Discovery of Catalysts for the Application of NO_3RR for Water Purification Using Machine Learning Techniques

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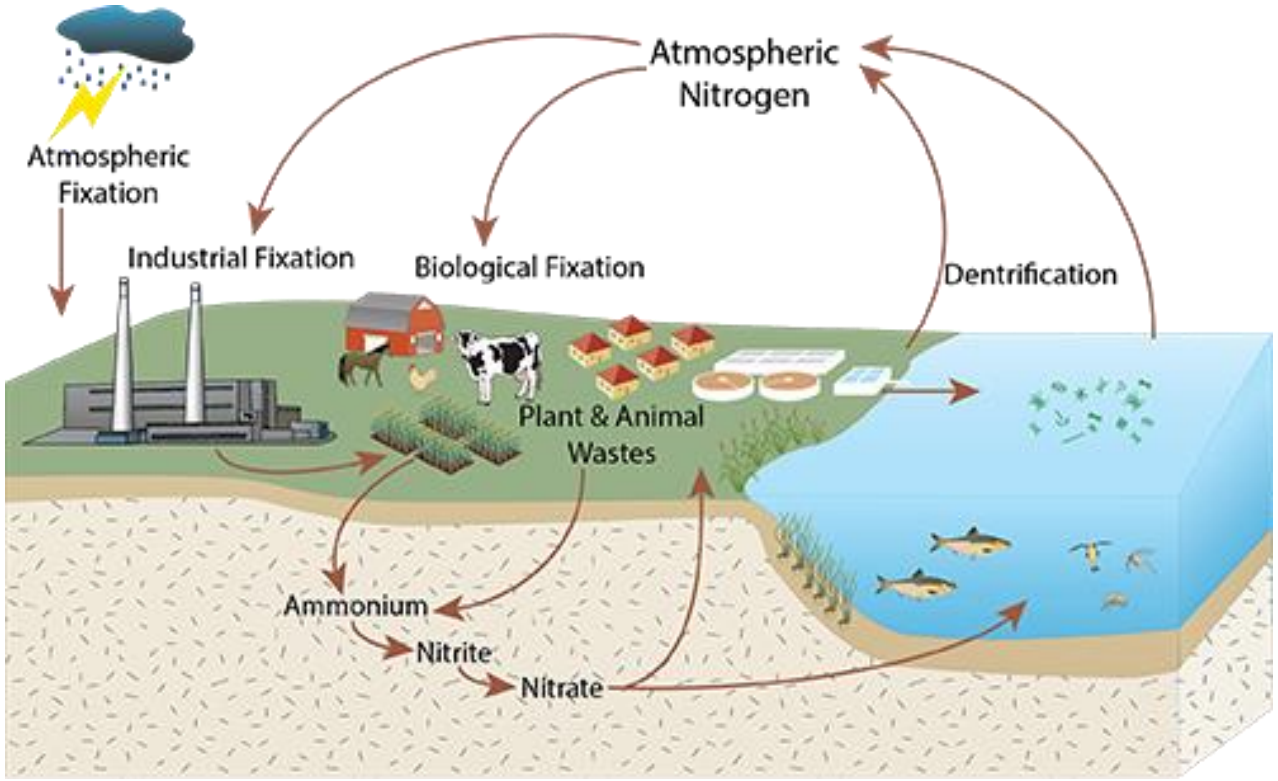
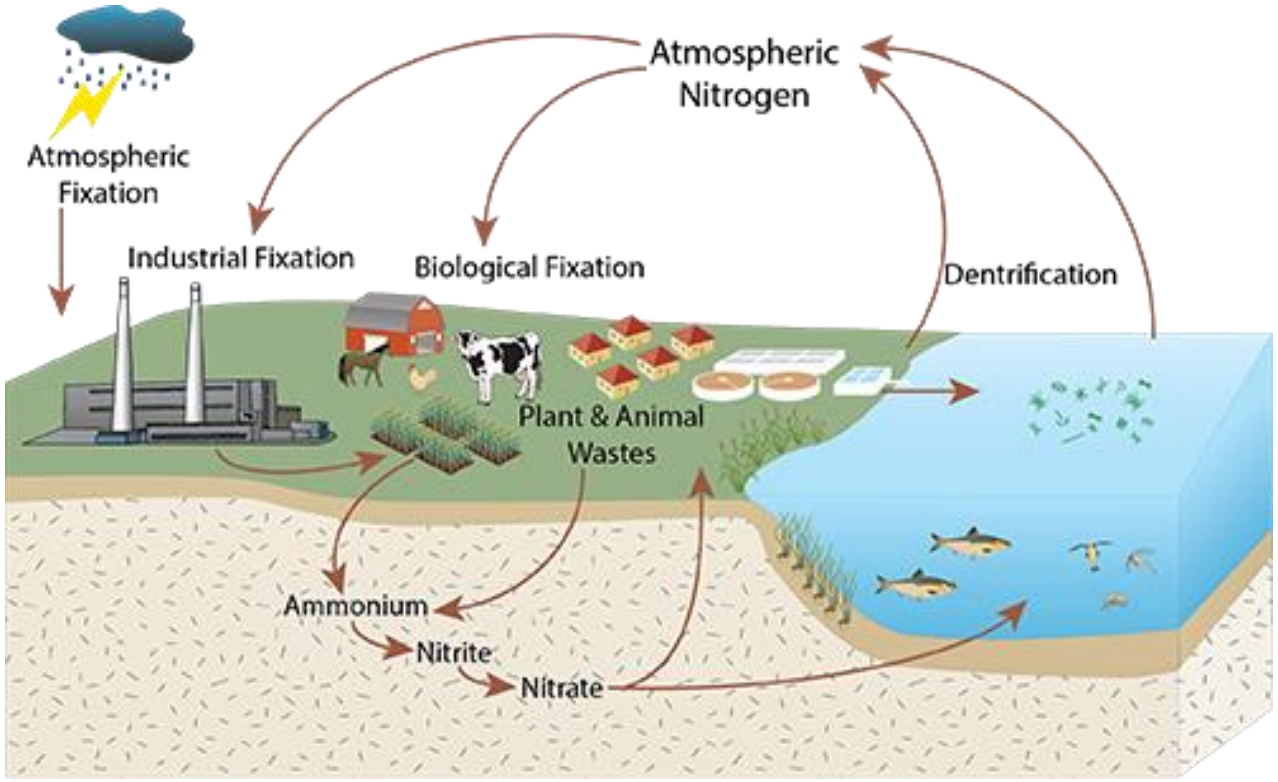
Water purification

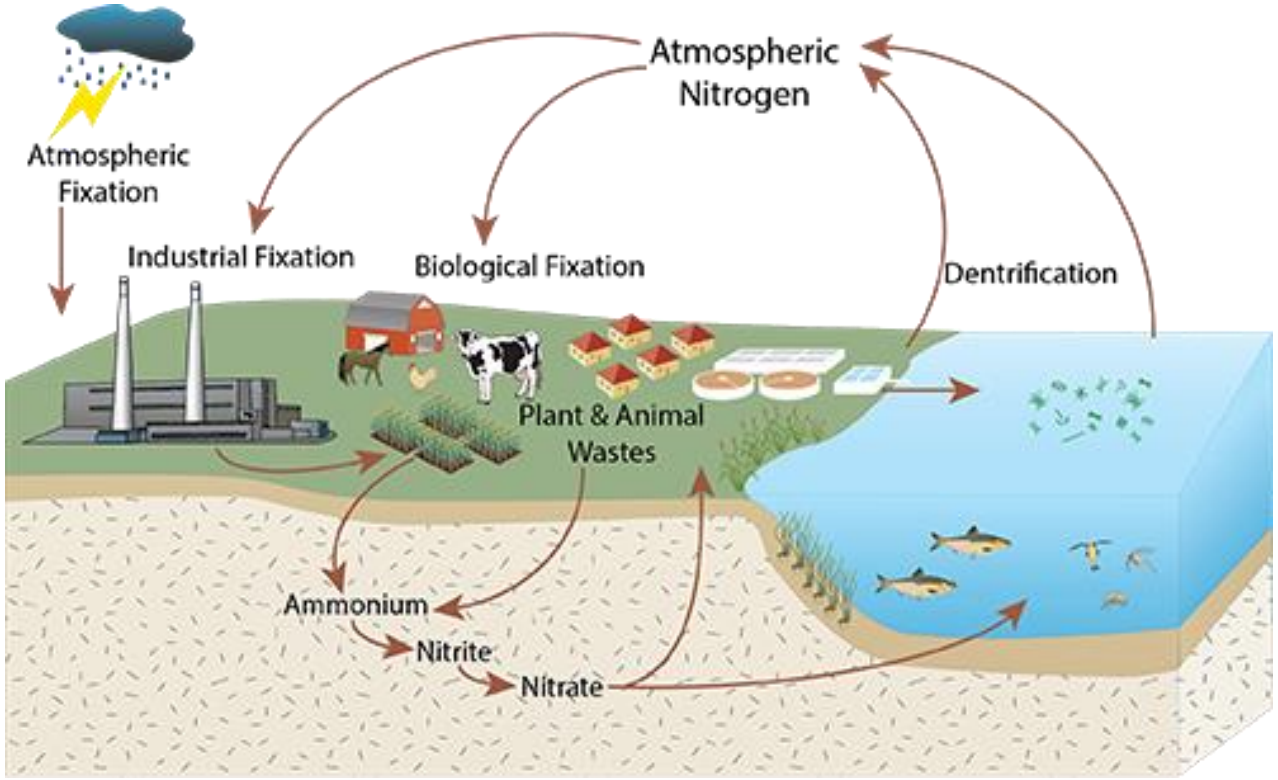


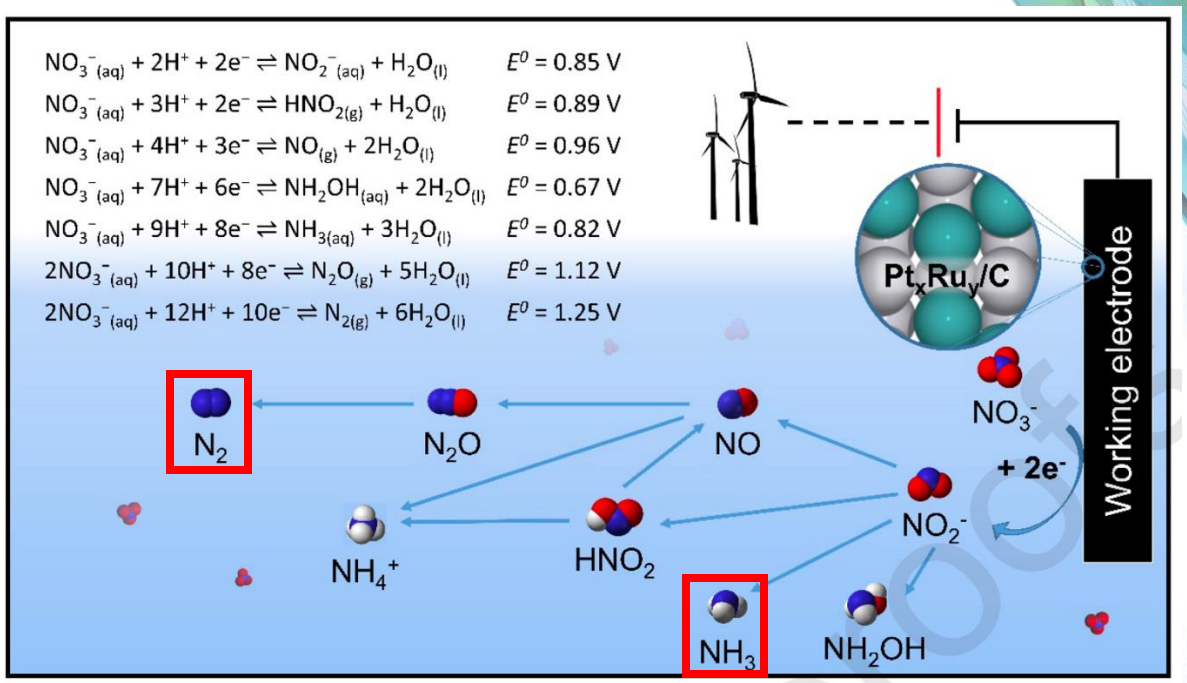
Water purification



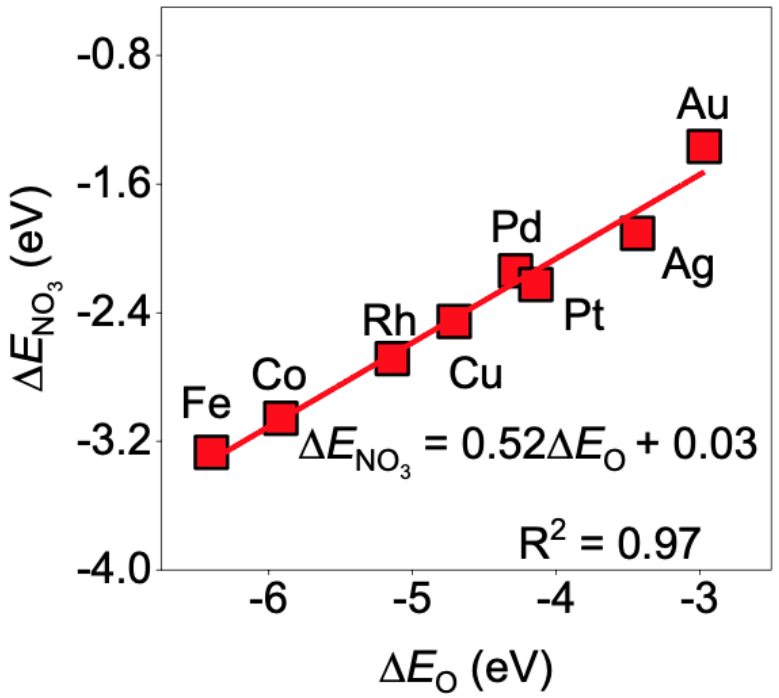
Process	Capital ($/1000 gal)	Operating ($/1000 gal)	Brine Disposal ($/1000 gal)	Total Cost ($/1000 gal)
Rev Osmosis	$0.44-0.88	$1.10-3.00	$0.40-2.60	$1.54-6.48
Ion Exchange	$0.24-1.18	$0.46-0.64	$0.04-0.32	$0.70-1.24
Bio Treatment	$0.40-0.90	$0.50-0.80	$0.01-0.02	$0.91-1.72
Electrocatalytic Treatment	***$?***	***$0.12-1.57***	***n/a***	***$?***

Water purification



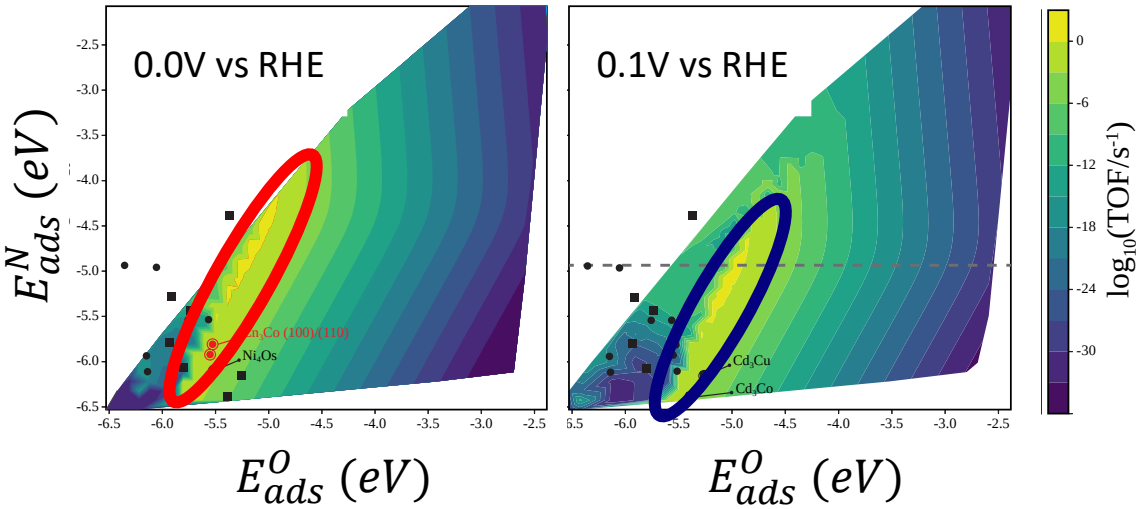


Screening with simple parameters



Liu, J. X., Richards, D., Singh, N., & Goldsmith, B. R. (2019). *ACS Catalysis*, *9*(8), 7052–7064.

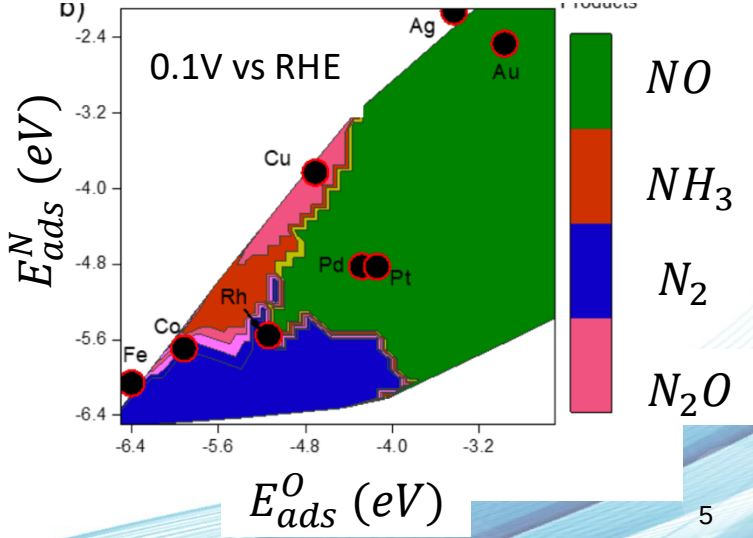
Activity for NO_3^- consumption



$$NO_3^* + * \leftrightarrow NO_2^* + O^*$$
$$E_a = 0.234E_O + 0.054E_N + 2.047$$
$$E_b = -0.593E_O - 0.064E_N - 1.324$$

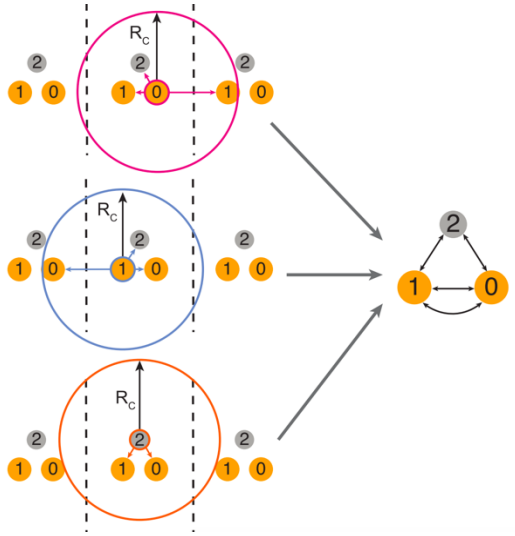
E_a and E_b can be used with microkintetic modelling to determine TOF and selectivity

Selectivity



Candidate screening via machine learning

Graph neural network (GNN) model:

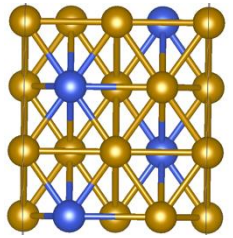


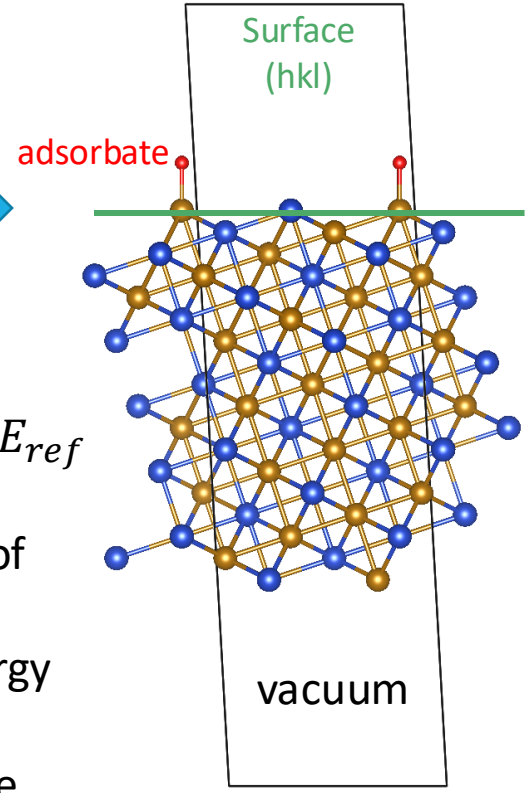
Specs:

- GNN model: DimeNet++
- MAE = ~0.3 eV
- Target: Initial structure (adsorbed slab)→E_{ads}
- Training data: ~100k (metals only)

Initial structure: Slab

Conventional unit cell





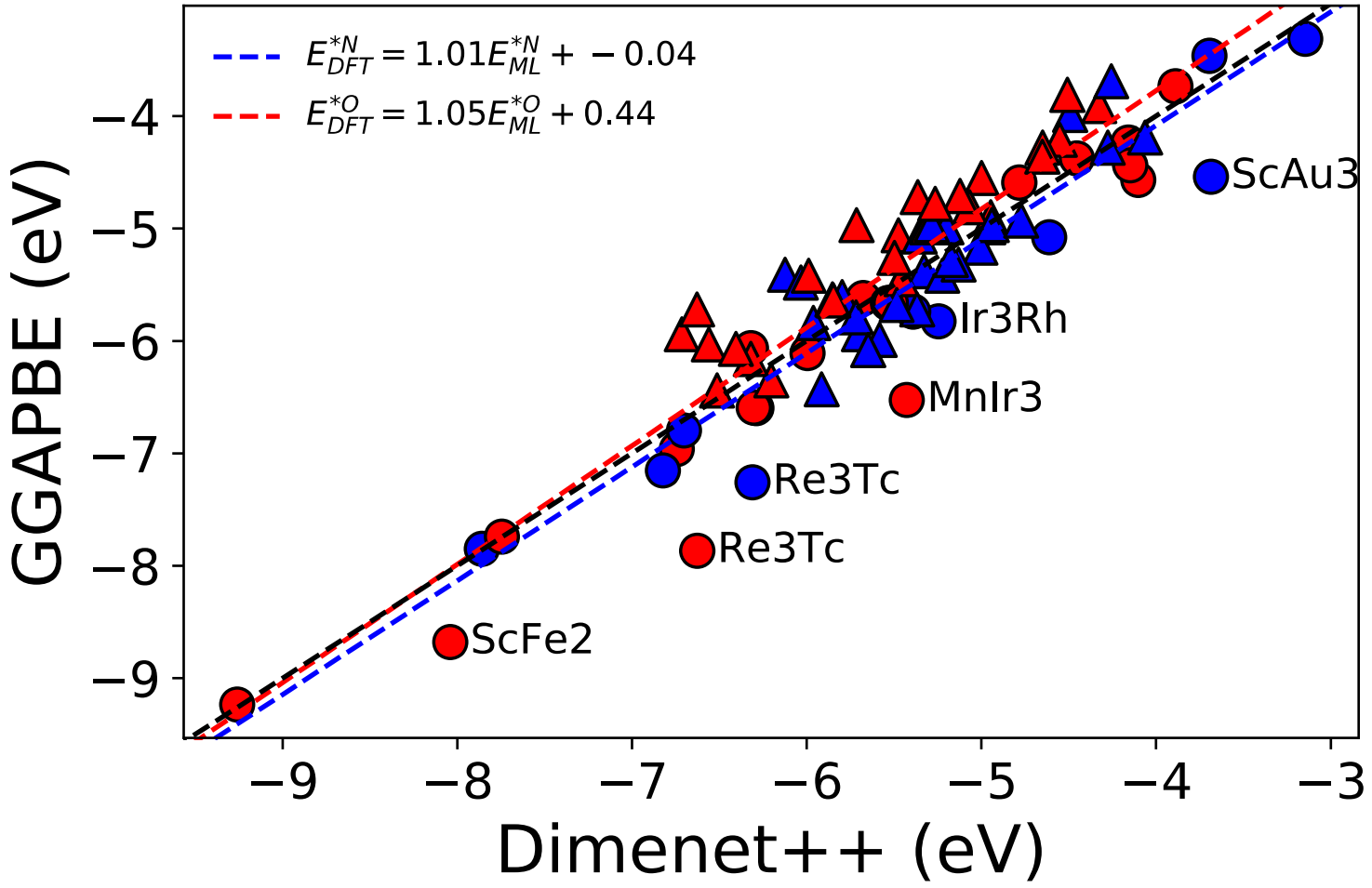
$E_{ads} = E_{slab} - E_{slab+ads} - E_{ref}$

- E_{slab} : Total DFT energy of slab model
- $E_{slab+ads}$: Total DFT energy of slab with adsorbate
- E_{ref}: Energy of adsorbate molecule

E_{slab} (without adsorbate)

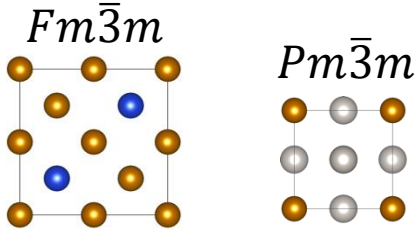
$E_{slab+ads}$

Validation and offset



△ Artificial intermetallics (A_3B) from Vegard's Law

$Fm\bar{3}m$ $Pm\bar{3}m$

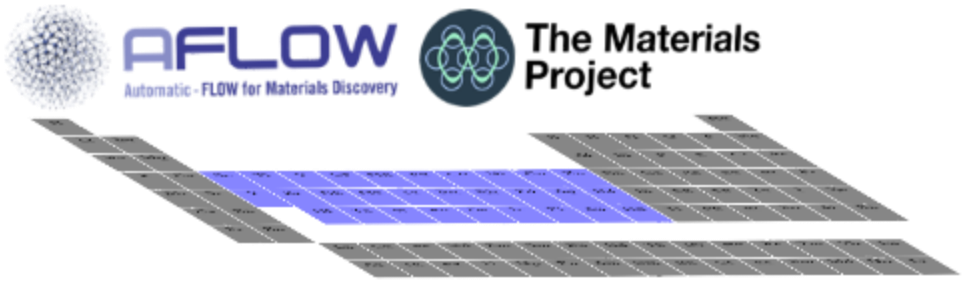


○ Intermetallics from AFLOW and MP

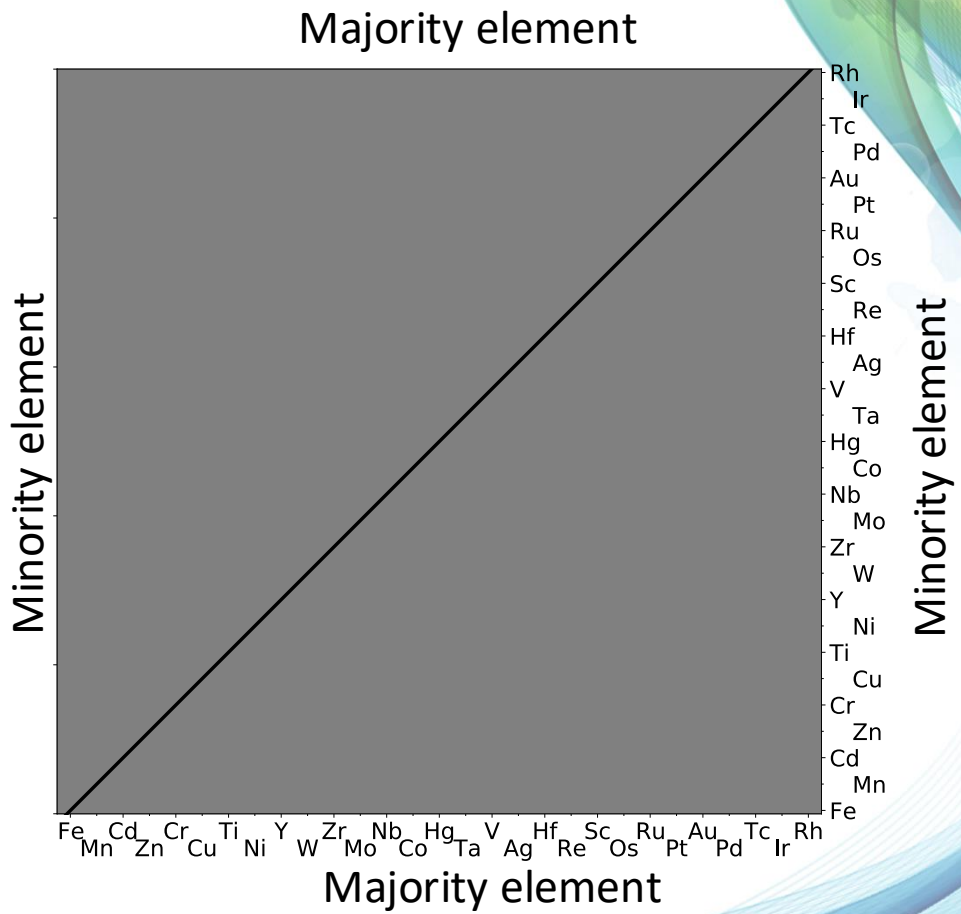
$R^2_N = 0.87, \mathrm{MAE} = 0.29$
$R^2_O = 0.88, \mathrm{MAE} = 0.36$

*Training dataset done with RPBE

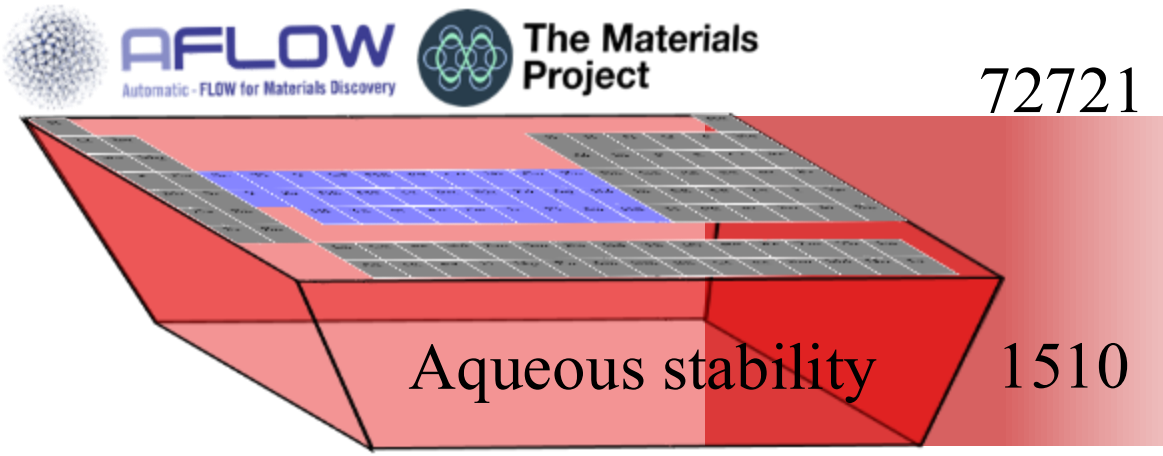
ML assisted screening

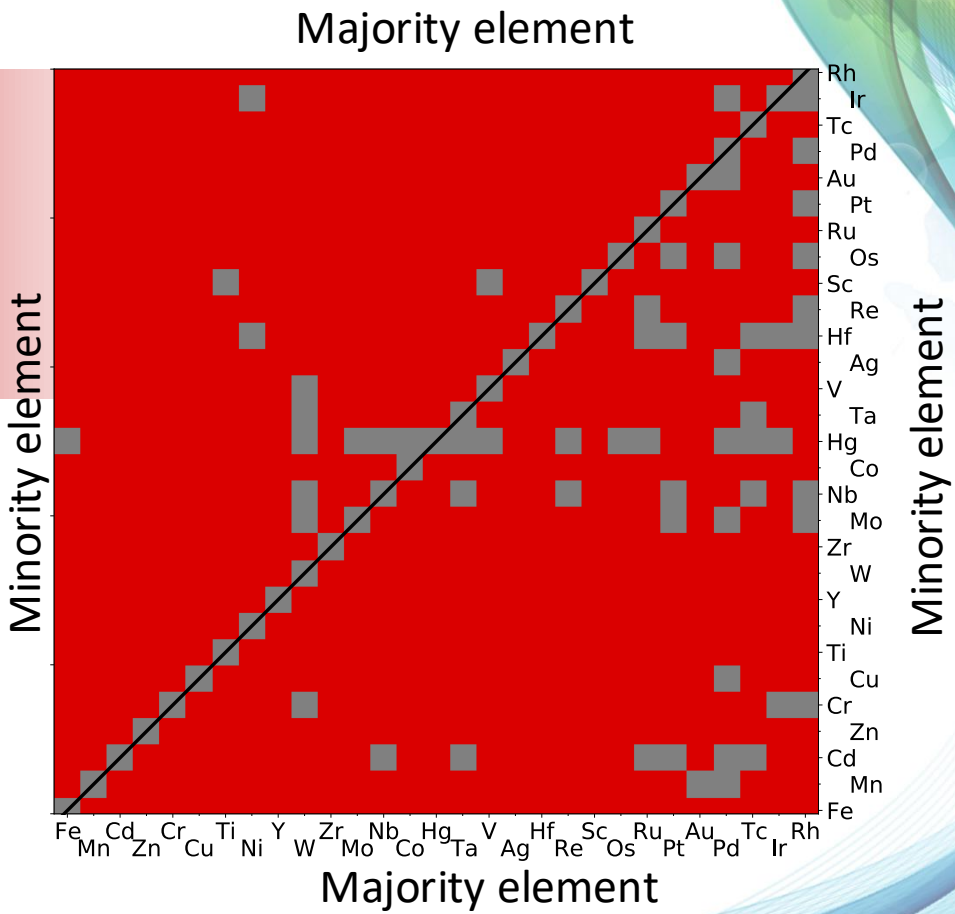


72721



ML assisted screening





ML assisted screening

AFLOW Automatic-FLOW for Materials Discovery

The Materials Project

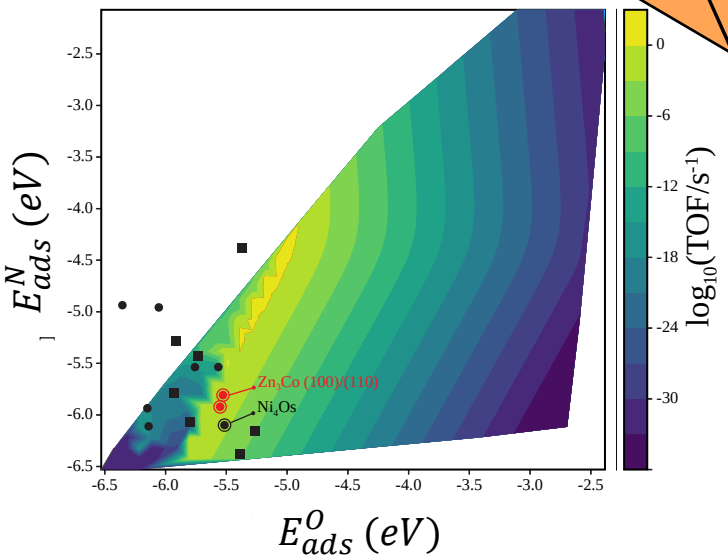
72721

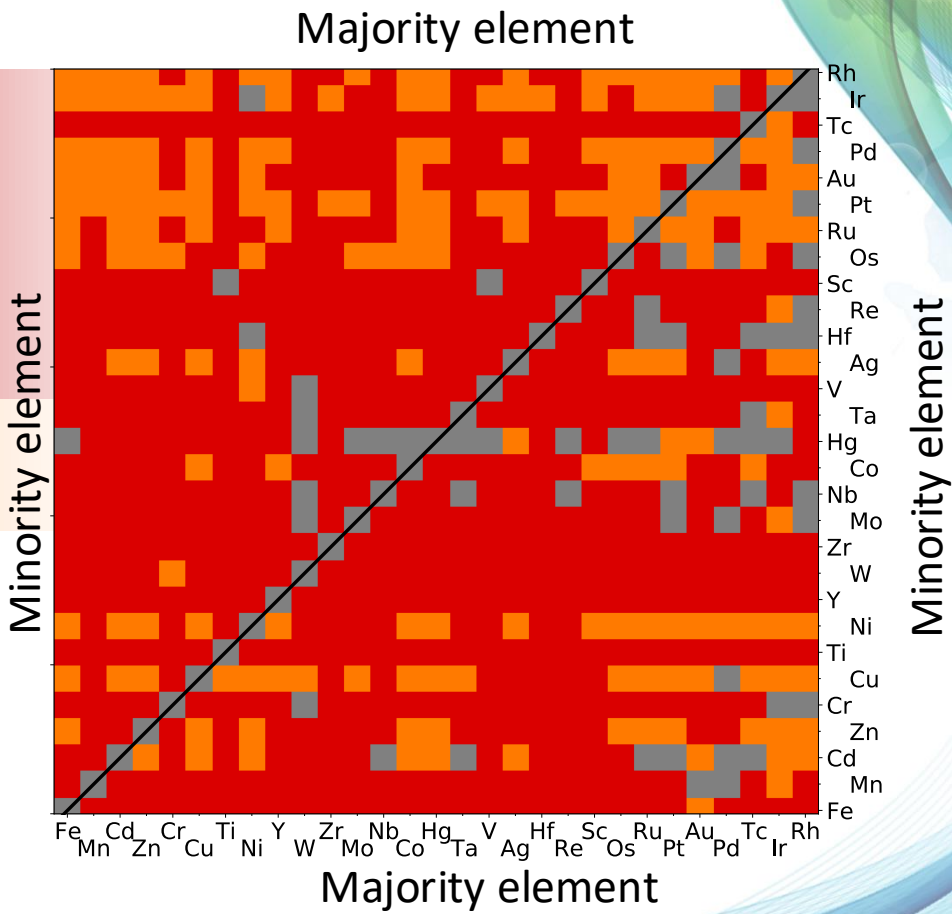
Aqueous stability

1510

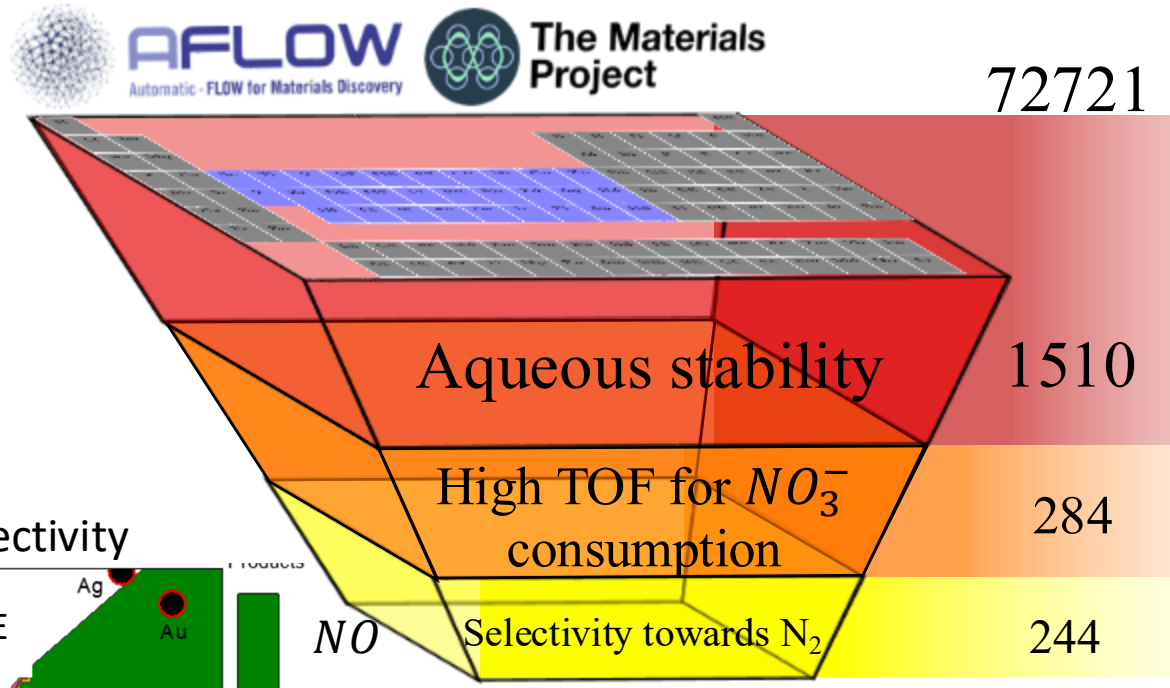
High TOF for NO_3^- consumption

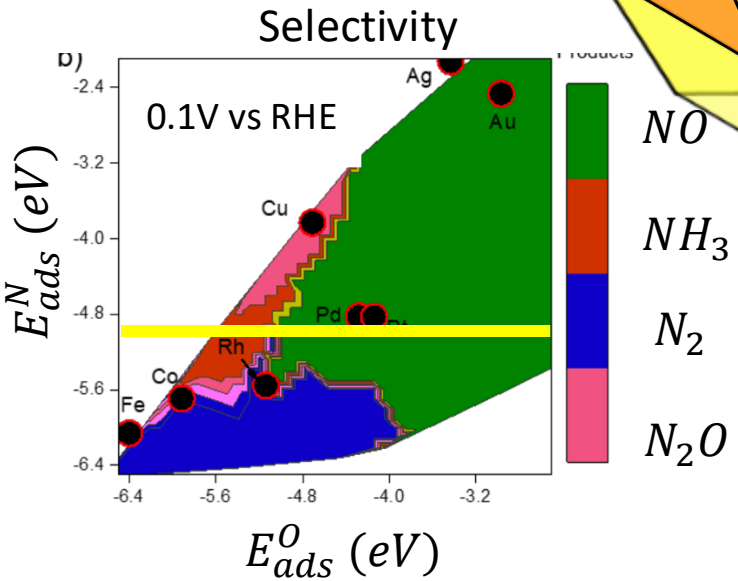
284

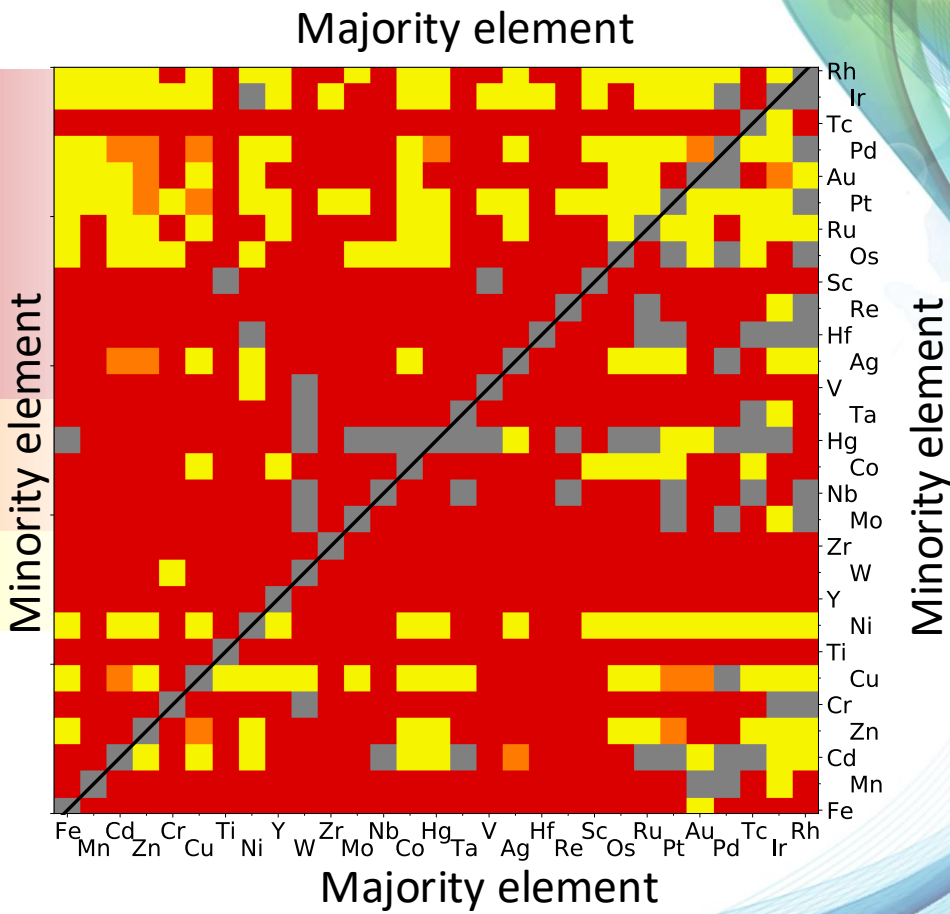




ML assisted screening







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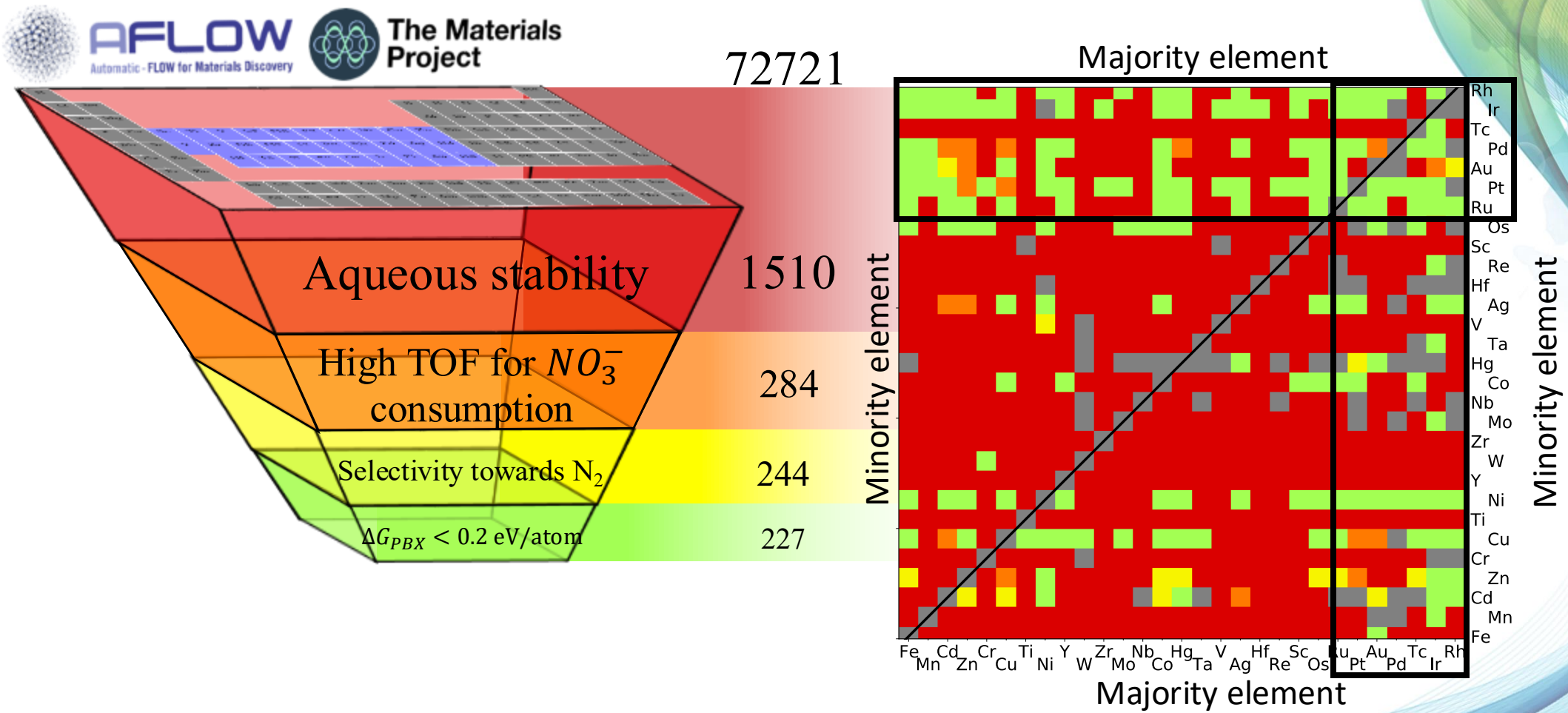

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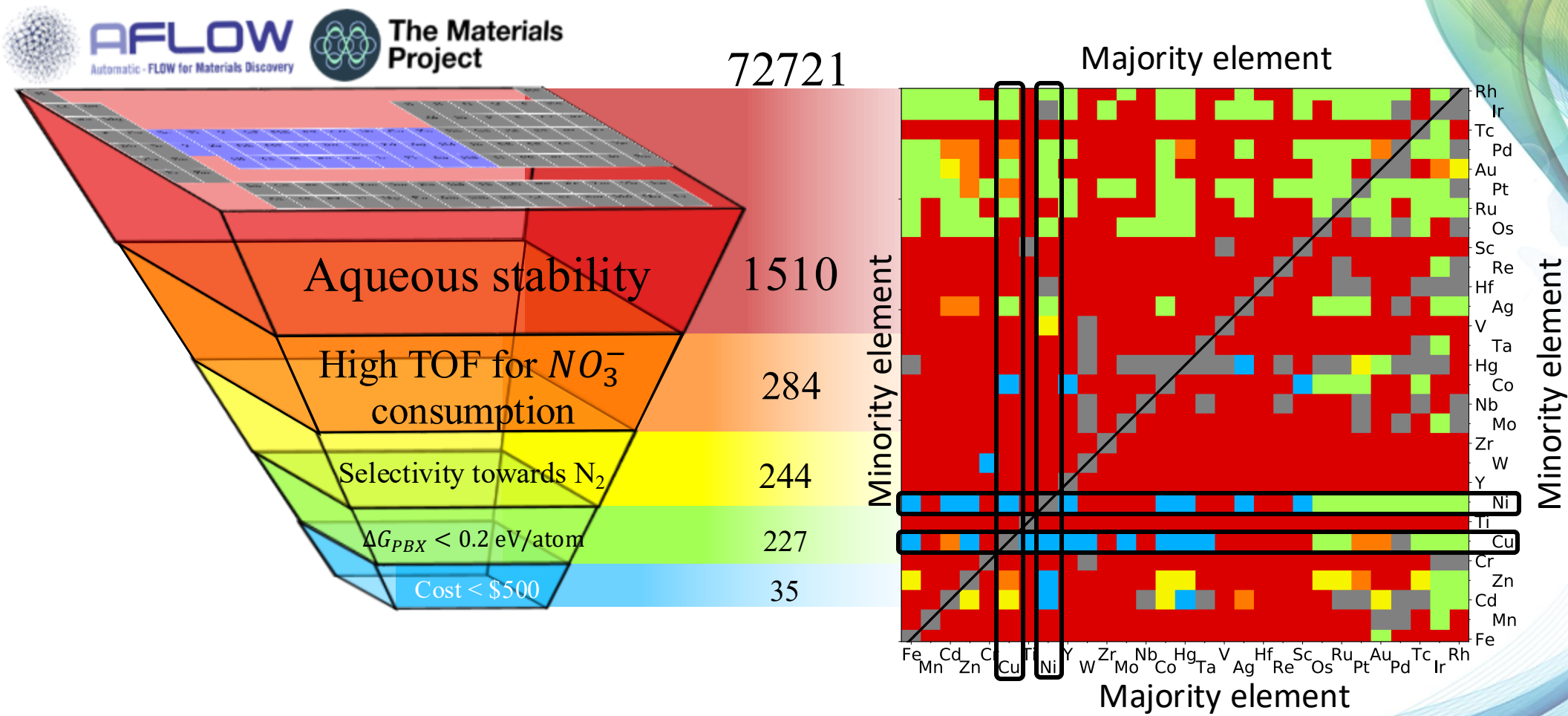
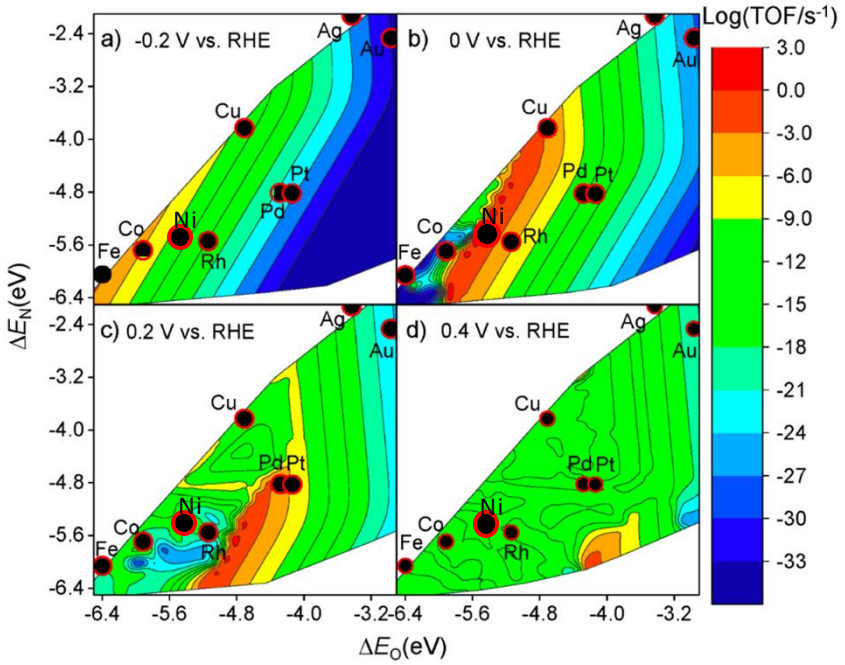
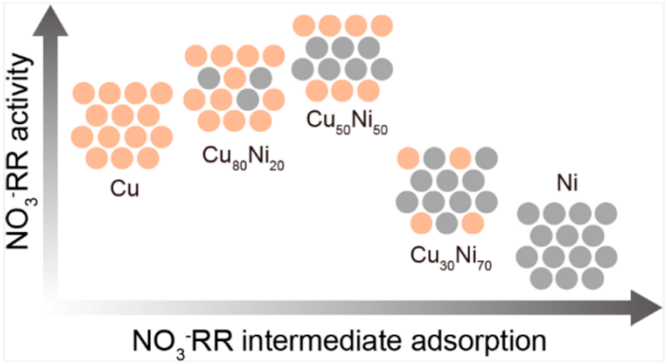
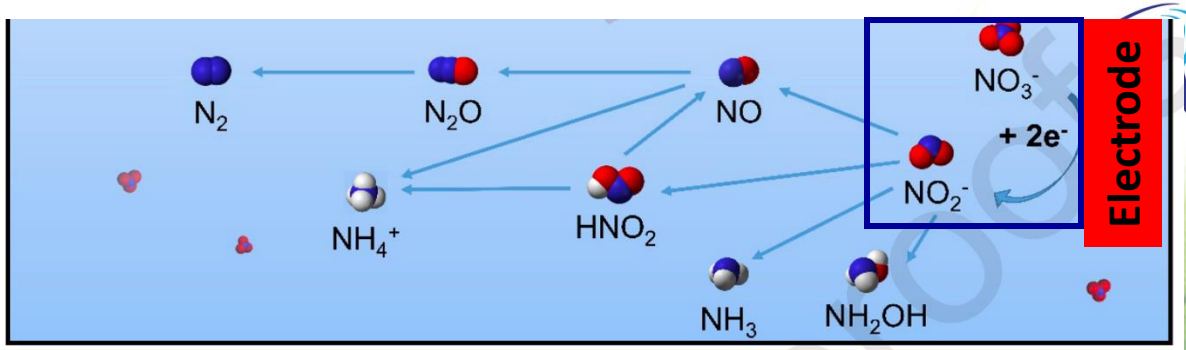
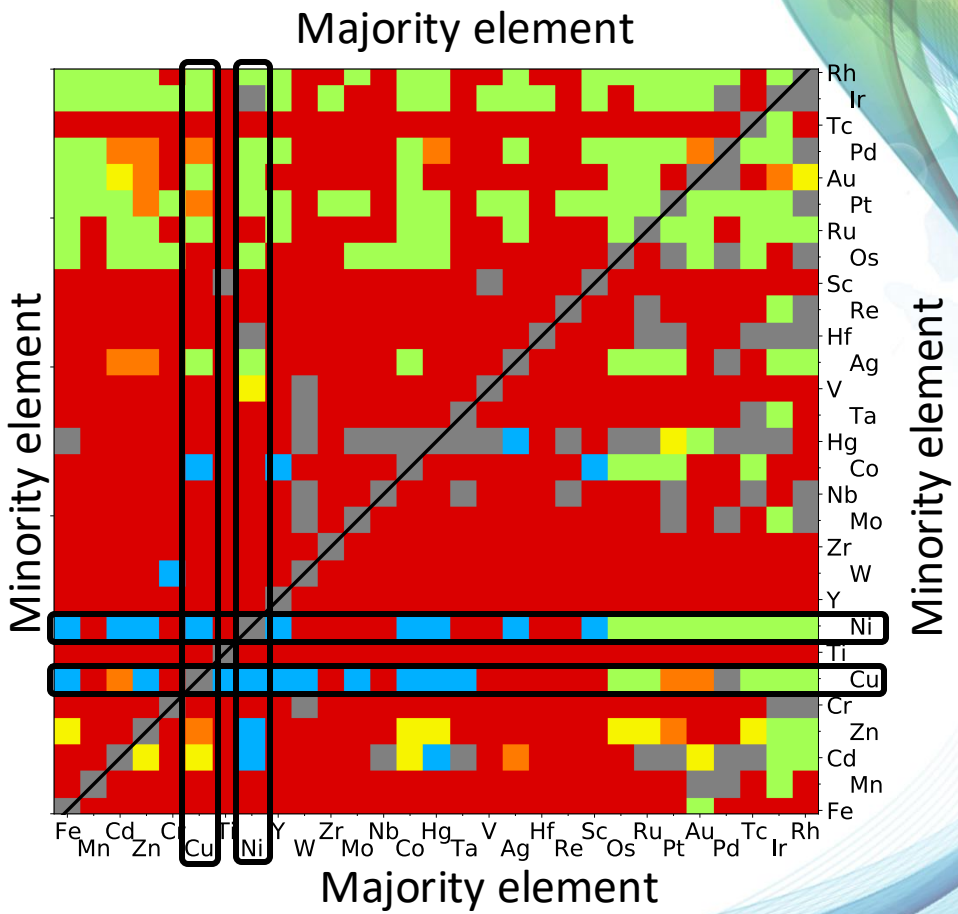


Cu-M and Ni-M

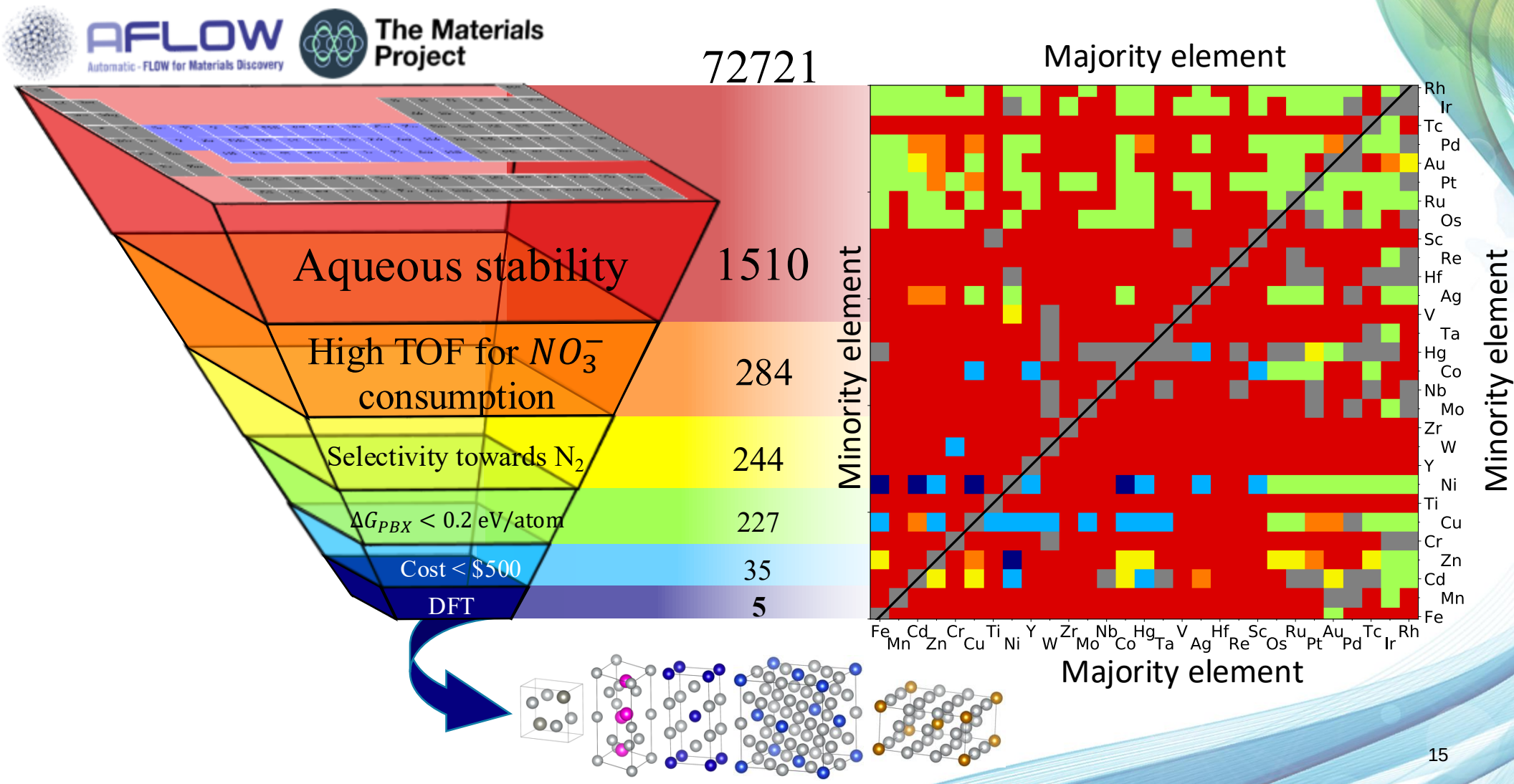

a) -0.2 V vs. RHE
b) 0 V vs. RHE
c) 0.2 V vs. RHE
d) 0.4 V vs. RHE
Log(TOF/s-1)
ΔEN(eV)
ΔEO(eV)


NO3-RR activity
Cu
Cu80Ni20
Cu50Ni50
Cu30Ni70
Ni
NO3-RR intermediate adsorption


N2
N2O
NO
NO3-
+ 2e-
NO2-
NH4+
HNO2
NH3
NH2OH
Electrode

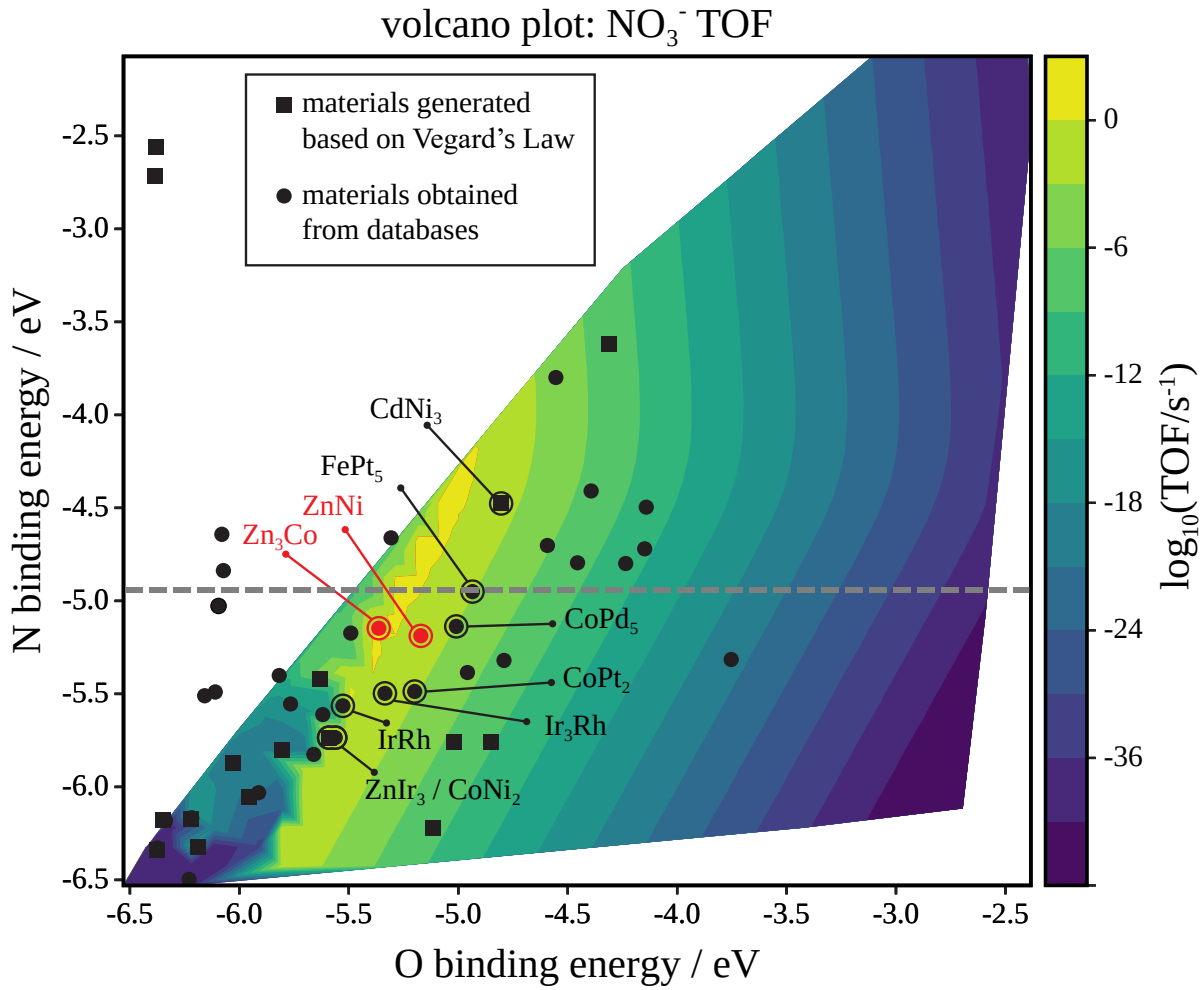

Majority element
Minority element
Minority element
Majority element

ML assisted screening



DFT validation

(0.0 V vs. RHE)

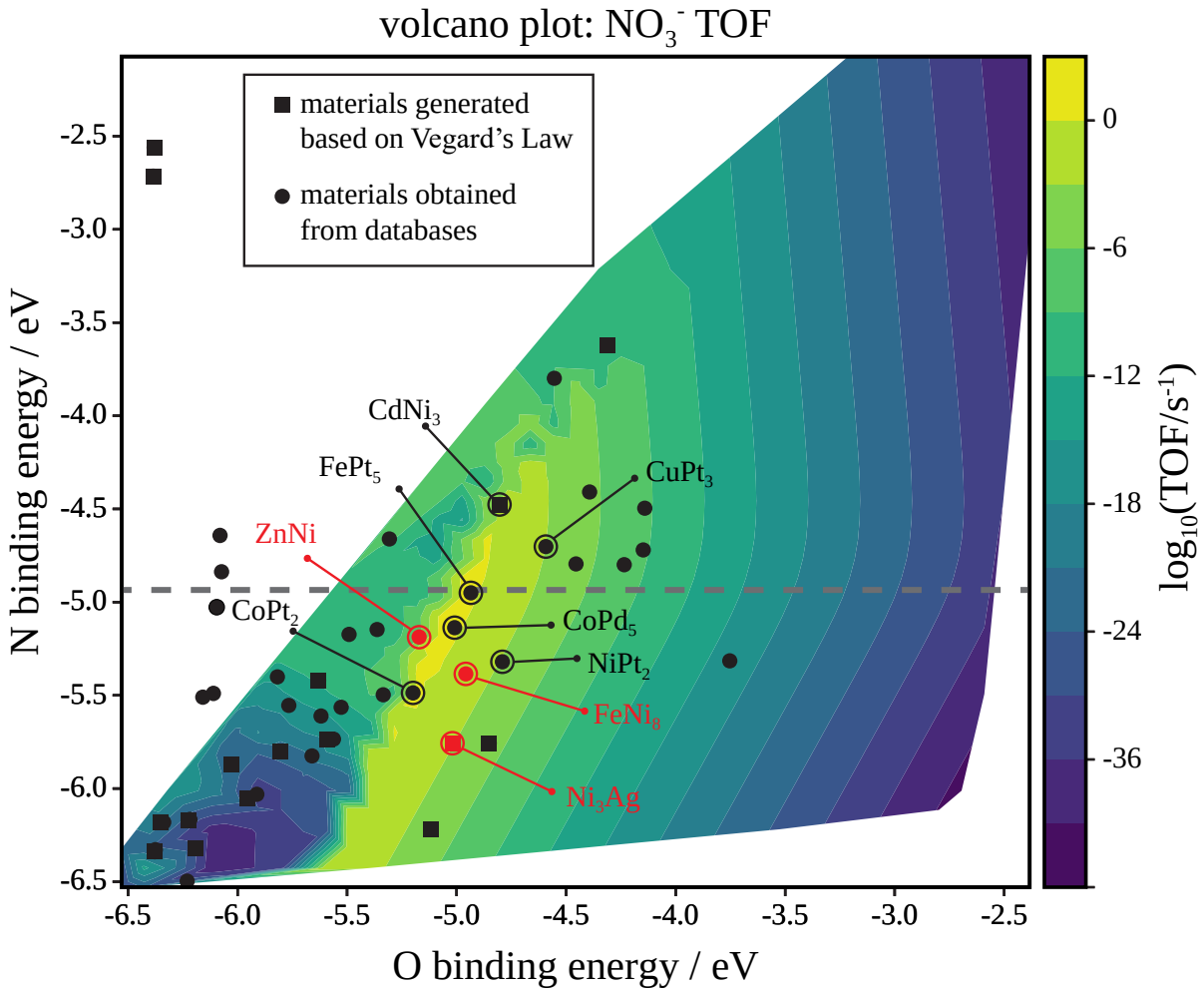


Calculated materials

ZnNi	AFLOW/MP
Zn_3Co	AFLOW/MP
$CdNi_3$	Template
$CoPt_2$	AFLOW/MP
$FePt_5$	AFLOW/MP
$CoPd_5$	Template
Ir_3Rh	AFLOW/MP
IrRh	AFLOW/MP
$ZnIr_3$	Template
$CoNi_2$	AFLOW/MP

DFT validation

(0.1 V vs. RHE)

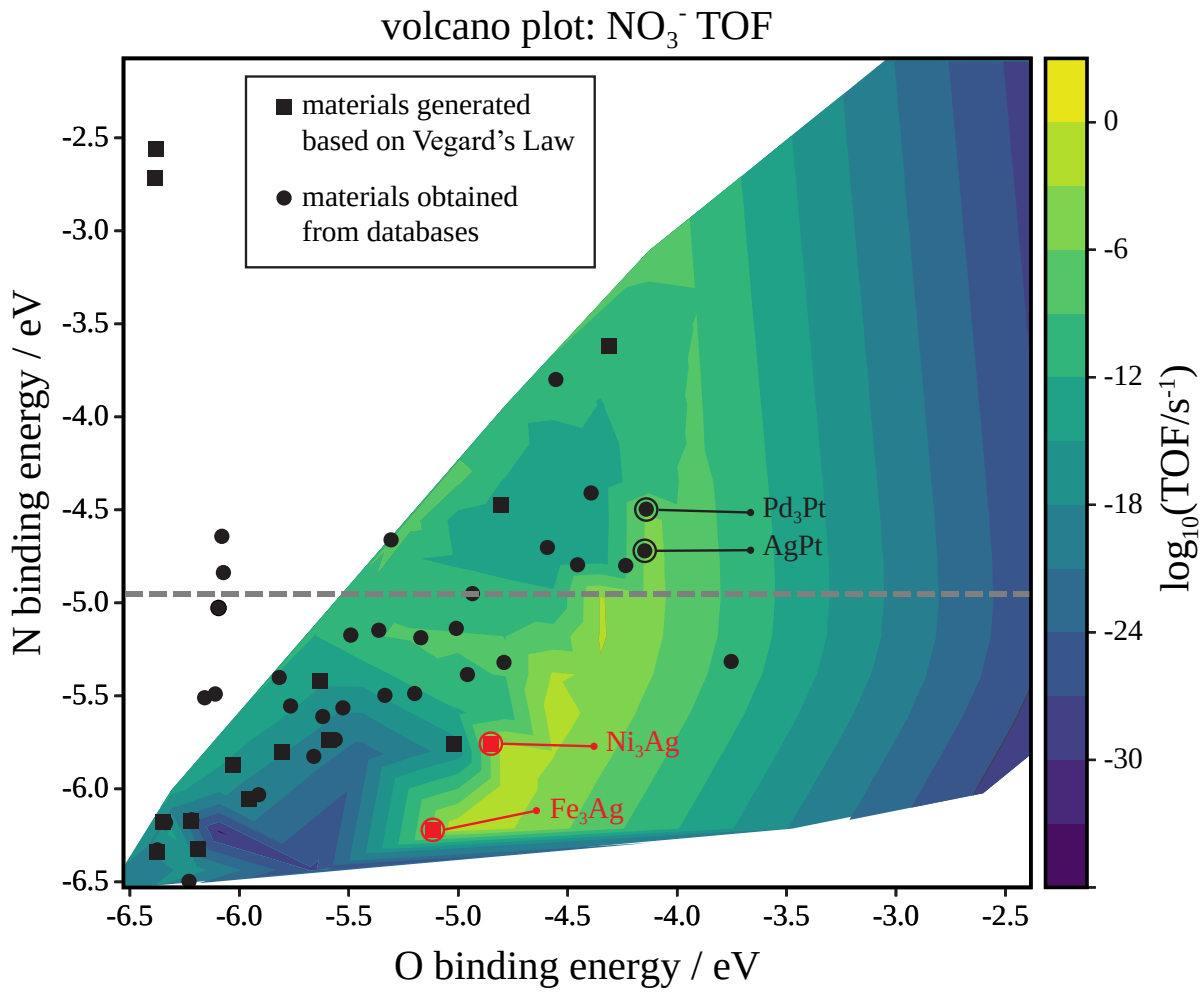


Calculated materials

ZnNi	AFLOW/MP
$FeNi_8$	AFLOW/MP
Ni_3Ag	Template
$CoPt_2$	AFLOW/MP
$FePt_5$	AFLOW/MP
$CdNi_3$	Template
$CuPt_3$	AFLOW/MP
$CoPd_5$	AFLOW/MP
$NiPt_2$	AFLOW/MP

DFT validation

(0.2 V vs. RHE)

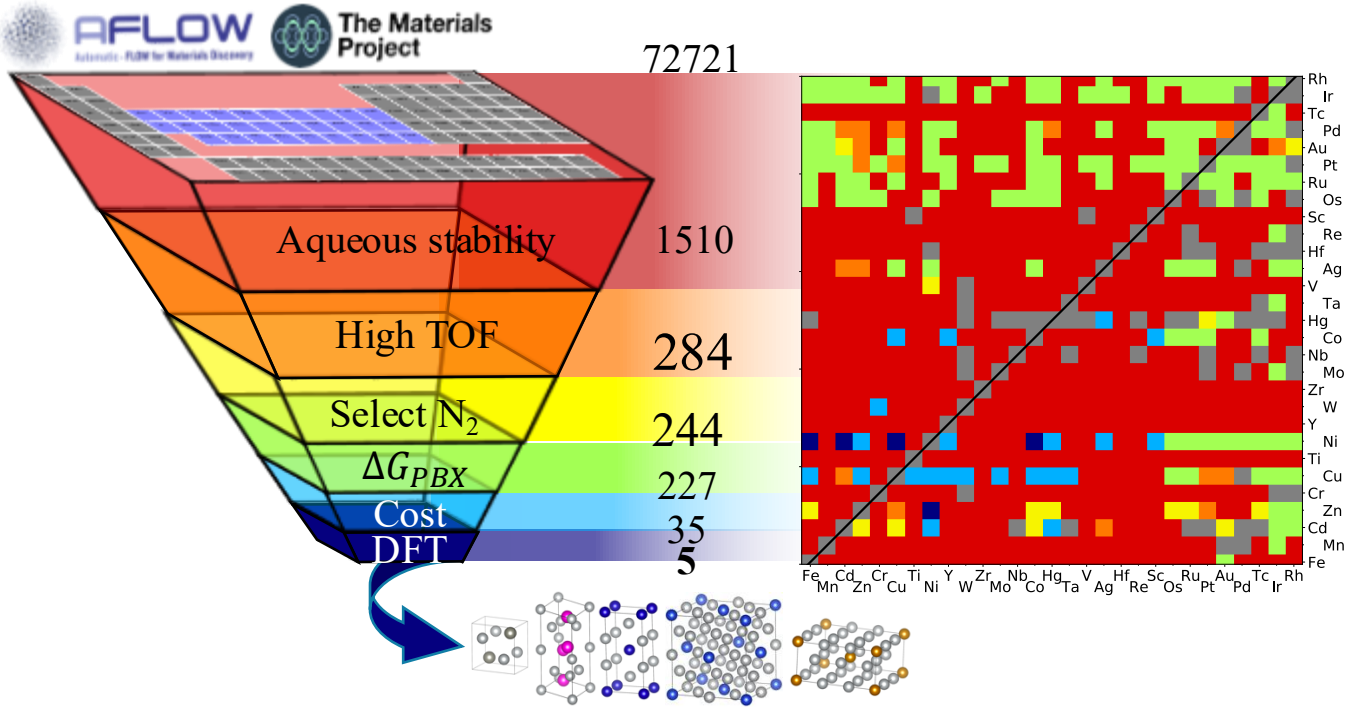


Calculated materials

Fe_3Ag	Template
Ni_3Ag	Template
$AgPt$	AFLOW/MP
Pd_3Pt	AFLOW/MP

Conclusion

- Screened the MP/AFLOW DBs for aqueously stable binary intermetallics.
- ML with OC20 provides a quick estimate of their adsorption energies which would otherwise be unfeasable with DFT
- Using microkinetic/scaling models from the literature, we found 35 economical bimetallics that can facilitate NO_3RR DFT verifying their E_{ads}



- Our results corroborate with previously known observations. Cu- and Ni- base intermetallics yield synergistic effects that makes them great for NO_3RR

Acknowledgements

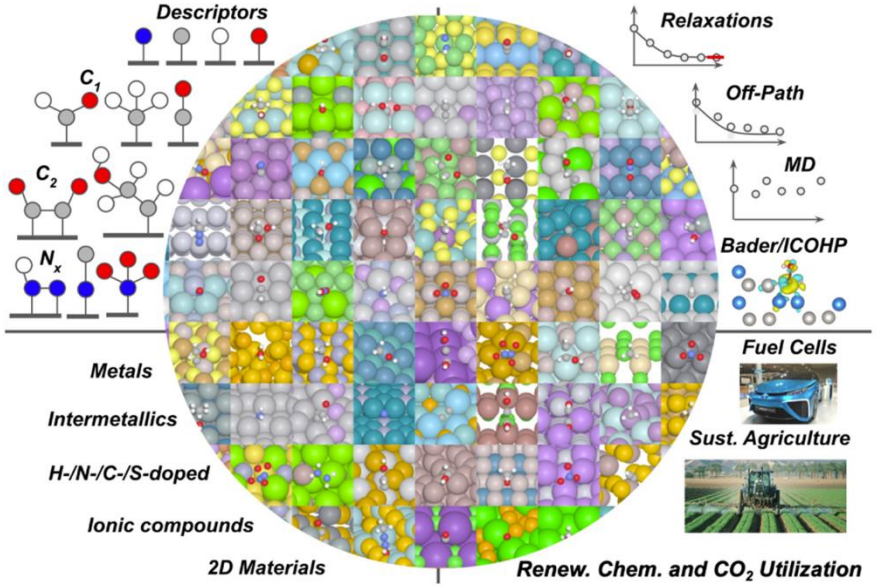
Funding

NAWI
National Alliance for Water Innovation

Computing resources

NREL
Transforming ENERGY

Open Catalyst Project



In collaboration with:

- Anubhav Jain (HT calculations)
- Jeffrey Preece (Techno-economic modeling)
- Wei Tong (Rapid materials synthesis and characterization)
- Bruce Moyer (Guidance in overall project)

Questions