ACS FALL 2023
HARNESSING THE POWER OF DATA

ACS Chemistry for Life
AMERICAN CHEMICAL SOCIETY
MEETINGS & EVENTS

Machine learning assisted screening of oxide catalysts for OER

Richard Tran, Liqiang Huang, Yuan Zi, Shengguang Wang, Benjamin Comer, Wu, Xuqing, Lars Grabow, Ligang Lu, Jiefu Chen

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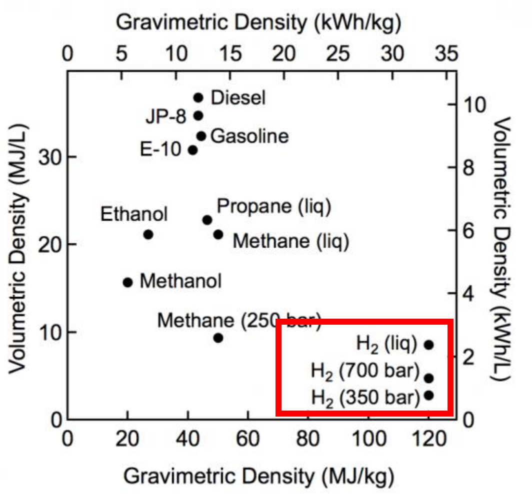
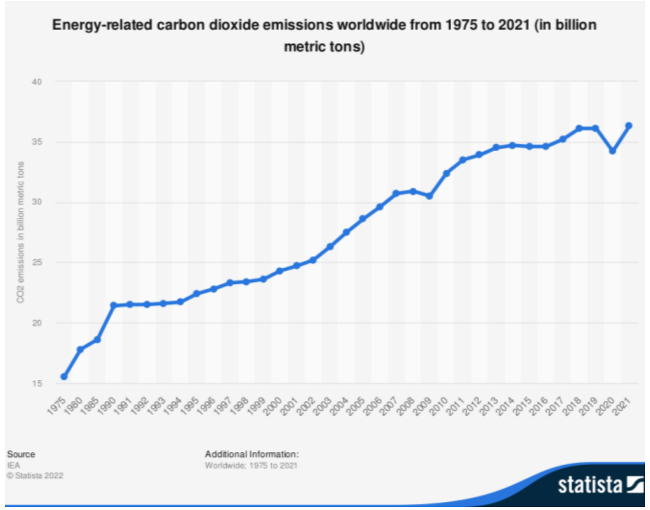
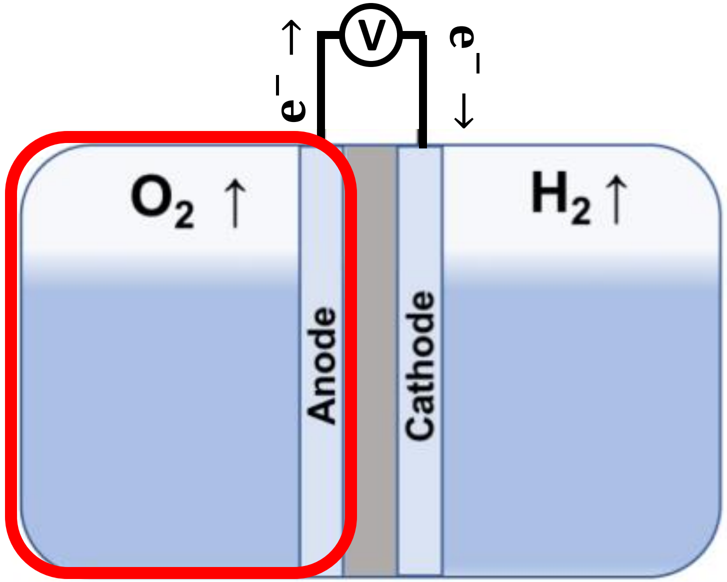
Shell

UNIVERSITY OF HOUSTON

HARNESSING THE **POWER** OF WATER







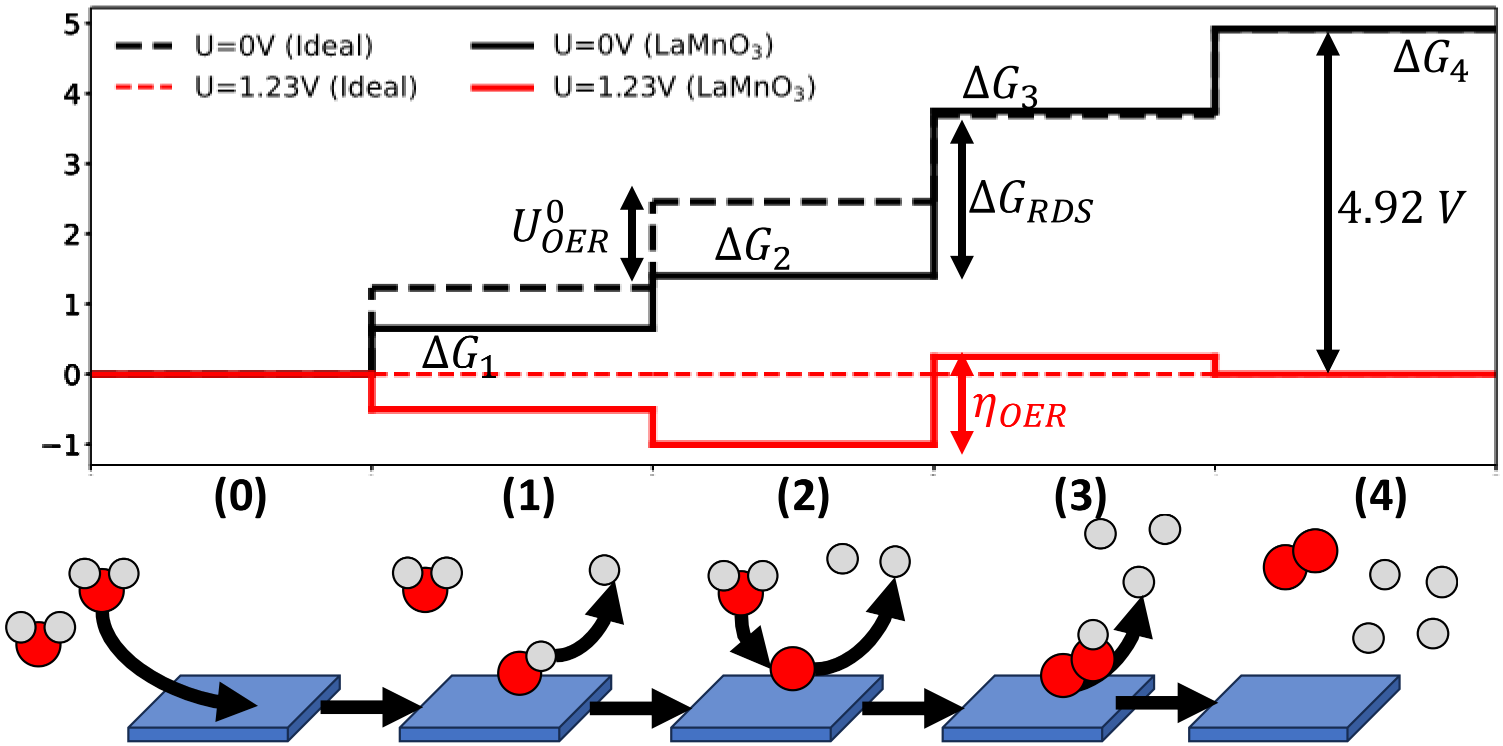
OER: $2H_2O \rightarrow O_2 + 4H^+ + 4e^-$

HER: $4H^+ + 4e^- \rightarrow 2H_2$

$2H_2O \rightarrow O_2 + \mathbf{2H_2}$

Oxygen evolution reaction and overpotential

Man, I. C., Su, H.-Y., Calle-Vallejo, F., Hansen, H. A., Martínez, J. I., Inoglu, N. G., Kitchin, J., Jaramillo, T. F., Nørskov, J. K., & Rossmeisl, J. (2011). *ChemCatChem*, *3*(7), 1159–1165. https://doi.org/10.1002/cctc.201000397



$U^0_{OER} = 1.23\ V\ vs\ SHE,$
$4U^0_{OER} = 4.92\ V$

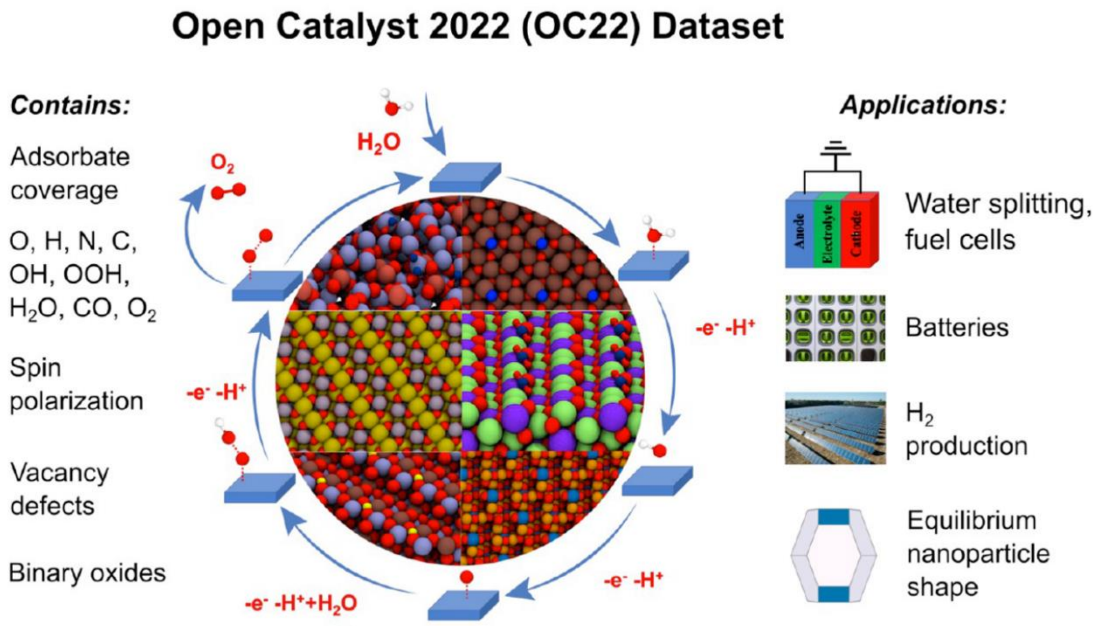
$$\Delta G_1 = E_{HO^*} + \frac{1}{2}\mu_{H_2} - \mu_{H_2O_{(l)}} - E^* + \Delta ZPE - T\Delta S^o$$
$$\Delta G_2 = E_{O^*} + \mu_{H_2} - \mu_{H_2O_{(l)}} - E^* + \Delta ZPE - T\Delta S^o$$
$$\Delta G_3 = E_{OOH^*} + \frac{3}{2}\mu_{H_2} - 2\mu_{H_2O_{(l)}} - E^* + \Delta ZPE - T\Delta S^o$$
$$\Delta G_4 = 2\mu_{H_2} - 2\mu_{H_2O_{(l)}}$$

$$\mu_{H_2} = \mu_{H^+} + \mu_{e^-}$$
$$\mu_{H^+} = \frac{1}{2}G_{H_{2(g)}} - k_B T pH ln(10)$$
$$\mu_{e^-} = -eU$$
$$\mu_{H_2O} = G_{H_2O_{(l)}}$$

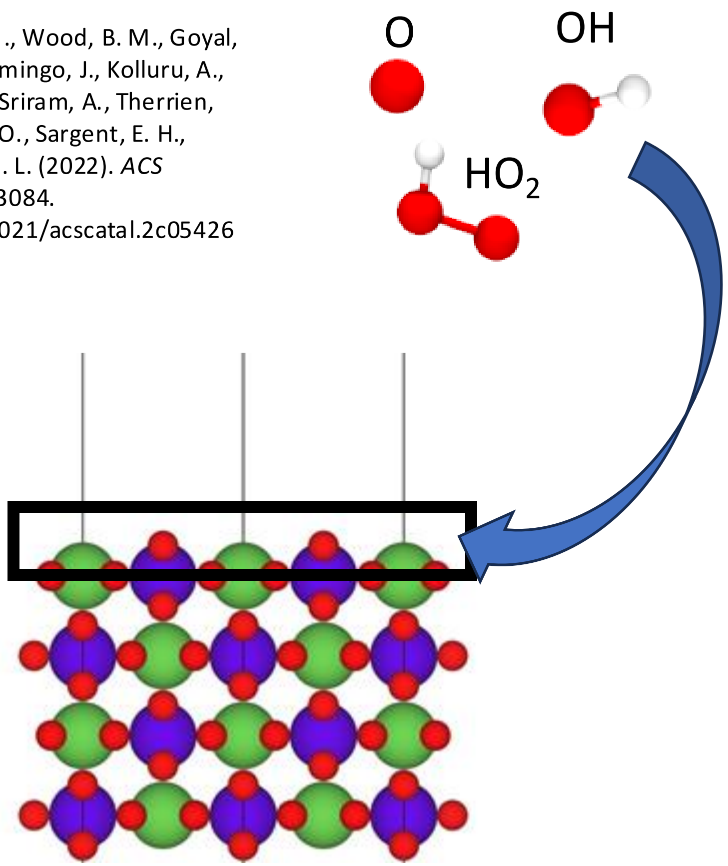
$$\Delta G_{RDS} = \max(\Delta G_1, \Delta G_2 - \Delta G_1, \Delta G_3 - \Delta G_2, \Delta G_4 - \Delta G_3,)$$

Overpotential: $\eta_{OER} = \Delta G_{RDS} - U^0_{OER}$

The Open catalyst project 2022



Tran, R., Lan, J., Shuaibi, M., Wood, B. M., Goyal, S., Das, A., Heras-Domingo, J., Kolluru, A., Rizvi, A., Shoghi, N., Sriram, A., Therrien, F., Abed, J., Voznyy, O., Sargent, E. H., Ulissi, Z., & Zitnick, C. L. (2022). *ACS Catalysis*, *13*, 3066–3084. https://doi.org/10.1021/acscatal.2c05426



Meta AI

Fundamental AI Research (FAIR)

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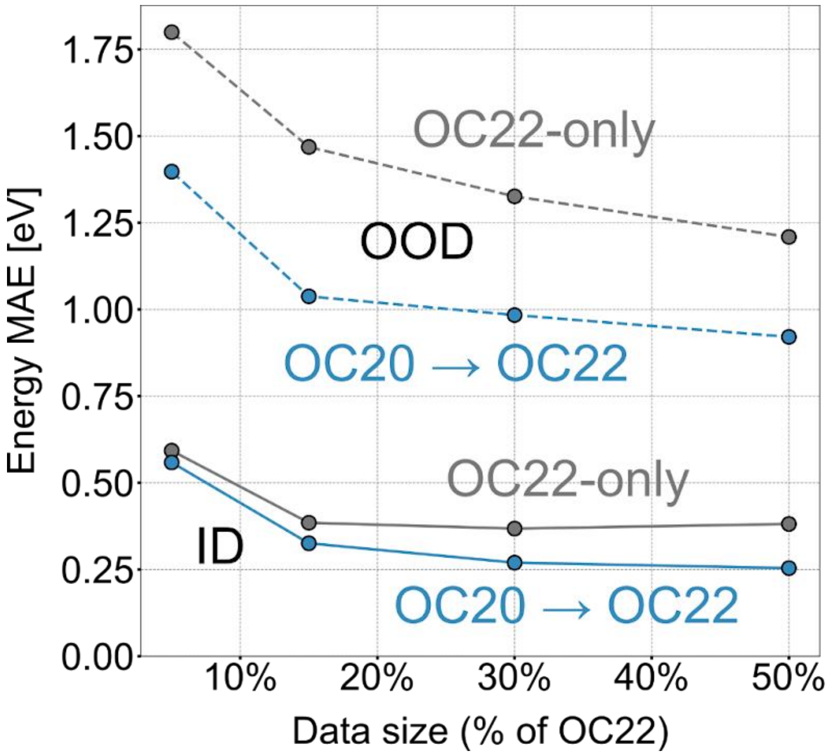
ML metric and performance

Tran, R., Lan, J., Shuaibi, M., Wood, B. M., Goyal, S., Das, A., Heras-Domingo, J., Kolluru, A., Rizvi, A., Shoghi, N., Sriram, A., Therrien, F., Abed, J., Voznyy, O., Sargent, E. H., Ulissi, Z., & Zitnick, C. L. (2022). *ACS Catalysis*, *13*, 3066–3084. https://doi.org/10.1021/acscatal.2c05426



- Model: GemNet-OC
- OC20 models finetuned with OC22 data
- Structure to energy and forces (S2EF) model
- Energy MAE: 0.260 eV (in domain), 0.943 eV (out of domain)
- Average prediction time: ***5 seconds per prediction***

Database scope for comprehensive set

	OC22 framework	Database so far	Complete database
Bare slab	19,142	49,018	240,000
Adsorbed slab (O*, OH*, OOH*)	43,189	1,763,134	7,000,000
Facet scope	max(hkl)=3 (rand)	max(hkl)=1 (all)	
Total	62,331	1,812,152	7,240,000

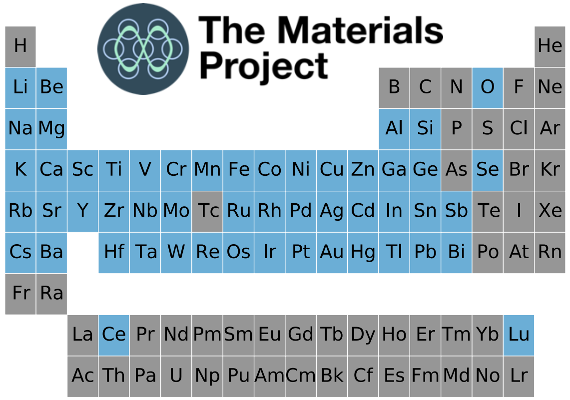
- **Purpose of OC22 dataset:** Ensemble of ***randomly*** chosen slabs/adsorbates/coverages/and sites used for training ML models
- **Purpose of current database:** Provided ***comprehensive*** ensemble of all slabs and sites for single molecule adsorption for analysis and screening

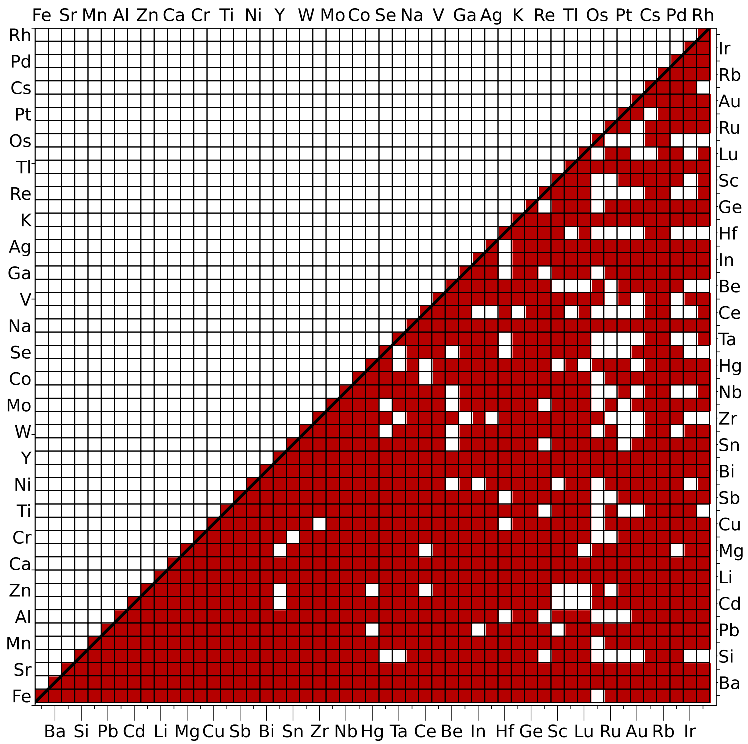
Application 1:
High-throughput screening

OC22 dataset

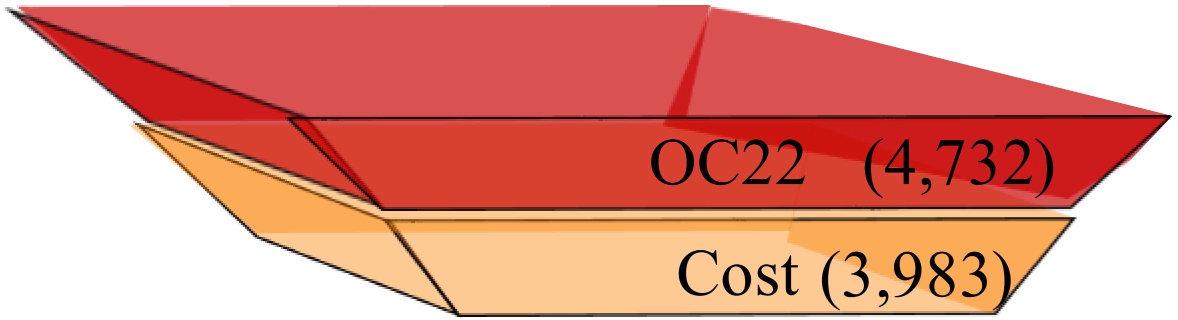
- Top 5 lowest E_{hull}
- Max # of atoms in bulk: 150
- 1720 bulks with U-values

- Unary bulks: 318
- Binary bulks: 4,414
- Total bulks: 4,732

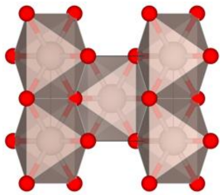




Material cost



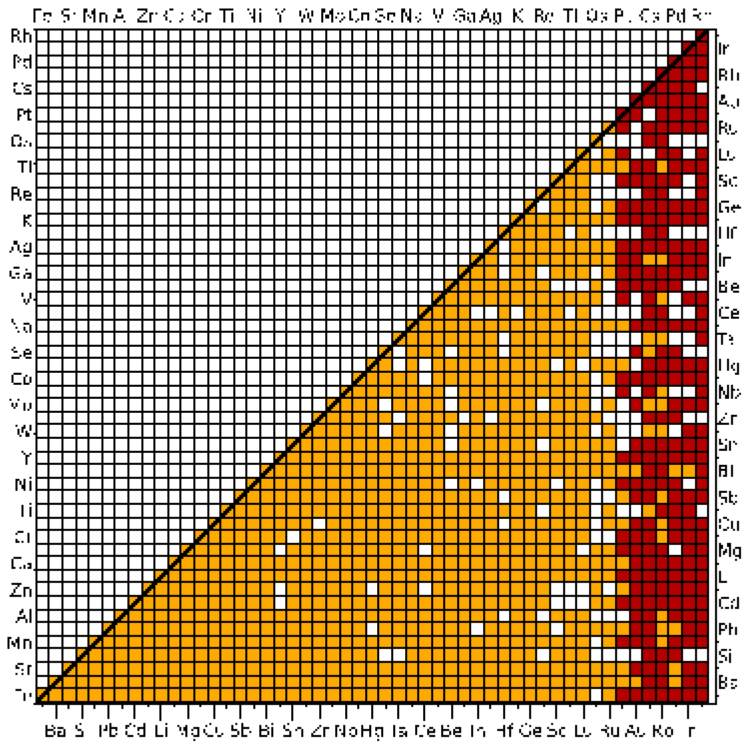
Cost < $18,315/kg



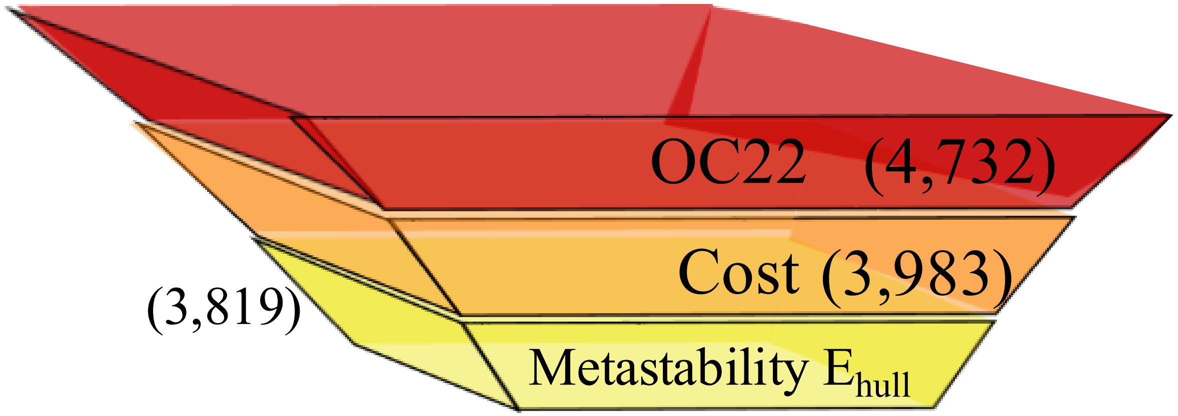
RuO_2

$18,315/kg

Ru: $24,113/kg



Metastability



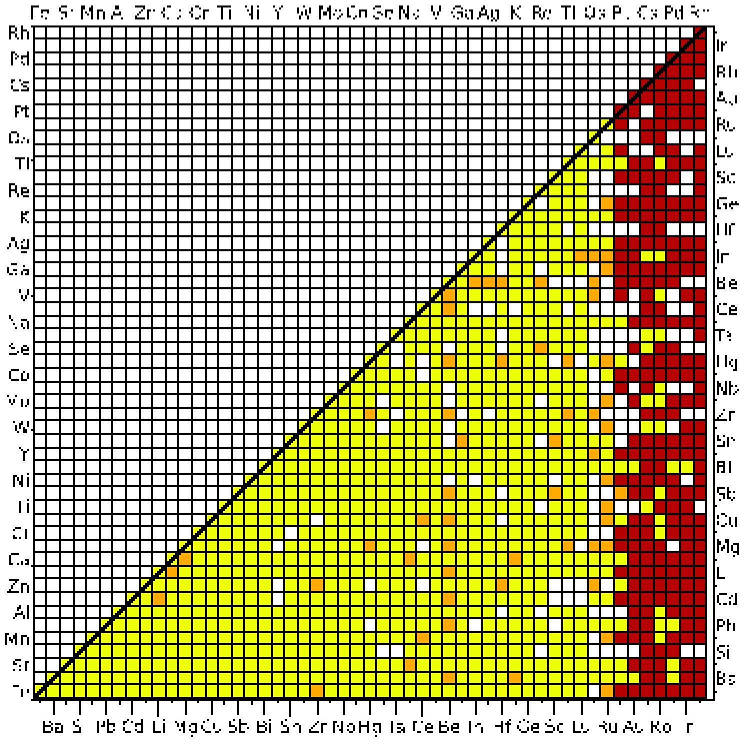
Aykol, M. *et al.* (2018). *Science Advances*, *4*(4), 1–8. doi.org/10.1126/sciadv.aaq0148

Metastability limits:

E_{hull} < 0.2 eV

E_{hull} from Materials Project:

Jain, A., Ong, S. P., Hautier, G., Chen, W., Richards, W. D., Dacek, S., Cholia, S., Gunter, D., Skinner, D., Ceder, G., & Persson, K. A. (2013). *APL Materials*, *1*(1), 011002 1. https://doi.org/10.1063/1.4812323

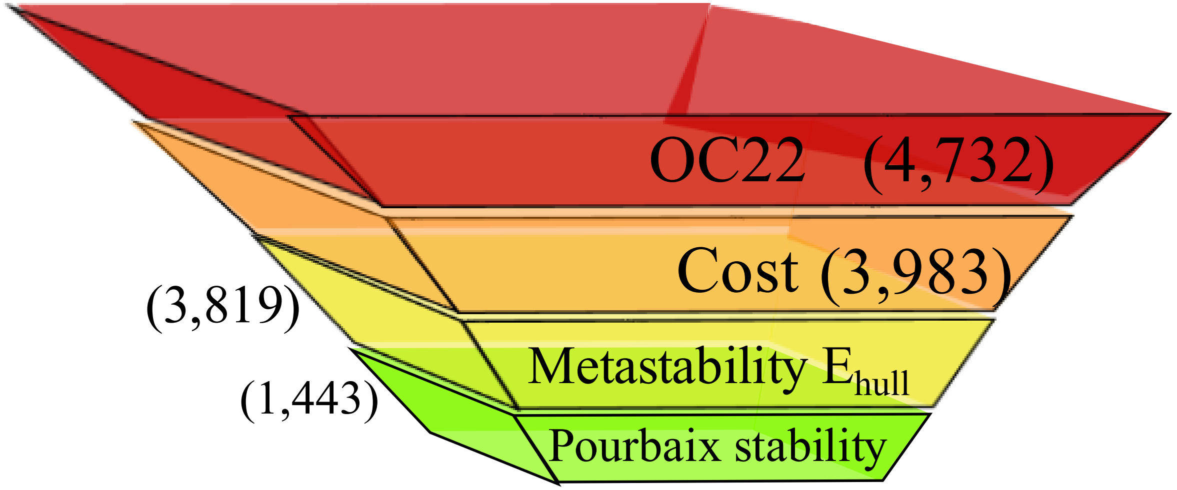


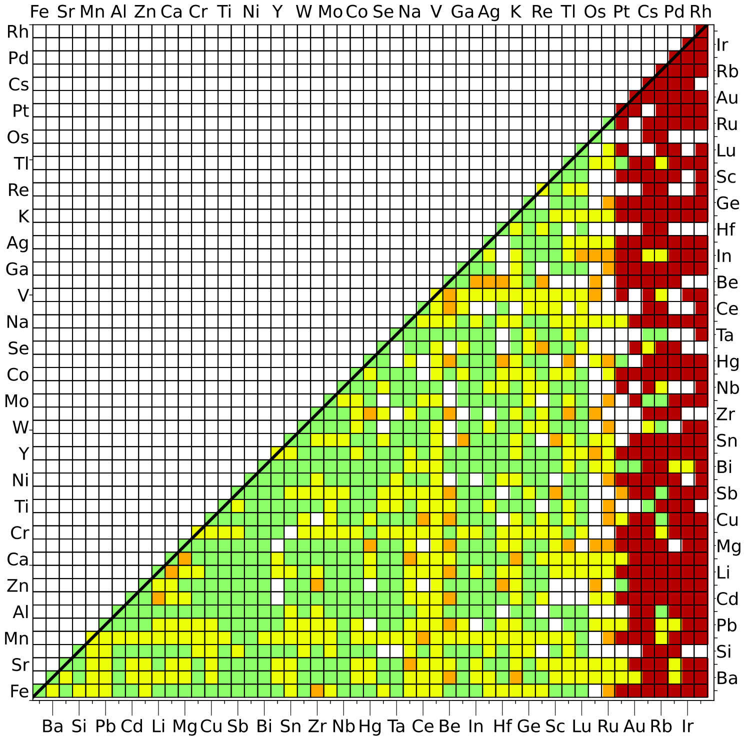
Pourbaix stability

Singh, A. K., Zhou, L., Shinde, A., Suram, S. K., Montoya, J. H., Winston, D., Gregoire, J. M., & Persson, K. A. (2017). *Chemistry of Materials*, *29*(23), 10159–10167. https://doi.org/10.1021/acs.chemmater.7b03980

surface passivation $\Delta G_{PBX} < 0.5\ eV$

metastability $\Delta G_{PBX} < 0.2\ eV$



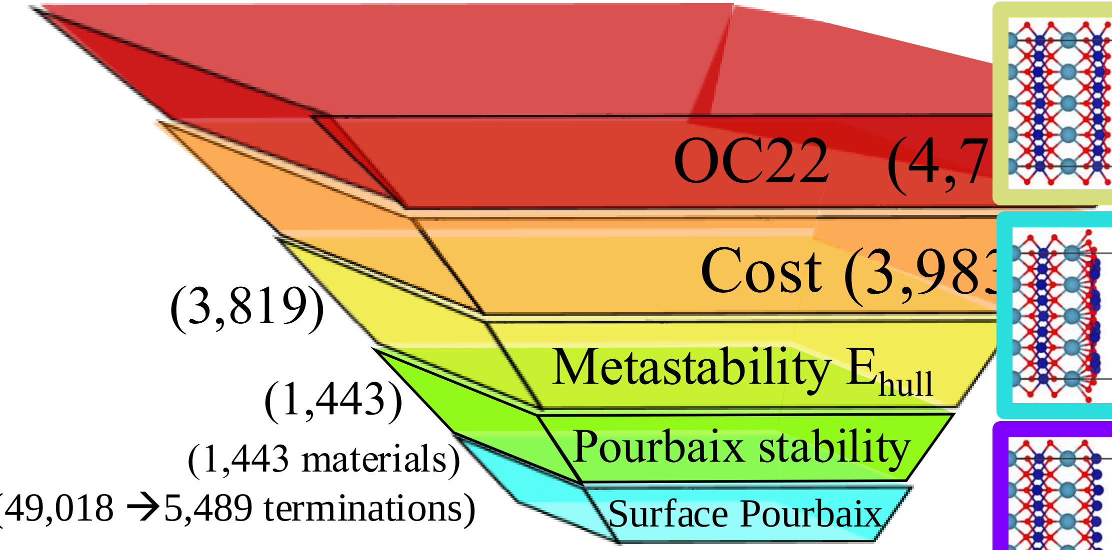


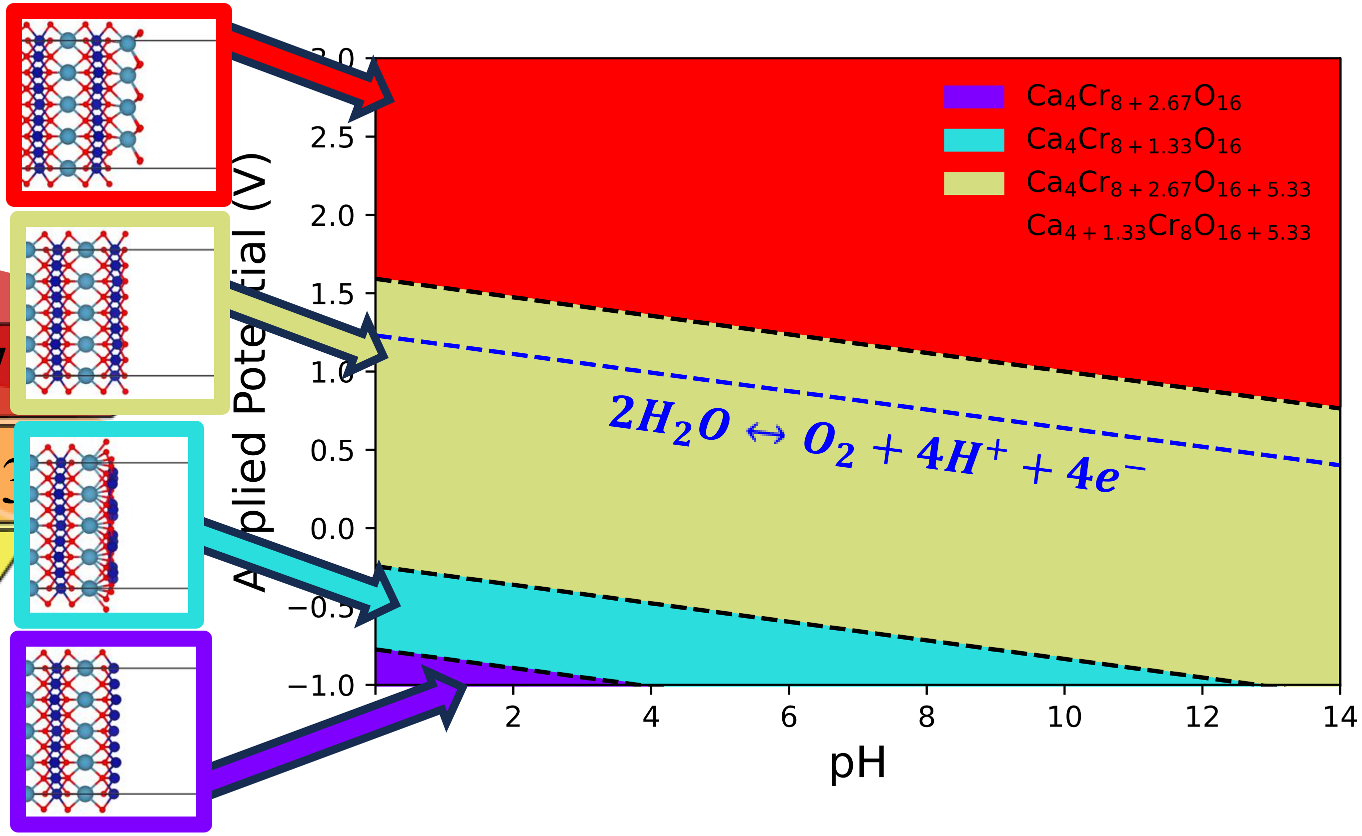
$$\Delta G_{PBX}\begin{pmatrix} \mathbf{pH = 0,\ \ U = 1.23\ V, or} \\ \mathbf{pH = 14, U = 0.404\ V} \end{pmatrix} < \mathbf{0.5\ eV}$$

$\mathbf{\Delta G_{PBX}}$ from Materials Project:

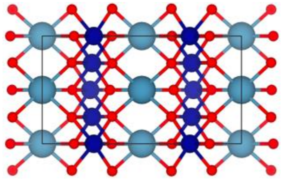
Jain, A., Ong, S. P., Hautier, G., Chen, W., Richards, W. D., Dacek, S., Cholia, S., Gunter, D., Skinner, D., Ceder, G., & Persson, K. A. (2013). *APL Materials*, *1*(1), 011002 1. https://doi.org/10.1063/1.4812323

Surface selection





Bulk $CaCr_2O_4$

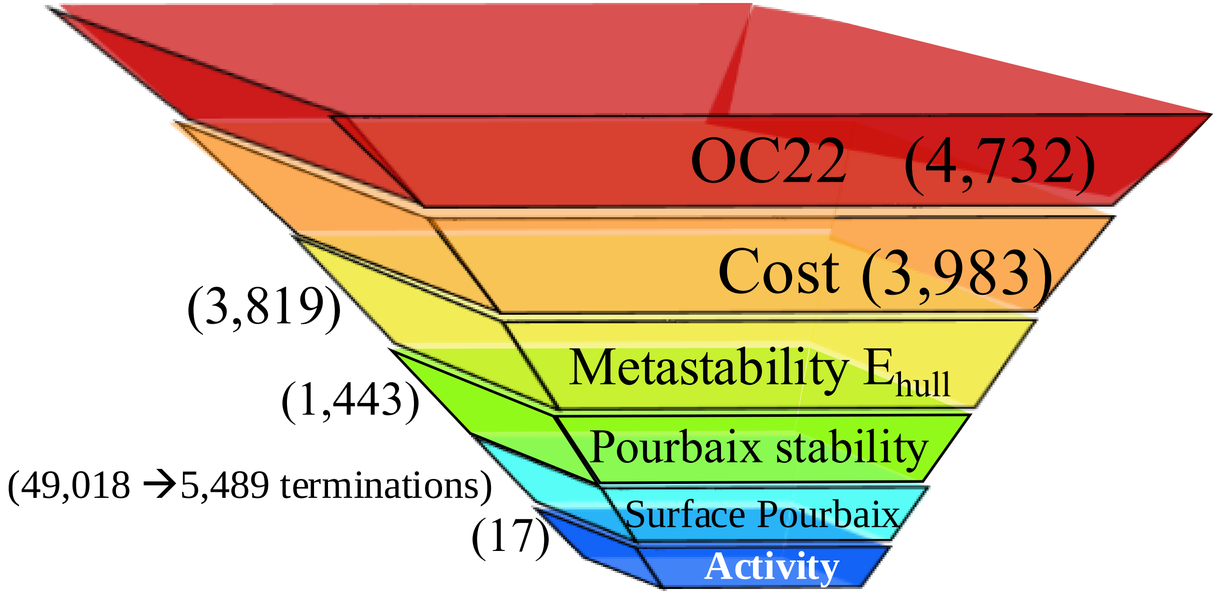


Approximately 36 bare slabs per material w/ max Miller index of 1

$$\gamma = E_{slab} - \sum_i n_i \mu_i = \mathrm{E_{slab}} - \mathrm{n_{Ca}}\mu_{Ca} - n_{Cr}\mu_{Cr} - n_O\mu_O$$

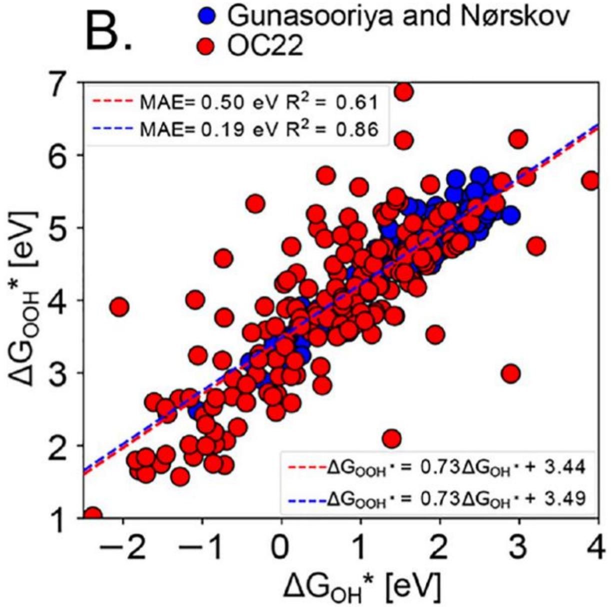
$$\mu_O = \mu_{H_2O} - \mu_{H_2} = E^{DFT}_{H_2O} - E^{DFT}_{H_2} - 2(-U - pH\, k_B T ln10)$$

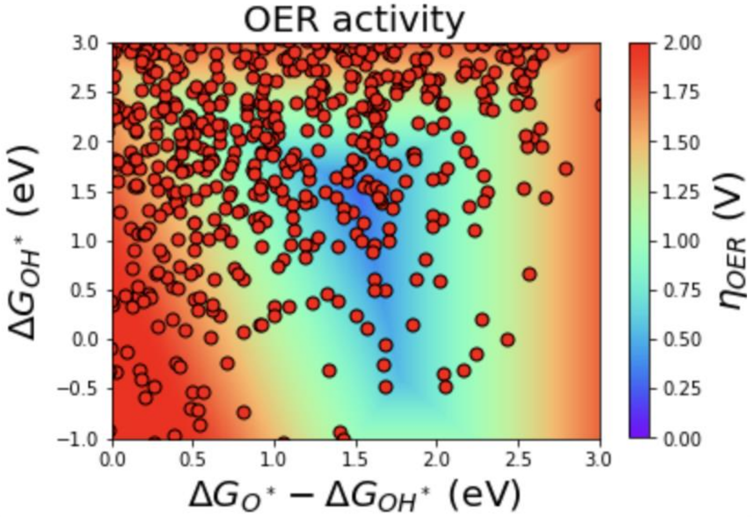
Overpotential / activity



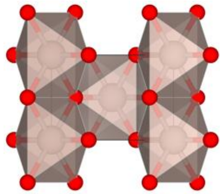
(value of $\eta_{OER}^{RuO_2}$ from OC22 predictions)

w/ at least 3 facets exhibiting
$\boldsymbol{\eta_{OER} \leq \eta_{OER}^{RuO_2} = 0.5 \pm 0.25}$ **eV**





$\eta_{OER}^{practical} \sim 0.4\ V\ to\ 0.3\ V$

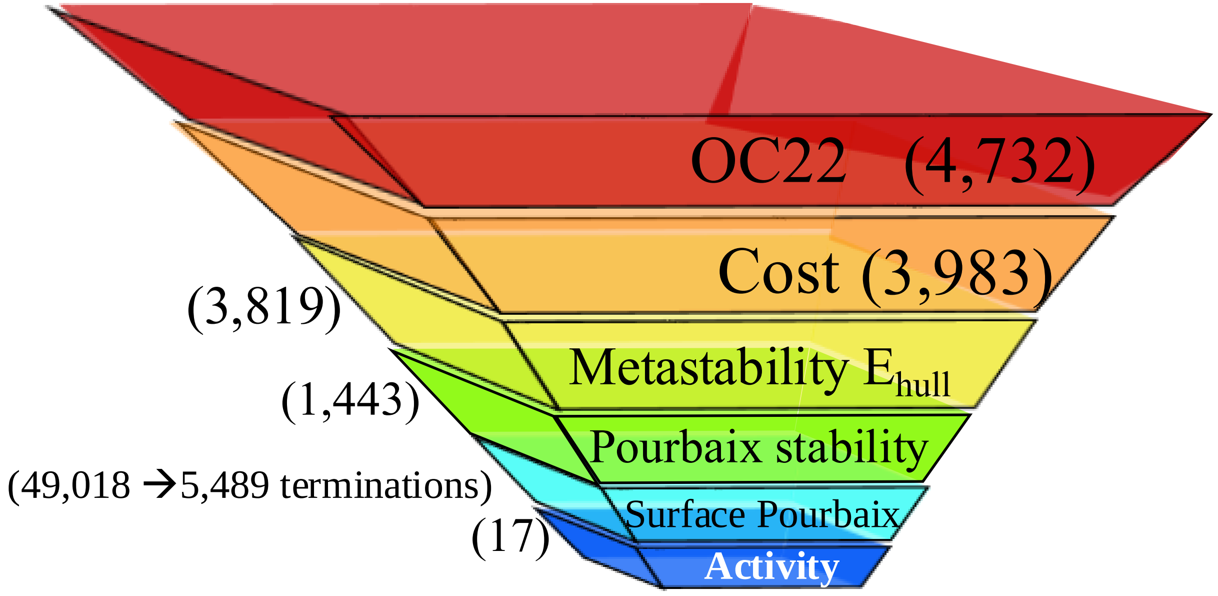


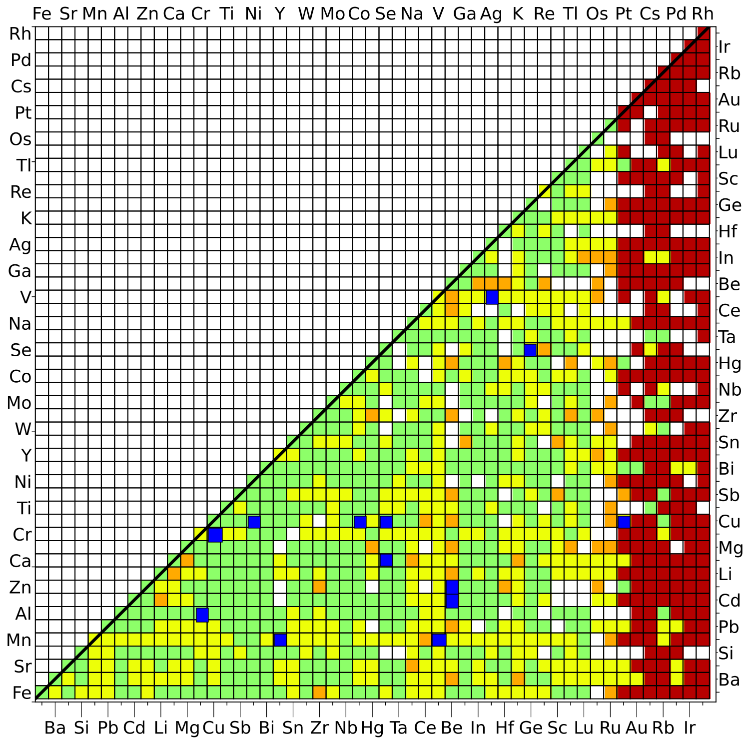
RuO_2

$18,315/kg
$\eta_{exp} = 0.25 - 0.5\ V$
$\eta_{DFT} = 0.50\ V$

$\boldsymbol{\eta_{OER}}$ predicted with OC22

Overpotential / activity



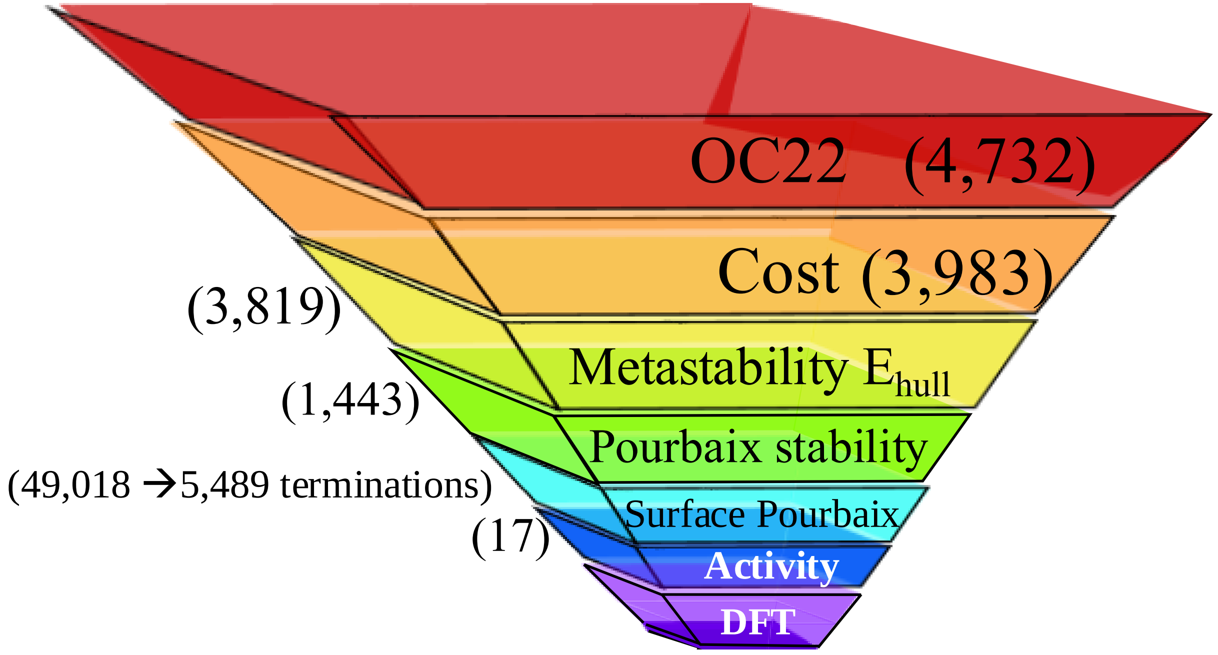


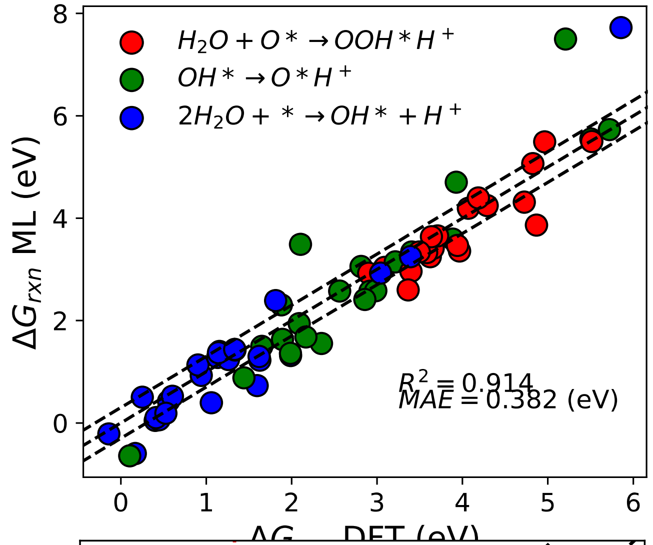
(value of $\eta_{OER}^{RuO_2}$ from OC22 predictions)

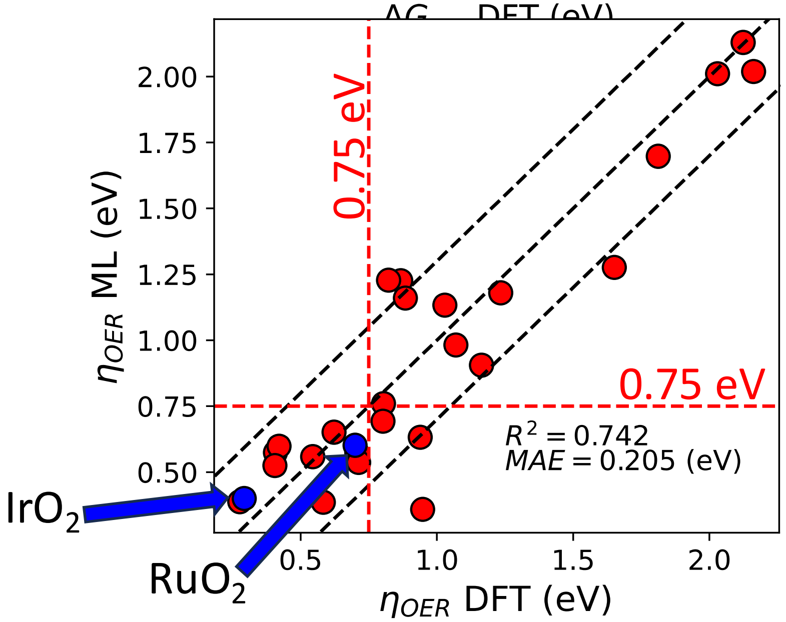
w/ at least 3 facets exhibiting

$$\eta_{OER} \leq \eta_{OER}^{RuO_2} = 0.5 \pm 0.25 \text{ eV}$$

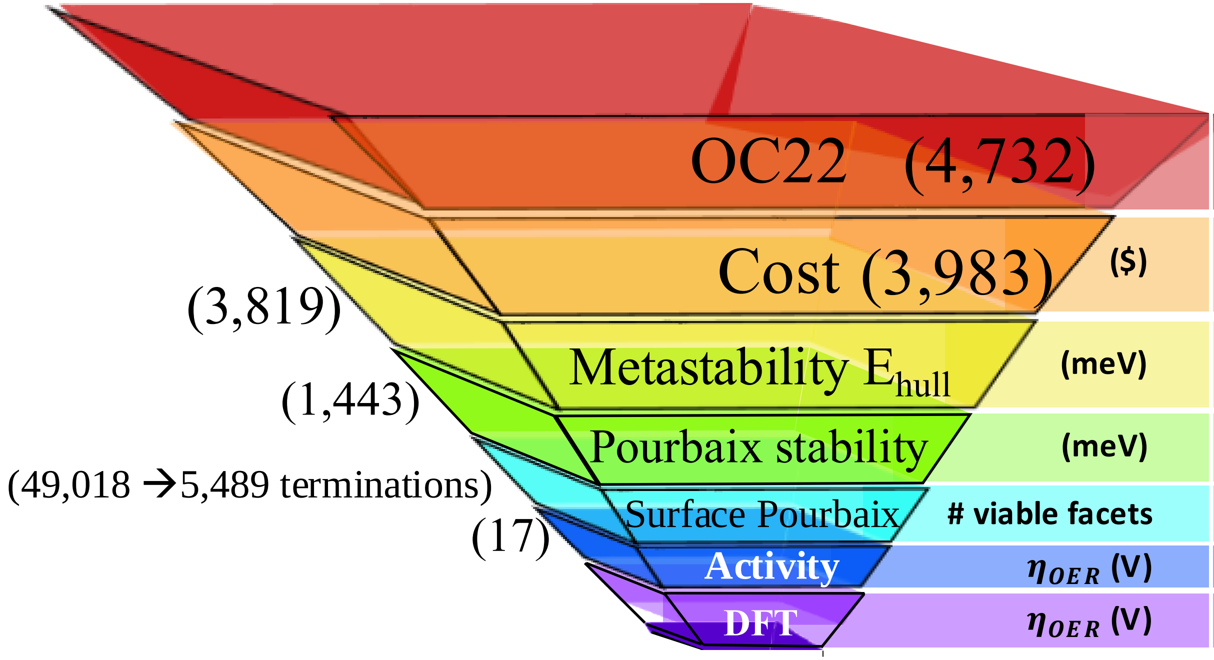
DFT validation



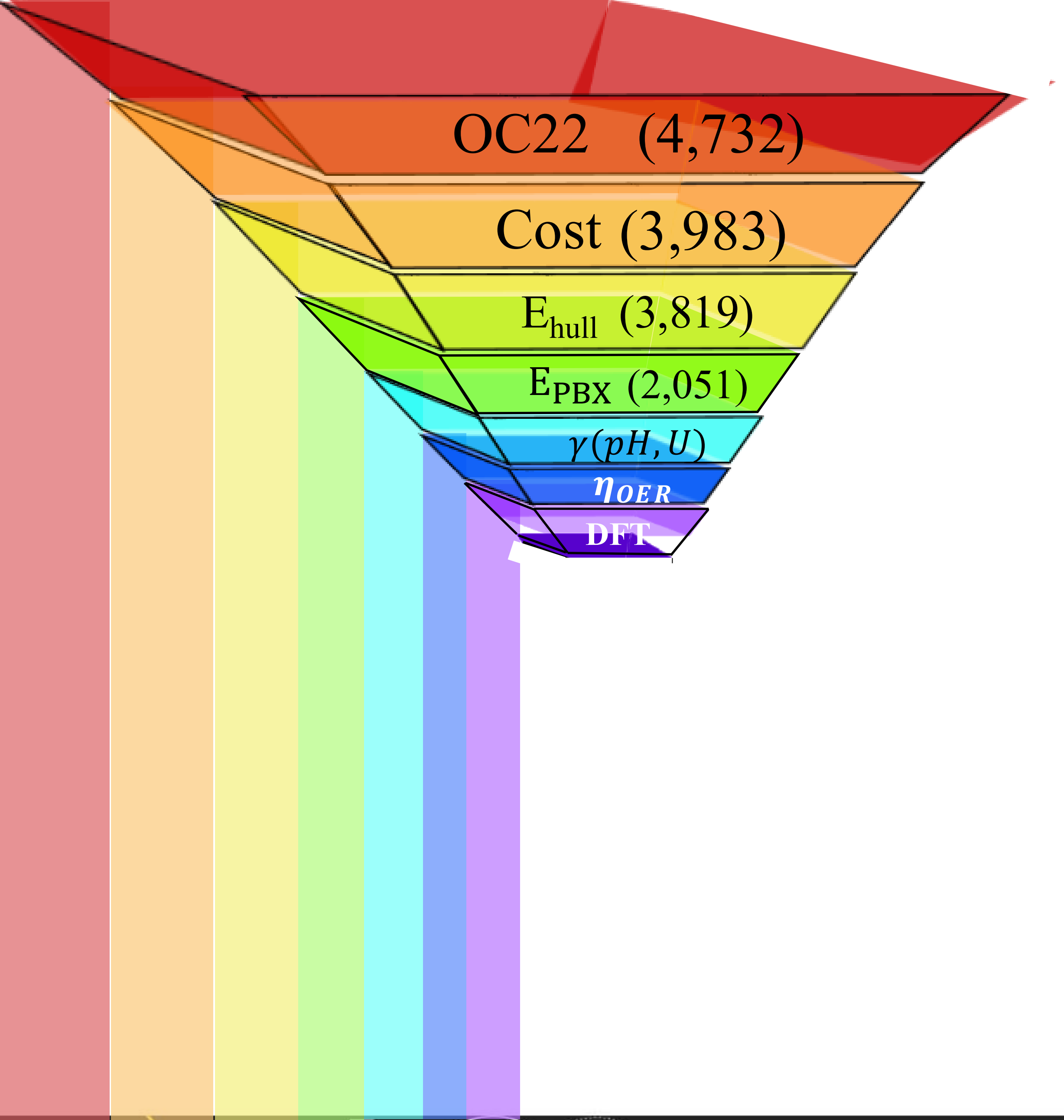




Candidates



	$CrCuO_2$	Co_5CuO_8	Be_3ZnO_4	$CaSeO_3$	VAg_3O_4	Cu_4PtO_5	$Mn_2V_2O_7$
($)	8.07	33.68	79.88	71.03	674.72	12,957	118.17
(meV)	0.00	13.32	90	11.9	0	10.09	0
(meV)	149	11	167	149	228	131	351
# viable facets	4	3	4	4	3	4	3
η_{OER} (V)	0.39	0.56	0.57	0.56	0.39	0.60	0.52
η_{OER} (V)	0.28	0.71	0.41	0.54	0.58	0.42	0.41

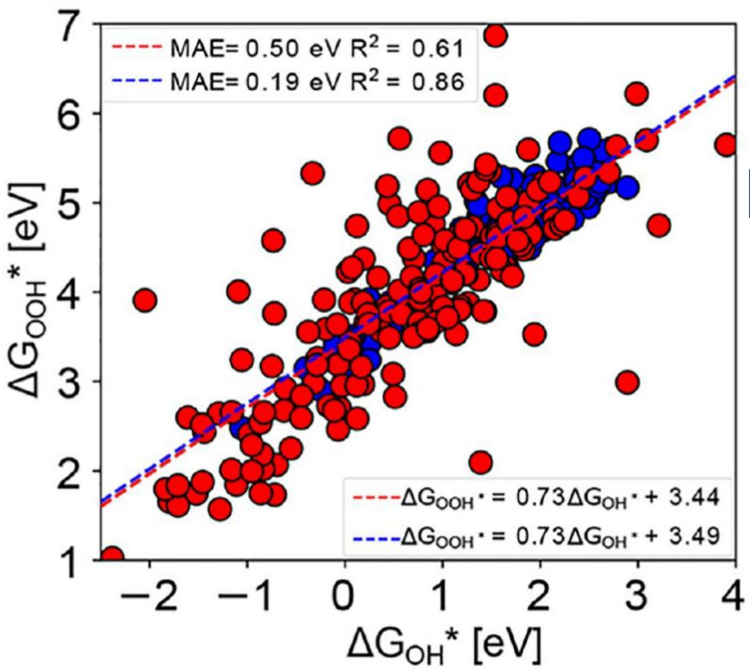


	$CrCuO_2$	Co
(\$)	8.07	3
(meV)	0.00	1
(meV)	149	
# viable facets	4	
η_{OER} (V)	0.39	0
η_{OER} (V)	0.28	0

Application 2:
Overcoming the overpotential wall

Dauenhauer, P. J., Ardagh, M. A., Shetty, M., Kuznetsov, A., Zhang, Q., Christopher, P., Vlachos, D. G., & Abdelrahman, O. A. (2020). *Chemical Science*, *11*(13), 3501–3510. https://doi.org/10.1039/c9sc06140a

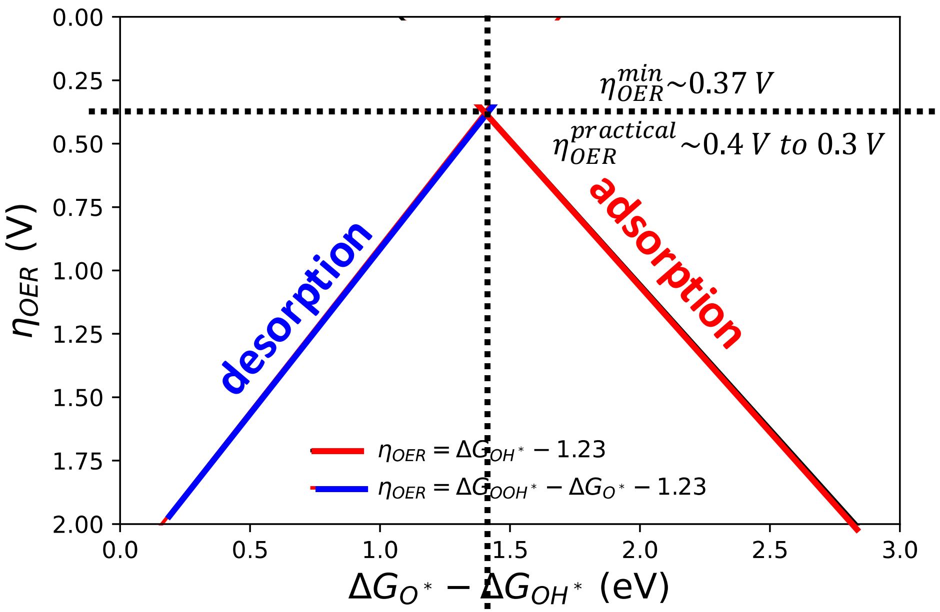
The Sabatier limit

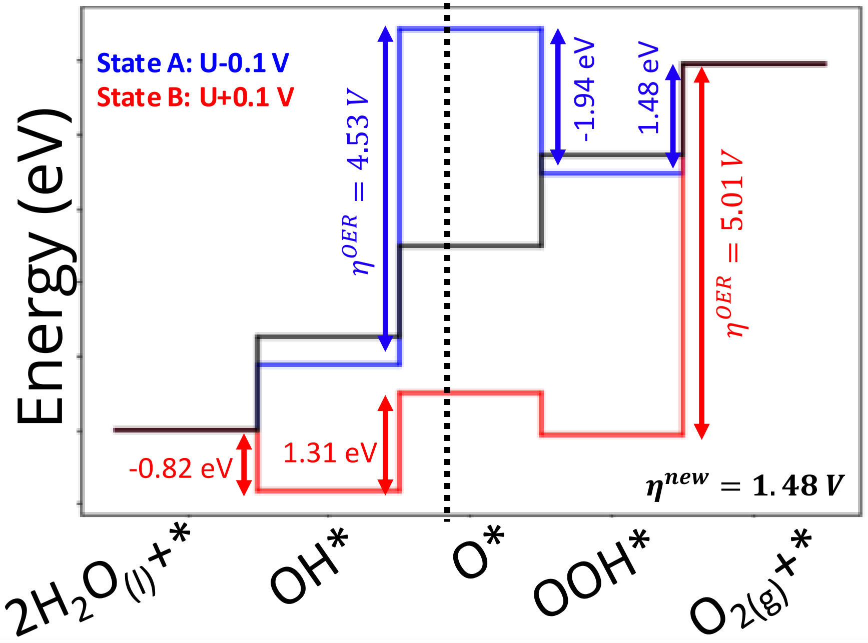


$$\Delta G_{OOH^*} \sim 0.73\Delta G_{OH^*} + 3.44$$
$$\Delta G_{O^*} \sim \Delta G_{OH^*} + 1.87$$

Solutions:

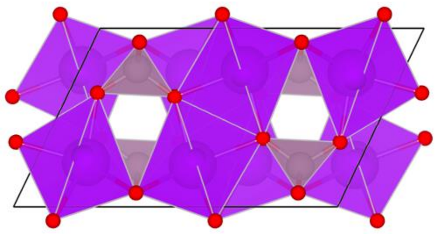
- Identify materials that do not follow conventional scaling (e.g. molecular catalysts or SACs)
- Create new reaction pathways with ***dynamic catalysis***

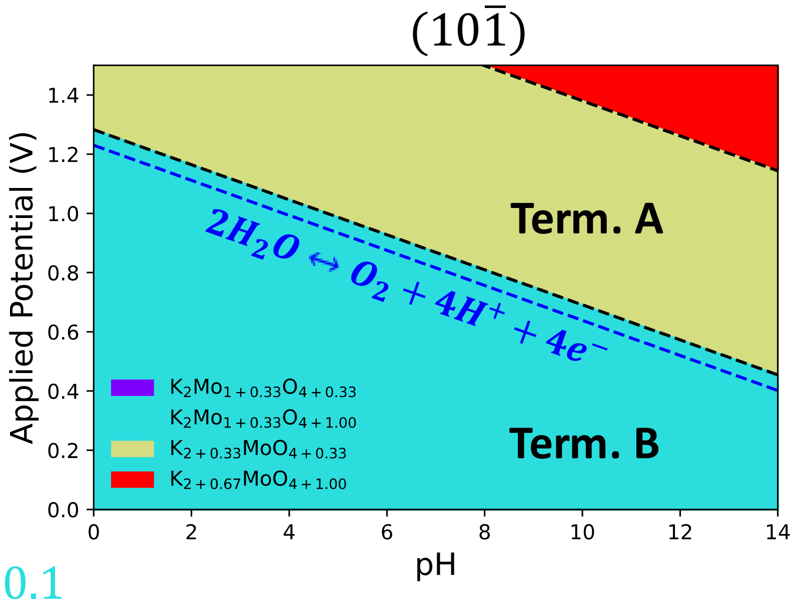


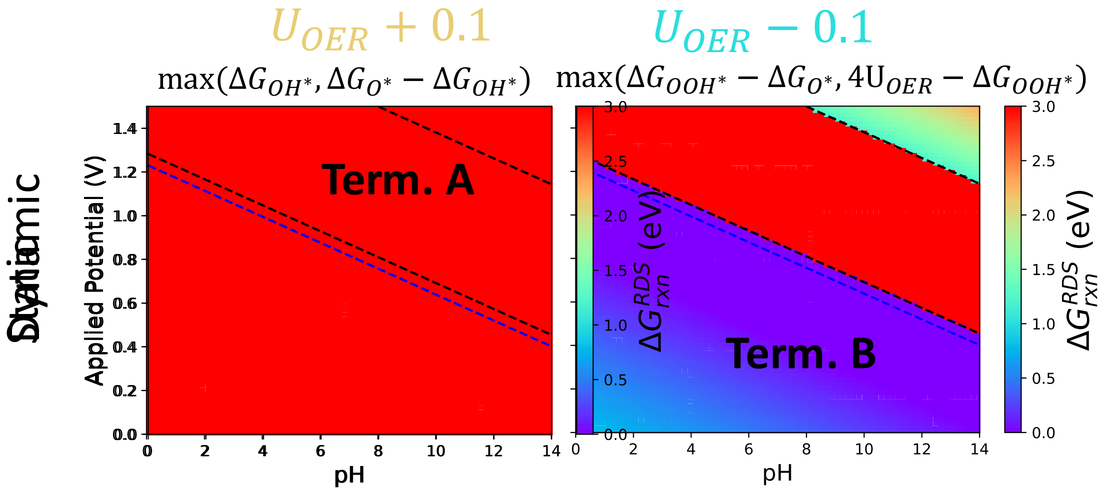


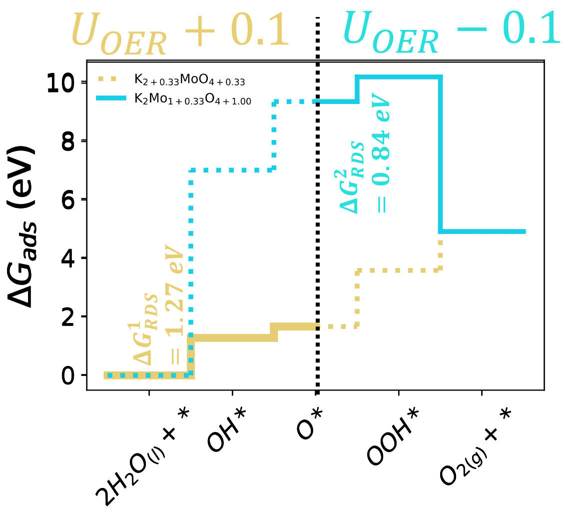
Dynamic catalysis

K_2MoO_4









Is the transition line between termination A and B coinciding with the OER equilibrium potential (± 0.1)?

Does dynamic switching between terminations result in a lower overpotential than the static case?

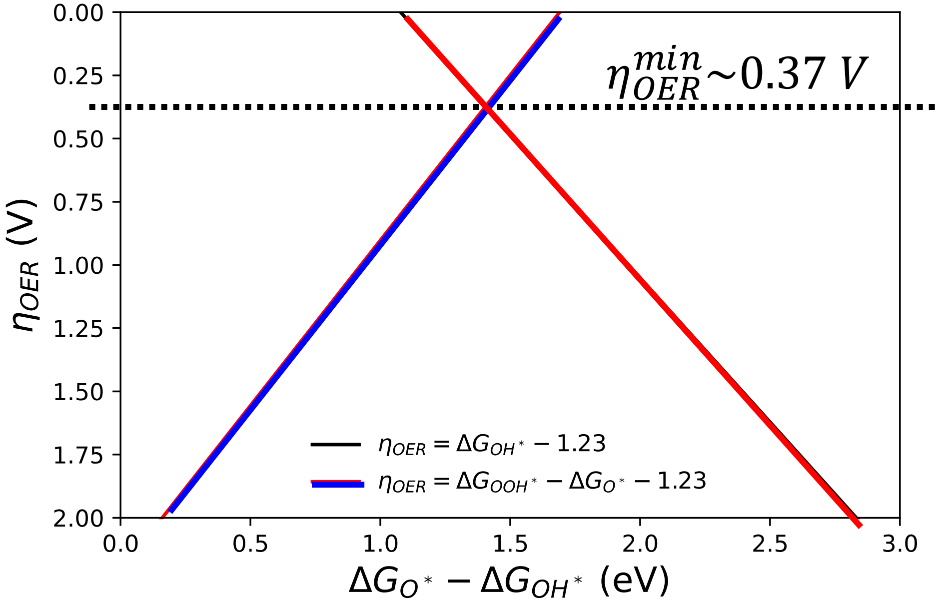
New overpotential:

$$\eta_{OER}^{dynamic} = \max(\Delta G_{RDS}^1, \Delta G_{RDS}^2) - 1.23 = 0.04\ V$$

Candidates

material	Static η_{OER} (V)	Dynamic η_{OER} (V)	(hkl)
$CaGe_2O_5$	6.86	0.30	(011)
Zn_2SiO_4	6.16	0.074	(001)
K_2MoO_4	5.79	0.05	$(10\bar{1})$
KV_3O_8	4.98	0.023	(101)

$\eta_{OER}^{practical} \sim 0.4\ V\ to\ 0.3\ V$



Summary

- Constructed a database of ML (OC22) predictions of total energy of surfaces and OER intermediates for all *in domain* materials considered in OC22.
- Implement high-throughput screening framework to identify 17 materials with competitive overpotential and cheaper material cost relative to IrO_2 and RuO_2
- Developed a framework for identifying 4 materials that can possibly exceed the Sabatier Limit via dynamic catalysis.

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Hewlett Packard Enterprise Data Science Institute

UH MODAL LAB

Multidisciplinary Modeling, Oilfield Data Analytics, and Well Logging Laboratory

Jiefu Chen

Wu Xuqing

Yuan Zi

Liqiang Huang

The Computational Catalysis and Interface Chemistry Group

Lars Grabow

Shengguang Wang

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