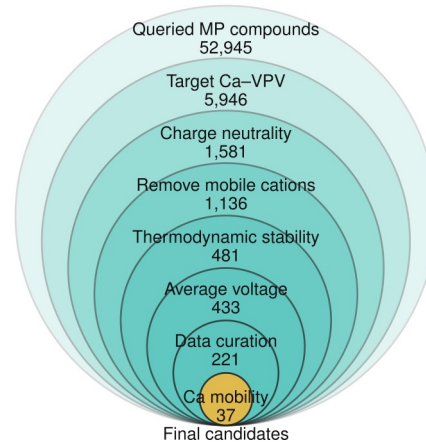




Machine learning accelerated screening of positive electrodes for calcium batteries

Sai Gautam Gopalakrishnan

Materials Engineering, Indian Institute of Science

saigautamg@iisc.ac.in; <https://sai-mat-group.github.io>

Acknowledgments



Jun 2026



Dereje



Achintha



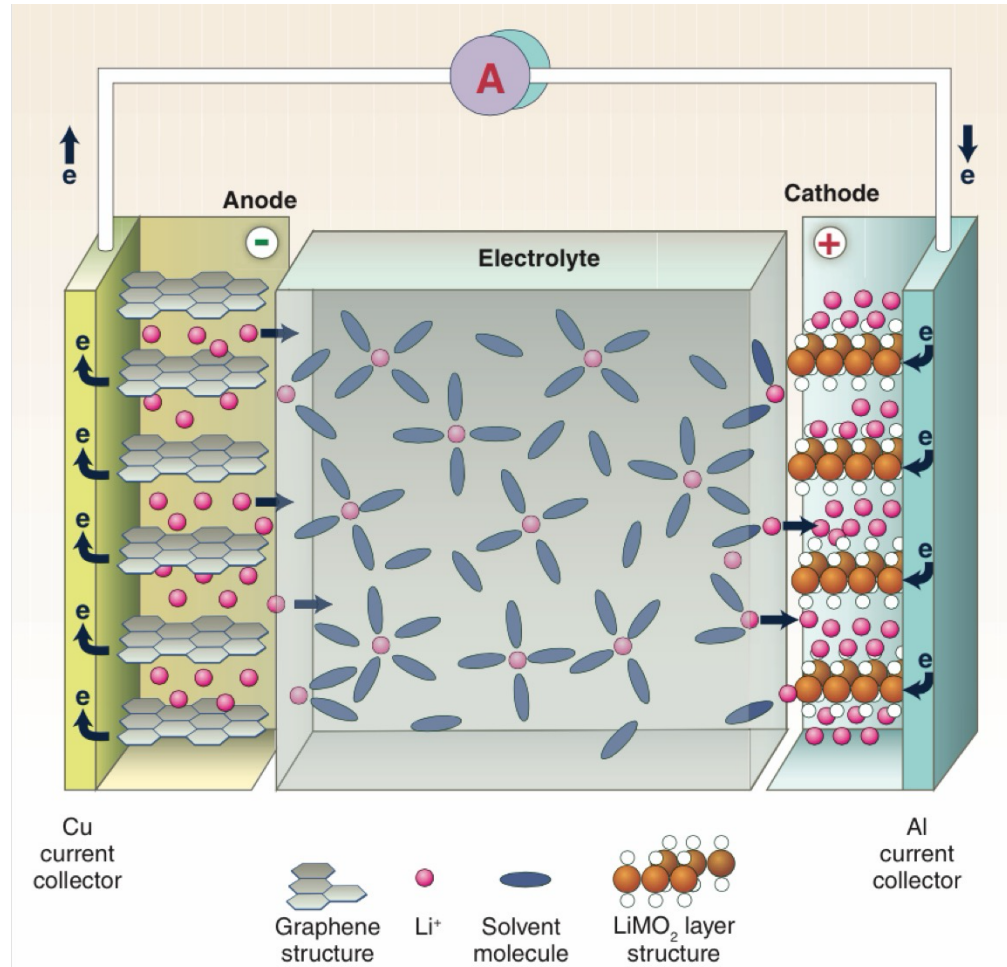
Jureca
(Germany)



SERC (IISc)



Li-ion batteries: state-of-the-art and headwinds

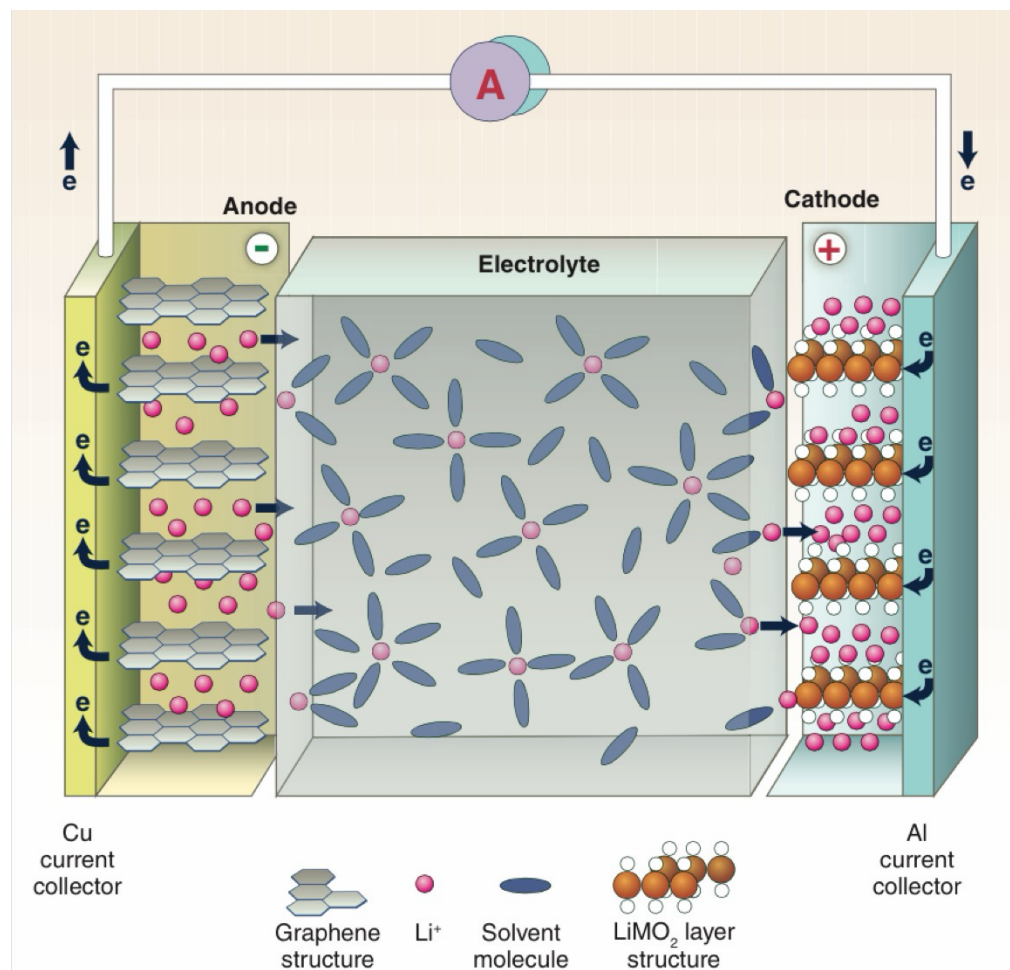


Li-ion batteries: state-of-the-art energy storage technology

- Supply chain constraints
- Safety
- Fundamental limits on energy density

Alternative energy storage technologies?

Li-ion batteries: state-of-the-art and headwinds



Li-ion batteries: state-of-the-art energy storage technology

- Supply chain constraints
- Safety
- Fundamental limits on energy density

Alternative energy storage technologies?

Voltage (V): Potential to do work

Capacity (Ah/g or Ah/l): Amount of charge stored

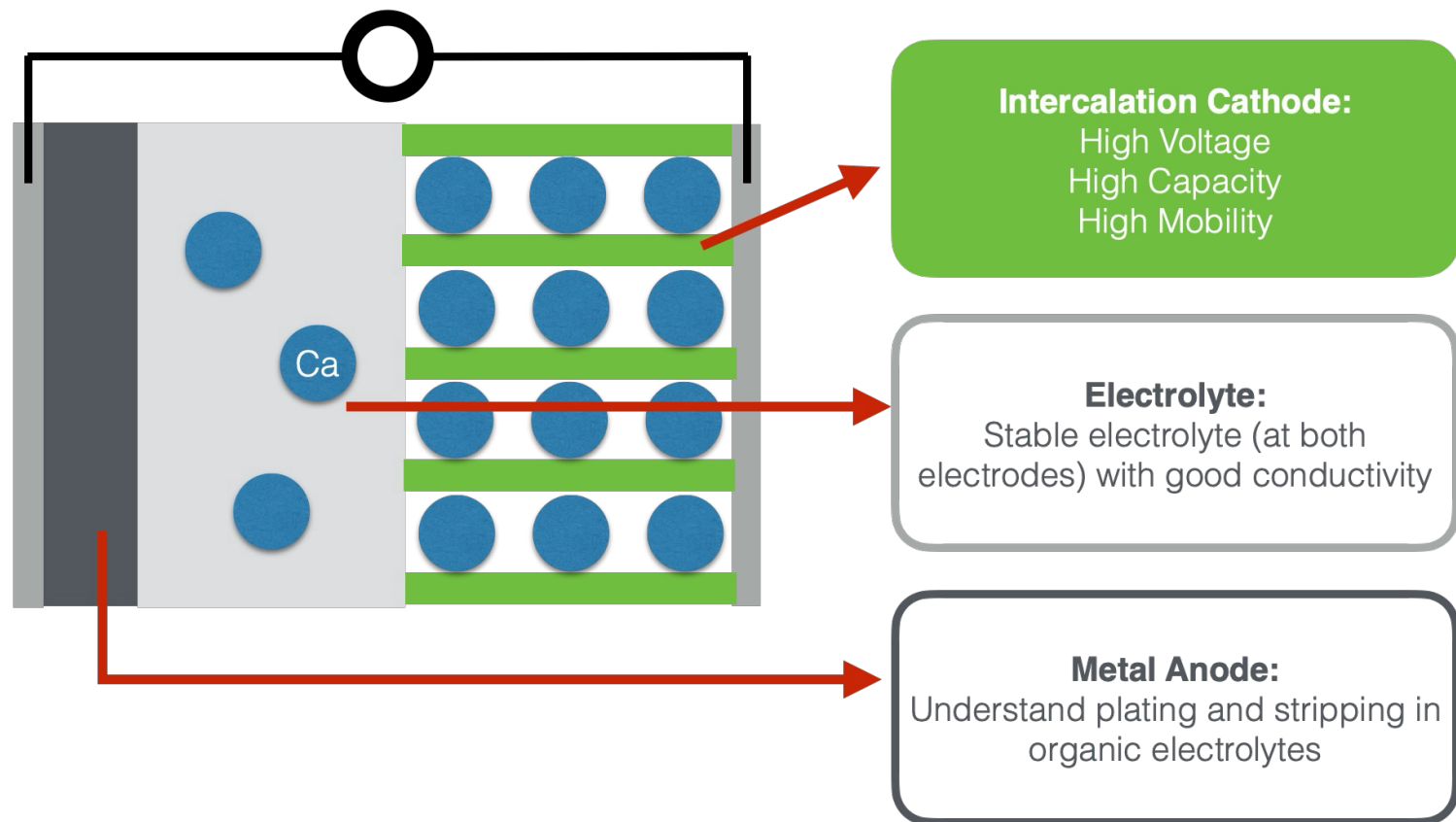
Stability (energy above convex hull, eV/atom): Reversibility, cycle life

Energy density (Wh/kg or Wh/l): Normalized energy stored

Power or rate performance (W/kg or W/l): Normalized power

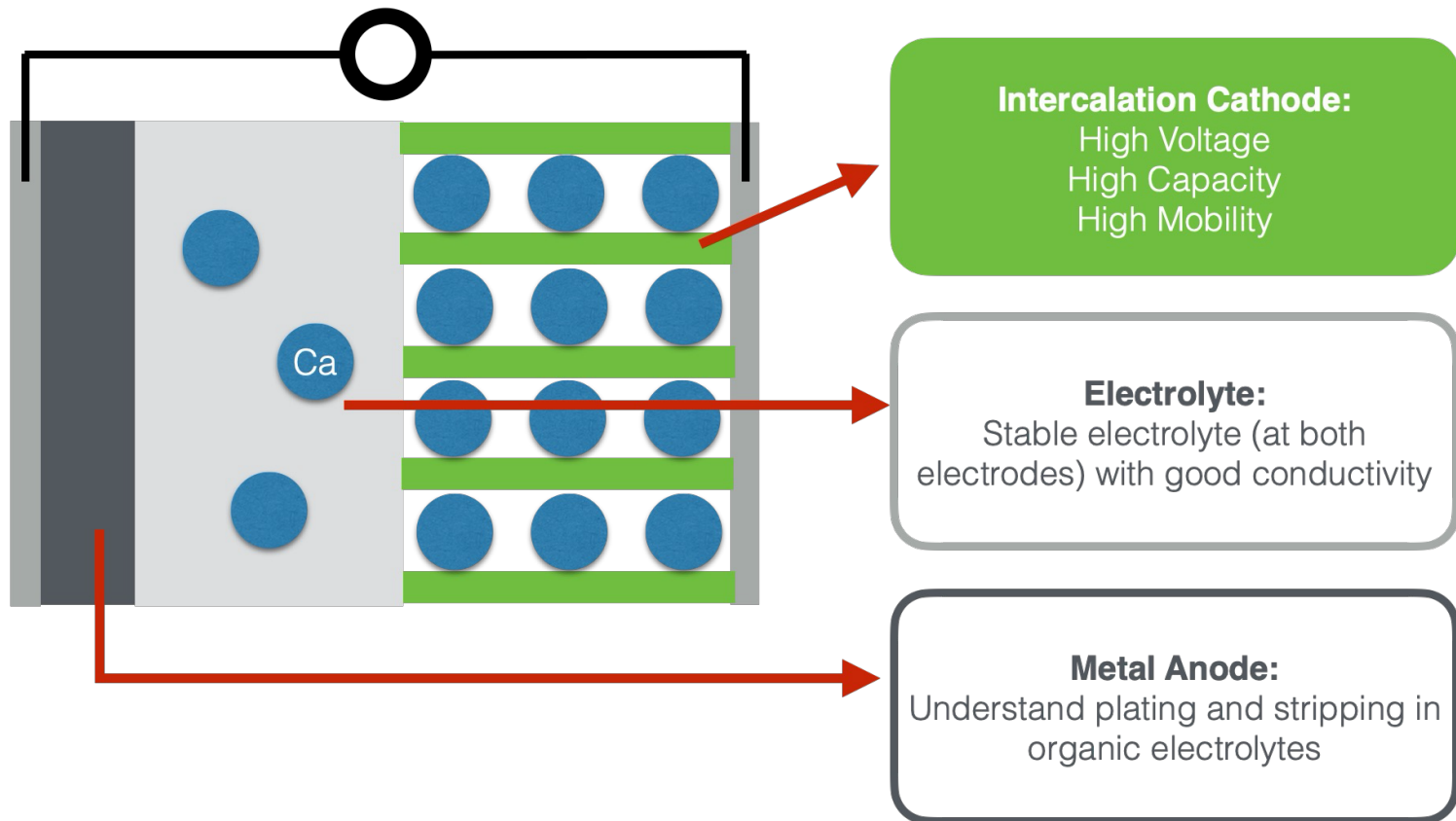
Charged/discharged structure: deintercalated/intercalated electrode

Ca batteries: alternative technologies that require cathodes



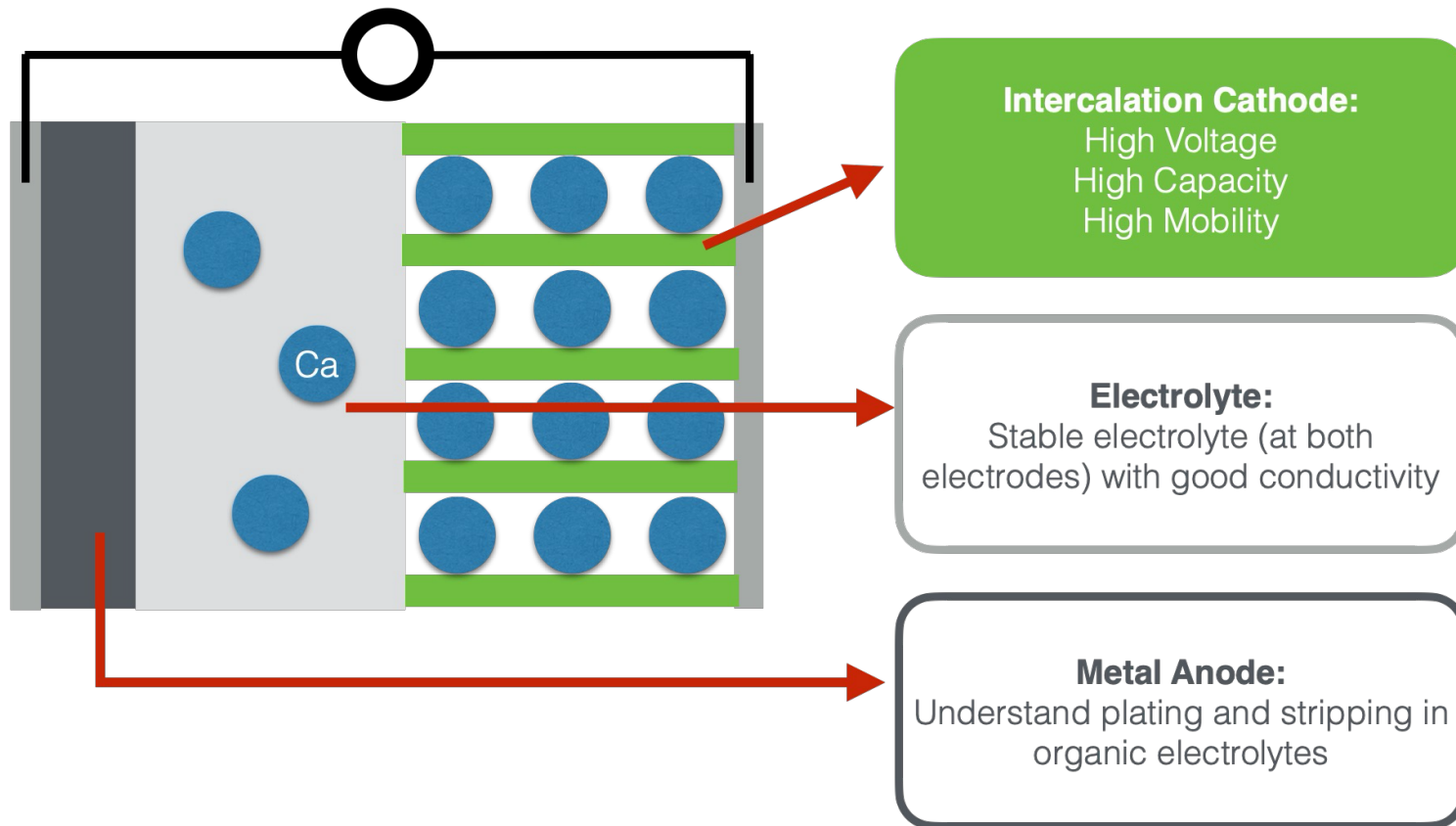
Ca batteries: alternative technologies that require cathodes

- Higher volumetric capacity (~ 2077 Ah/l) for Ca vs. Li in graphite (~ 800 Ah/l) $\rightarrow$ higher energy densities
- Less constrained geopolitically; similar standard redox potential to Li
- Need thermodynamically (meta)stable cathodes with high **voltage** and high *mobility*



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Computationally screen for promising cathodes?

Previous screening efforts: low success

Filtering through
ternary Ca-containing
compounds

(1) 210,000+

(2) 365

(3) 181

(4) 66

(5) 10

2

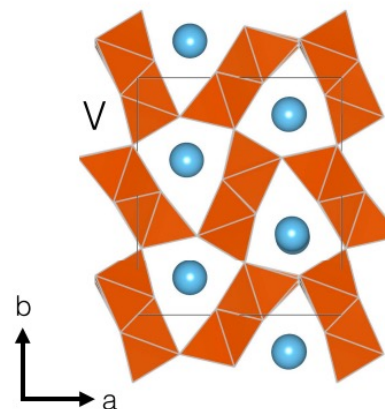
(1) Compounds in ICSD

(2) Ternary $\text{Ca}_i\text{M}_j\text{Z}_k$

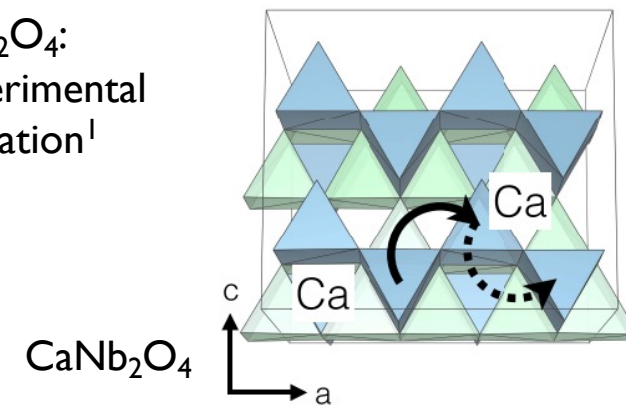
(3) $\text{Ca}_i\text{TM}_j\text{Z}_k$ without undesired
elements & polyanions

(4) $\text{Ca}_i\text{TM}_j\text{Z}_k$ polymorph
matched by charged TM_jZ_k

(5) Stable $\text{Ca}_i\text{TM}_j\text{Z}_k$ selected



CaV_2O_4 :
experimental
validation¹



Previous screening efforts: low success

Filtering through ternary Ca-containing compounds

(1) 210,000+

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2

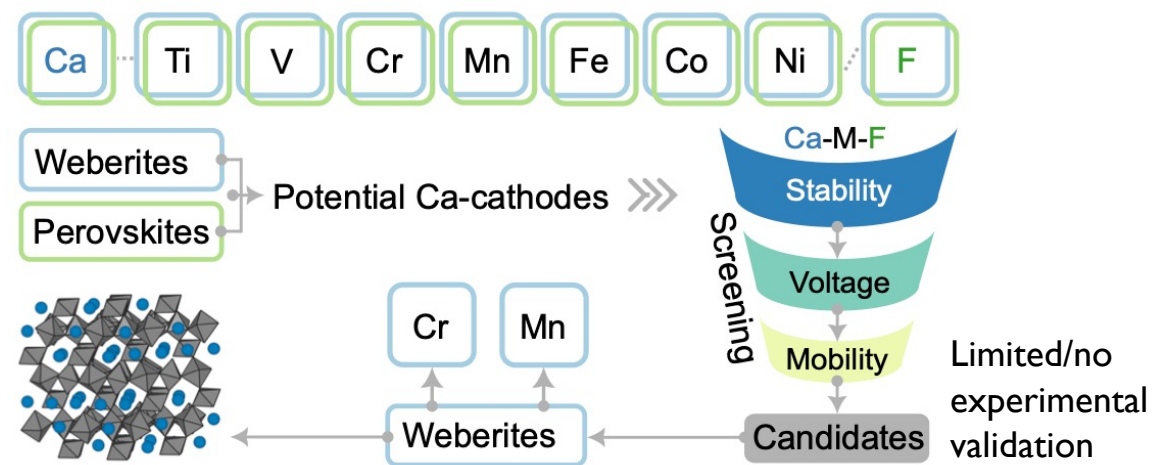
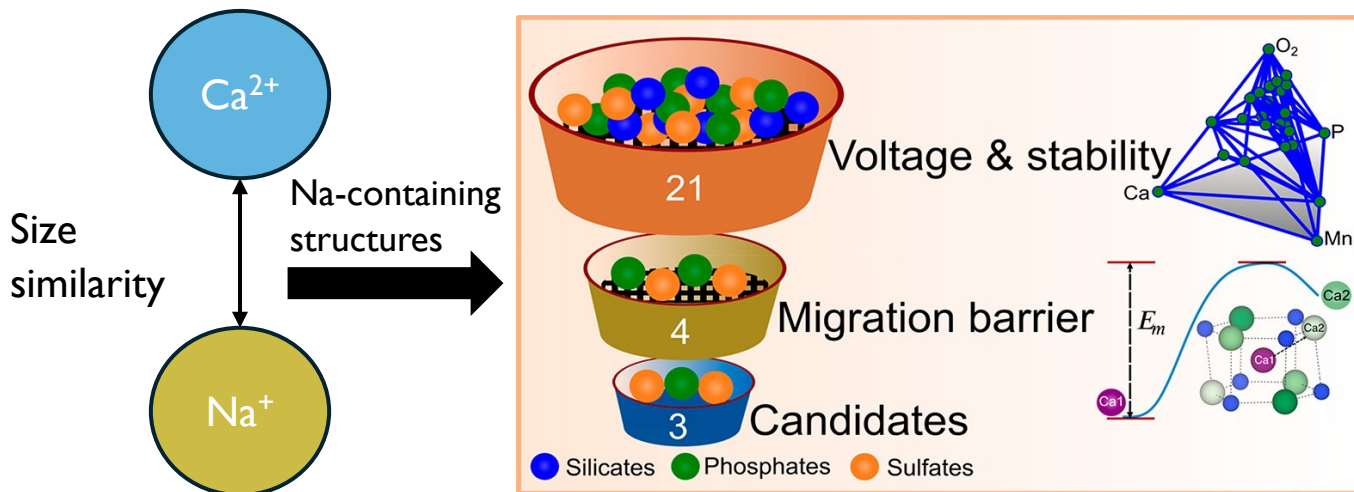
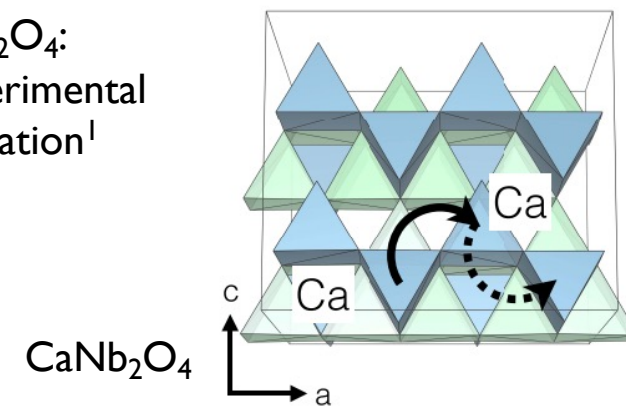
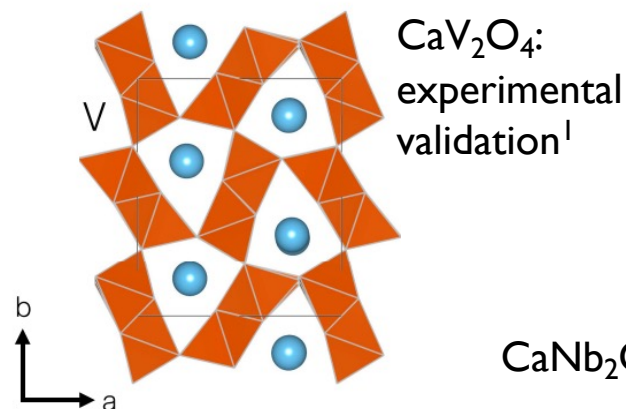
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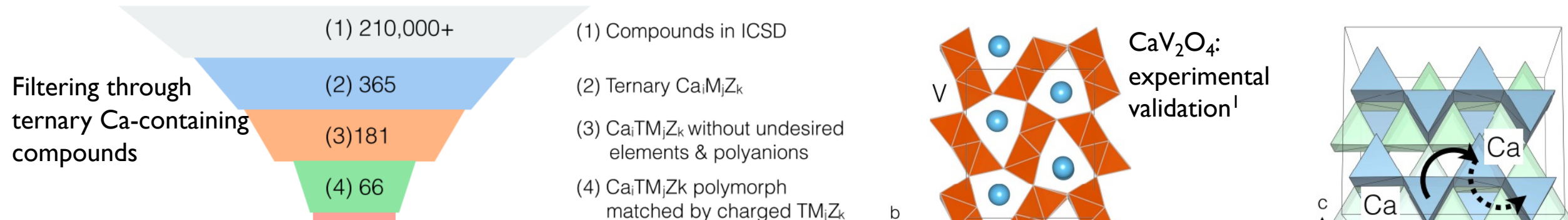
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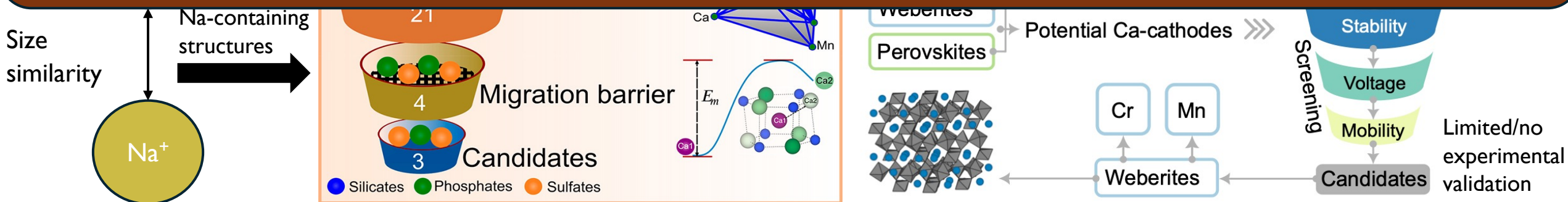


Previous screening efforts: low success



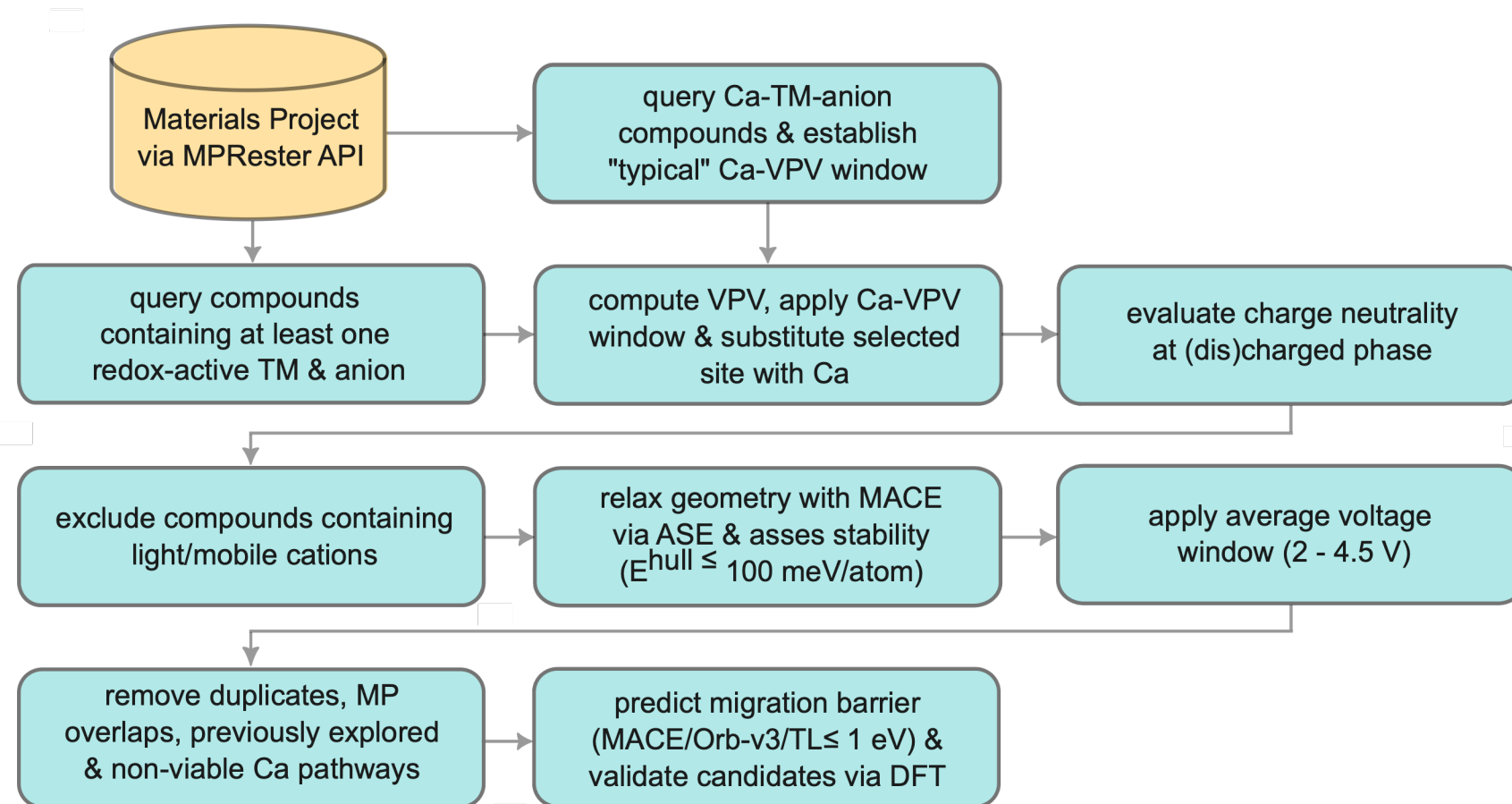
Need to re-define screening strategy to have a higher success rate

- Look at chemical spaces that do not have *Ca a priori*?
- Use machine learning (ML) techniques to expand search space?



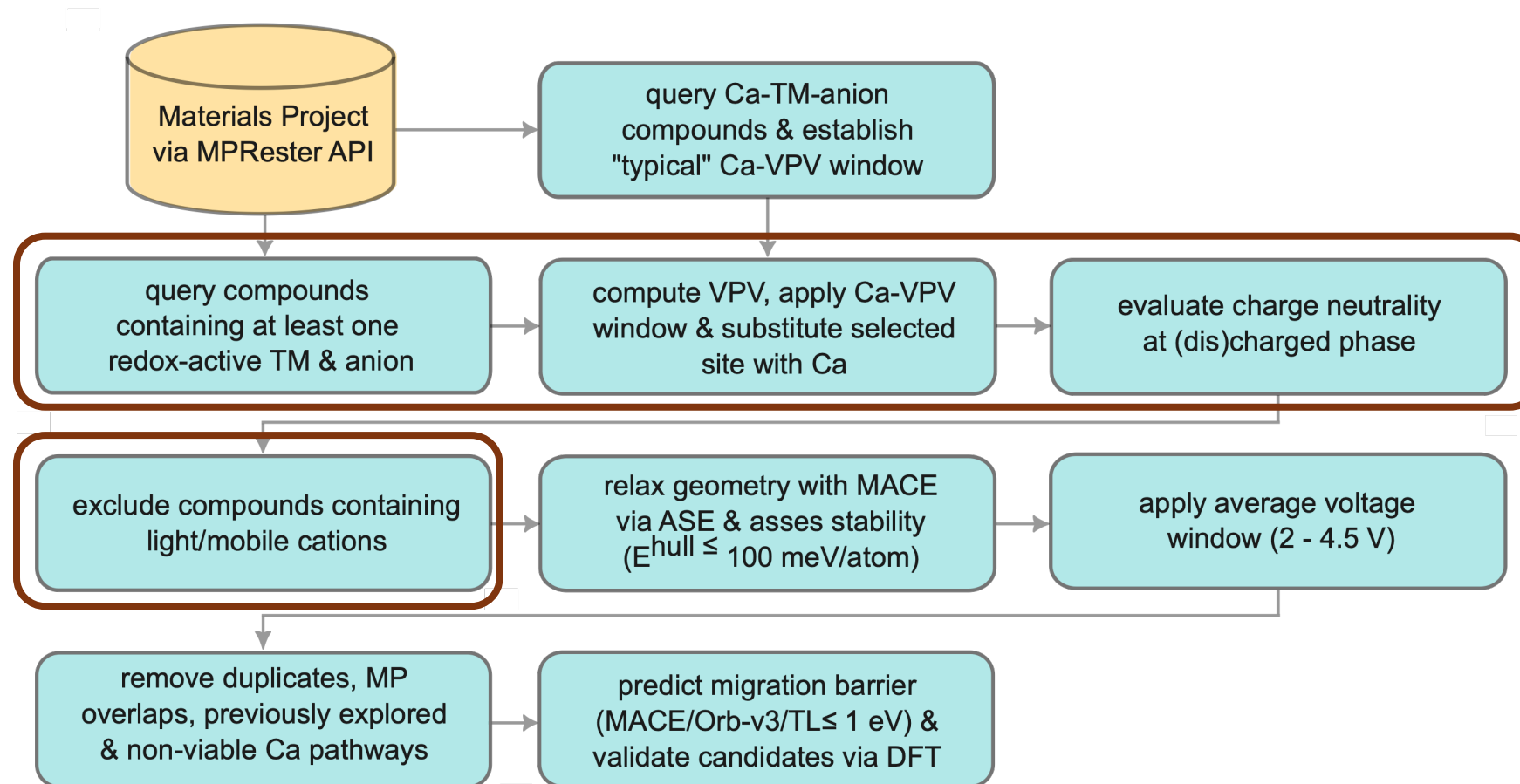
Objective: find cathodes without *a priori* Ca

Workflow: (human) innovation + (ML) speed



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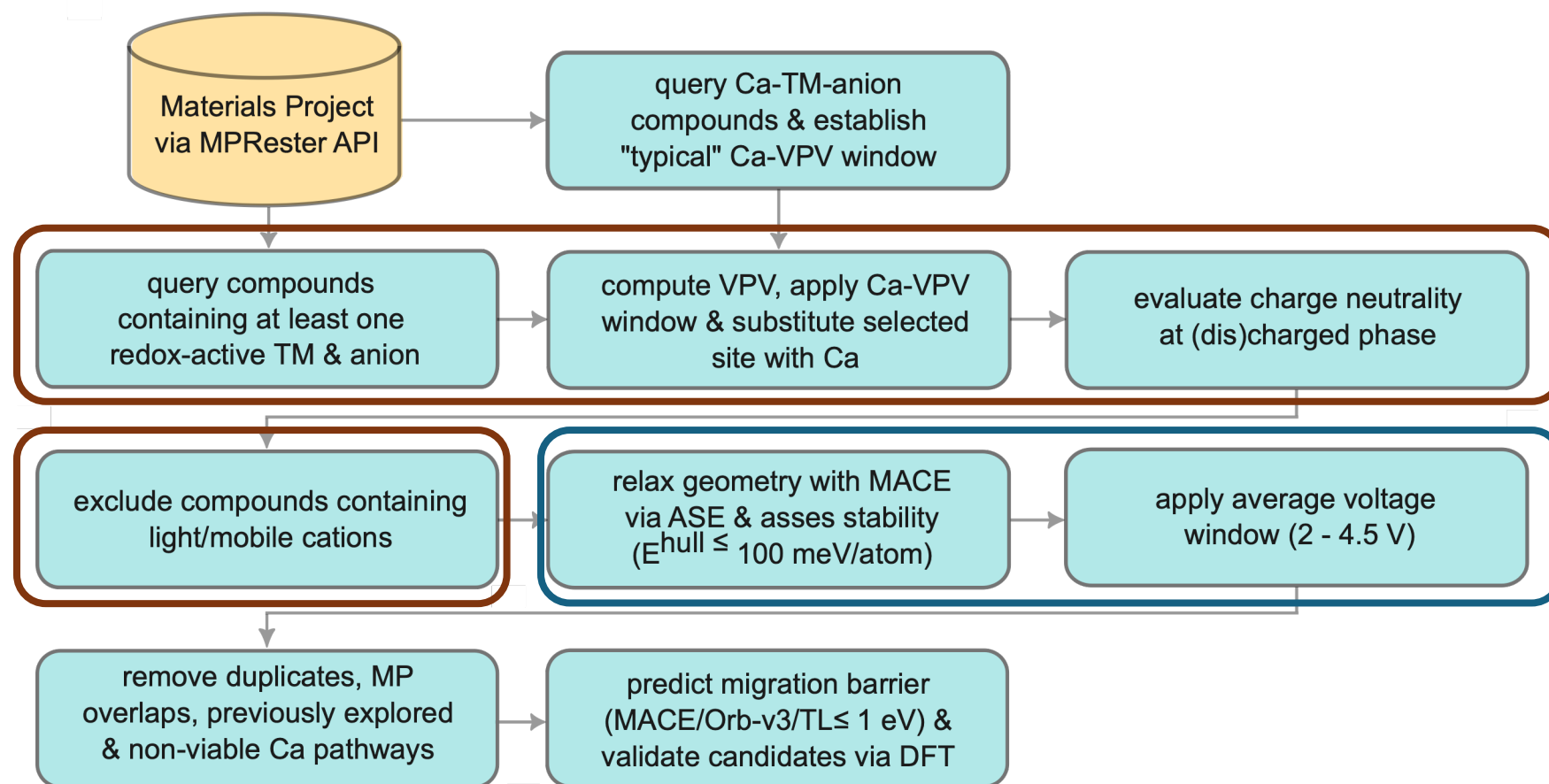


Geometric and chemical criteria: human inputs

- Removes 'unphysical' and 'non-realistic' candidates

Objective: find cathodes without *a priori* Ca

Workflow: (human) innovation + (ML) speed



Geometric and chemical criteria: human inputs

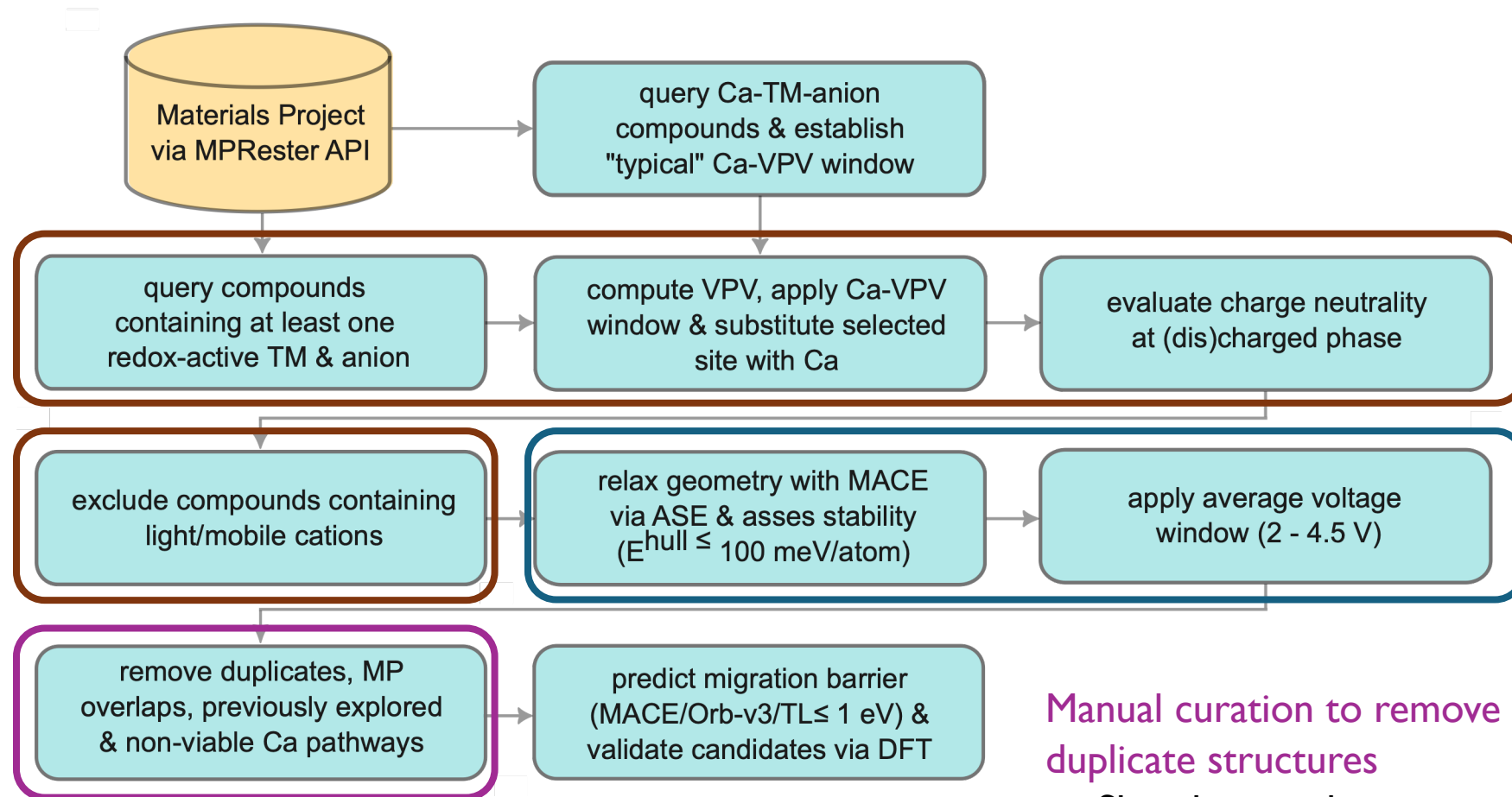
- Removes 'unphysical' and 'non-realistic' candidates

ML inputs: improves speed

- Typically done with high-throughput density functional theory (DFT)
- Foundational machine learned interatomic potentials (MLIPs) used

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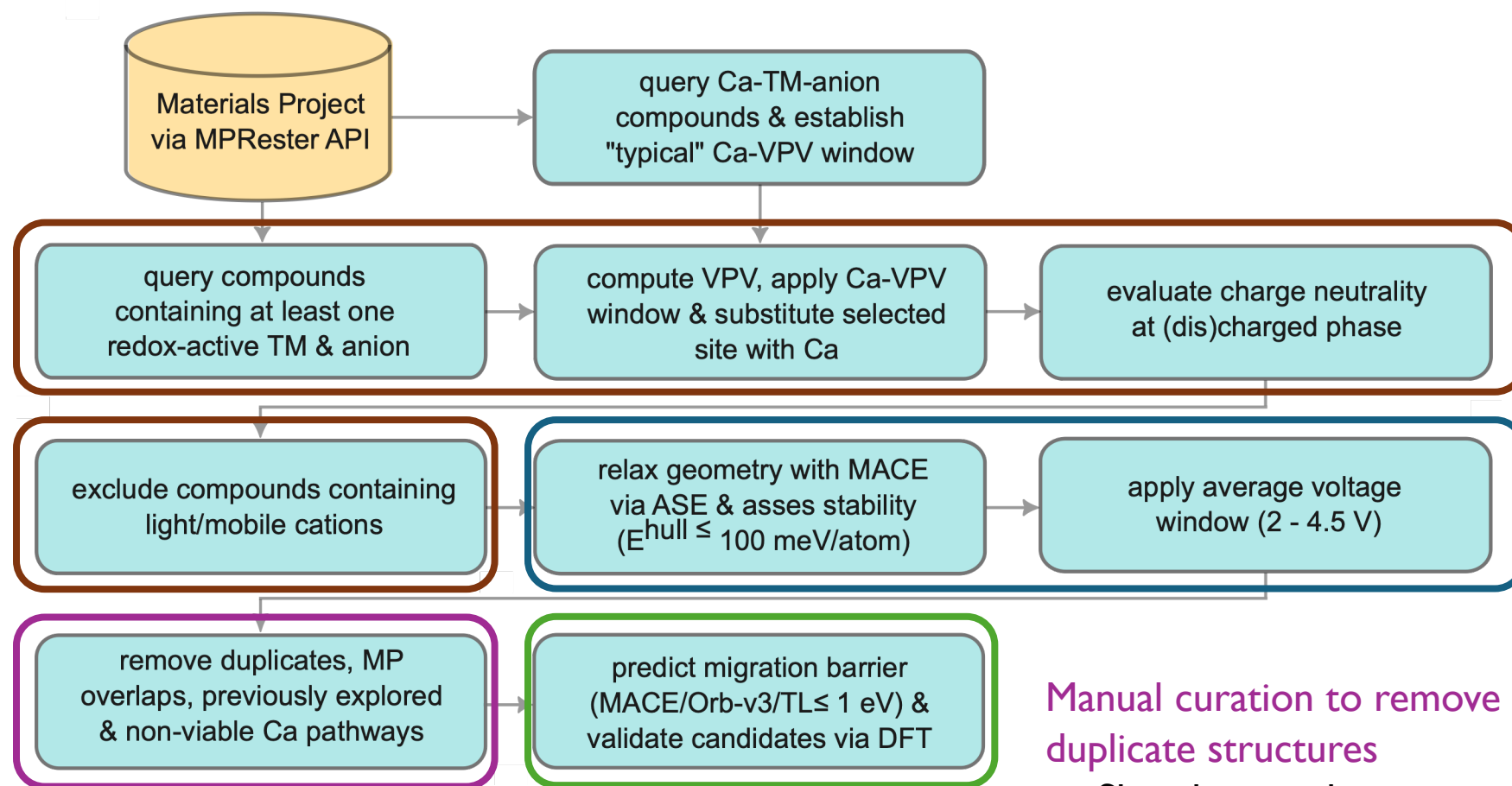
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Manual curation to remove duplicate structures

- Slow, but can be automated

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ML-filtered candidates, DFT validation

- Validation is computationally heavy

Manual curation to remove duplicate structures

- Slow, but can be automated

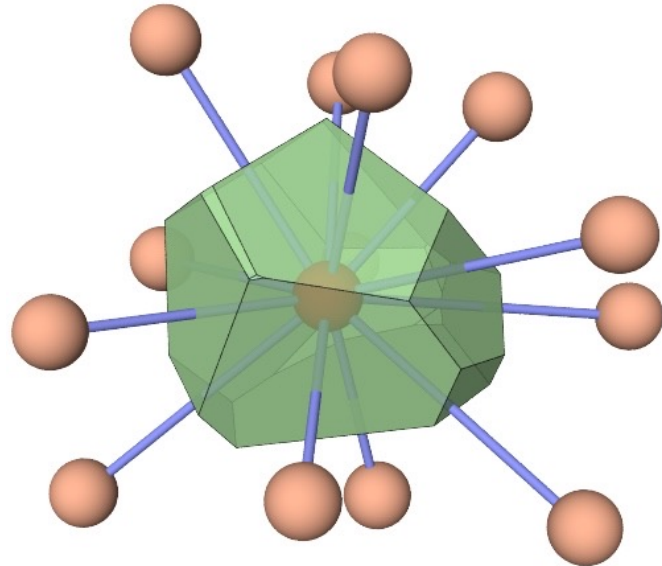
Geometry + chemical + MLIP screening

Find suitable structures: use geometry

- Need structure to have suitable cation sites or interstitial/void sites to host Ca
 - Check geometric compatibility via Voronoi polyhedral volume (VPV)

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- Need structure to have suitable cation sites or interstitial/void sites to host Ca
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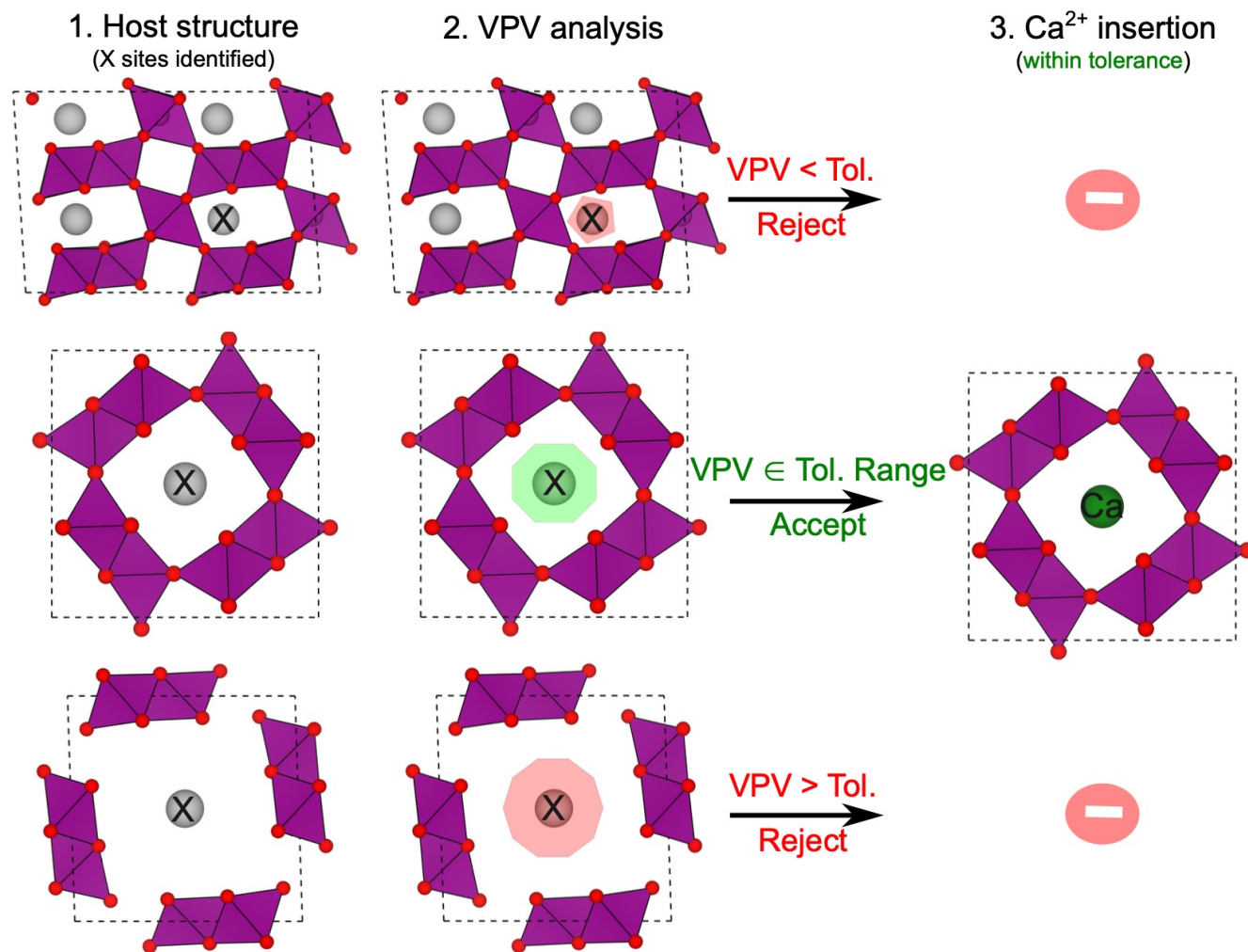
Polyhedral volume that is 'similar' across different coordination environments

- Possibly more robust than using ionic or atomic radius

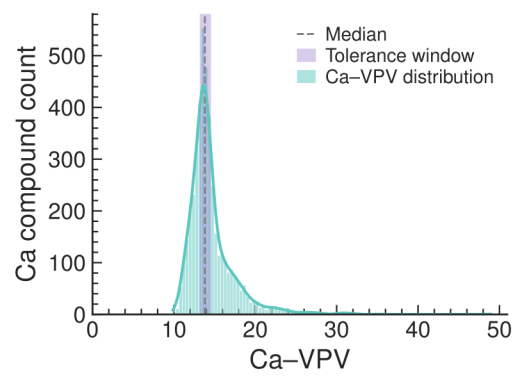
Define an 'ideal' VPV for Ca

- Based on known/explored (at least ternary) structures in MP that contain Ca

Find suitable structures: use geometry



Apply VPV on **52,945** structures: **5,946** filtered

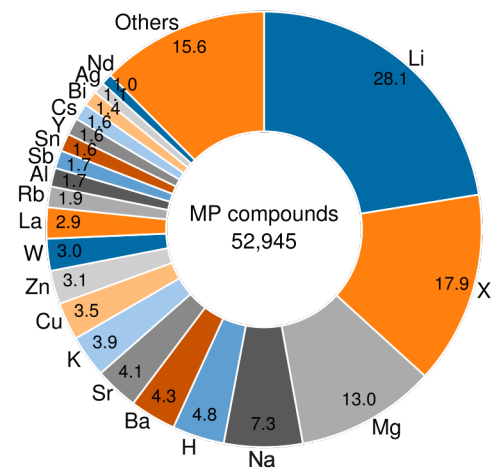
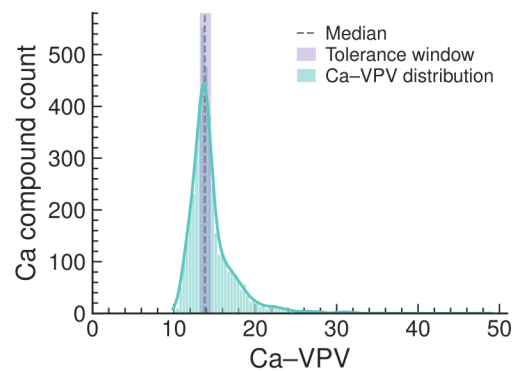


Median: 13.86 Å³

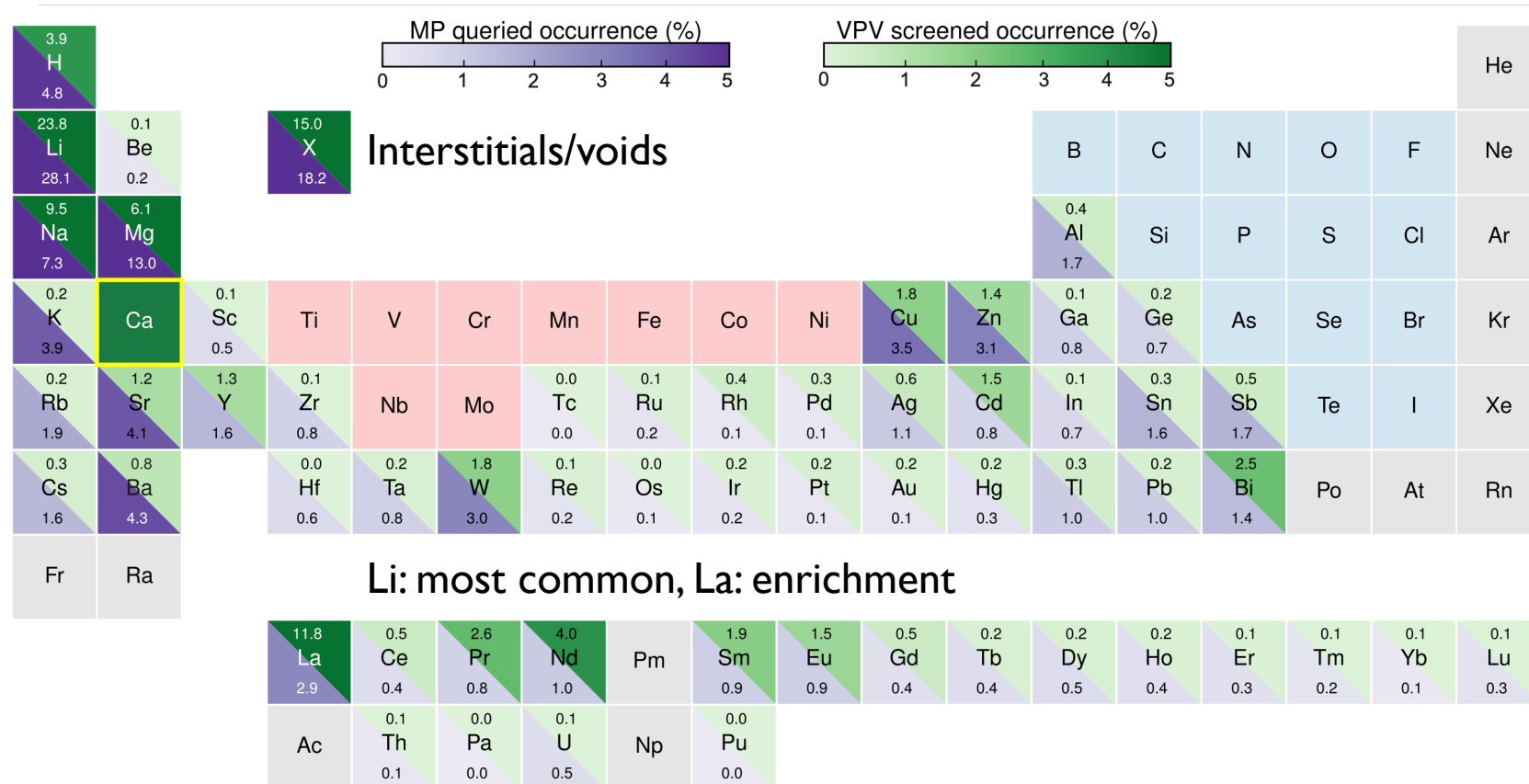
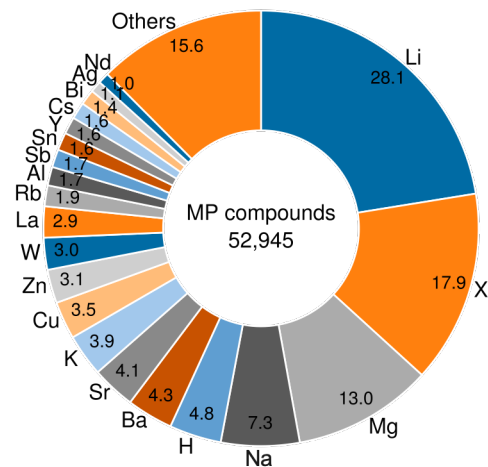
Window: 13.17-14.56 Å³

Median VPV: obtained from 4,350 Ca-containing structures in MP

Apply VPV on **52,945** structures: **5,946** filtered

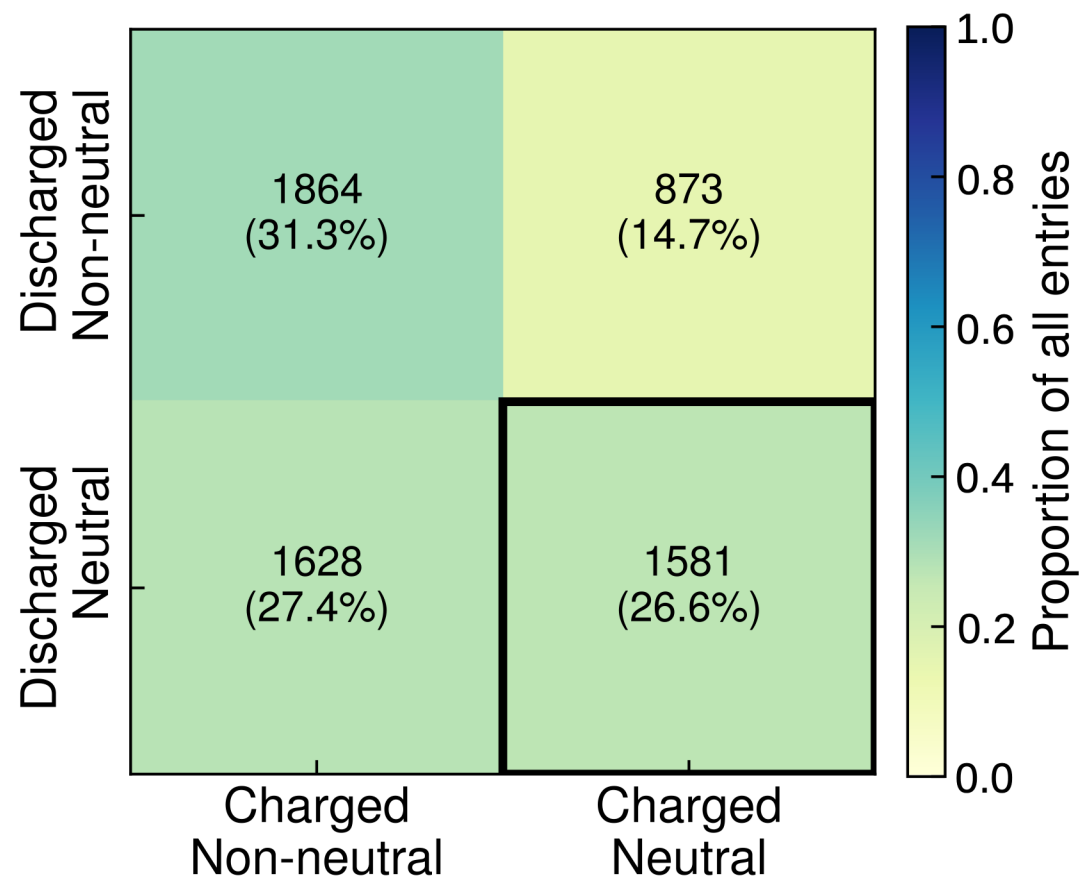


52,945: at least one redox-active transition metal and at least one anion



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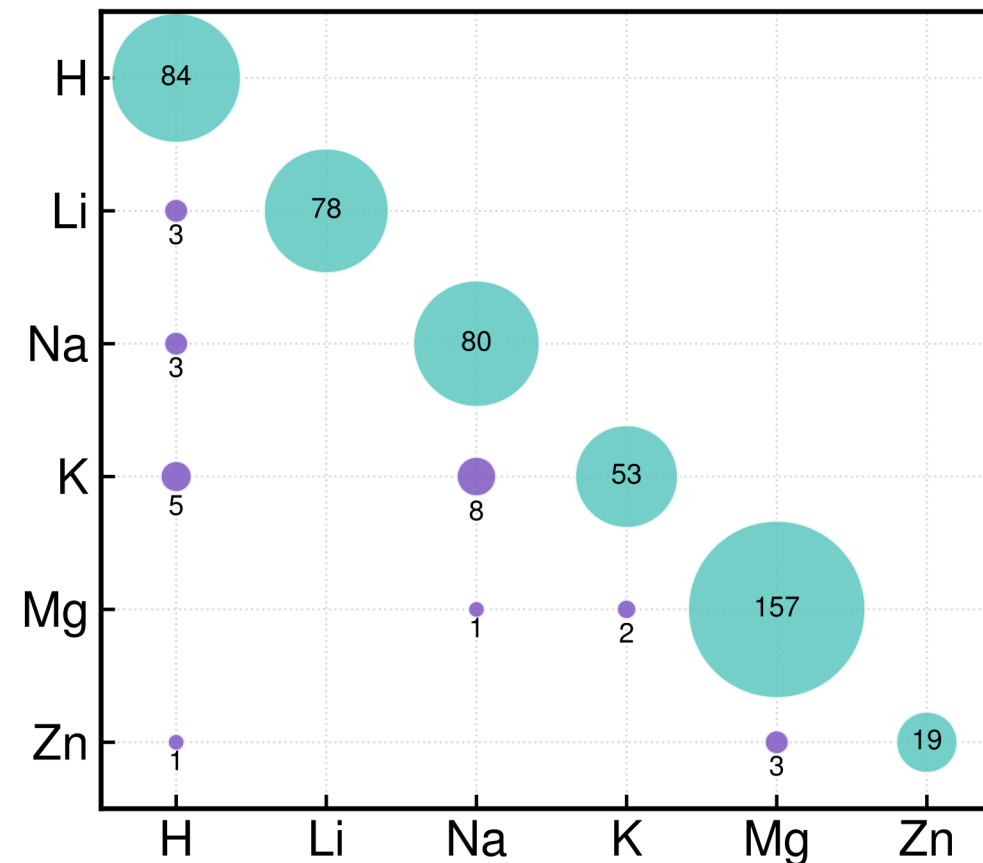
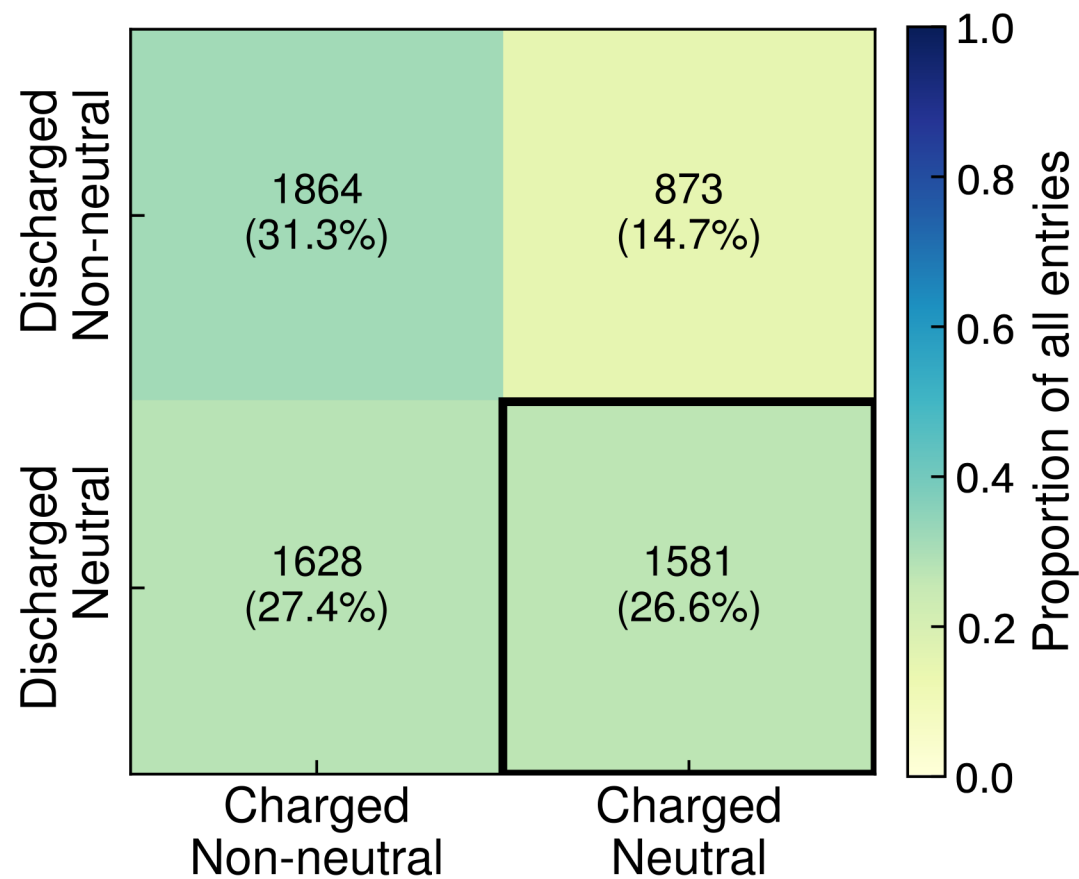
Charge neutrality and non-Ca mobile cation filters: 1,136



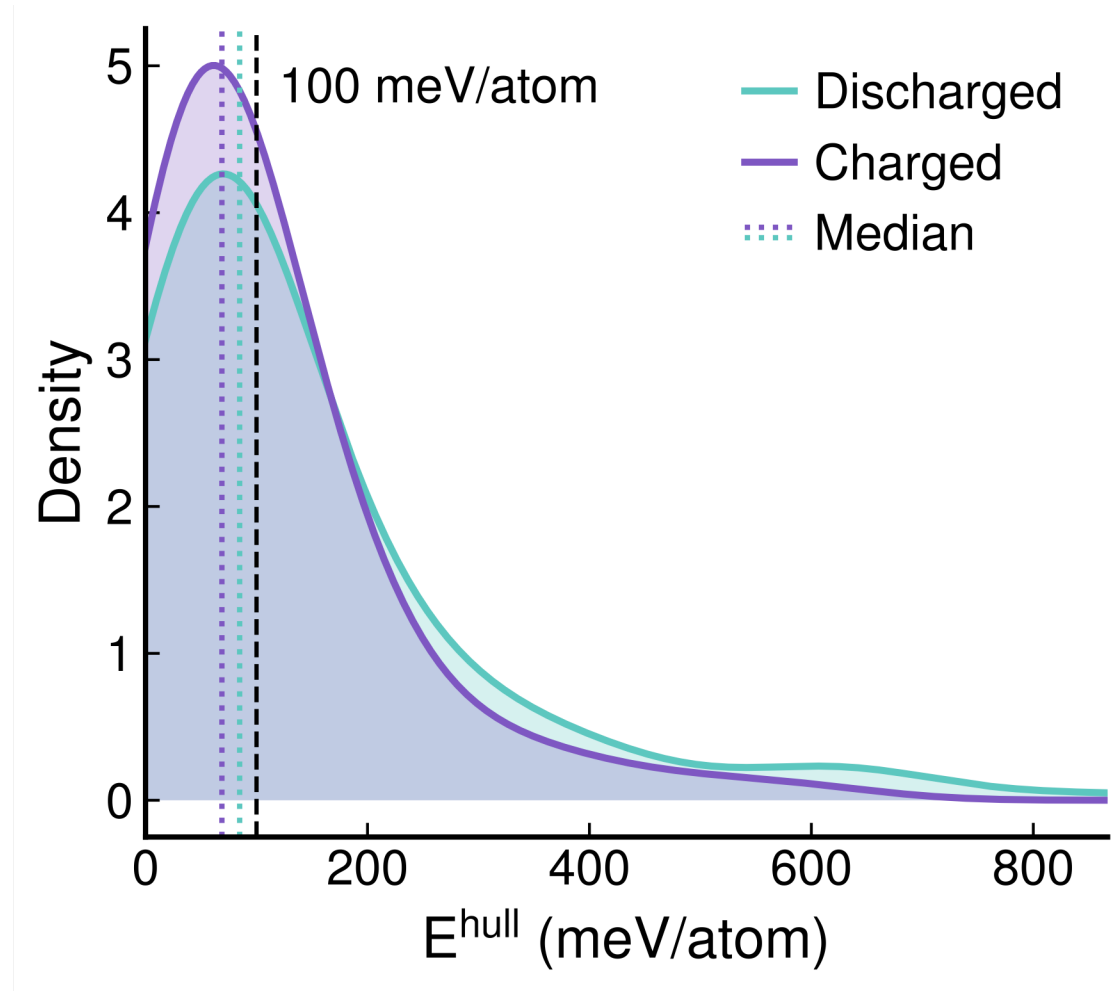
Assign oxidation states per chemical rules to determine charge neutrality

- Redox-active transition metals allowed any fractional oxidation state between +2 to +4, except V (+2 to +5)

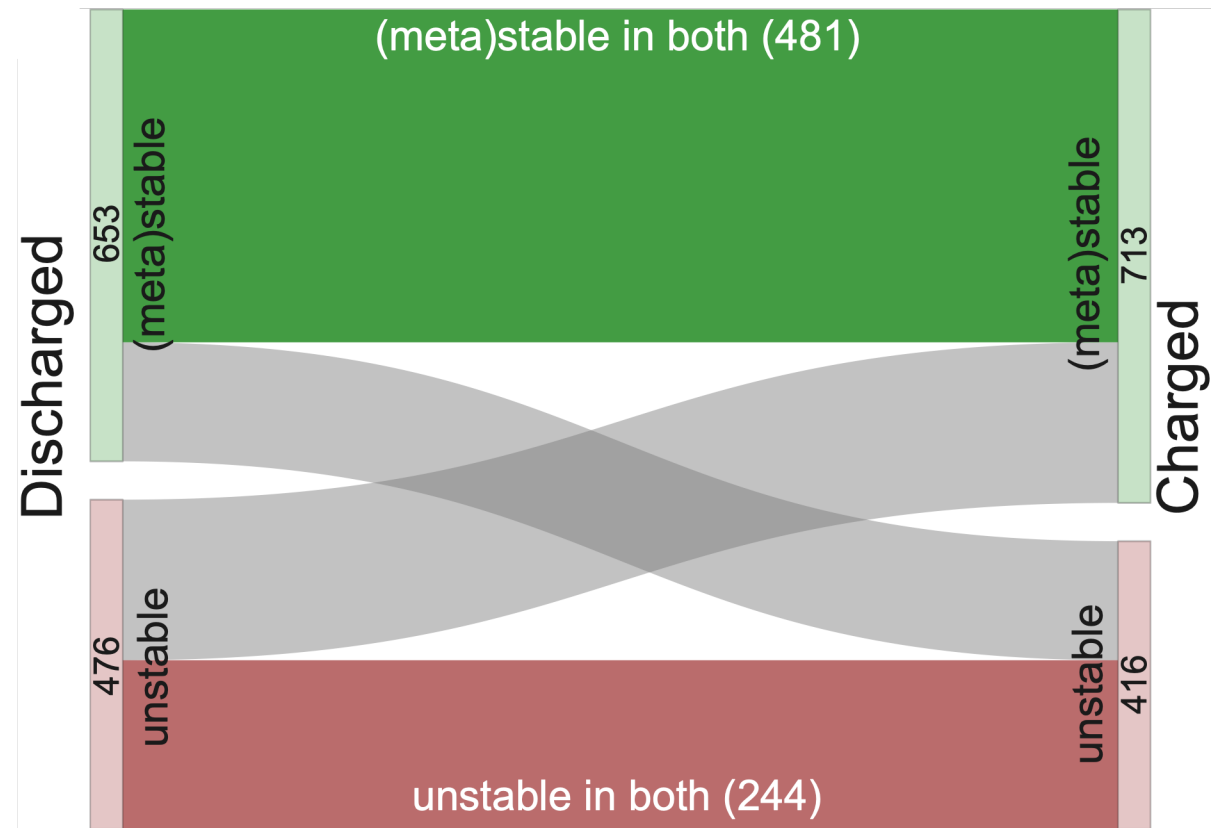
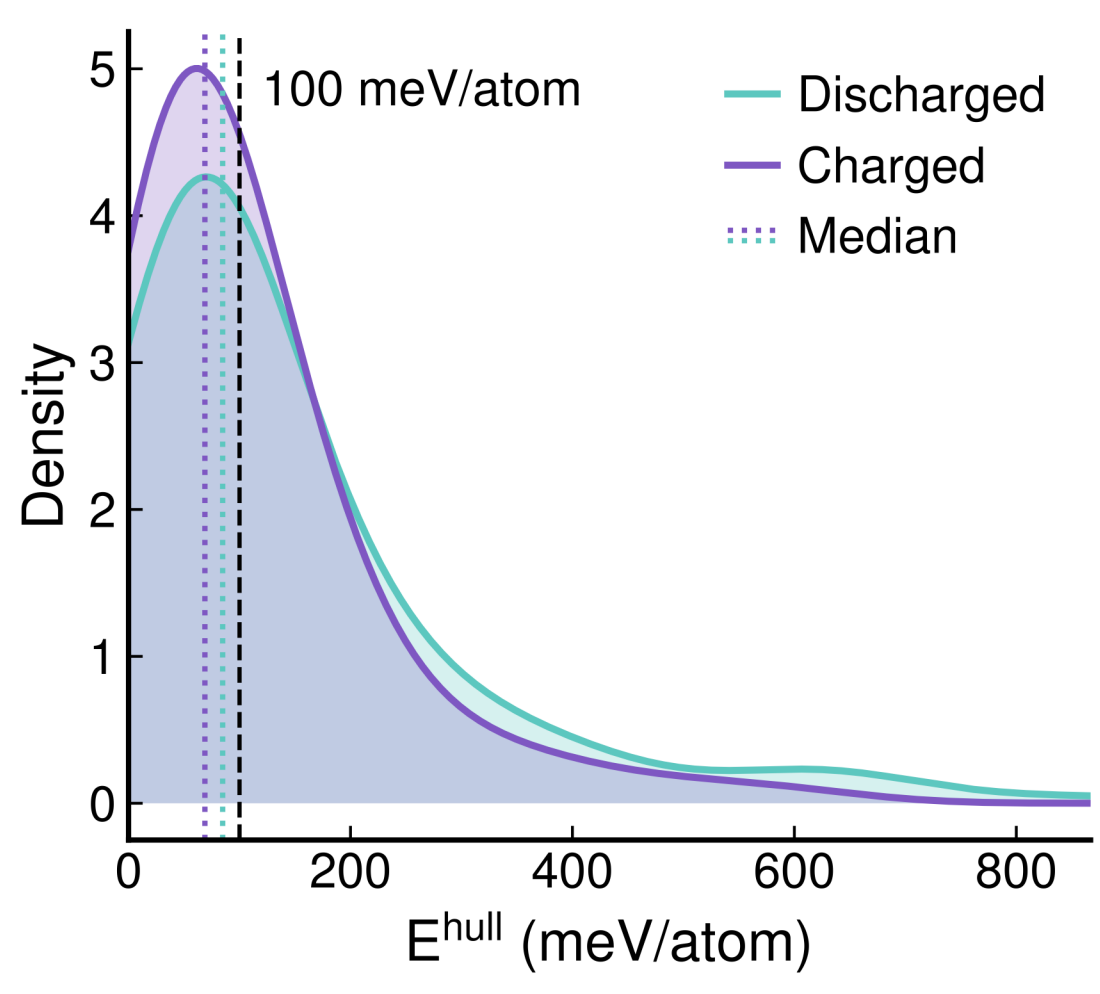
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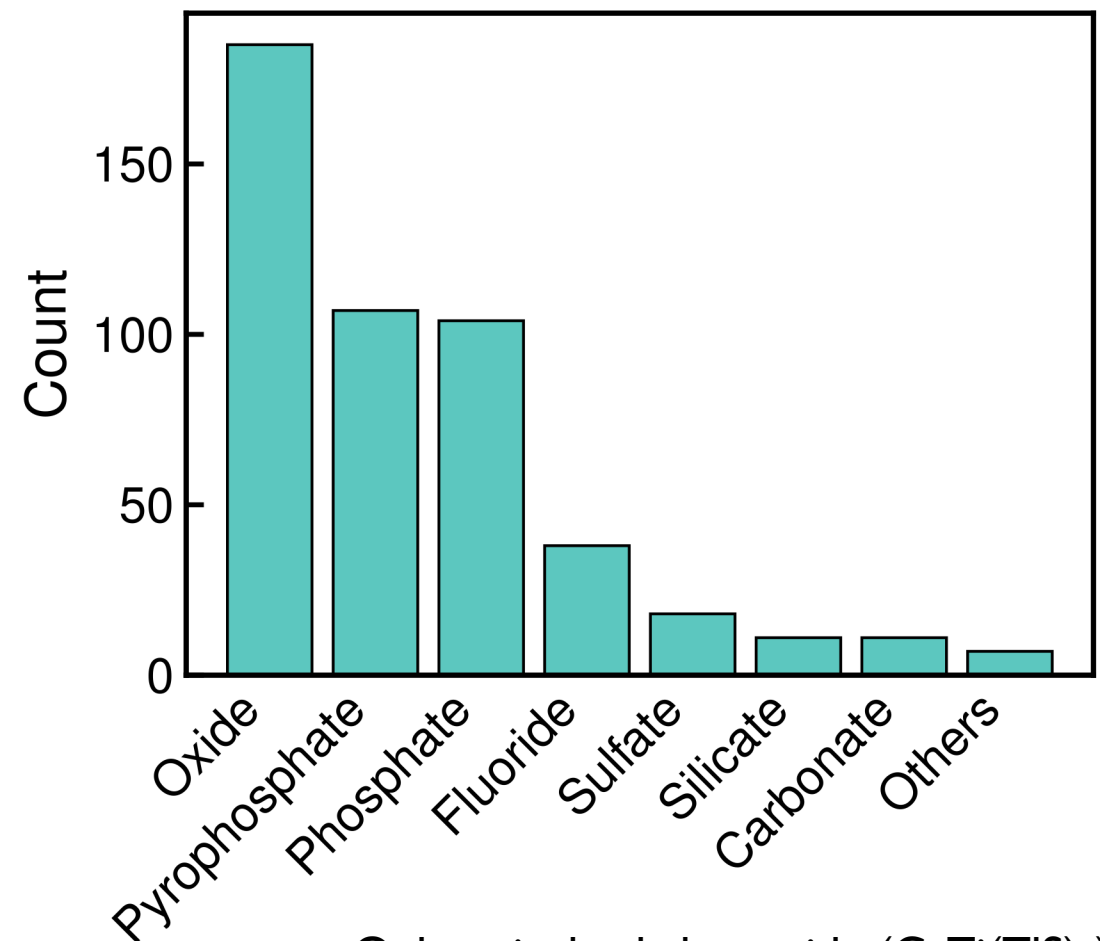
(Meta)stability and voltage*: 433



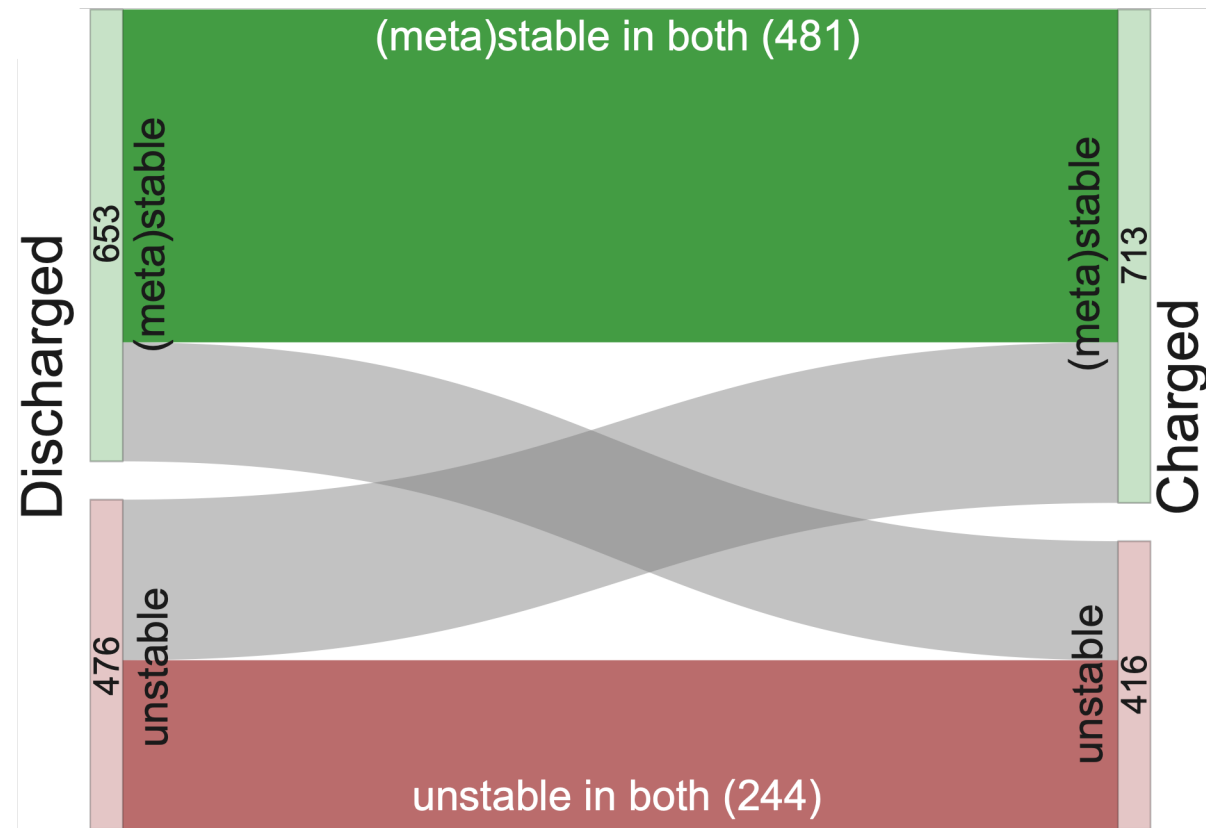
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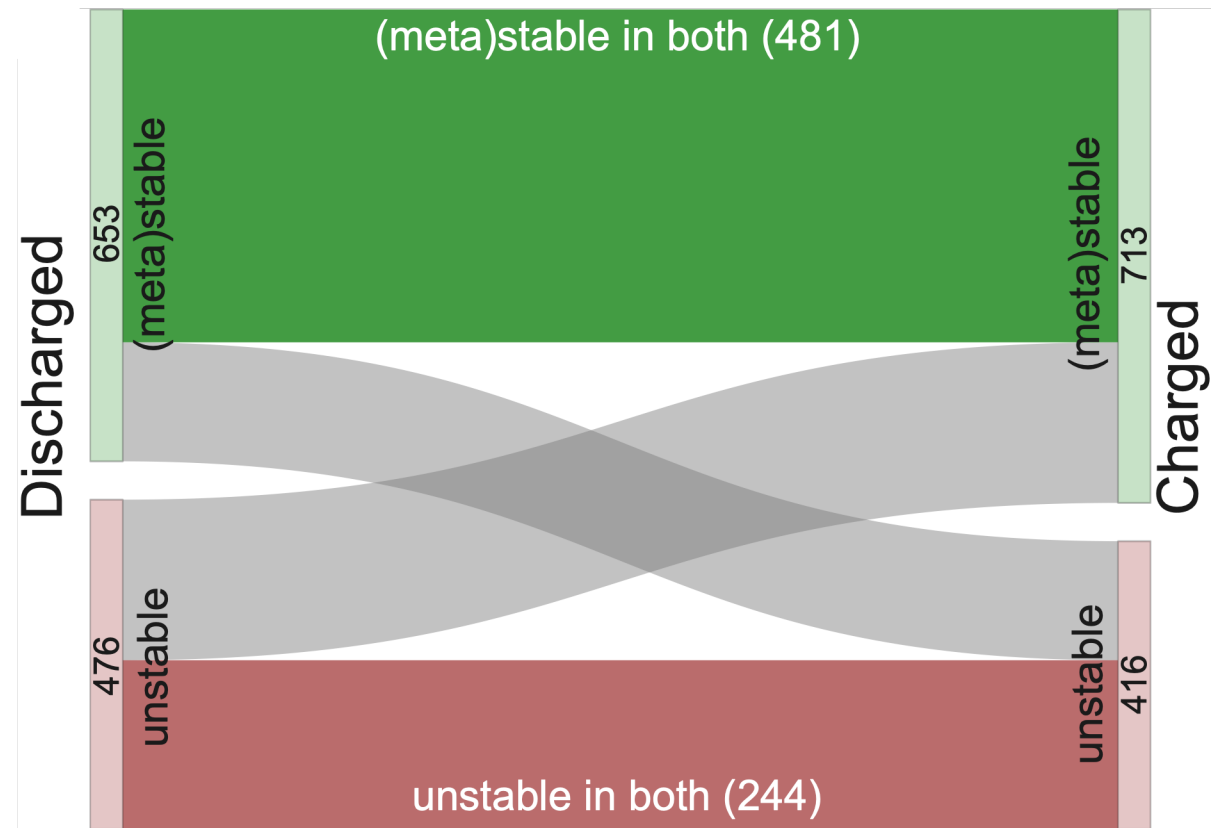
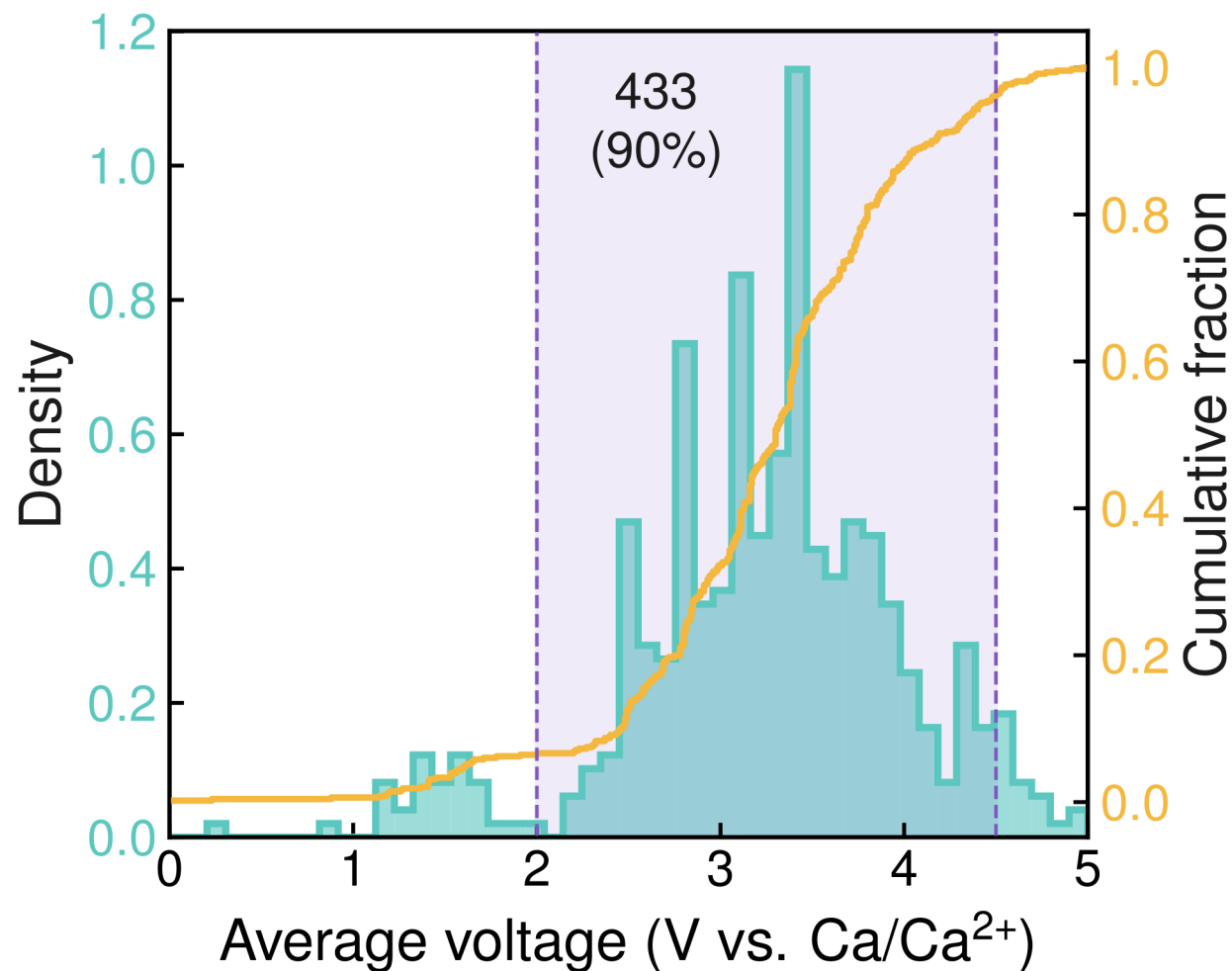
(Meta)stability and voltage*: 433



Only a single chalcogenide ($\text{CaTi}(\text{TIS})_4$)!

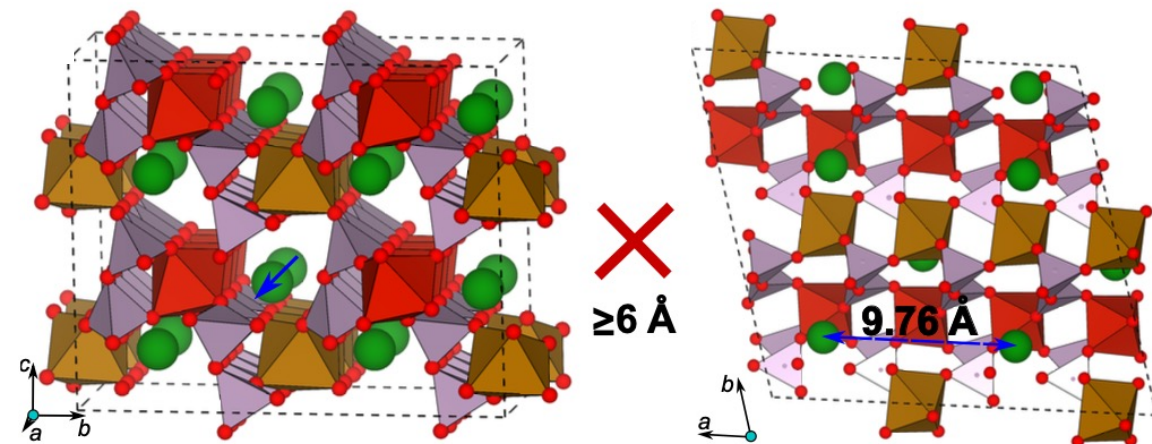


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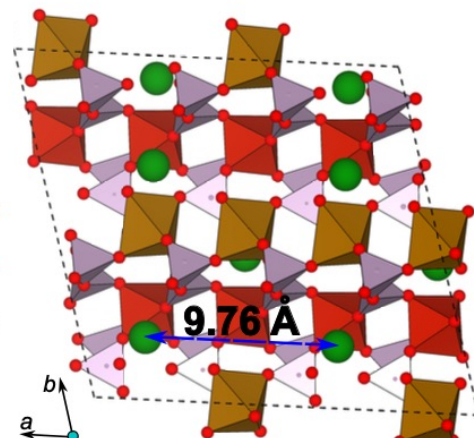
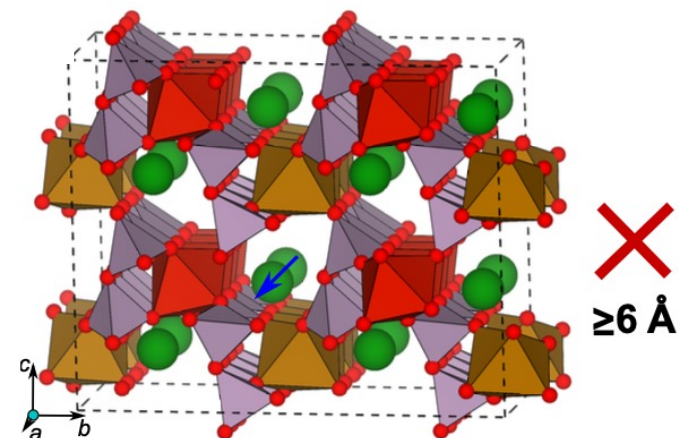
Manual curation + mobility + DFT validation

(Manual) data curation: eliminate structures with unfeasible migration pathways

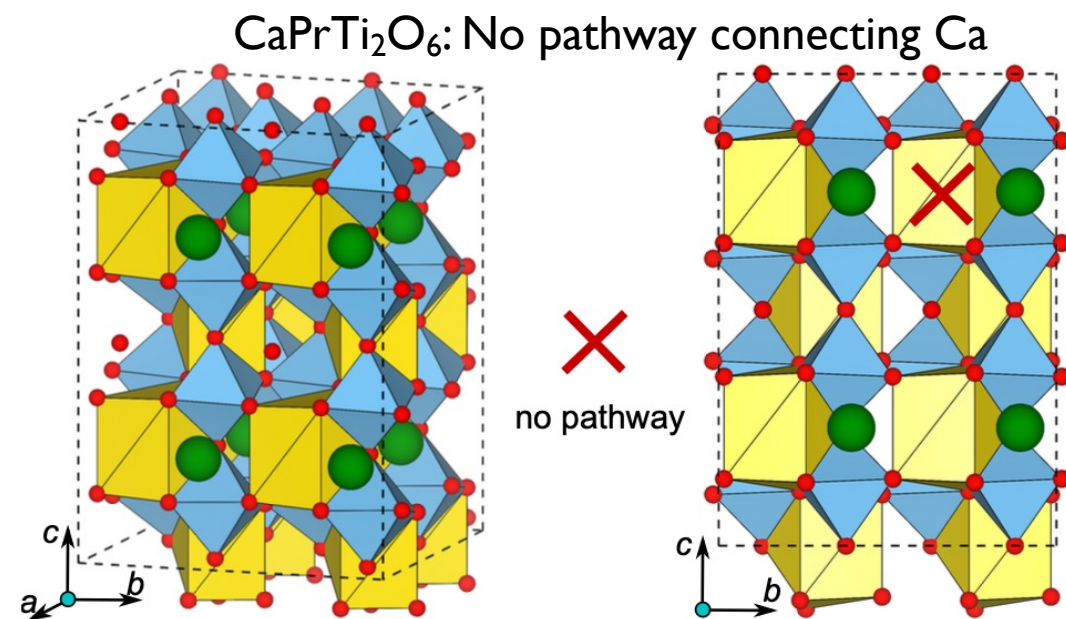


$\text{CaVFe}(\text{P}_2\text{O}_7)_2$: Long pathway

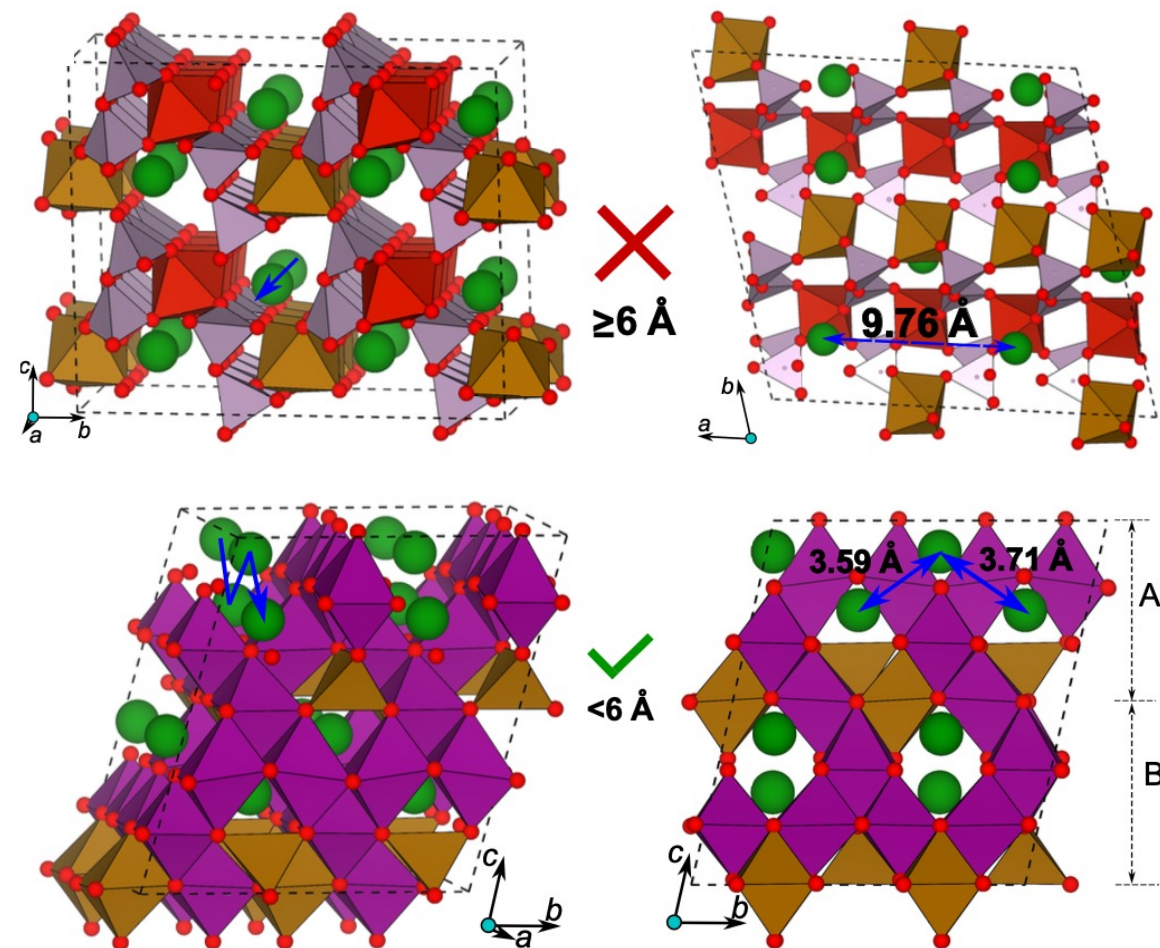
(Manual) data curation: eliminate structures with unfeasible migration pathways



CaVFe(P₂O₇)₂: Long pathway

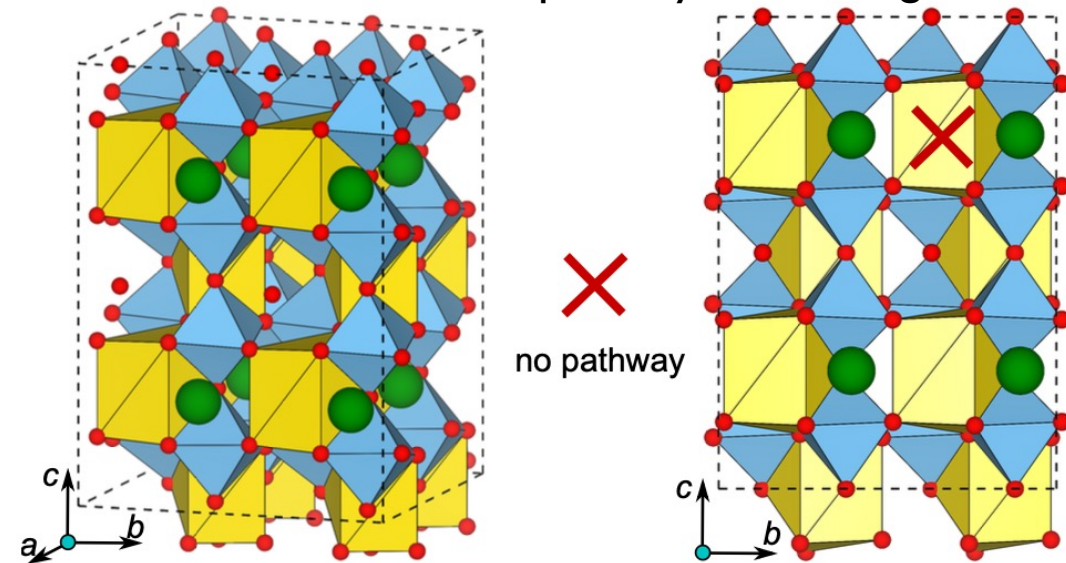


(Manual) data curation: eliminate structures with unfeasible migration pathways



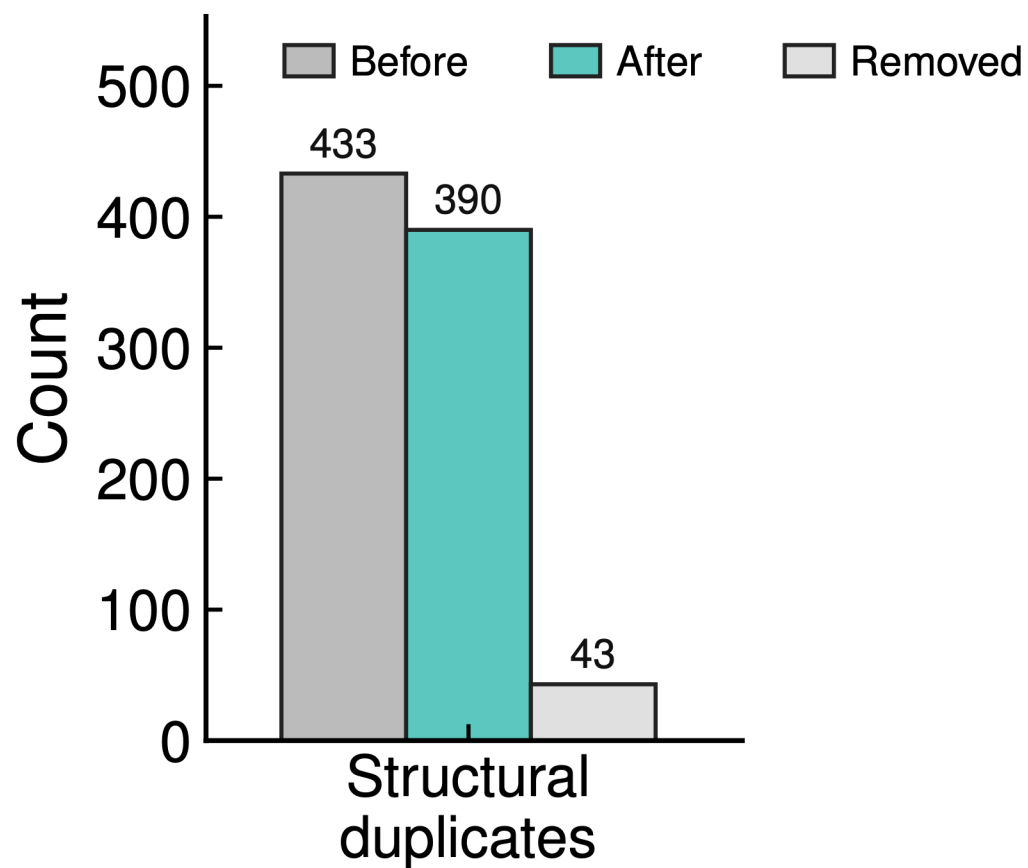
$\text{CaVFe(P}_2\text{O}_7)_2$: Long pathway

$\text{CaPrTi}_2\text{O}_6$: No pathway connecting Ca

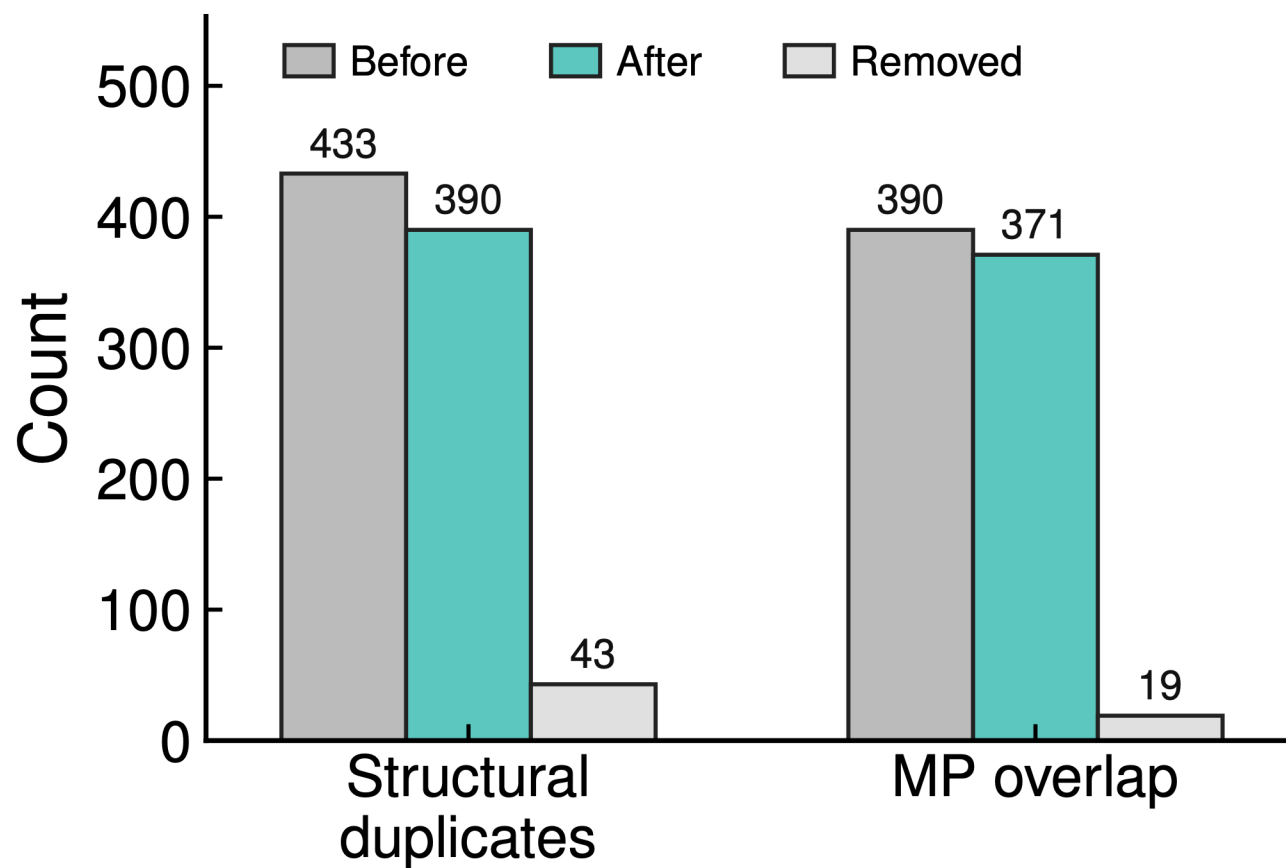


$\text{Ca}_2\text{Mn}_5(\text{FeO}_6)_2$: Allowed 'zig-zag' pathway

Data curation: remove duplicates, remove overlap with MP, remove non-viable pathways

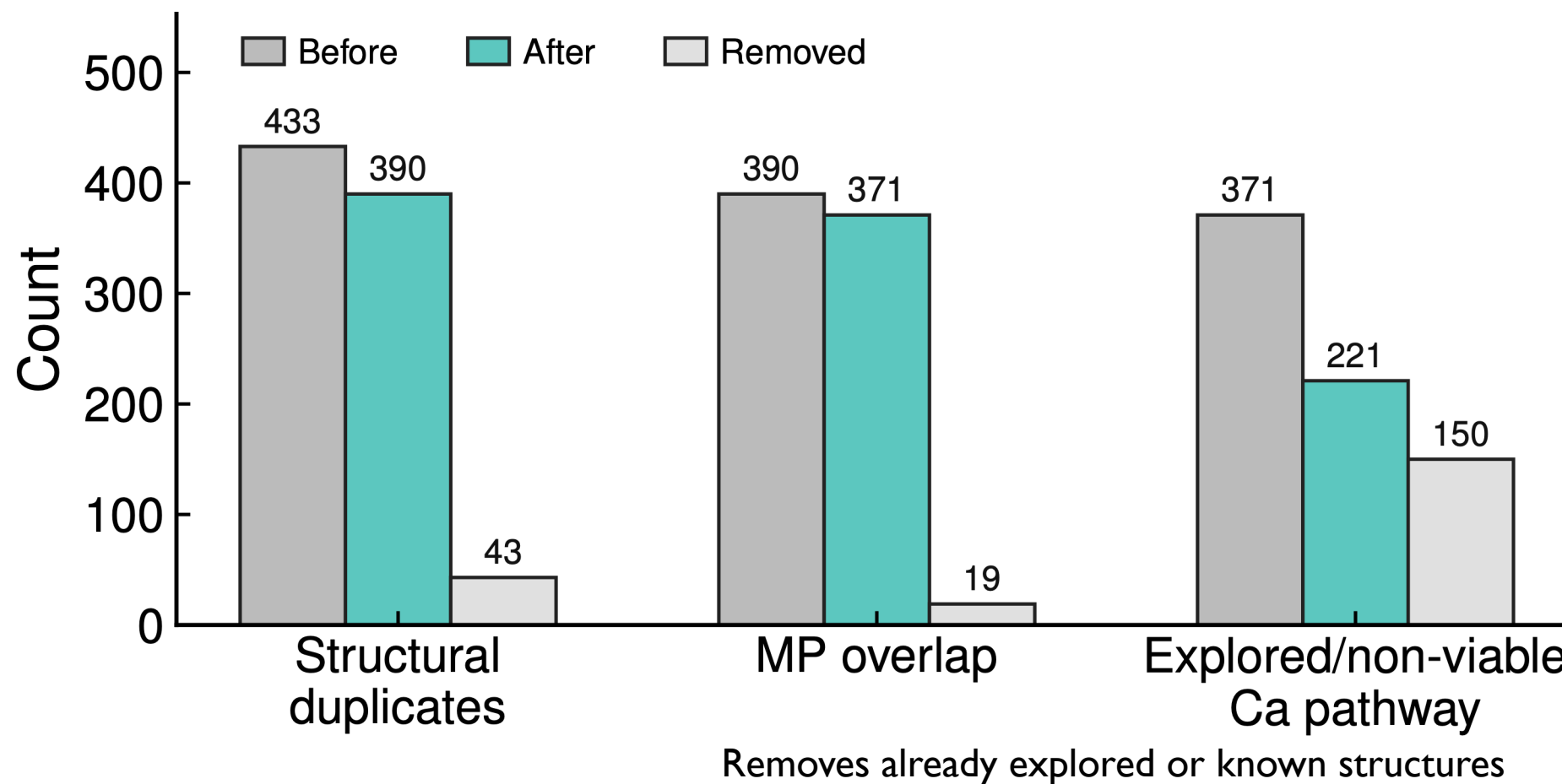


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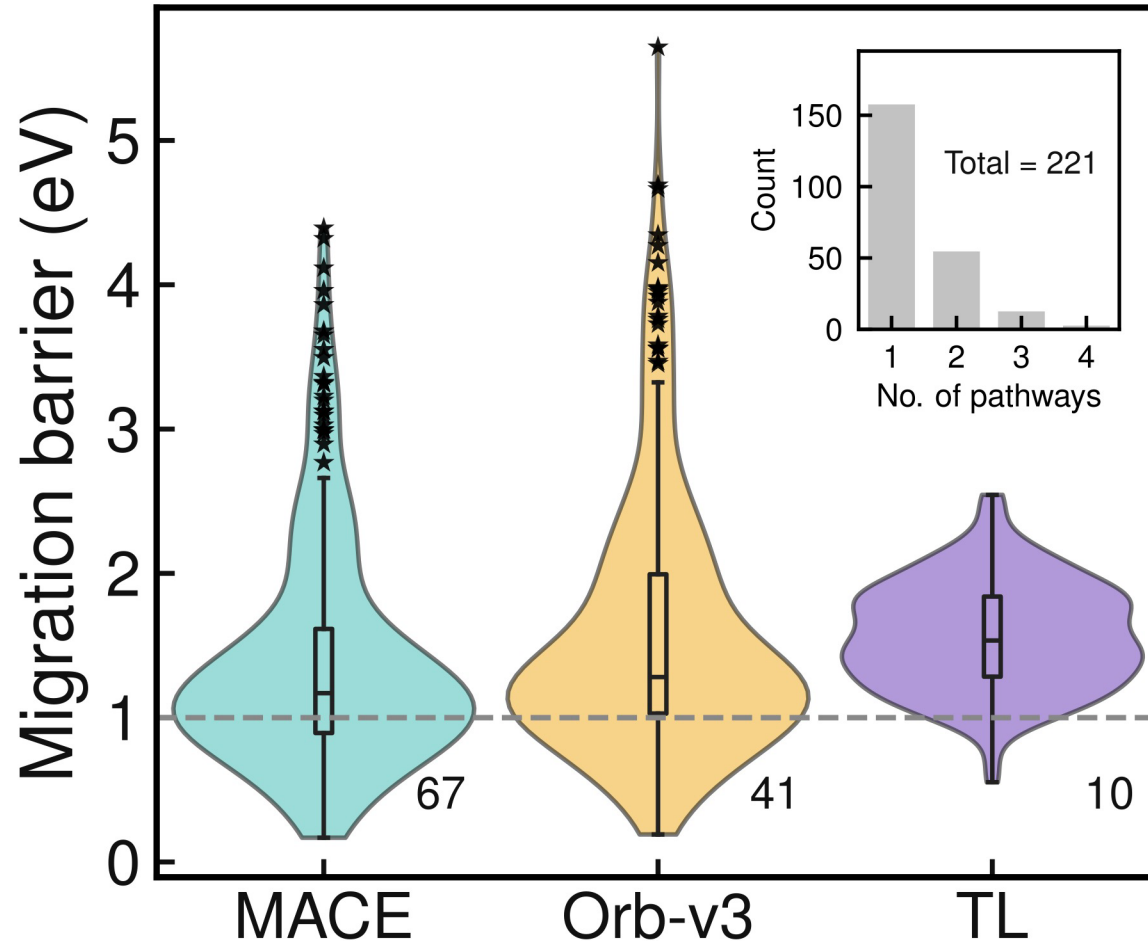


Removes already explored or known structures

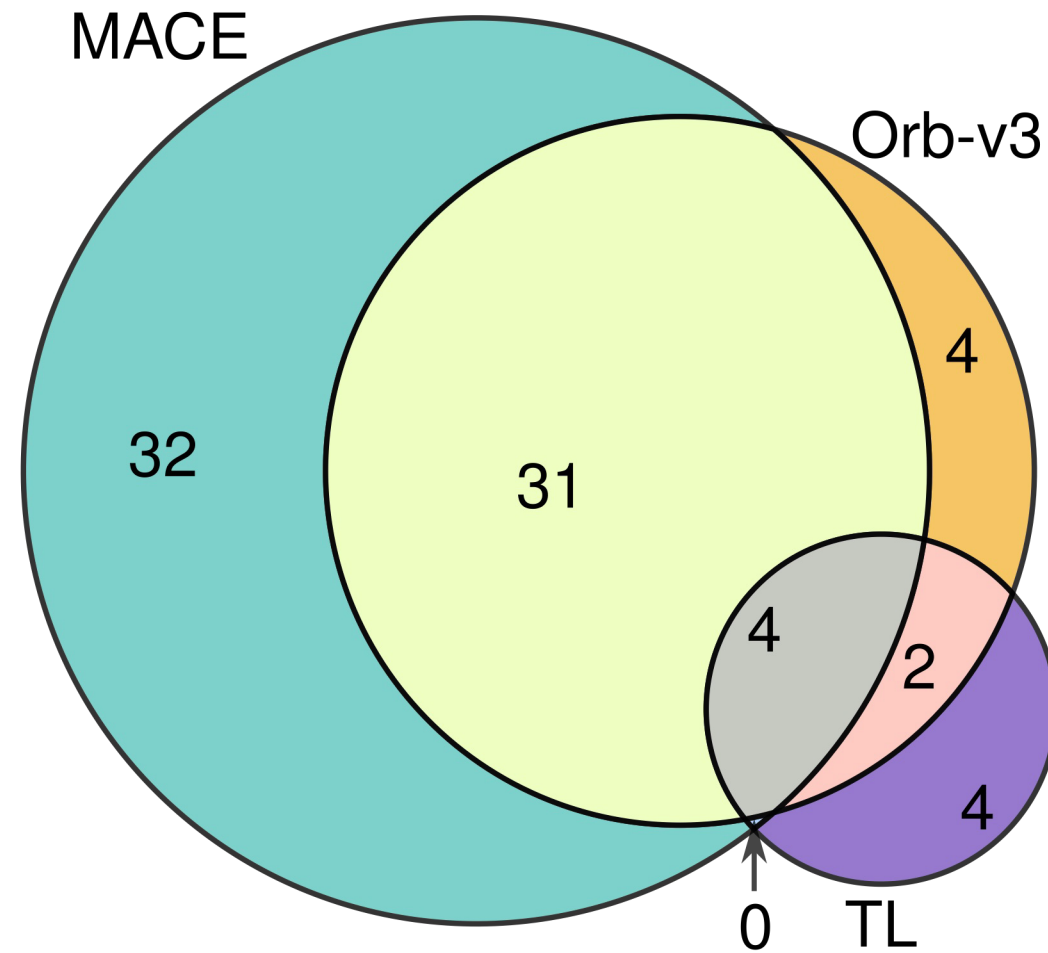
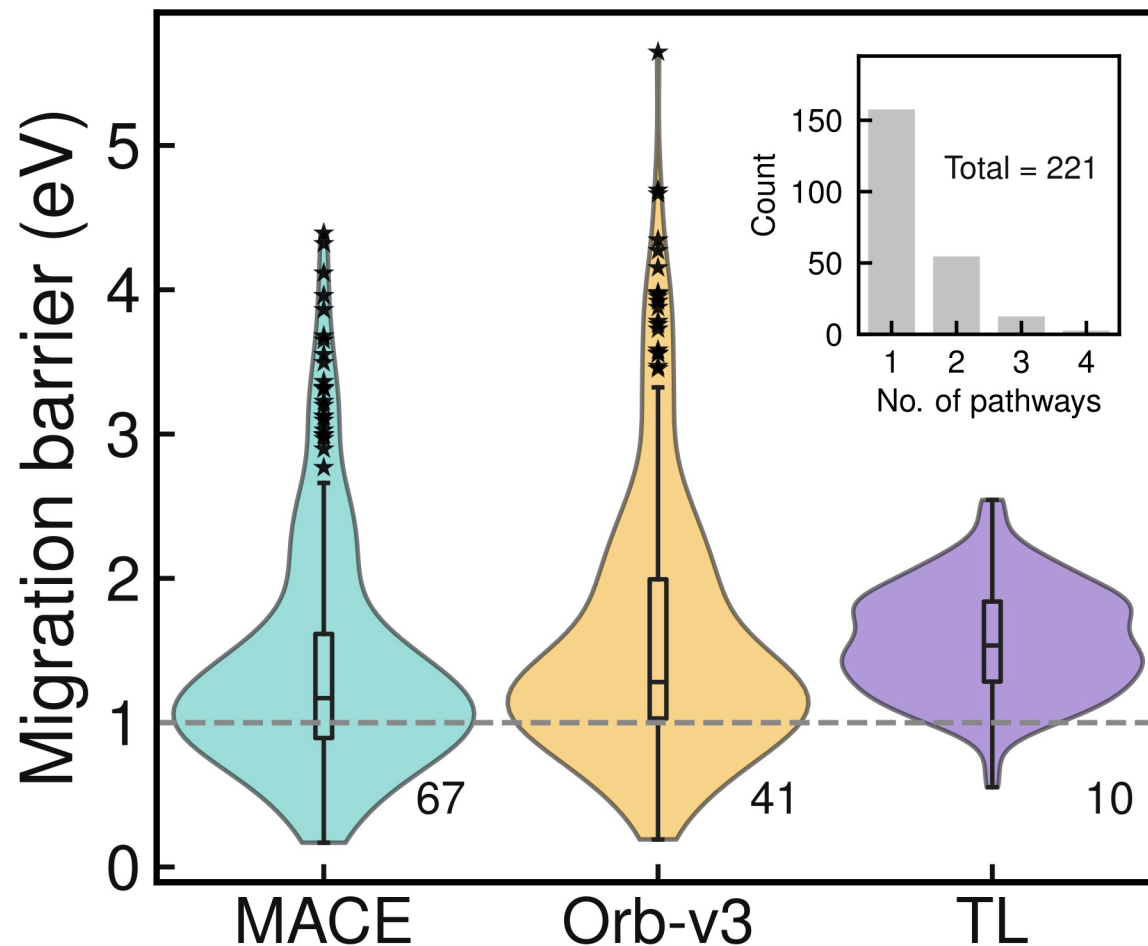
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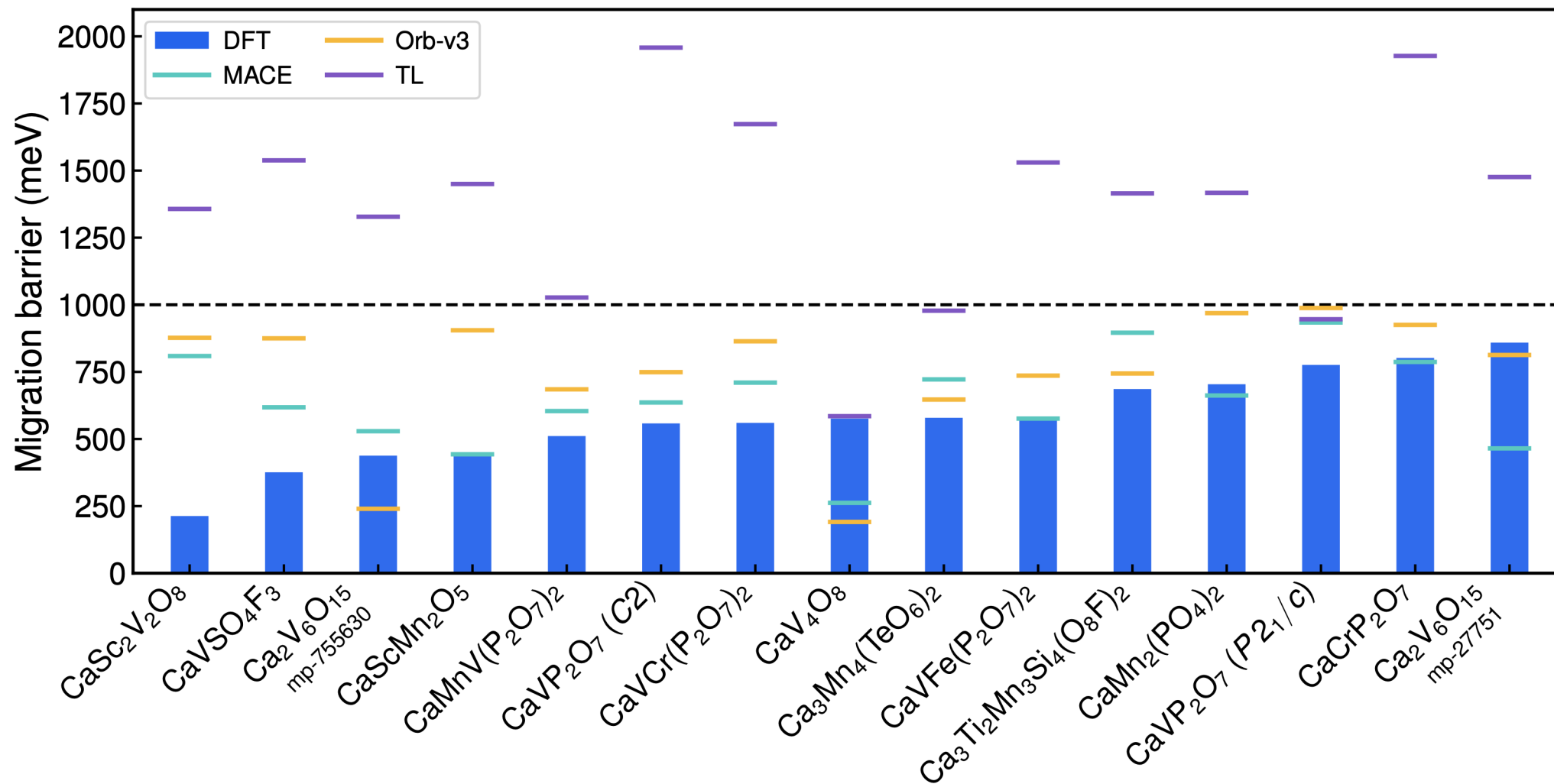
'Ensemble' predictions of Ca mobility: 37 candidates



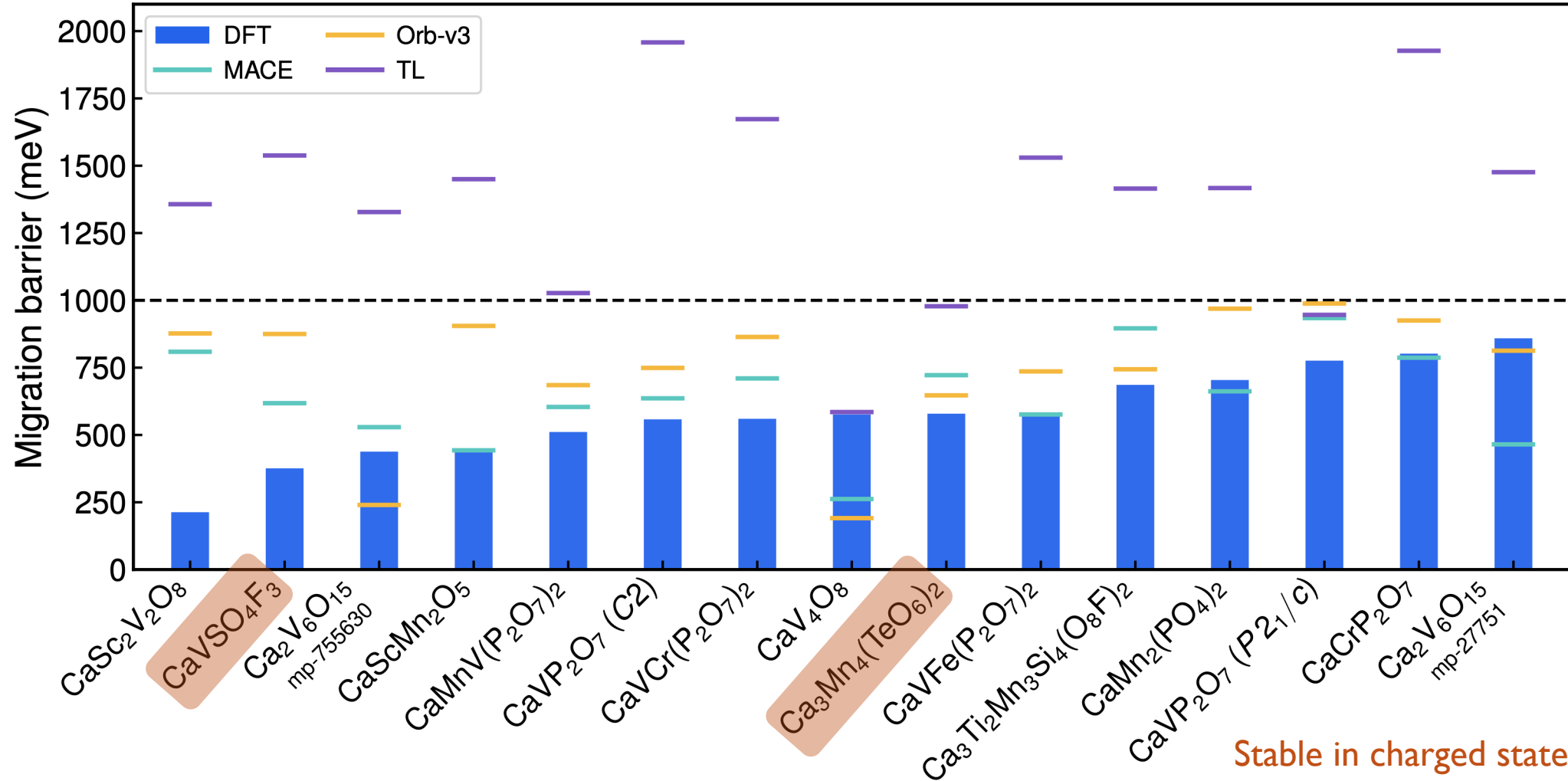
'Ensemble' predictions of Ca mobility: 37 candidates



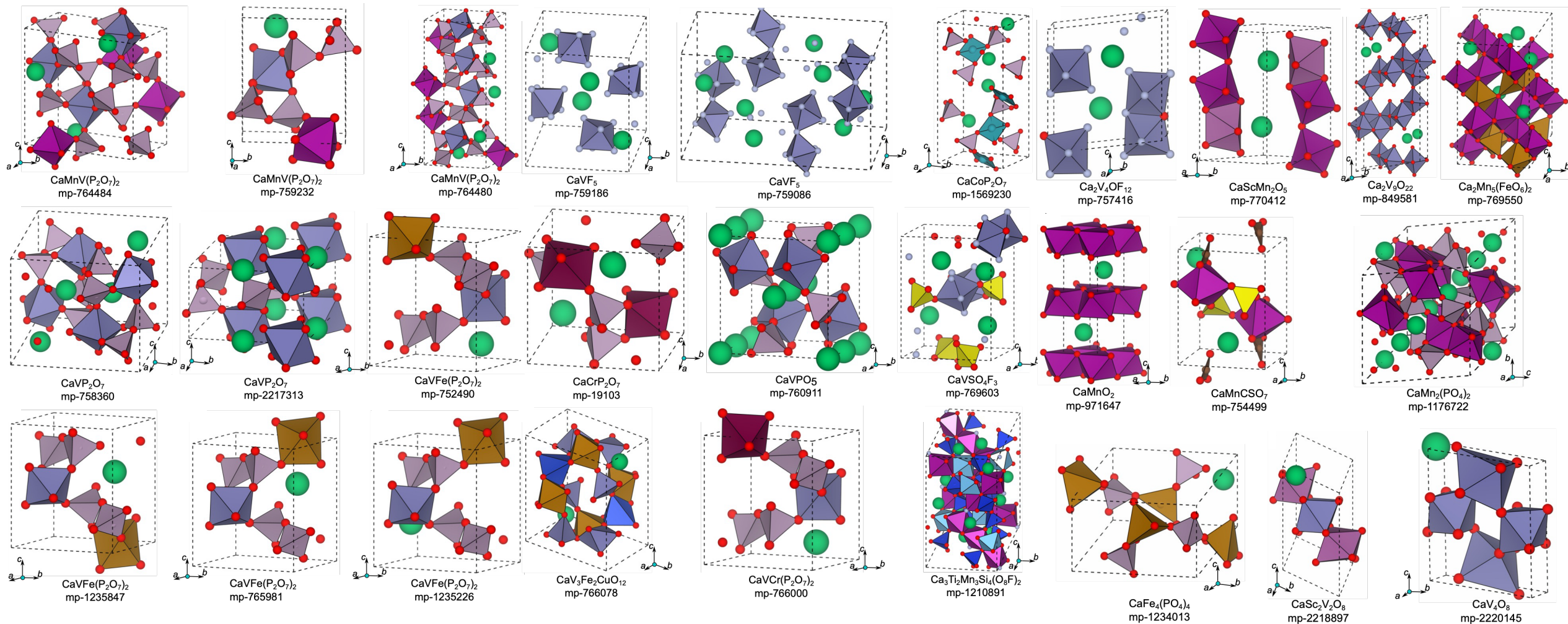
DFT validation for 15 candidates



DFT validation for 15 candidates



Candidates: **37** structures, **26** compositions

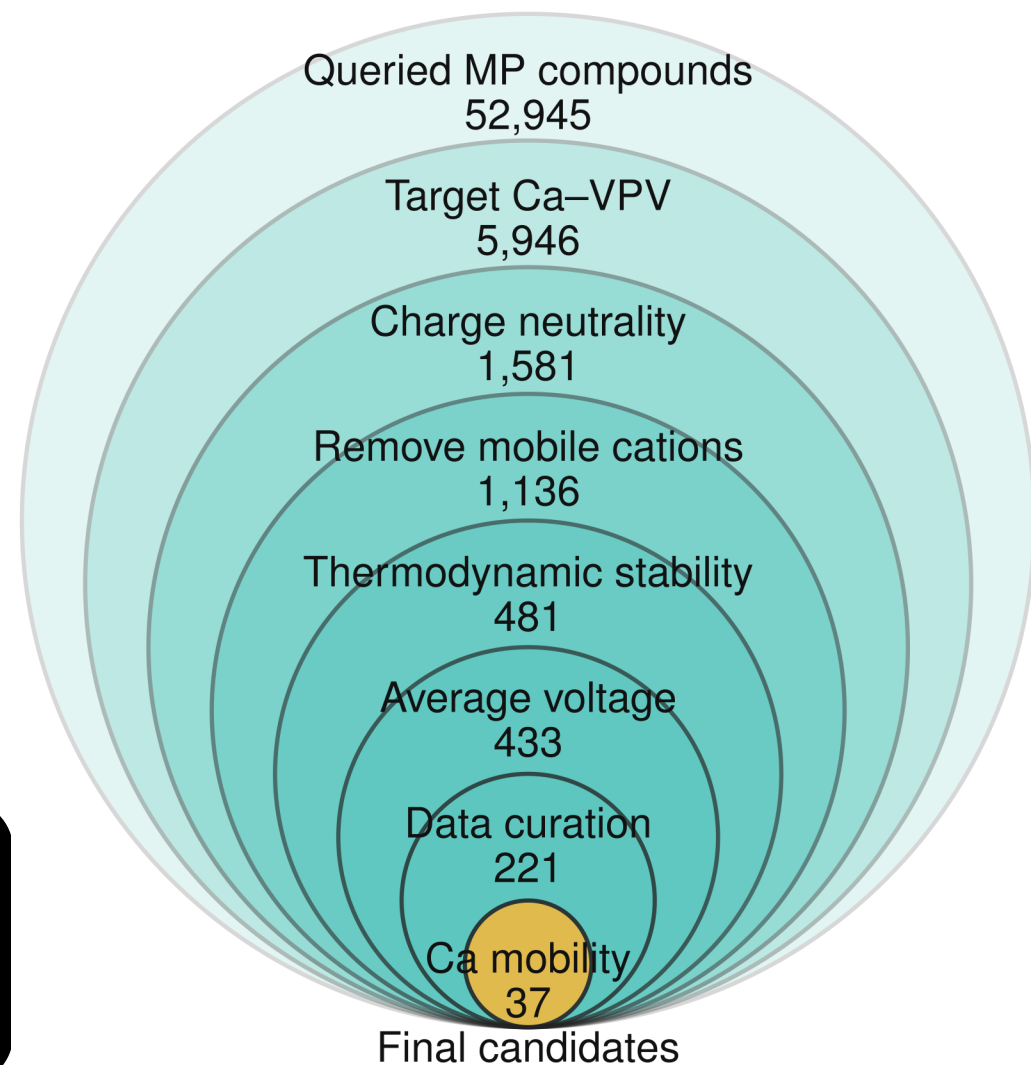


Largest collection of candidate cathodes for Ca batteries, so far!

Conclusions

- Ca batteries: promising beyond Li-ion technology requiring good cathodes
- Geometric + Chemical descriptors + filters on (meta)stability, voltage, and mobility – **37** non-Ca containing candidate structures
- Foundational MLIPs can accelerate material screening
 - Execution < 1 year (possibly faster now)
- Ensemble predictions improve reliability
 - (Meta)stable candidates with high Ca mobility!

“Geometry-based Discovery of Calcium Battery Cathodes Accelerated by Foundational Machine-Learned Models”, D.B. Tekliye, A.K. Bheemaguli, and G. Sai Gautam, coming soon on *npj Comput. Mater.* [arXiv 2605.29029 (2026)].



saigautamg@iisc.ac.in; <https://sai-mat-group.github.io>; <https://github.com/sai-mat-group>