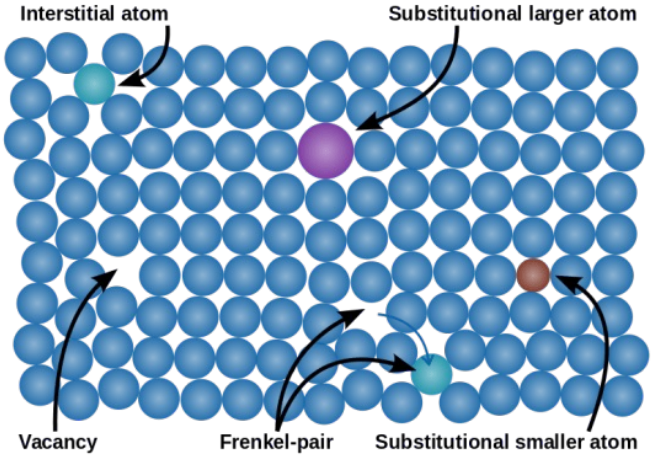
Materials science is data limited

Several key material properties that govern performance in applications have limited data

- 'Small' datasets (< 10^4 datapoints)
  - Ionic mobilities, defect formation energies, adsorption energies,…
- Limits application of deep learning (DL) frameworks

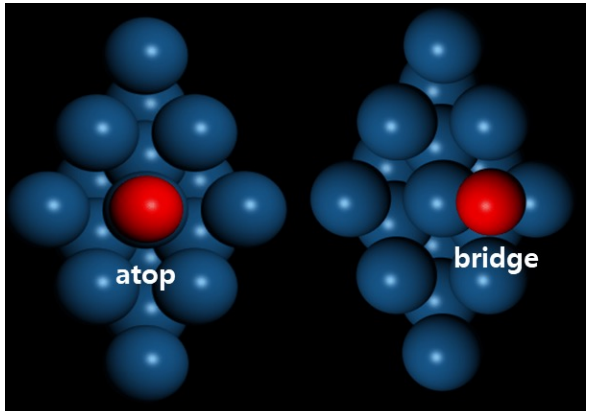


Devi et al., **npj Comput. Mater.** 2022



https://www.differencebetween.com/difference-between-point-defect-and-line-defect/

https://sites.psu.edu/



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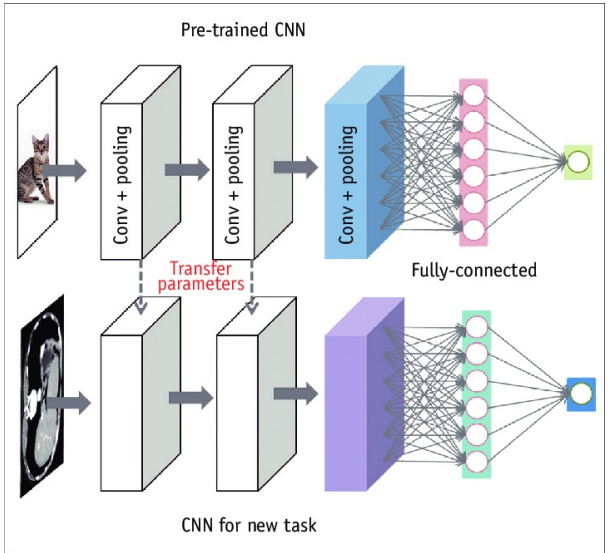
Transfer learning: efficiently use DL frameworks on small datasets

- Pre-train (**PT**) on 'large' dataset, fine-tune (**FT**) on 'small' dataset



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

Materials science is data limited

Several key material properties that govern performance in applications have limited data

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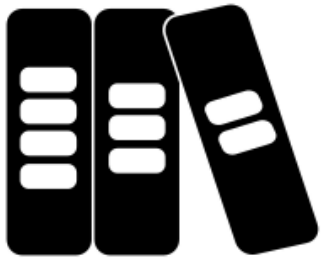
Transfer learning: efficiently use DL frameworks on small datasets

- Pre-train (**PT**) on 'large' dataset, fine-tune (**FT**) on 'small' dataset

How useful is transfer learning in materials science?

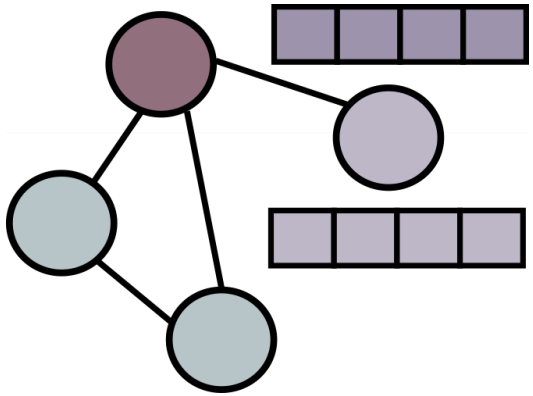
- Optimal ways to use?
- Ways to generate 'generalizable' models?

Handles to consider



Dataset(s)

- What, how, how many?



Architecture

- Graph neural network

Frozen

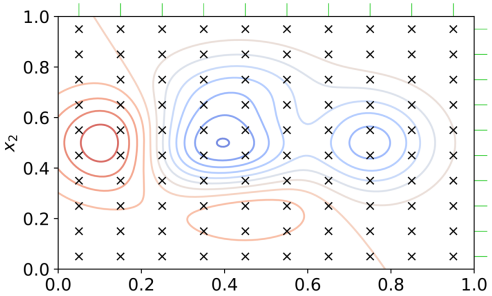
Unfrozen

Strategy

- FT techniques in pair-wise PT/FT models
- Multi-property PT (**MPT**) models

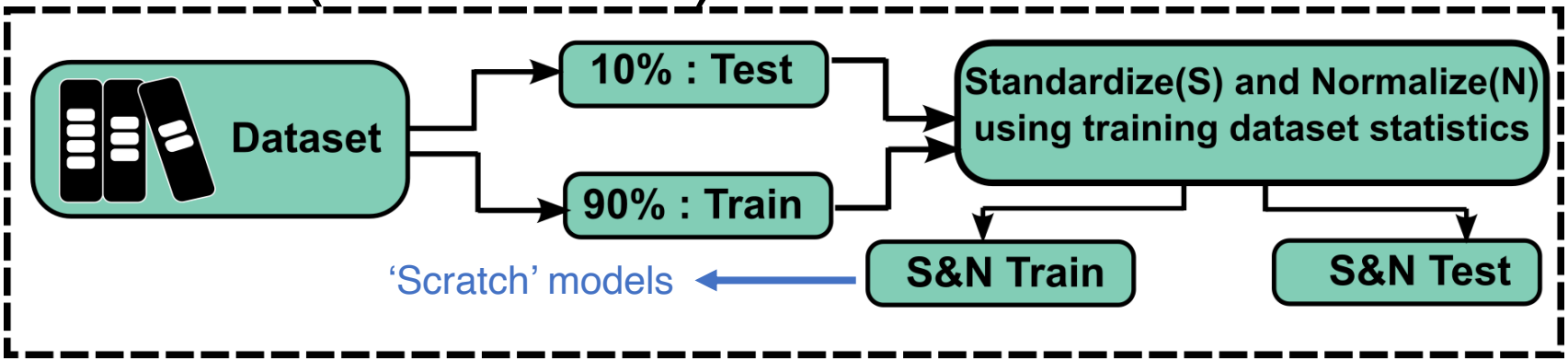
(Learning) Hyperparameters

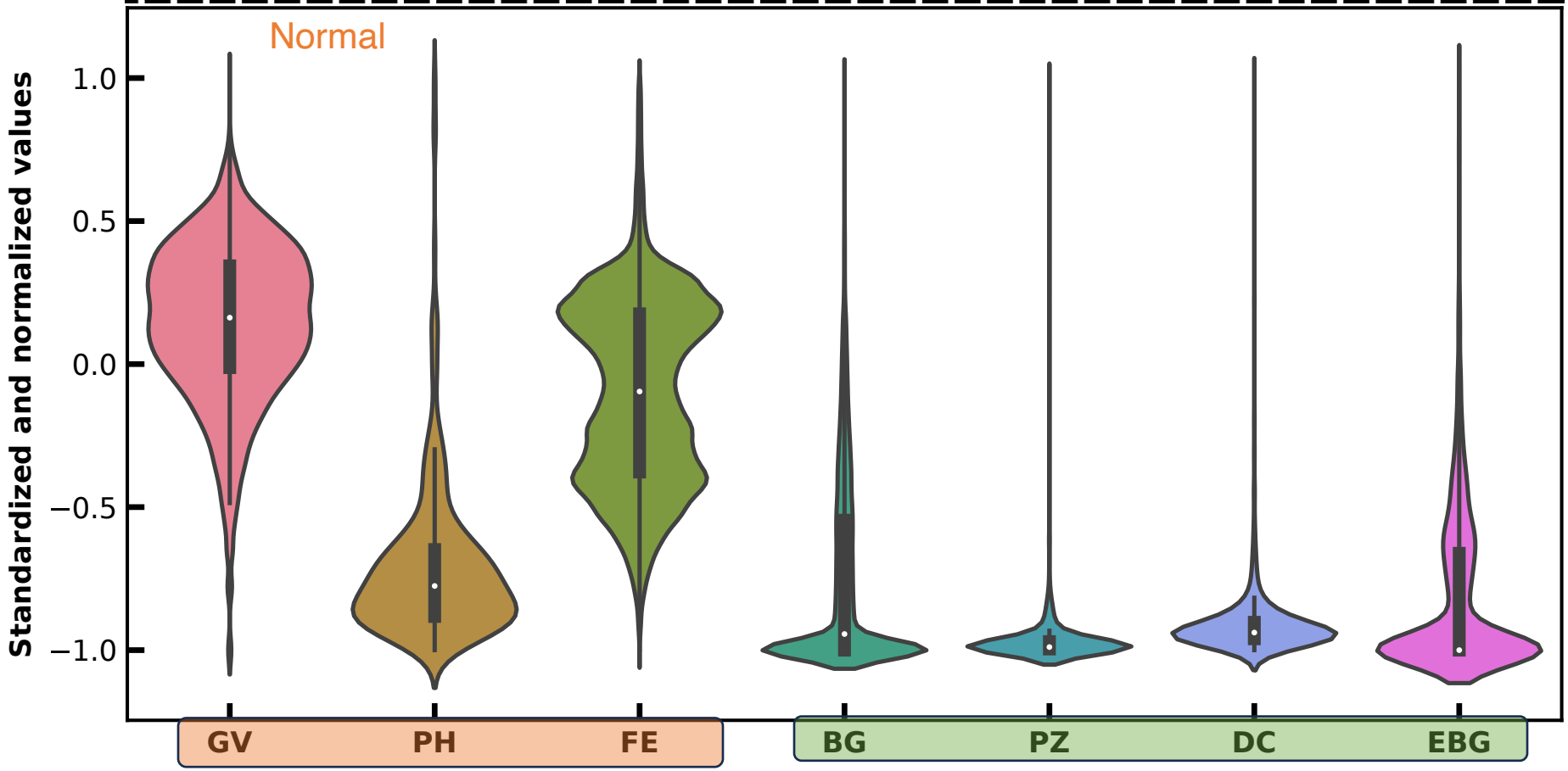
- Data sampling
- Learning rate
- Number of datapoints during PT, FT



https://en.wikipedia.org/

7 datasets (Matminer)

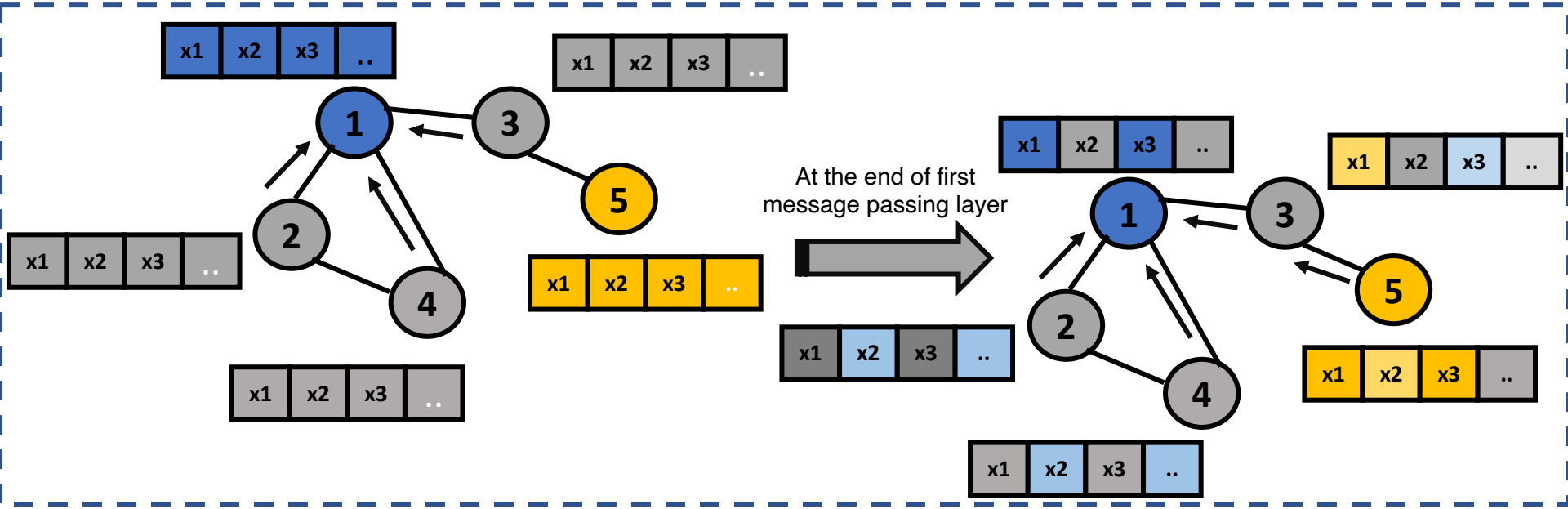


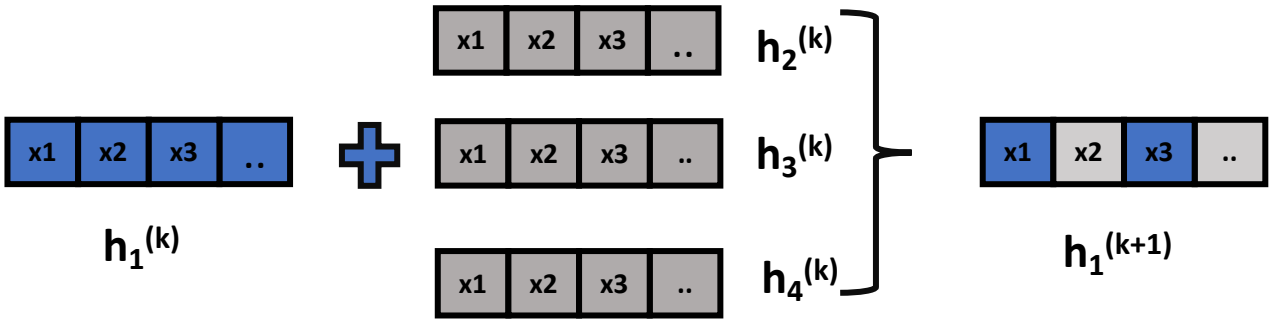


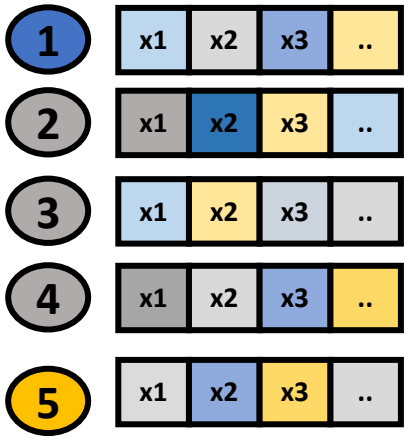
1. Ward *et al.*, **Comput. Mater. Sci.** *152*, 60–69 (2018).

Base model architecture: graph neural networks

Specific architecture: ‘ALIGNN’



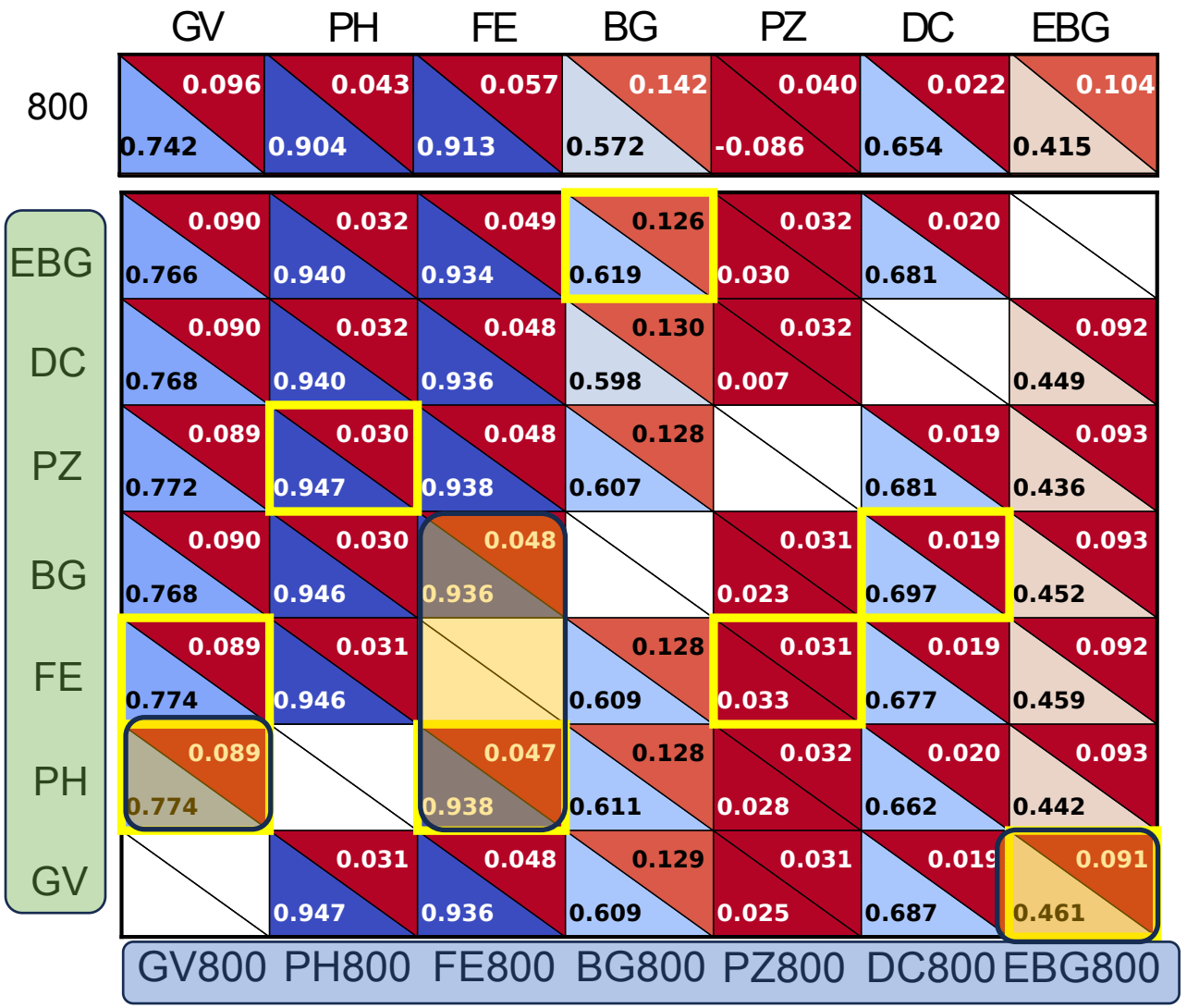




After multiple message passing layers

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

7×6 combinations of pair-wise models



Pair-wise models: better than scratch

- Average increase in R^2: 25%
- Average decrease in MAE: 16%

Best models: GV, PH, FE ($R^2 > 0.75$)

Average models: BG, DC, EBG

Specific PT property: little influence on FT

No symmetry

FT dataset+size

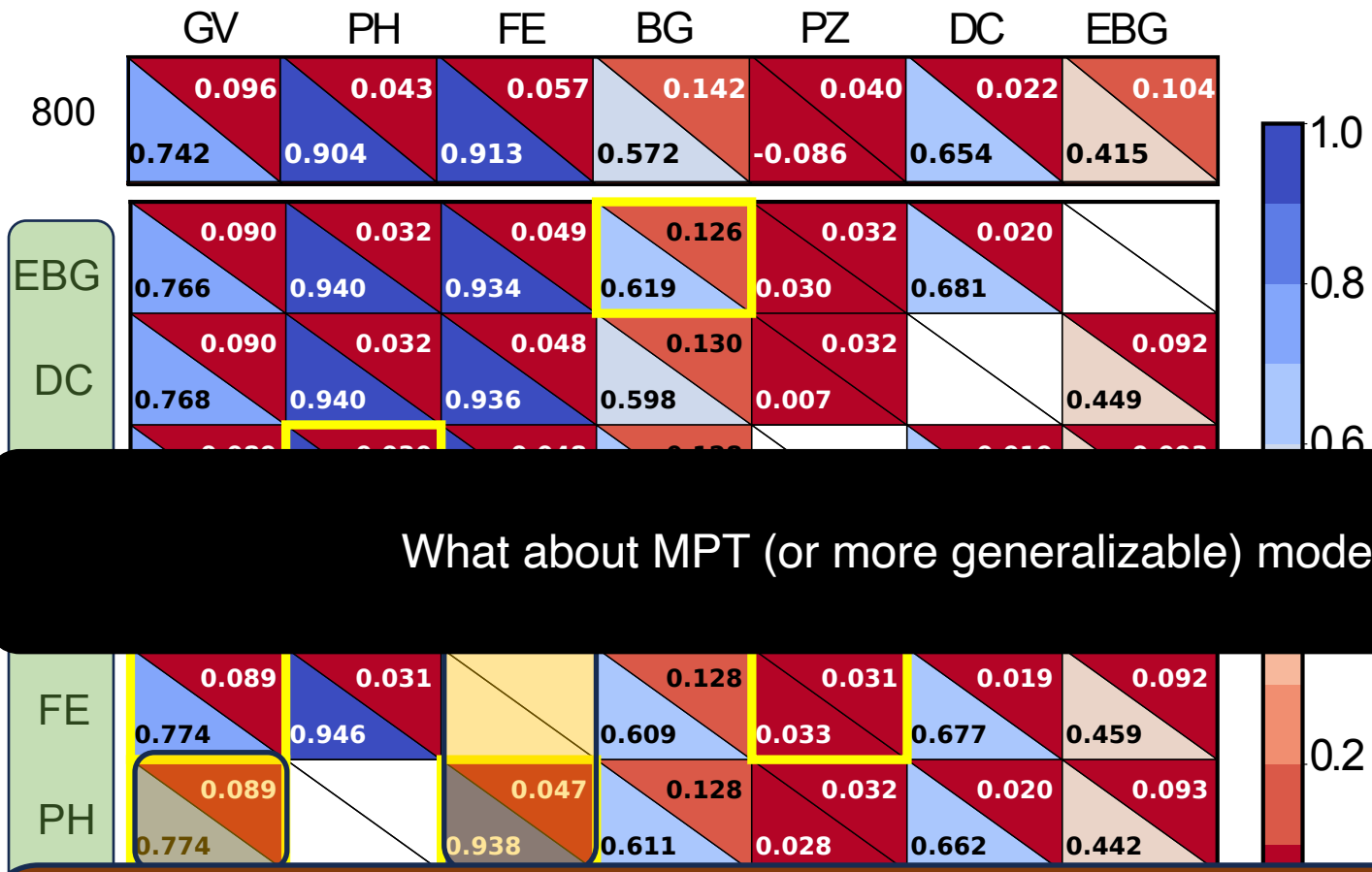
PT dataset (941)

Best model

Test scores

GV: Shear modulus; PH: Phonons: FE: Formation energy; BG: Band gap
PZ: Piezoelectric modulus; DC: Dielectric constant; EBG: Experimental band gap

7×6 combinations of pair-wise models



Pair-wise models: better than scratch

- Average increase in R^2: 25%
- Average decrease in MAE: 16%

Best models: GV, PH, FE ($R^2 > 0.75$)

What about MPT (or more generalizable) models?

property: little influence on FT

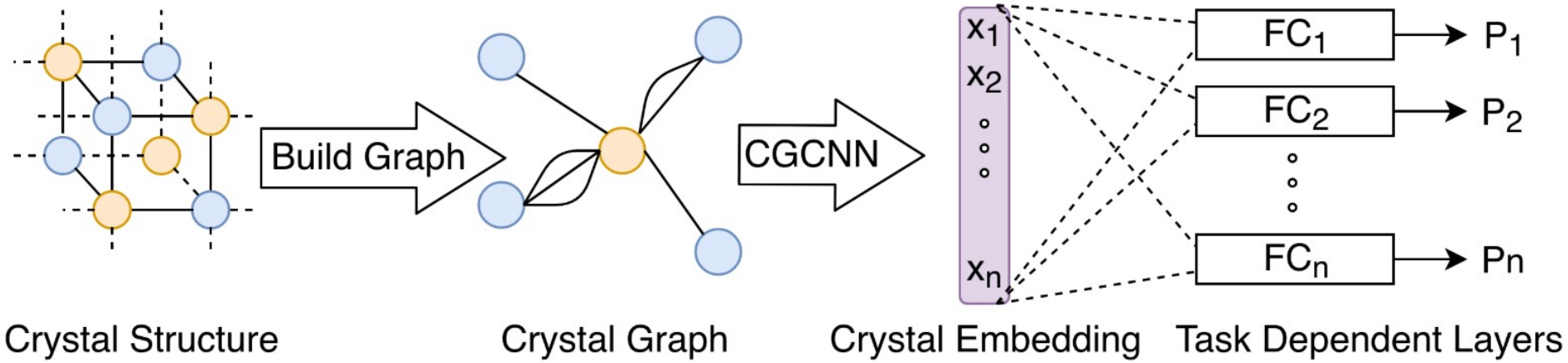
No symmetry

At capped dataset size, specific PT property is a weak handle; Normal distribution is better

Pair-wise transfer learning has significant utility

MPT: (Beta) Generalized models

Inspiration from literature: multi-task crystal graph convolutional neural network[1]



MPT models can generalize dependence of several properties on the structure

- Build cumulative dataset: 132,270 points
  - Remove overlaps
- Add task-dependent prediction heads with a one-hot encoded vector
  - Presence/absence of property
- Modify loss function

$$\mathcal{L} = \frac{1}{N}\sum_{i=1}^{N} |y_p^i - y_t^i| \delta^i$$

- PT on all (but one) property, FT on one property

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

MPT: better out-of-domain than PT-FT

Band gap of 2D materials (1,103 datapoints) from JARVIS-DFT[1]

Model	Test R^2	Test MAE
Scratch	0.635	0.148
MPT (all seven datasets)	**0.671**	**0.125**
FE(100K)	0.670	0.127
BG(50K)	0.617	0.138
PH(1256)	0.628	0.145
GV(10,987)	0.626	0.143
EBG(2,481)	0.619	0.143

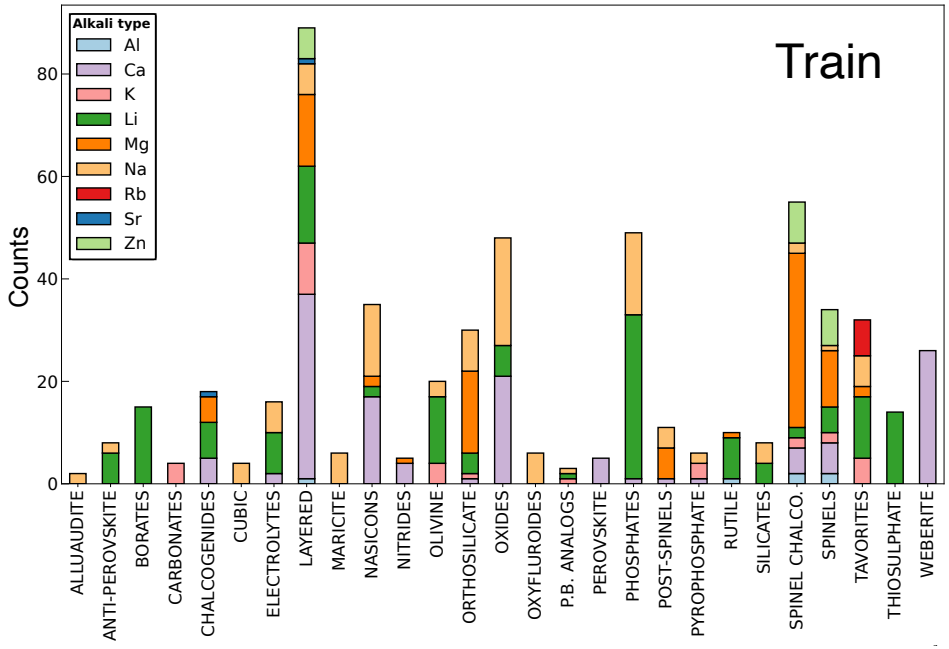
On average, MPT is 6% and 10% better on R^2 and MAE than PT-FT
Closest performer to MPT is FE: largest dataset within MPT
MPT models: may generalize quite well with more properties

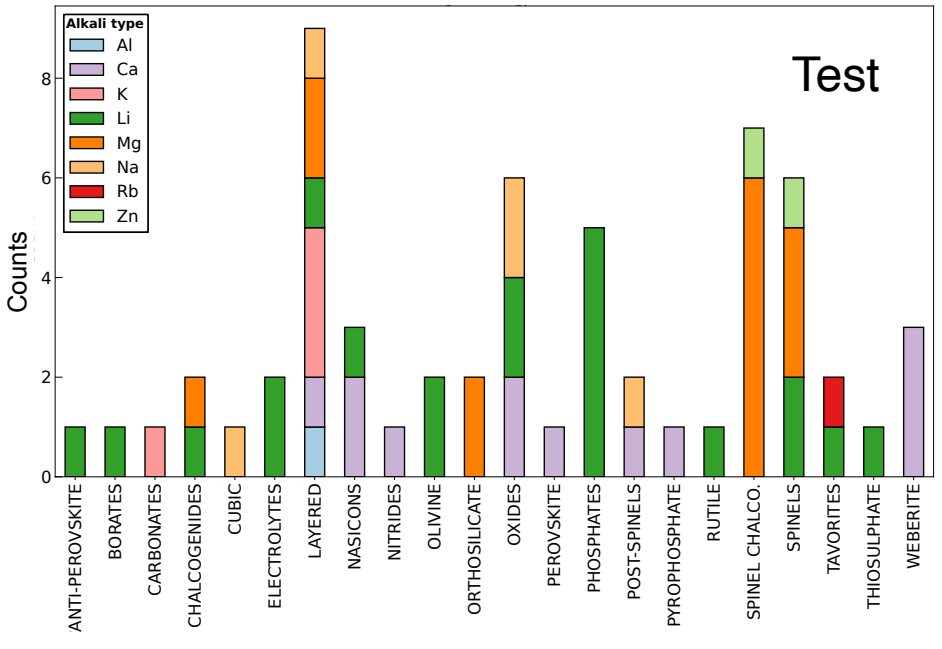
FE: Formation energy; BG: Band gap; GV: Shear modulus;
EBG: Experimental band gap

1. Choudhary et al. **npj Comput. Mater.** *6*, 173 (2020)

How to predict migration barriers?
Using the MPT model

Careful split of dataset



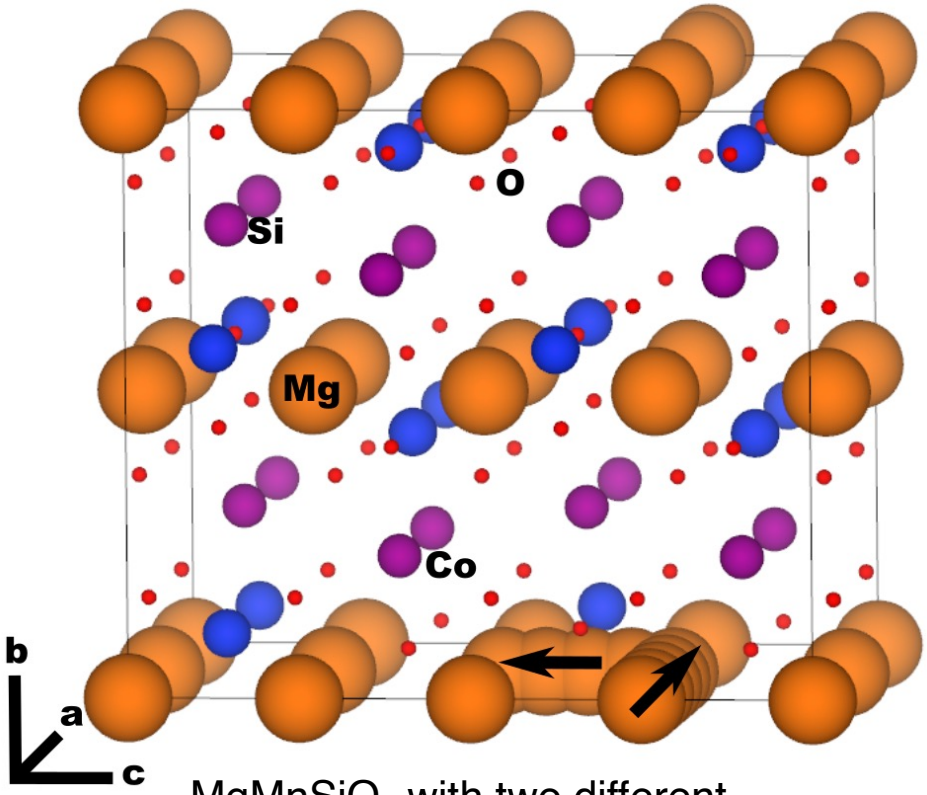


Structural groups

559 datapoints in train; 60 in test

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

Distinguish multiple paths in a structure: use modified model architectures

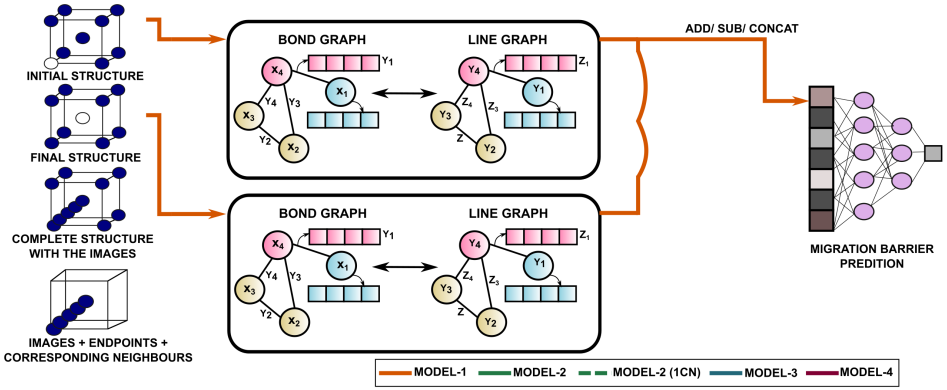


$MgMnSiO_4$ with two different pathways in the same structure

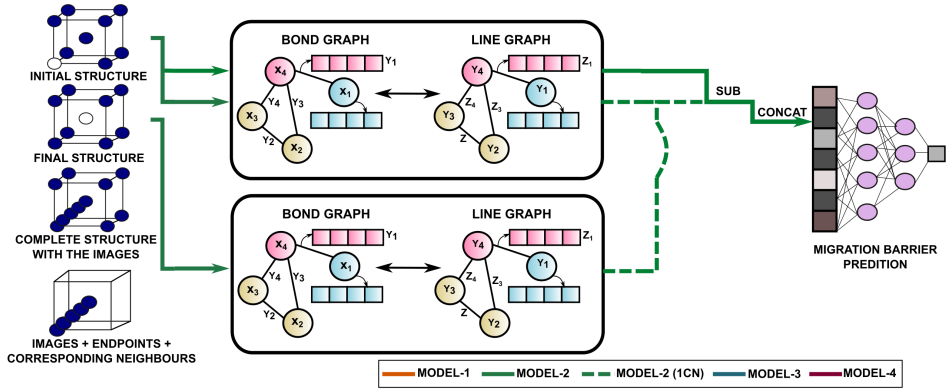
PT: MPT, FT: E_m

Distinguish multiple paths in a structure: use modified model architectures

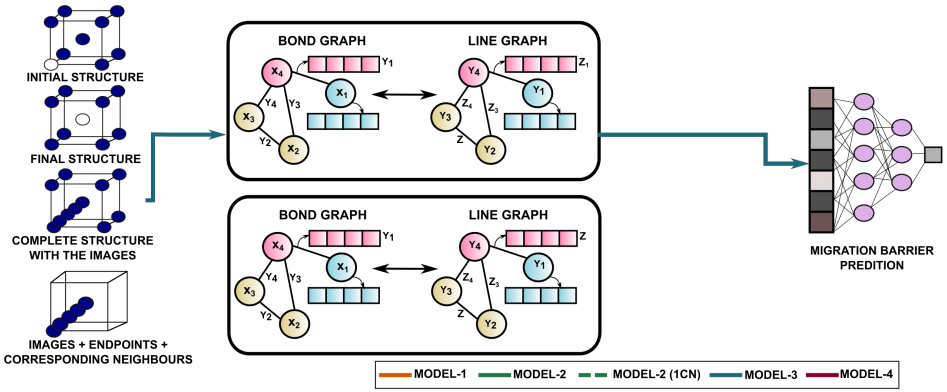
Model 1: Take initial and final as input



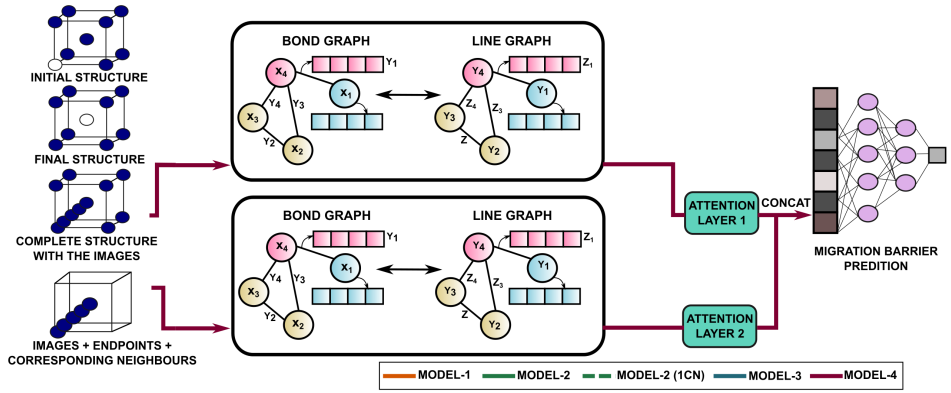
Model 2: Take initial and delta as input



Model 3: Band as input

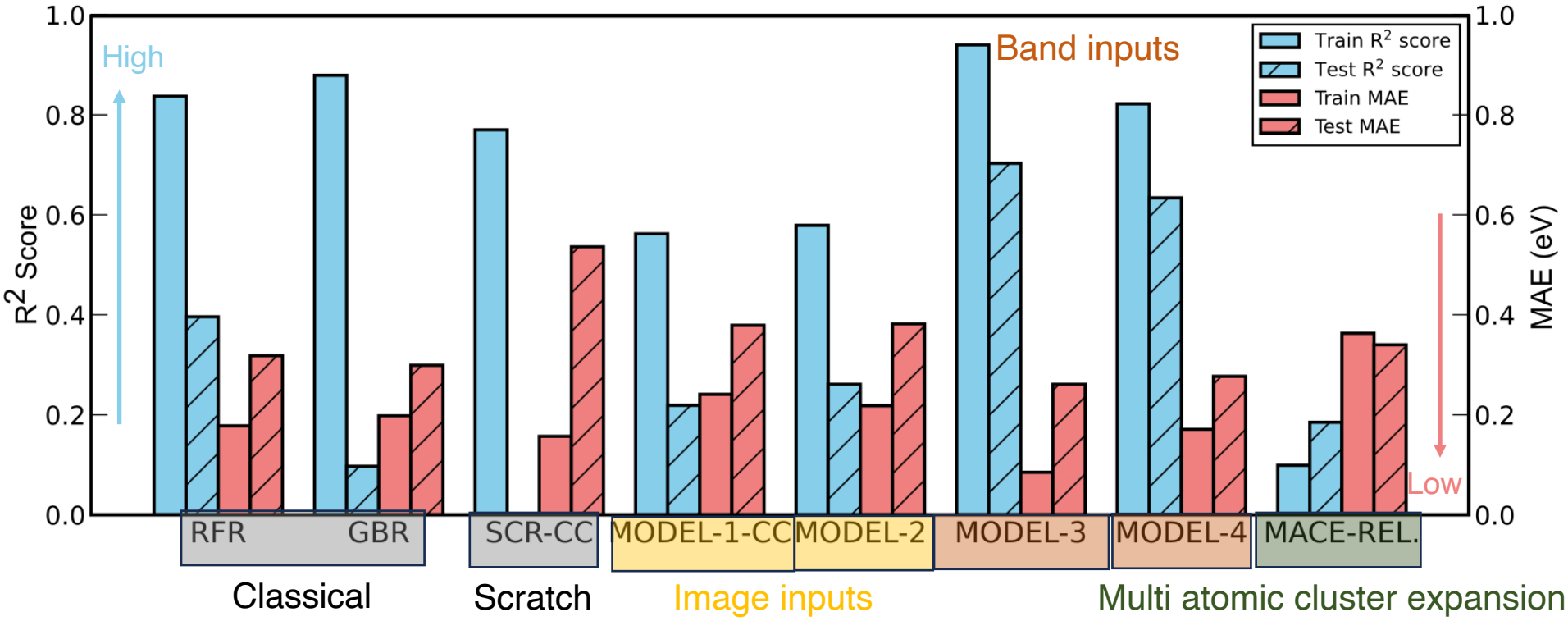


Model 4: Band with attention



PT: MPT, FT: E_m

Model-3: best performing model

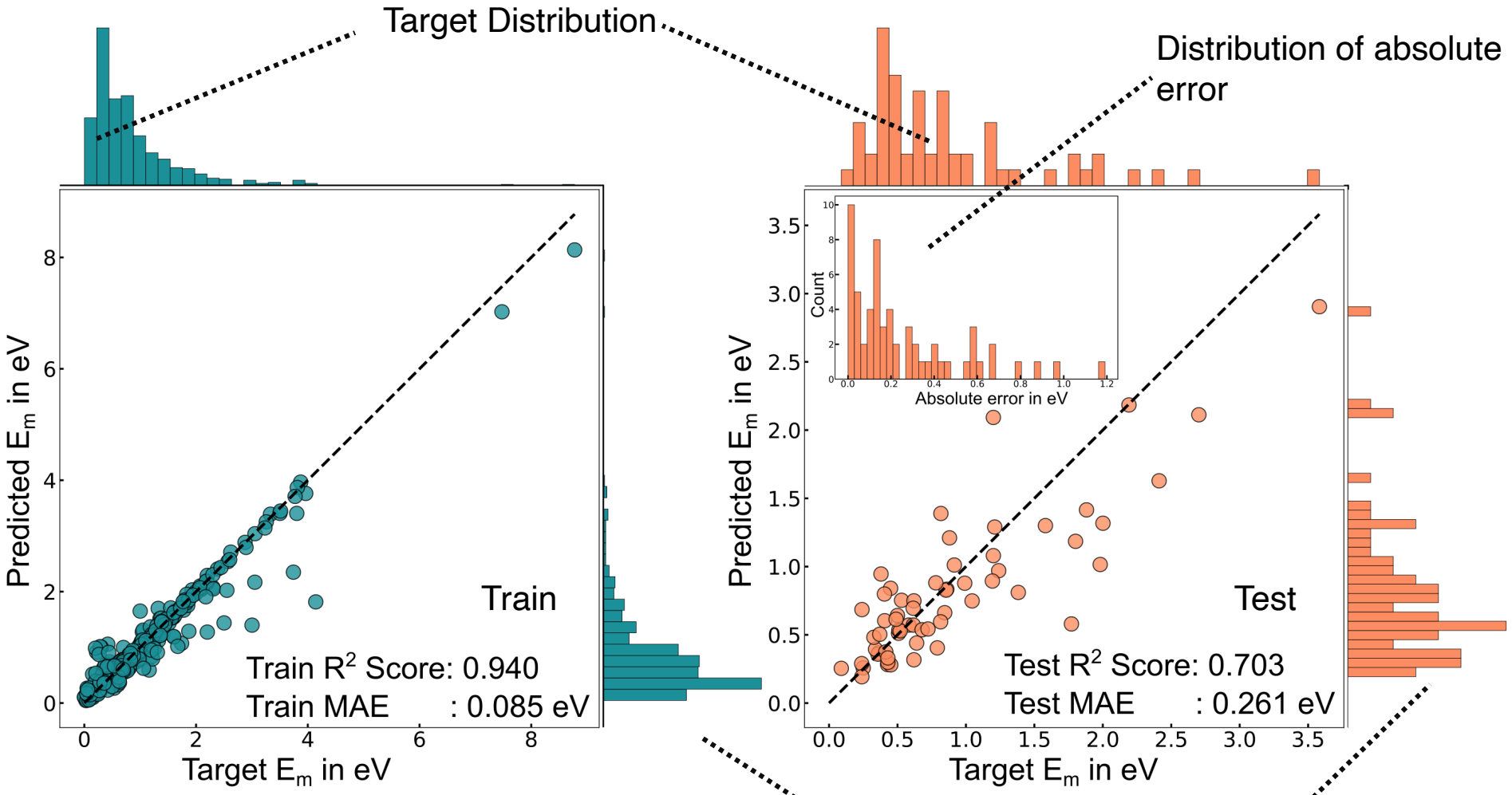


Model-3 performance (test set statistics, hashed bars):

- R^2: 0.703, MAE: 0.261 eV (~1.4-1.8× DFT error)
- Better scores than classical and scratch models
- Versus MACE: slightly better MAEs, significantly better R^2
- Identifies multiple migration pathways in a structure (band inputs > image inputs)
- Model-4 is closest performer to Model-3

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

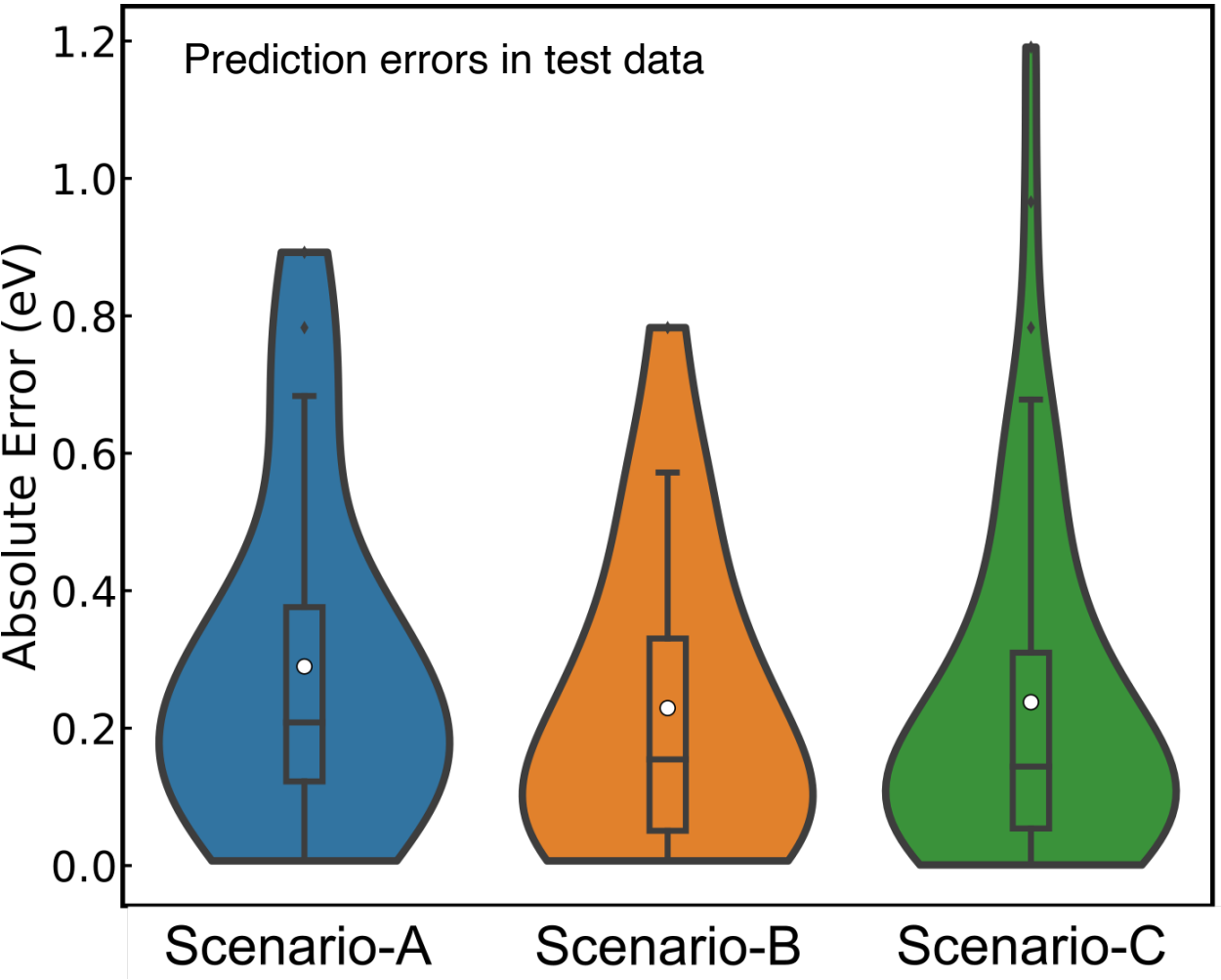
Model-3 performance



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

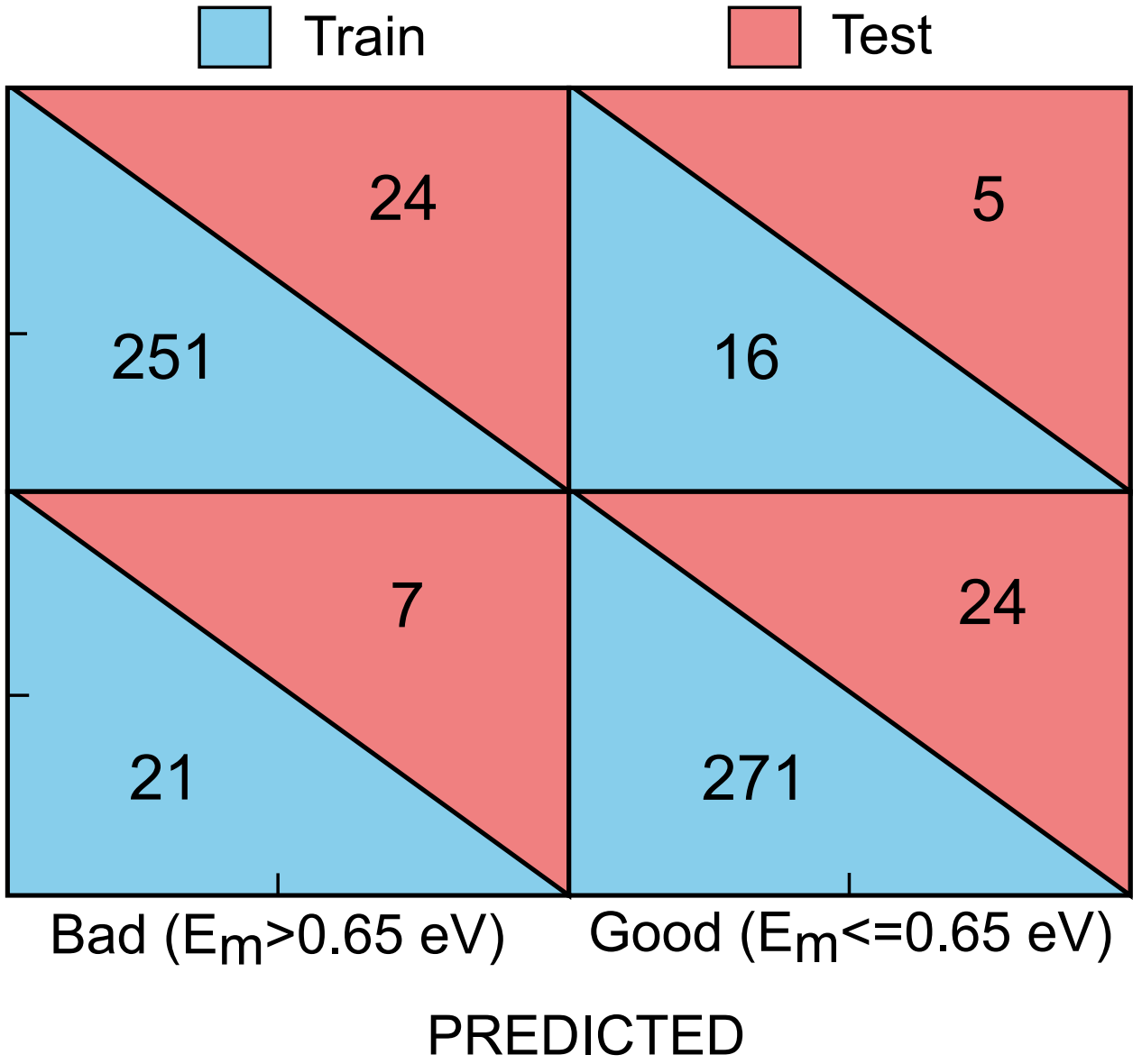
Generalization across intercalant composition (charged vs. discharged)

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

Lower mean/median errors for Model-3 in scenarios B & 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

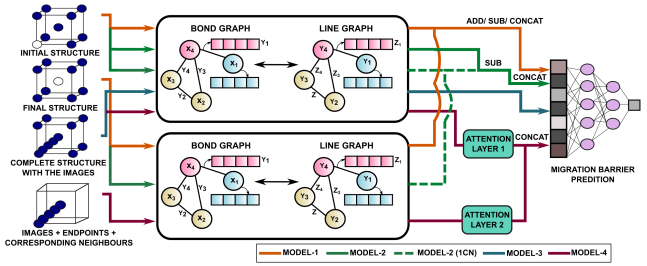
Accuracy: 80%

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



"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, **arXiv**, 2508.06459 (2025) [under review].

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