Carnegie Mellon University

Data-Driven Assisted Quantum Chemistry of Catalytic Materials

MAY 11 2022

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2022 MRS® SPRING MEETING & EXHIBIT
May 8–13, 2022 I Honolulu, Hawai‘i
May 23–25, 2022 I Virtual

The


LOCAL NEWS ›
Earth 365: Is climate change causing more severe weather in Pittsburgh?
CBS NEWS PITTSBURGH
BY KRISTIN EMERY
APRIL 18, 2022 / 5:41 PM / CBS PITTSBURGH
NEW AT 5:00
SCAN HERE FOR KDKA NEWS APP
Climate Change & Extreme Weather
5:30 37°
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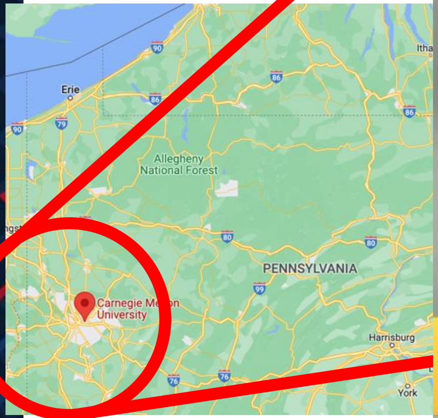
@KDKA
CTL 0:01:32:10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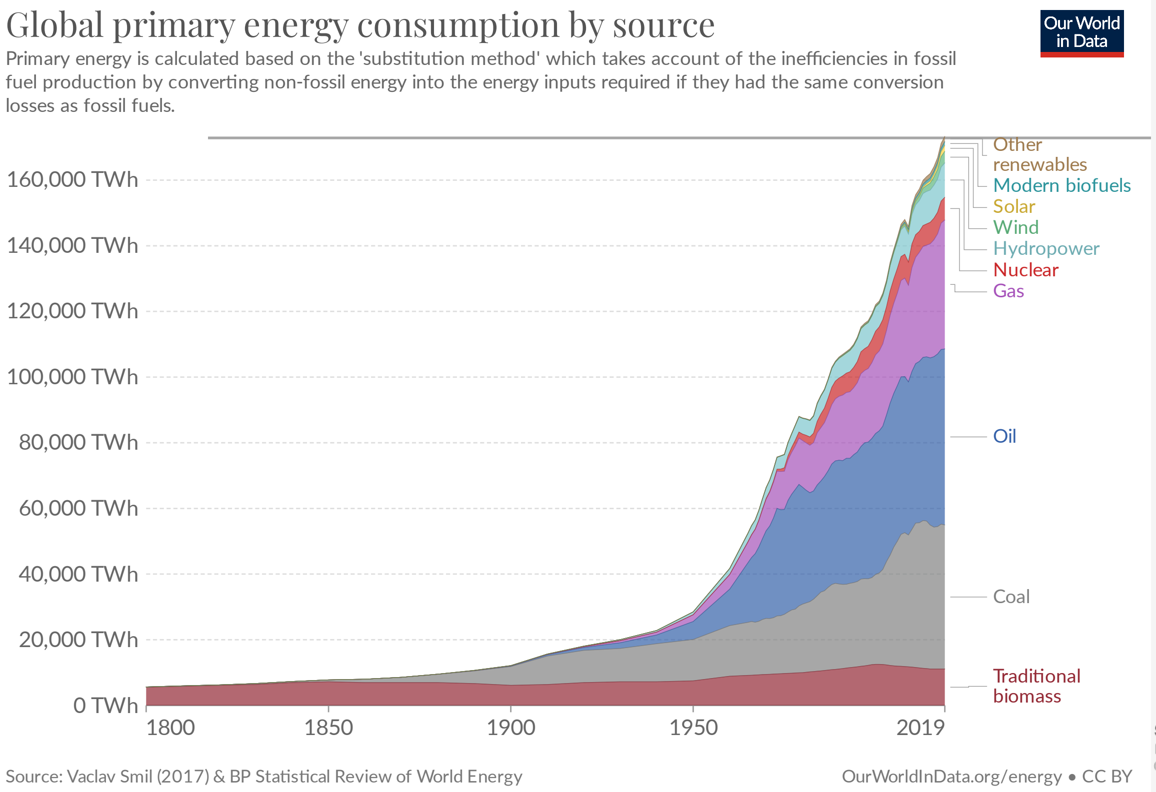
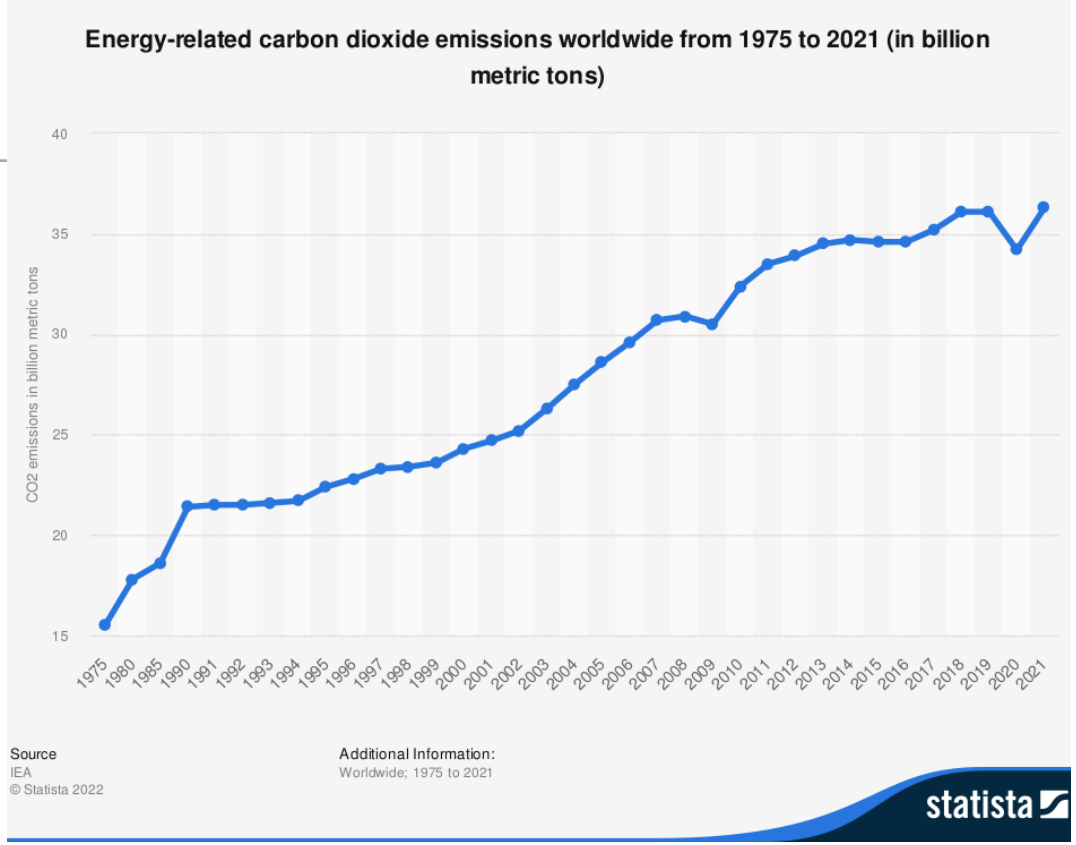

Global primary energy consumption by source
Our World in Data
Primary energy is calculated based on the 'substitution method' which takes account of the inefficiencies in fossil fuel production by converting non-fossil energy into the energy inputs required if they had the same conversion losses as fossil fuels.
160,000 TWh
140,000 TWh
120,000 TWh
100,000 TWh
80,000 TWh
60,000 TWh
40,000 TWh
20,000 TWh
0 TWh
1800
1850
1900
1950
2019
Other renewables
Modern biofuels
Solar
Wind
Hydropower
Nuclear
Gas
Oil
Coal
Traditional biomass
Source: Vaclav Smil (2017) & BP Statistical Review of World Energy
OurWorldInData.org/energy • CC BY


Energy-related carbon dioxide emissions worldwide from 1975 to 2021 (in billion metric tons)
CO2 emissions in billion metric tons
40
35
30
25
20
15
1975
1980
1985
1990
1991
1992
1993
1994
1995
1996
1997
1998
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Source
IEA
© Statista 2022
Additional Information:
Worldwide; 1975 to 2021
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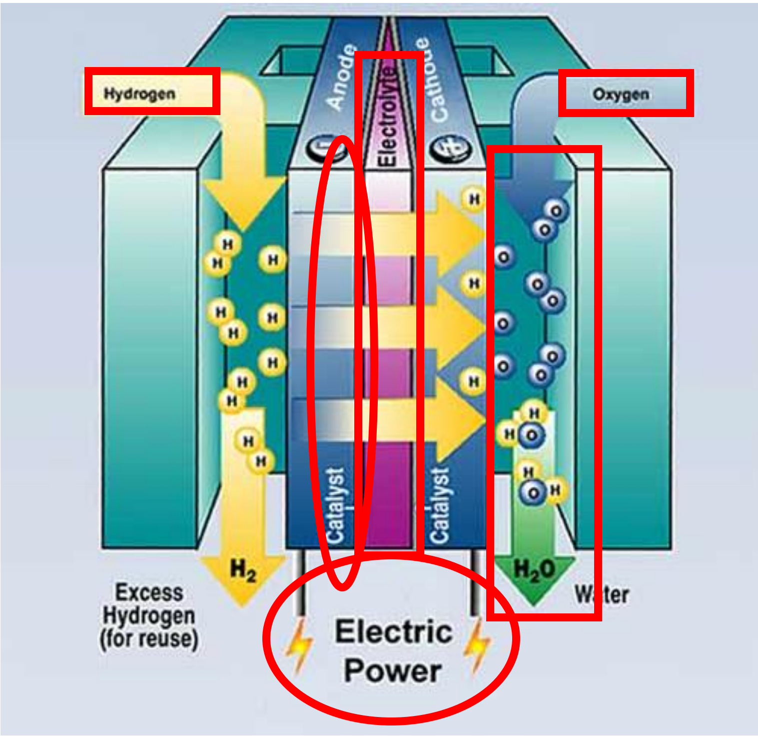
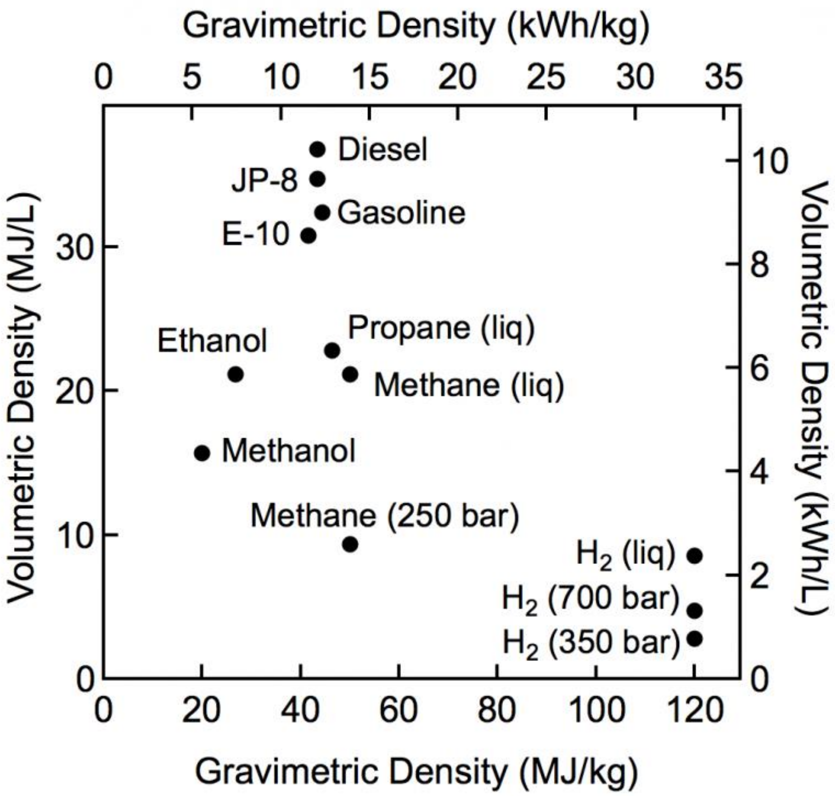
Hydrogen fuel cell



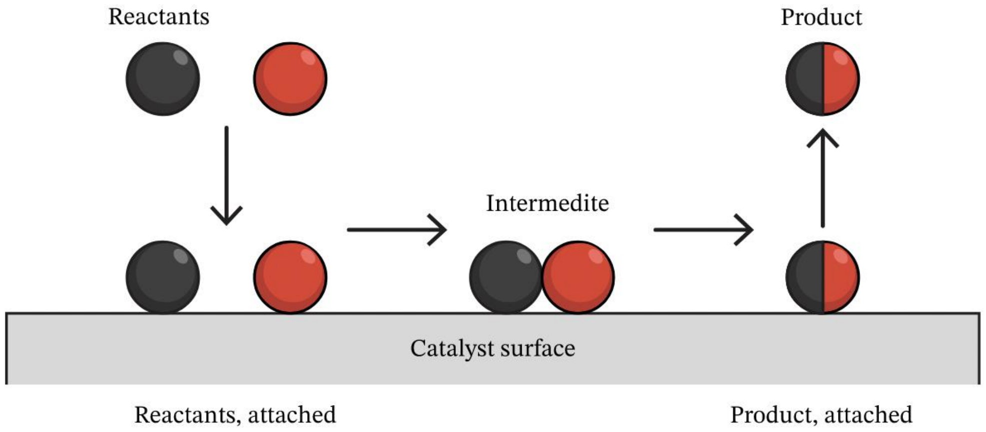


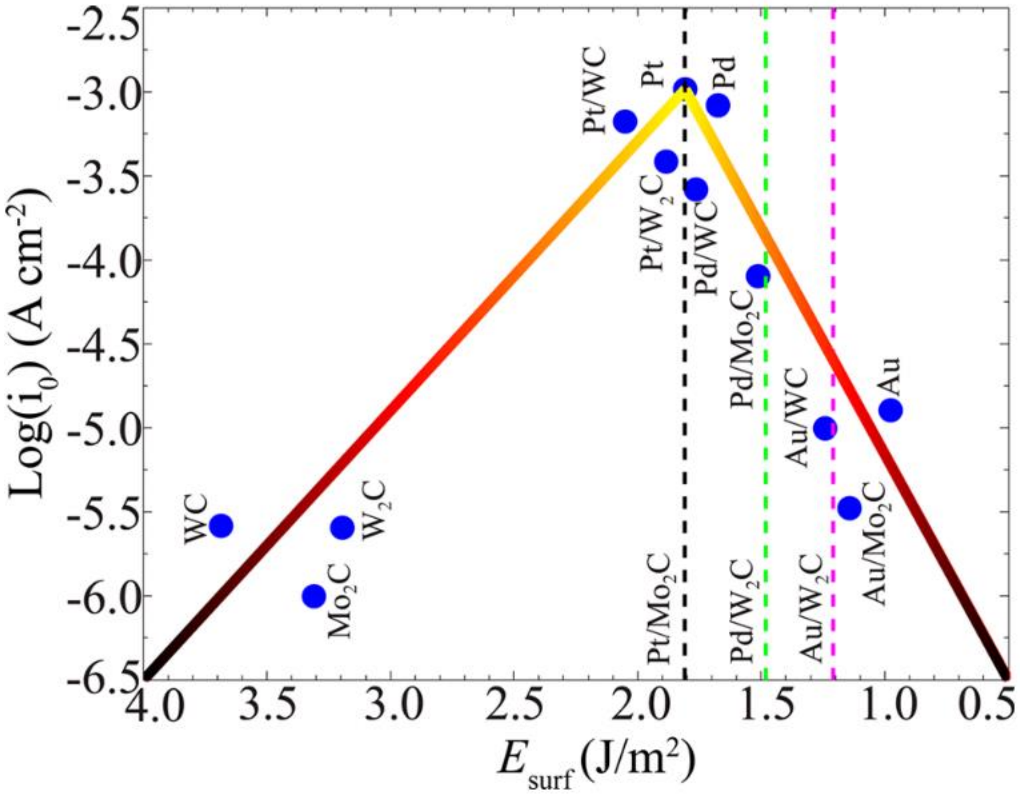
Data driven surface science in recent years

Zhuang, H., Tkalych, A. J., & Carter, E. A. (2016). Surface Energy as a Descriptor of Catalytic Activity. *Journal of Physical Chemistry C*, *120*(41), 23698–23706. https://doi.org/10.1021/acs.jpcc.6b09687

Surface energy

$$\gamma = \frac{E_{slab} - NE_{bulk}}{2A}$$





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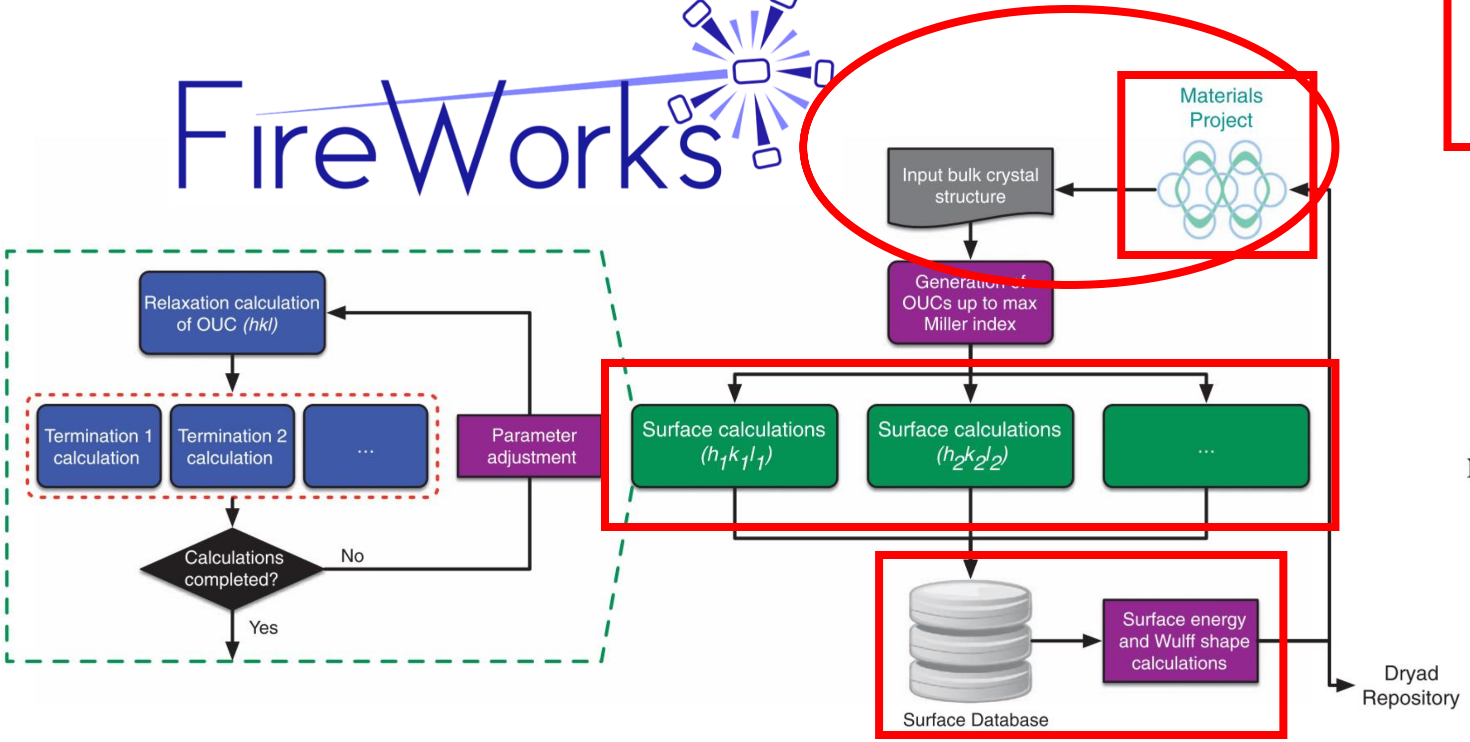
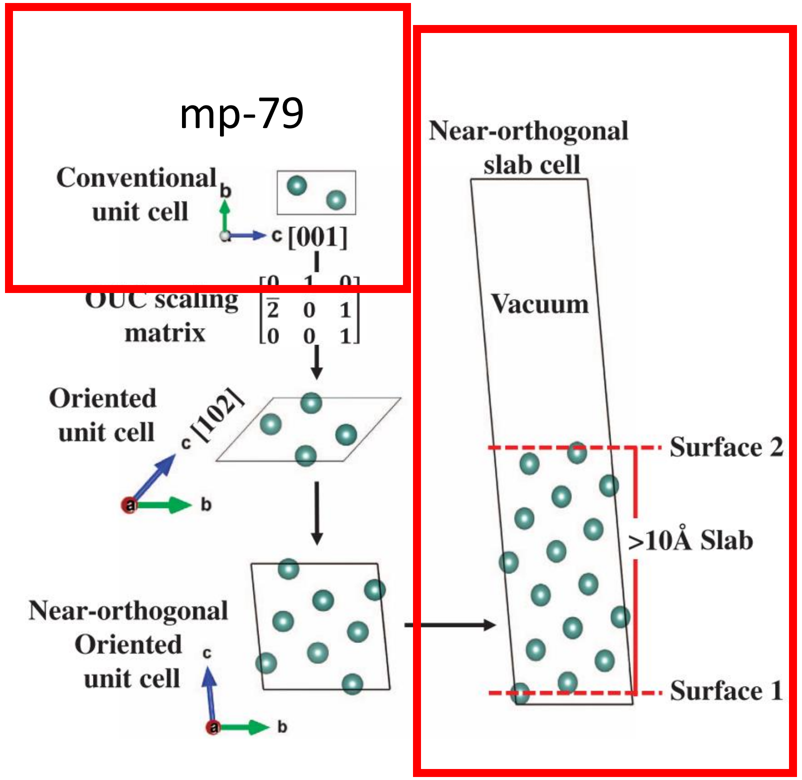
Tran, R., Li, X.-G., Montoya, J., Winston, D., Persson, K. A., & Ong, S. P. (2019). Anisotropic work function of elemental crystals. *Surface Science*, *687*(September), 48–55.
Tran, R., Xu, Z., Radhakrishnan, B., Winston, D., Sun, W., Persson, K. A., & Ong, S. P. (2016). Data Descripter: Surface energies of elemental crystals. *Scientific Data*, *3*(160080), 1–13.

High throughput efforts and databases

pymatgen

FireWorks





Tran, R., Li, X.-G., Montoya, J., Winston, D., Persson, K. A., & Ong, S. P. (2019). Anisotropic work function of elemental crystals. *Surface Science*, *687*(September), 48–55.
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High throughput efforts and databases

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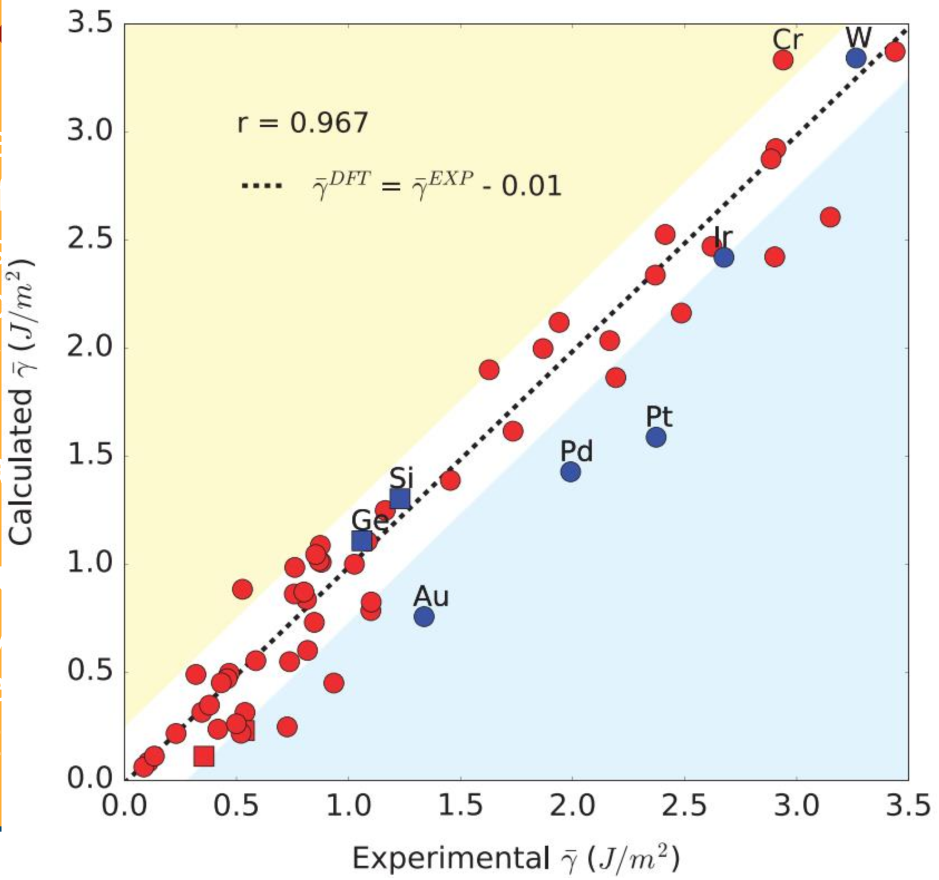
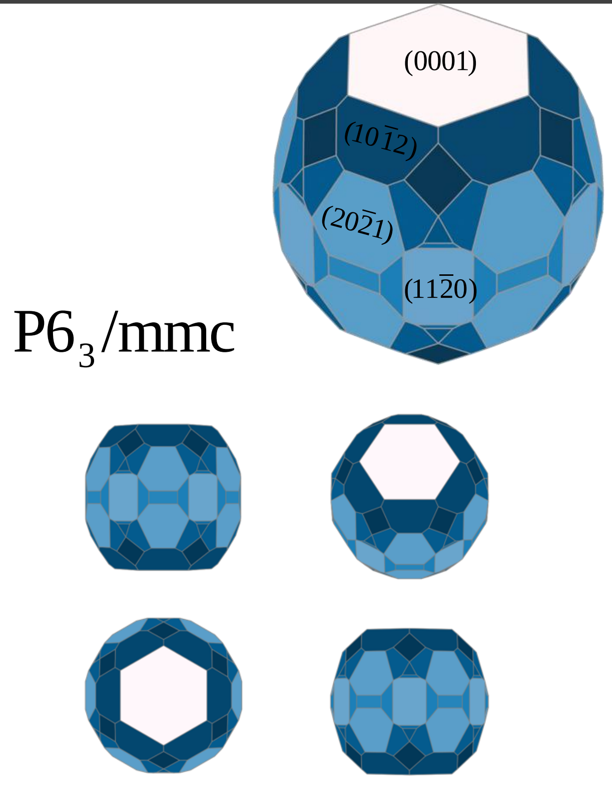


N
J/m²
1.50
1.55
1.66
1.67
1.68
1.69
1.70
1.71
1.72
1.73
1.74
1.75
1.77



$\boxed{\bar{g} = 1.86\ J/m^2}$ $\quad \eta = 5.10$

$\bar{\phi} = 3.51\ eV \quad \alpha_\gamma = 0.05$

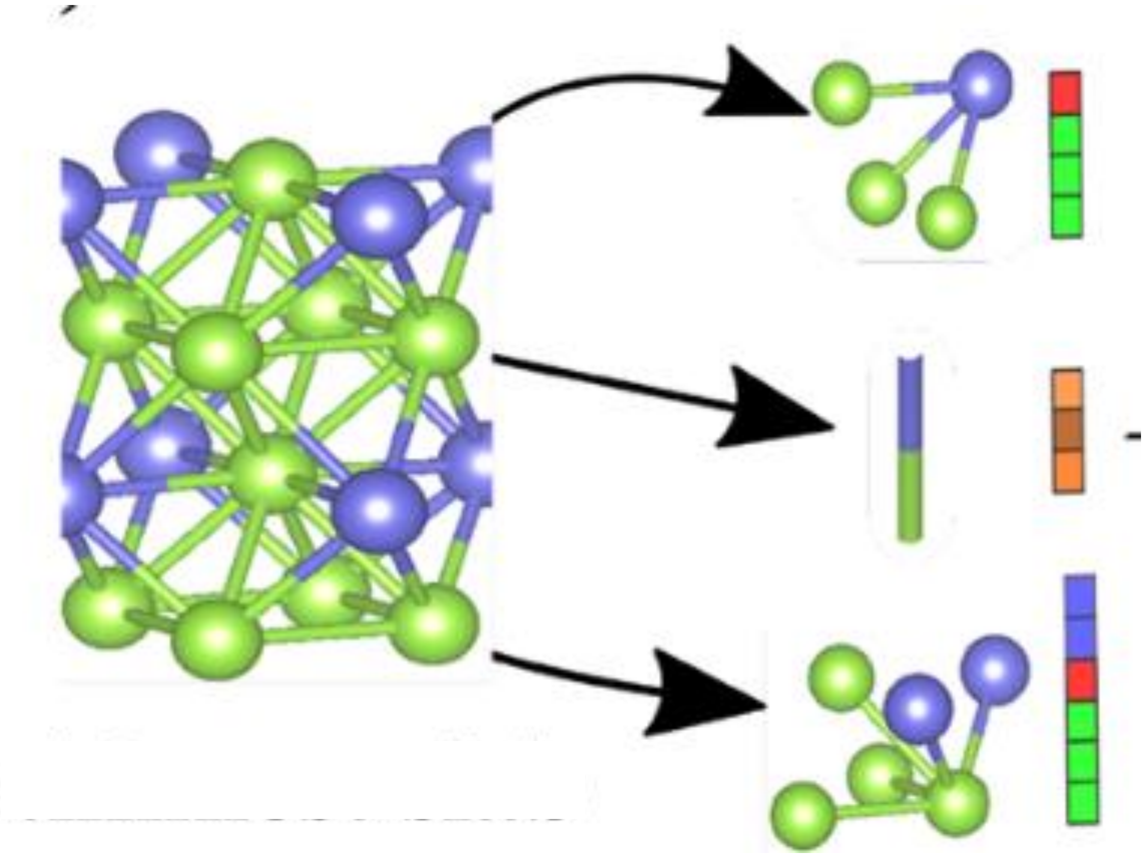
$$\gamma = \frac{1}{2A}(E_{slab} - NE_{bulk})$$

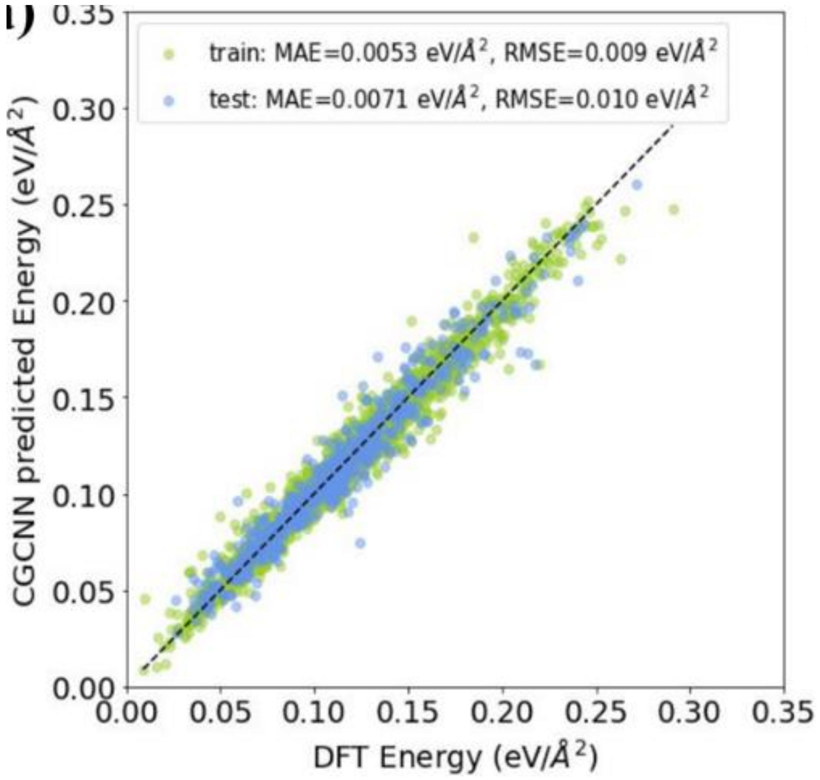
$$\phi = V_{vac} - E_F$$

http://crystalium.materialsvirtuallab.org

Palizhati, A., Zhong, W., Tran, K., Back, S., & Ulissi, Z. W. (2019). Toward Predicting Intermetallics Surface Properties with High-Throughput DFT and Convolutional Neural Networks. *Journal of Chemical Information and Modeling*, *59*(11), 4742–4749.

Machine learning

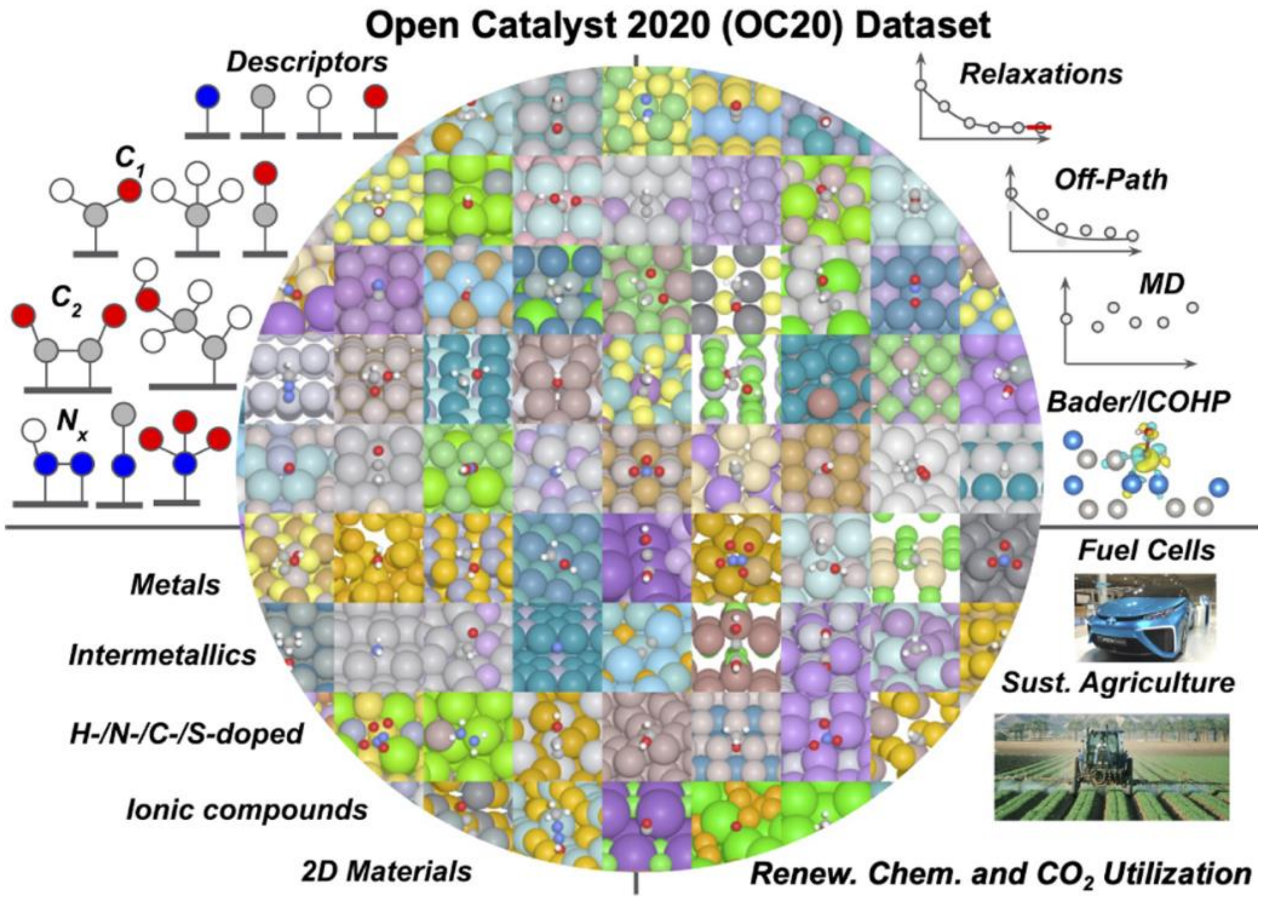




Cleavage energy of 3,000 intermetallic surfaces

Chanussot, L., Das, A., Goyal, S., Lavril, T., Shuaibi, M., Riviere, M., Tran, K., Heras-Domingo, J., Ho, C., Hu, W., Palizhati, A., Sriram, A., Wood, B., Yoon, J., Parikh, D., Zitnick, C. L., & Ulissi, Z. (2021). Open Catalyst 2020 (OC20) Dataset and Community Challenges. *ACS Catalysis*, *11*(10), 6059–6072.

Open Catalyst Project



Scope of data

- GNN-based models for adsorption energy and adsorption induced relaxation
- 82 adsorbates
- 55 elements
- 11,451 materials (unary/binary/ternary)
- Miller index up to 2
- 872,000 ionic steps

Chanussot, L., Das, A., Goyal, S., Lavril, T., Shuaibi, M., Riviere, M., Tran, K., Heras-Domingo, J., Ho, C., Hu, W., Palizhati, A., Sriram, A., Wood, B., Yoon, J., Parikh, D., Zitnick, C. L., & Ulissi, Z. (2021). Open Catalyst 2020 (OC20) Dataset and Community Challenges. *ACS Catalysis*, *11*(10), 6059–6072.

Models and metrics

Model	S2EF test			
	ID	OOD Ads	OOD Cat	OOD Both
	Energy MAE [eV] ↓			
median baseline	2.0430	2.4203	1.9916	2.5770
CGCNN[86]	0.5272	0.6322	0.5372	0.7675
SchNet[87]	0.4426	0.4907	0.5288	0.7161
SchNet[87] —force-only	34.0316	33.769	35.2982	38.4652
SchNet[87] —energy-only	0.3948	0.4460	0.5510	0.7031
DimeNet++[88,89]	0.4858	0.4702	0.5331	0.6482
DimeNet++[88,89] —force-only	28.2134	28.9428	28.9069	34.9049
DimeNet++[88,89] —energy-only	0.3586	0.4022	0.5060	0.6540
DimeNet++[88,89]-large—force-only	29.3382	30.0365	30.0461	36.7537

Model	Approach	IS2RE est			
		Energy MAE [eV] ↓			
		ID	OOD Ads	OOD Cat	OOD Both
Median baseline		1.7489	1.8911	1.7107	1.6807
CGCNN (1)	Direct	0.6135	0.9155	0.6211	0.8506
SchNet (2)	Direct	0.6372	0.7342	0.6611	0.7035
DimeNet++ (3,4)	Direct	0.5605	0.7252	0.5750	0.6613
SchNet (2)	Relaxation	0.7088	0.7741	0.7665	0.8055
SchNet (2) force-only + energy-only	Relaxation	0.7066	0.7420	0.7966	0.7493
DimeNet++ (3,4)	Relaxation	0.6687	0.6864	0.6858	0.6835
DimeNet++ (3,4) force-only + energy-only	Relaxation	0.5112	0.5744	0.5922	0.6130
Dimenet++ (3,4) – large force-only + energy-only	Relaxation	0.5022	0.5430	0.5780	0.6117

IS2RE: Initial structure to relaxed adsorption energy
S2EF: Structure to adsorption energy and force

Materials discovery:
Nitrate reduction reaction

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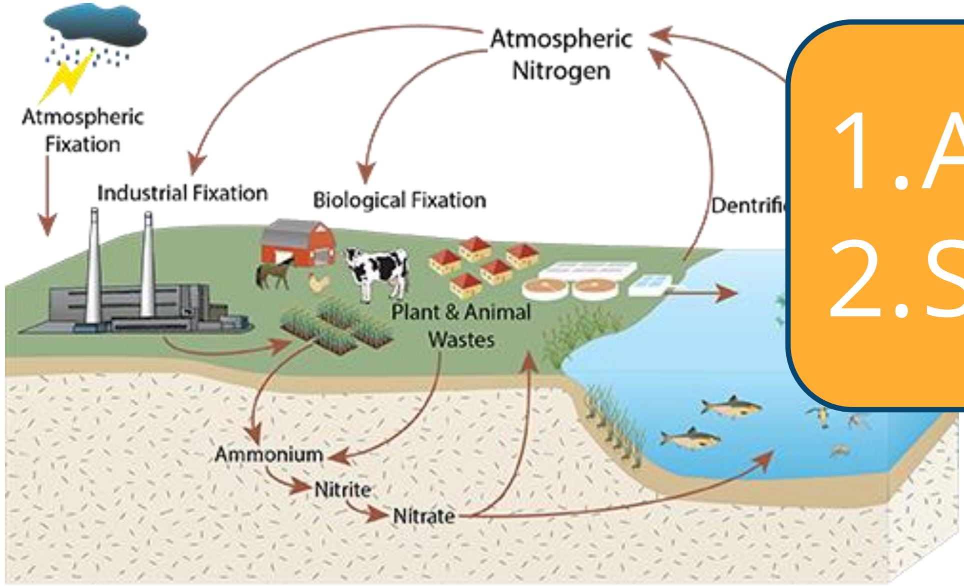
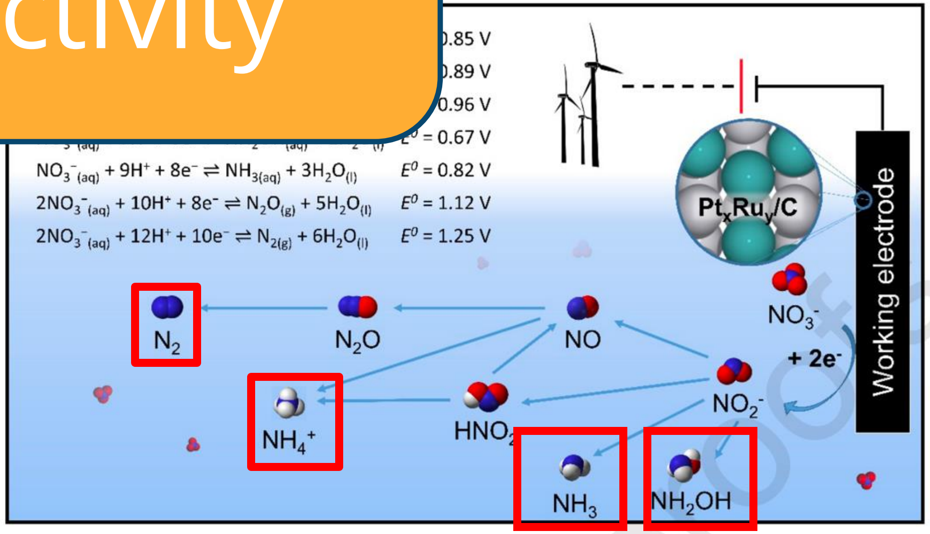
Wang, Z., Young, S. D., Goldsmith, B. R., & Singh, N. (2021). Increasing electrocatalytic nitrate reduction activity by controlling adsorption through PtRu alloying. *Journal of Catalysis*, *395*(3), 143–154.

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
		.46-0.64	$0.04-0.32	$0.70-1.24
		.50-0.80	$0.01-0.02	$0.91-1.72
		.12-1.57	*n/a*	*$?*



1. Activity
2. Selectivity

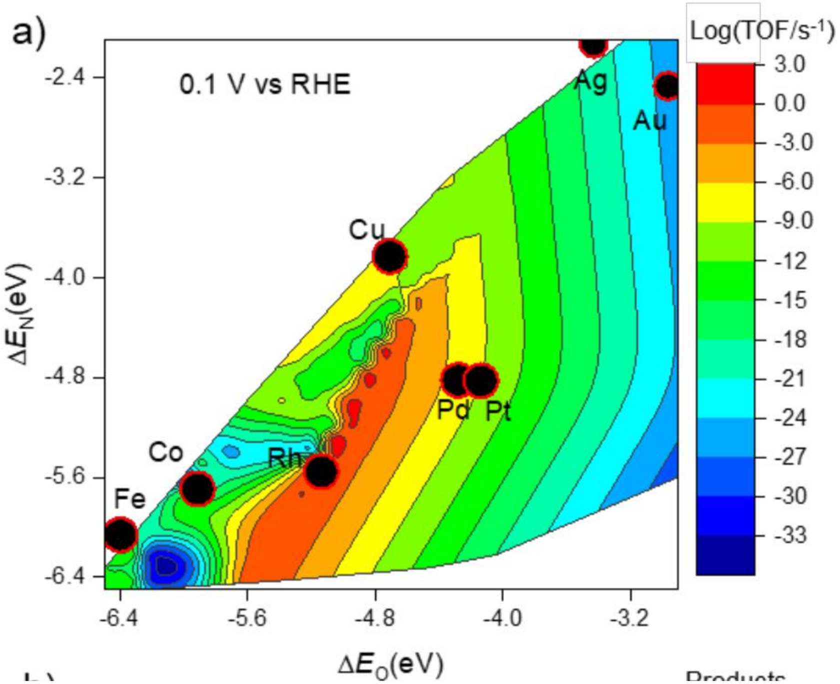


Liu, J. X., Richards, D., Singh, N., & Goldsmith, B. R. (2019). Activity and Selectivity Trends in Electrocatalytic Nitrate Reduction on Transition Metals. *ACS Catalysis*, *9*(8), 7052–7064.

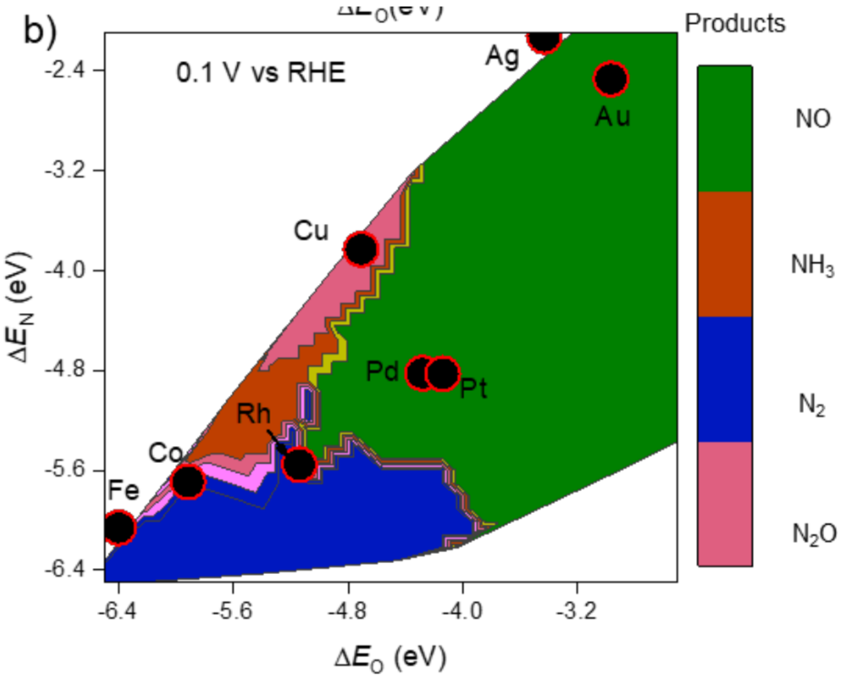
Application to catalyst discovery

Energy barriers and therefore *k and activity* is now a function of adsorption energy

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

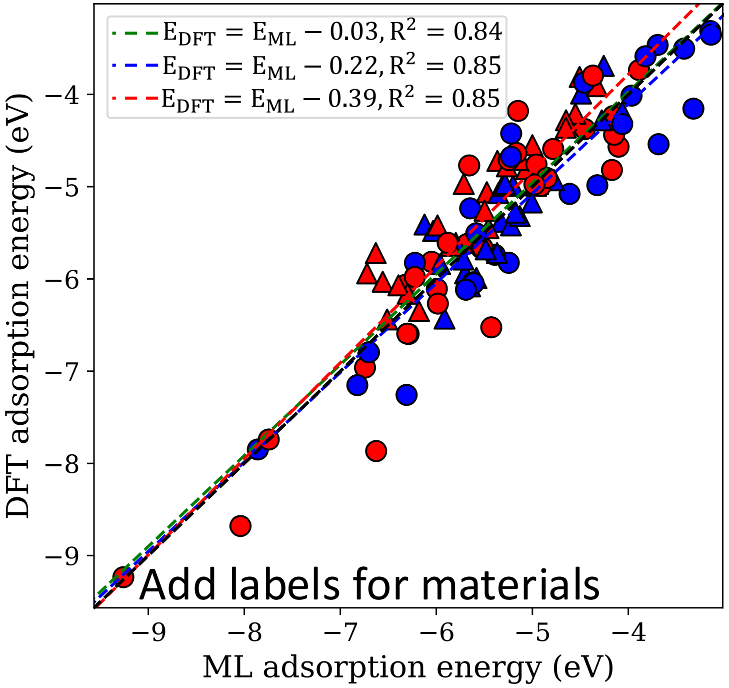


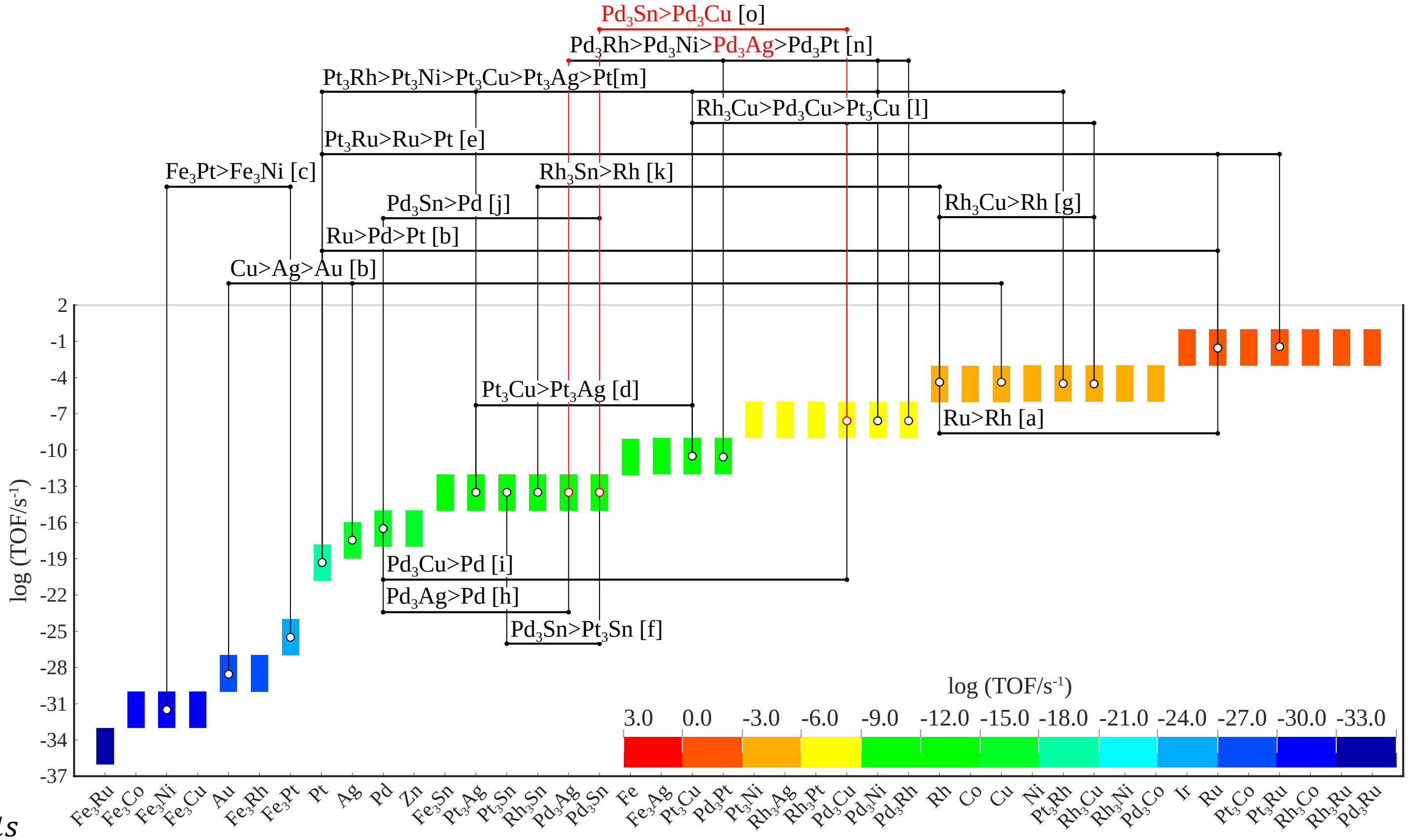
Activity map



Selectivity map

DFT and ML verification

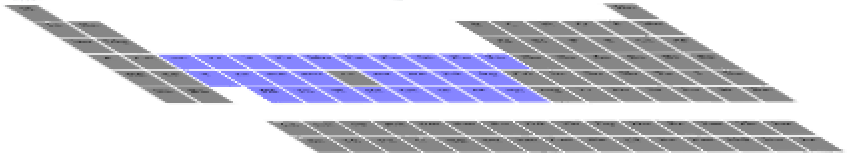




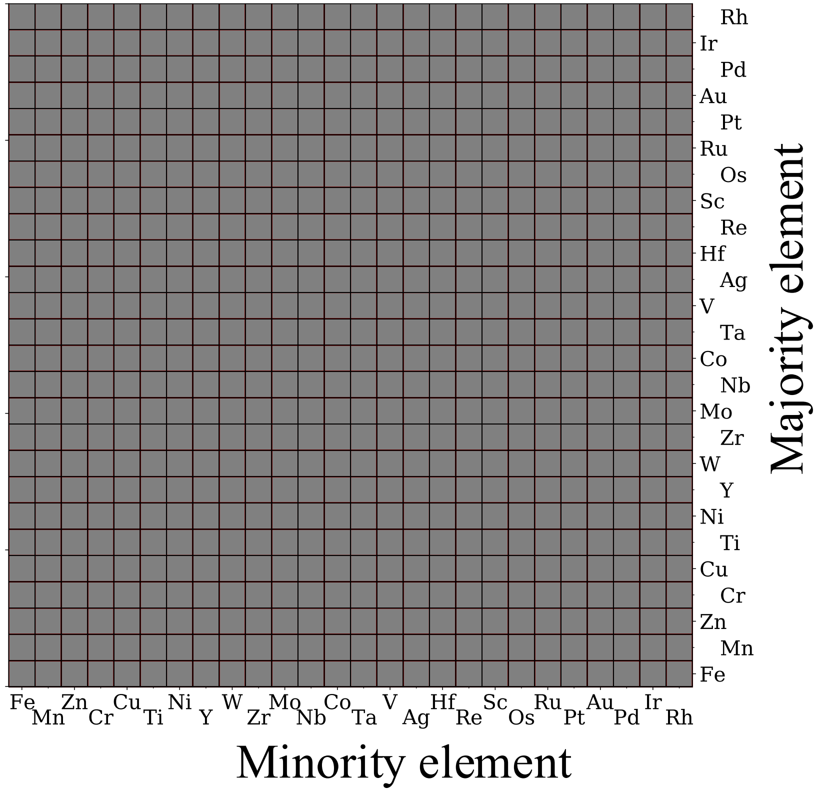
- GNN model: DimeNet++
- MAE = ~0.3 eV
- Target: Initial structure (adsorbed slab)→E_{ads}
- Training data: ~100k (metals only)

Under revision

ML assisted screening

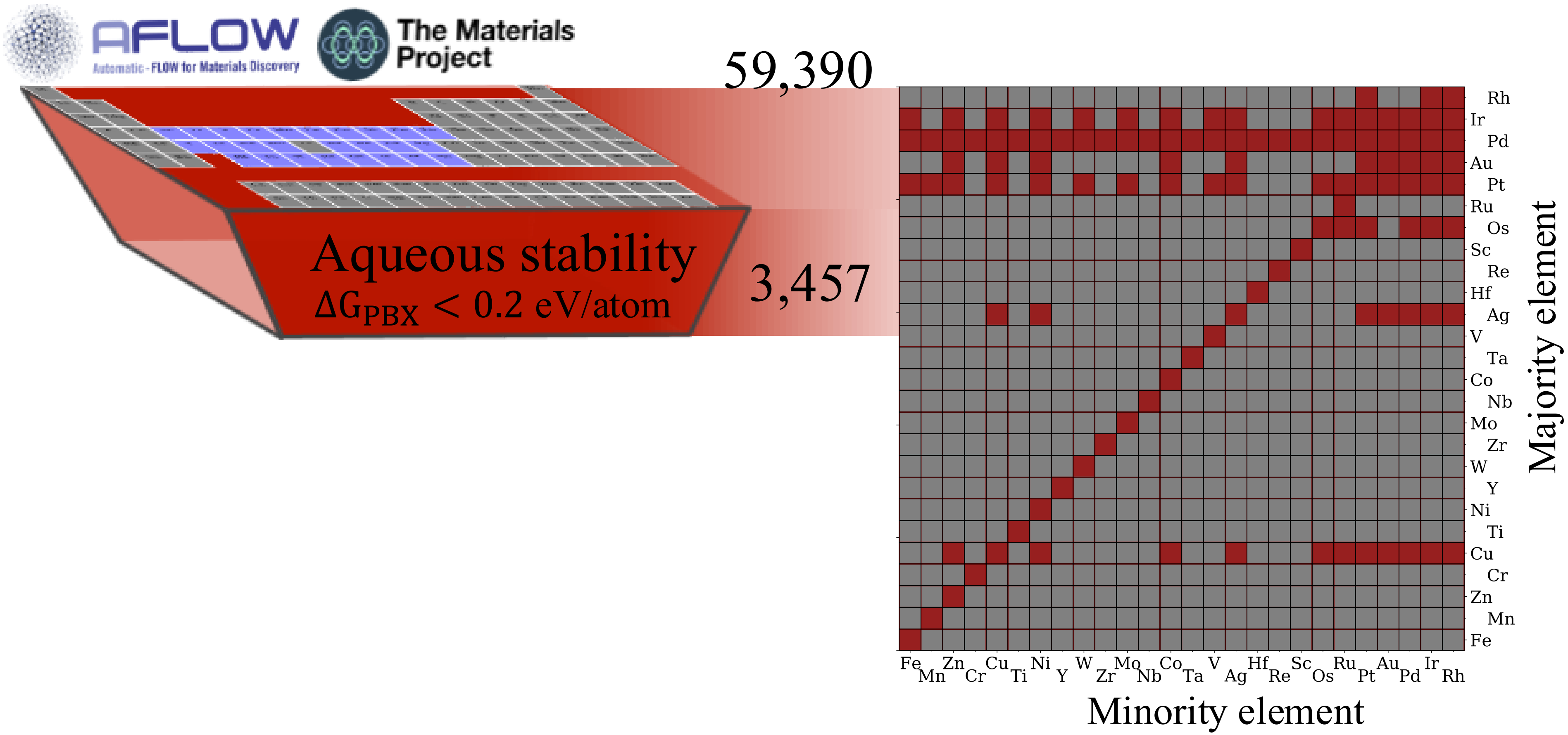


59,390



Under revision

ML assisted screening



Under revision

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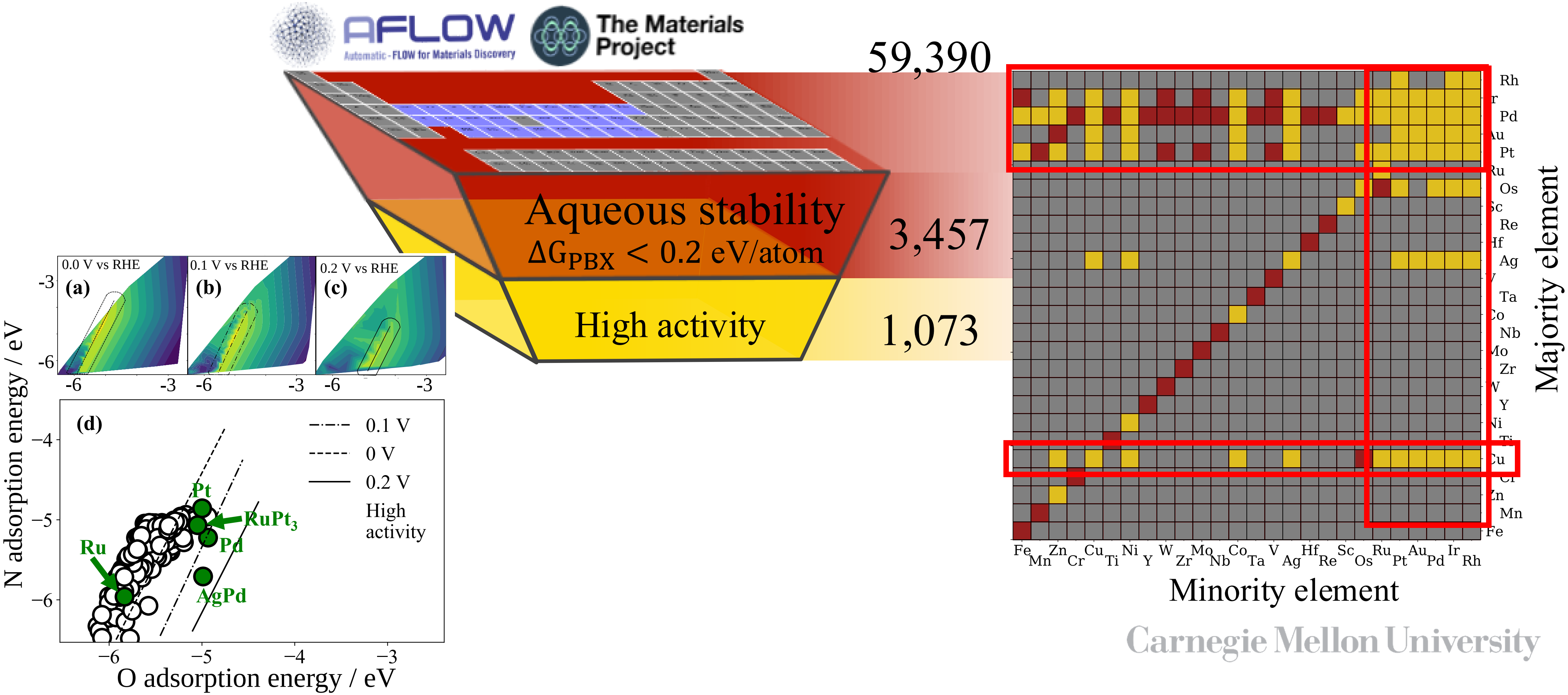
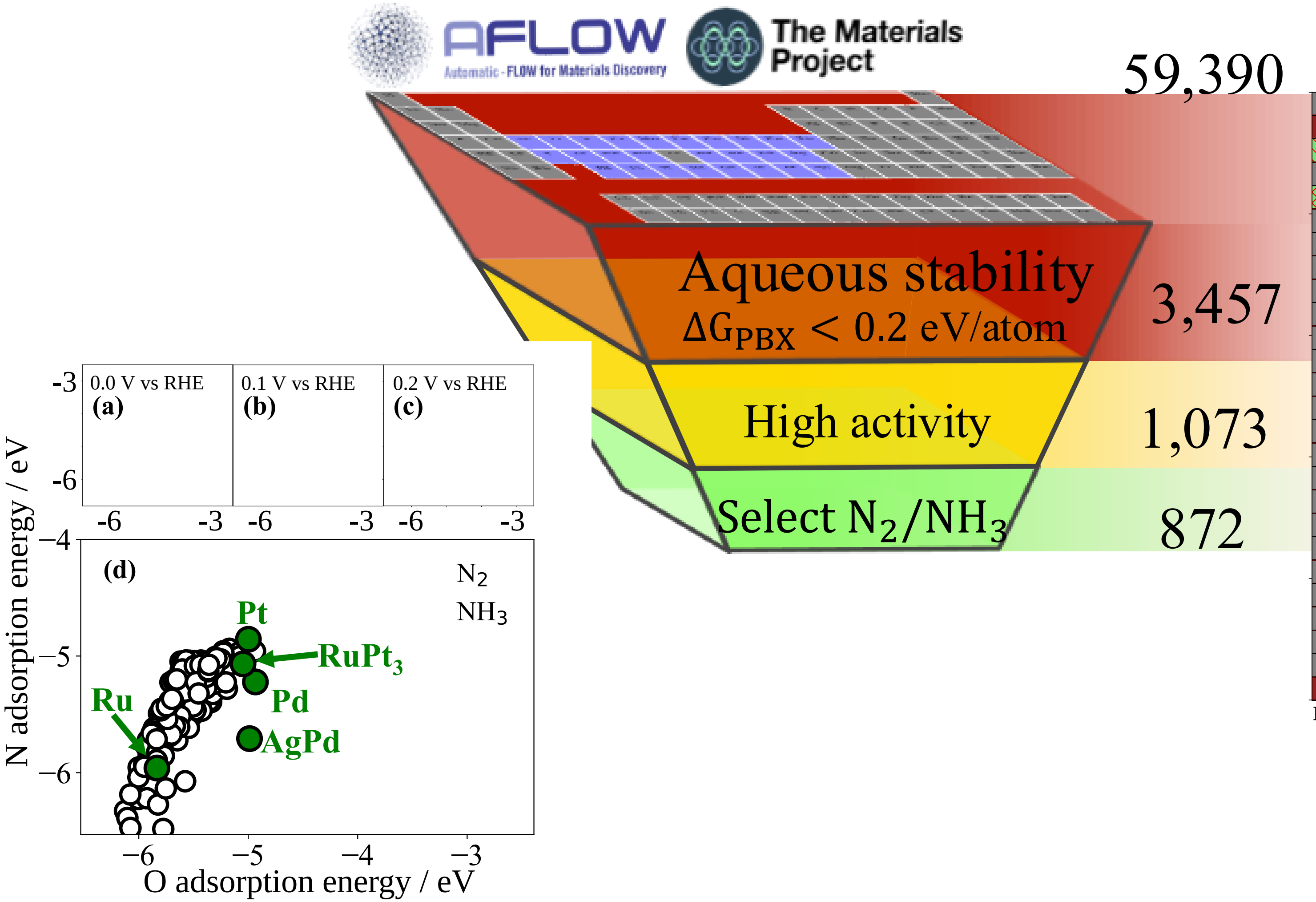
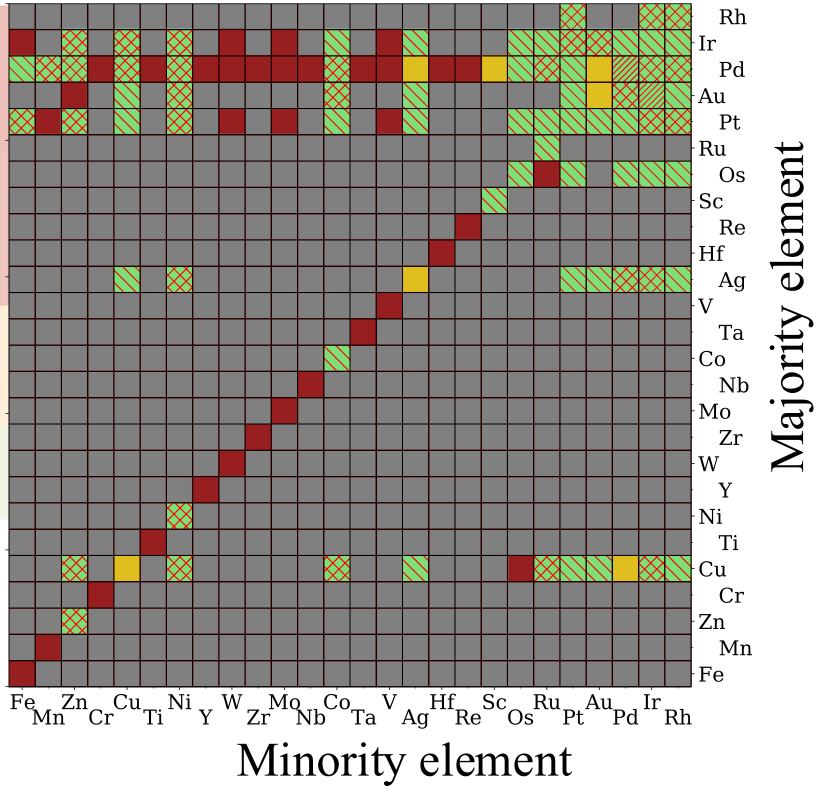


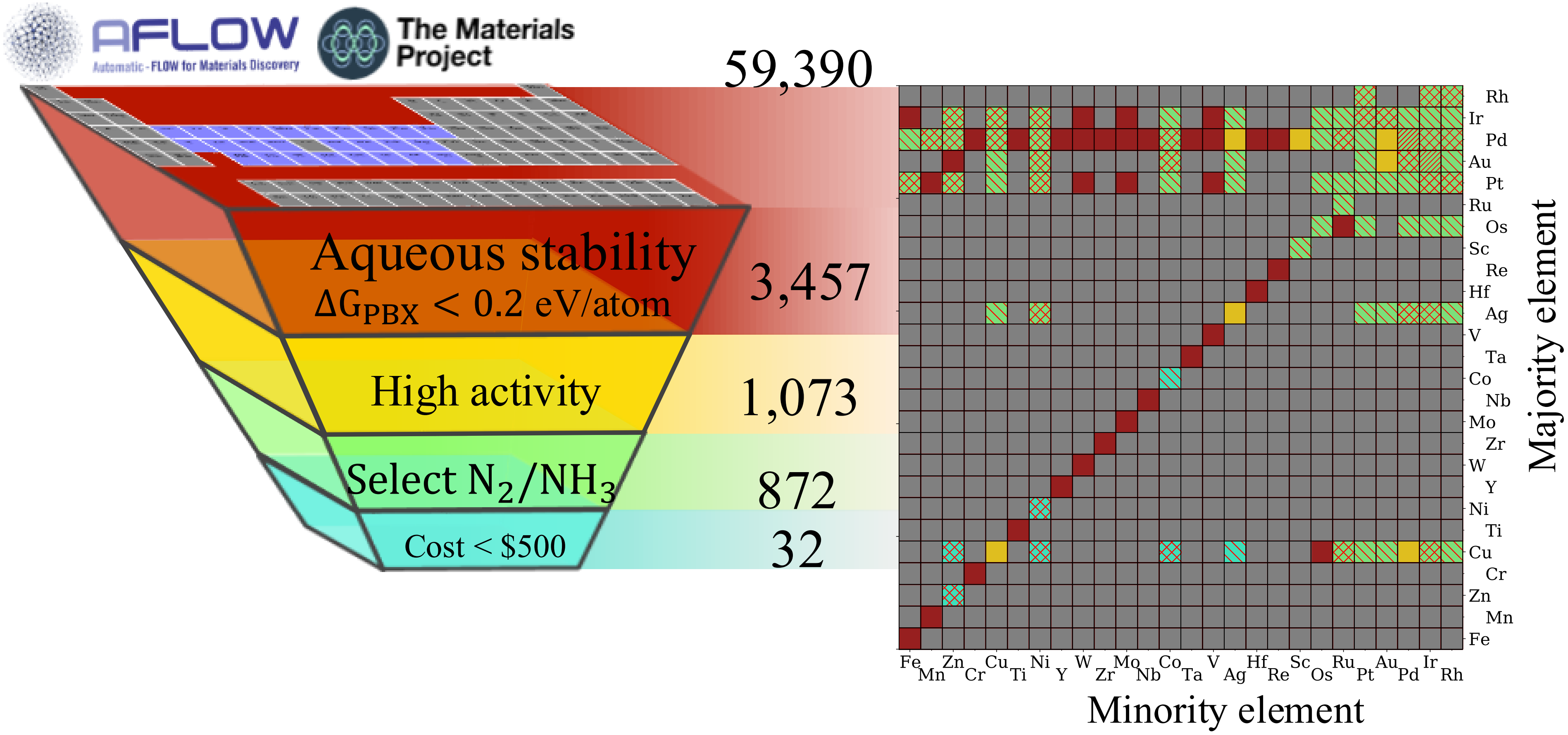
Under revision

ML assisted screening



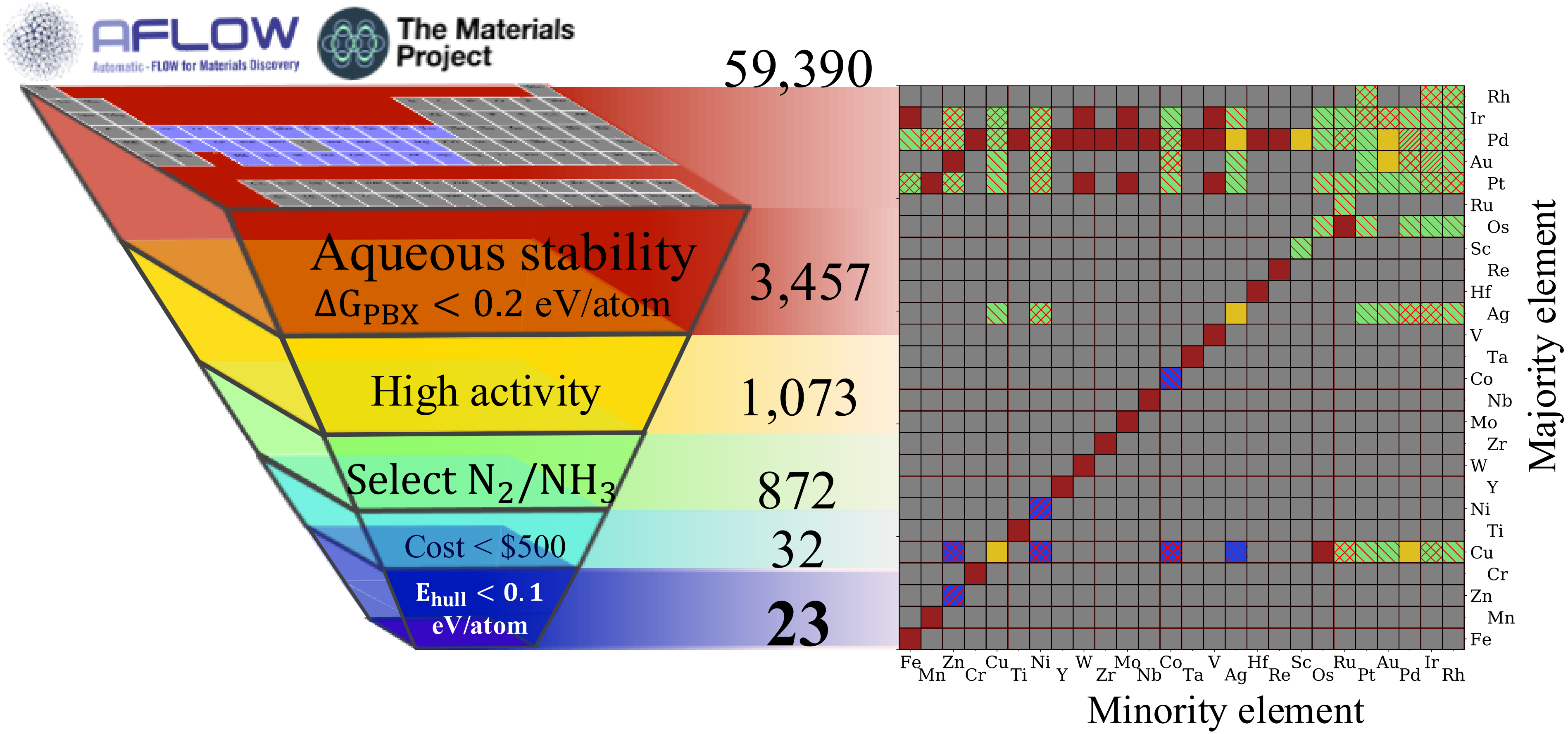


ML assisted screening



Under revision

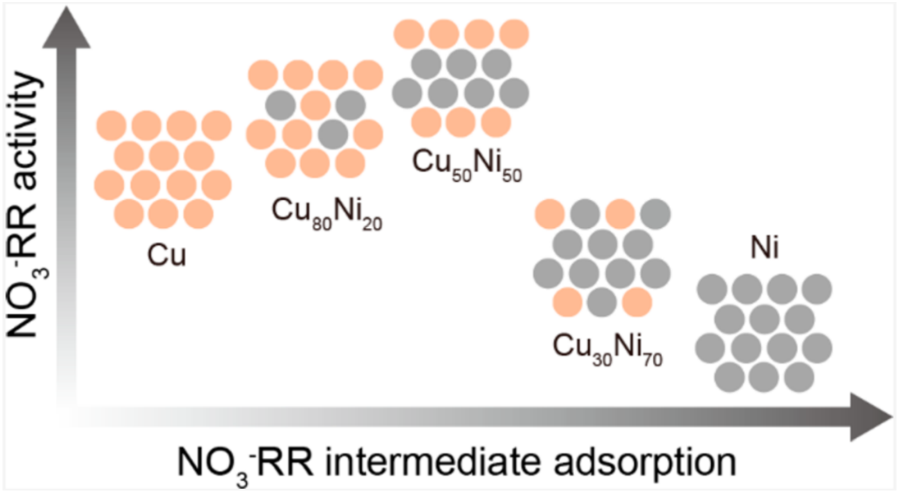
ML assisted screening



Under revision

Final candidates

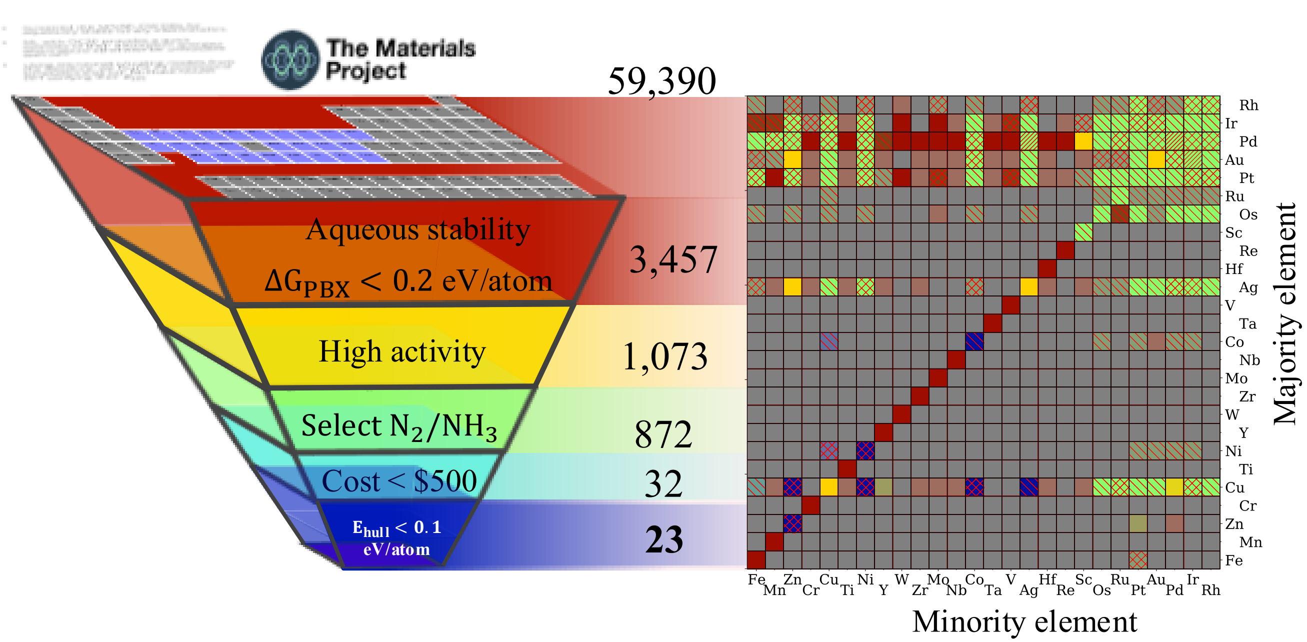
Formula	Space group	Cost ($\$$/kg mol^{-1})	ΔG_{aq}(0.0 V) (eV/atom)	ΔG_{aq}(0.1 V) (eV/atom)	Active at 0.1 V	N_2	NH_3
Zn	$P6_3/mmc$	2.95	0.00	0.00	No	✓	✓
$ZnCu_8$	$I4/mmm$	8.78	0.18	0.29	No	✓	✓
Cu_5Ni	Cm	10.93	0.15	0.27	Yes	✓	✓
Cu_5Ni	$Amm2$	10.93	0.15	0.27	Yes	✓	✓
Cu_4Ni	$I4/m$	11.22	0.17	0.29	Yes	✓	✓
Cu_3Ni	$R\bar{3}m$	11.65	0.20	0.33	Yes	✓	✓
Cu_3Ni	$I4/mmm$	11.65	0.21	0.33	Yes	✓	✓
Cu_3Ni	$Cmmm$	11.65	0.21	0.33	Yes	✓	✓
$CoCu_7$	$Fm\bar{3}m$	14.52	0.19	0.30	No	✓	✓
Ni	$Fm\bar{3}m$	18.54	0.00	0.00	Yes	✓	✓
Co	$P6_3/mmc$	52.21	0.00	0.00	No	✓	
Cu_4Ag	$I4/m$	261.58	0.08	0.16	No	✓	✓
Cu_3Ag	$P4/mmm$	315.24	0.08	0.15	Yes	✓	✓
Cu_3Ag	$Pmmm$	315.24	0.09	0.16	No	✓	✓
Cu_3Ag	$Pmmn$	315.24	0.10	0.18	No	✓	✓
Cu_3Ag	$C2/m$	315.24	0.10	0.18	Yes	✓	✓
Cu_3Ag	$I4/mmm$	315.24	0.10	0.17	No	✓	✓
Cu_3Ag	$Pmmn$	315.24	0.10	0.17	No	✓	✓
Cu_2Ag	$P6_3/mmc$	397.92	0.07	0.14	No	✓	✓
Cu_2Ag	$C2/m$	397.92	0.09	0.15	No	✓	
Cu_2Ag	$P6_3/mmc$	397.92	0.07	0.14	No	✓	✓
Cu_2Ag	$C2/m$	397.92	0.10	0.17	No	✓	✓
Cu_5Ag_4	$I4/mmm$	496.75	0.10	0.15	No	✓	✓



Wang, Y., Xu, A., Wang, Z., Huang, L., Li, J., Li, F., Wicks, J., Luo, M., Nam, D. H., Tan, C. S., Ding, Y., Wu, J., Lum, Y., Dinh, C. T., Sinton, D., Zheng, G., & Sargent, E. H. (2020). Enhanced Nitrate-to-Ammonia Activity on Copper-Nickel Alloys via Tuning of Intermediate Adsorption. *Journal of the American Chemical Society*, *142*(12), 5702–5708. https://doi.org/10.1021/jacs.9b13347

Recap

- 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 23 economical bimetallics that can facilitate NO_3RR DFT verifying their E_{ads}



Outlook and Future works

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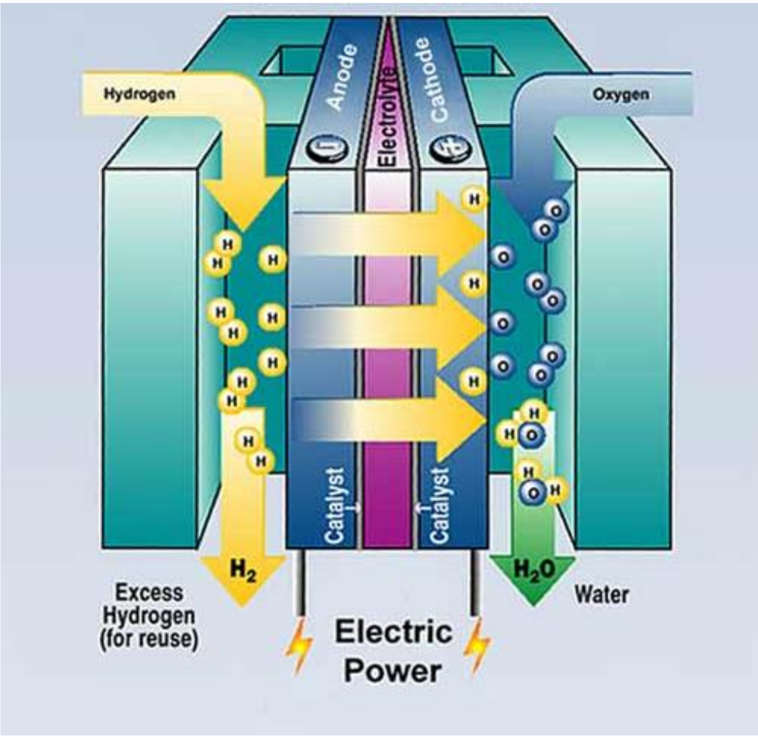
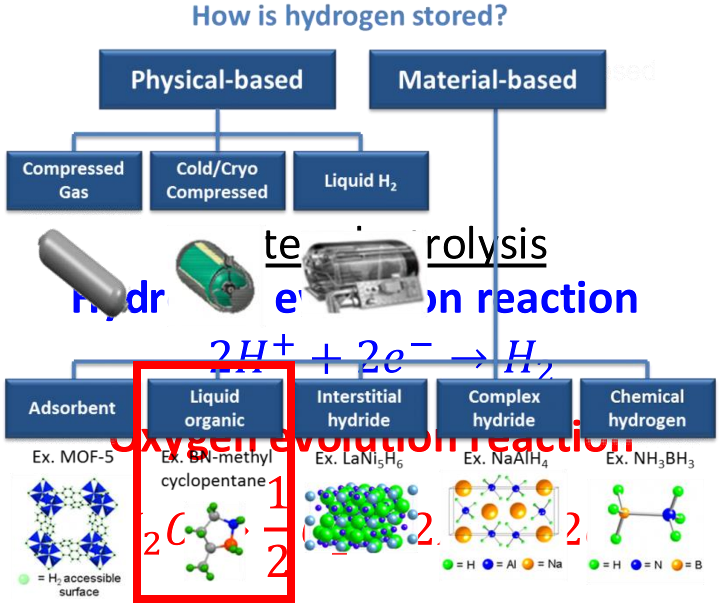
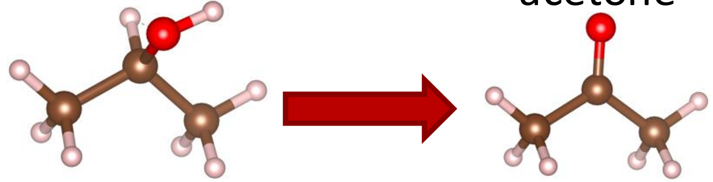
Hydrogen fuel source





Isopropyl alcohol



acetone

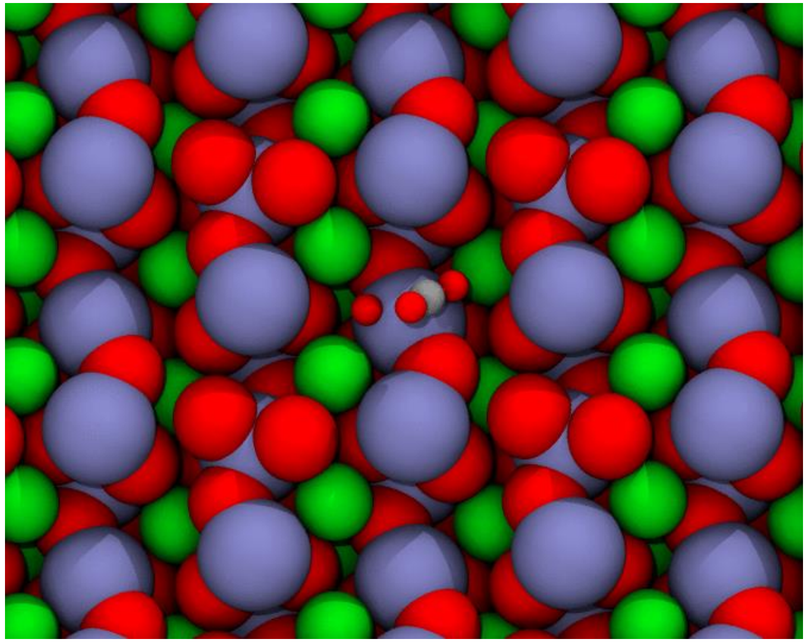
González, D., Heras-Domingo, J., Sodupe, M., Rodríguez-Santiago, L., & Solans-Monfort, X. (2021). Importance of the oxyl character on the IrO2 surface dependent catalytic activity for the oxygen evolution reaction. *Journal of Catalysis*, *396*, 192–201.

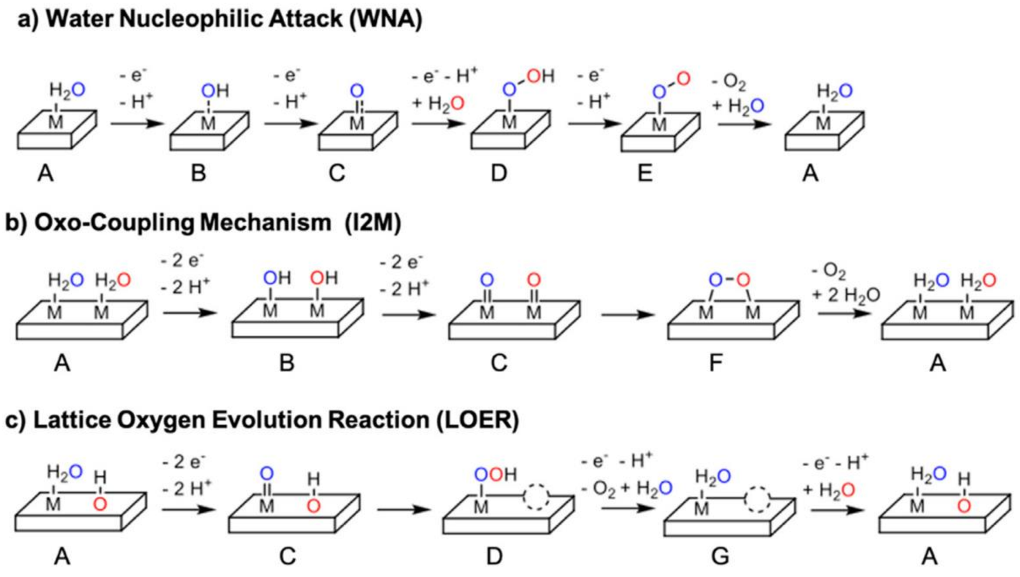
Meta AI

RESEARCH

Accelerating renewable energy with new data set for green hydrogen fuel

April 18, 2022





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Acknowledgements

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Funding

NAWI
National Alliance for Water Innovation

Meta AI

NSF Designing Materials to Revolutionize and Engineer our Future (DMREF)

Computing resources

NREL
Transforming ENERGY

NERSC

Zachary Ulissi

Nitrate reduction reaction

Duo Wang

Jain Anubhav

Ryan Kingsbury

Aini Palizhati

Kristin A. Persson

Isopropyl alcohol dehydrogenation

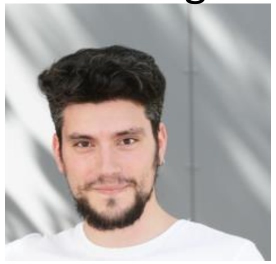
Hilda Mera

Open Catalyst 2022 (OER)

Adeesh Kolluru

Muhammed Shuaibi

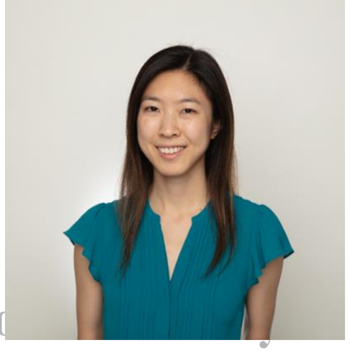
Javier Heras Domingo



Larry Zitnick

Brandon Wood

Siddharth Goyal



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Questions