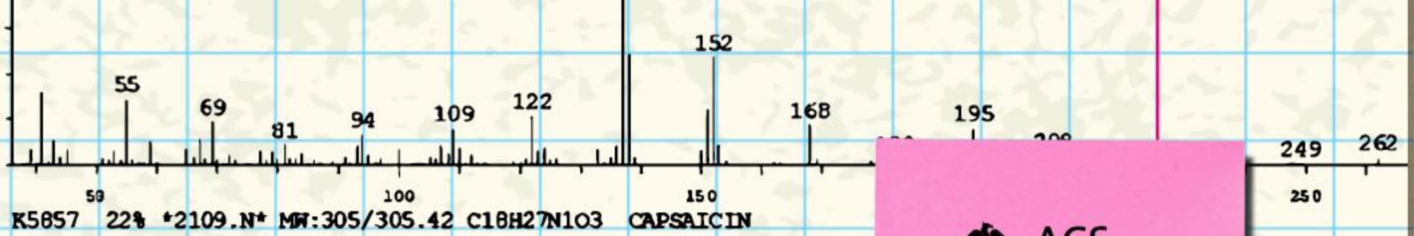

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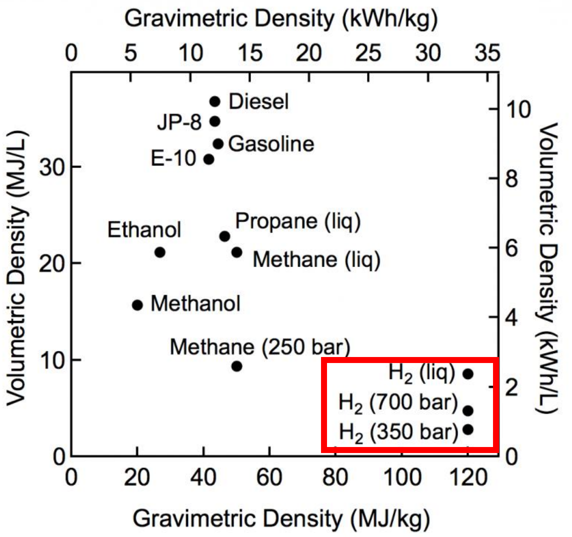
Applications of the Open Catalyst Project: High-throughput screening and design of heterogeneous catalysts

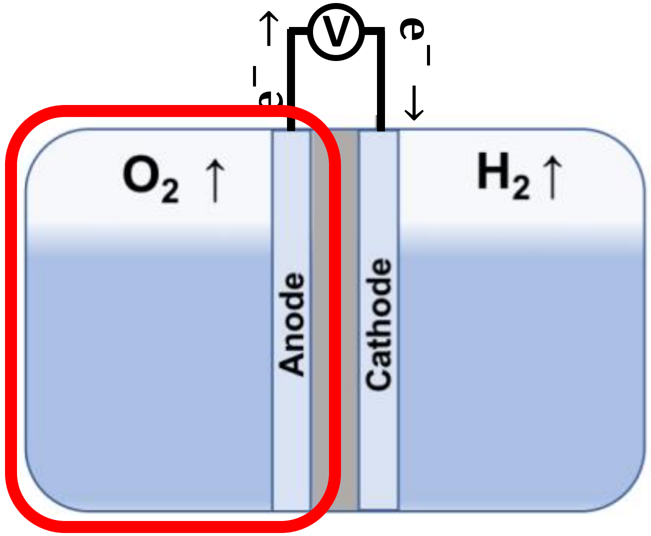
Richard Tran

3990961

rtran25@cougarnet.uh.edu

Harnessing the power of water

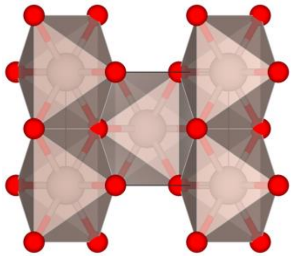




OER: $2H_2O \rightarrow O_2 + 4H^+ + 4e^-$

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

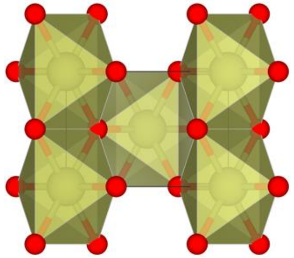
$2H_2O \rightarrow O_2 + \mathbf{2H_2}$



RuO_2

\$18,315/kg

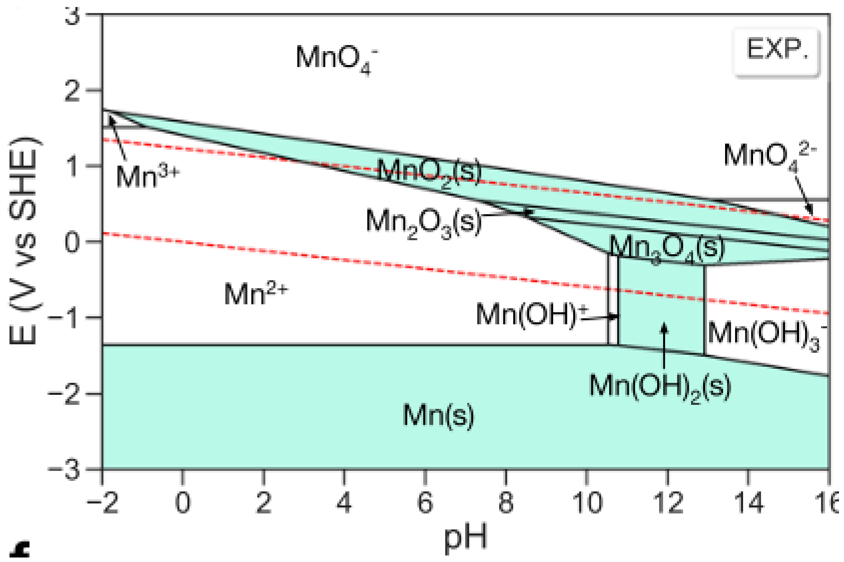
Ru: \$24,113/kg



IrO_2

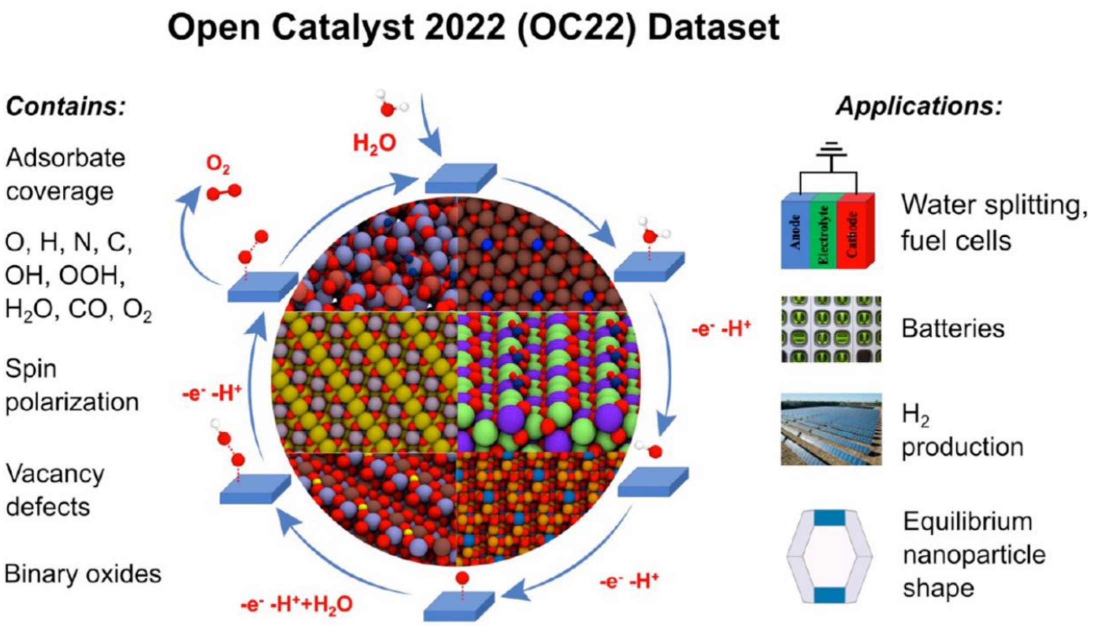
\$155,727/kg

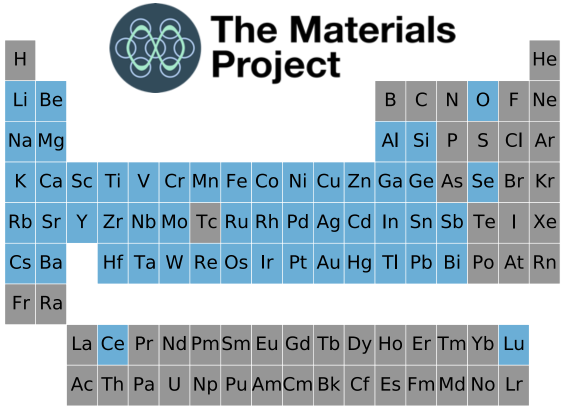
Ir: \$181,651/kg

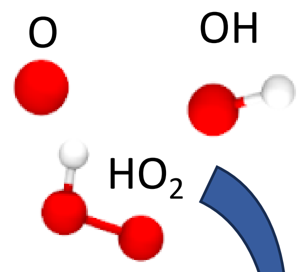


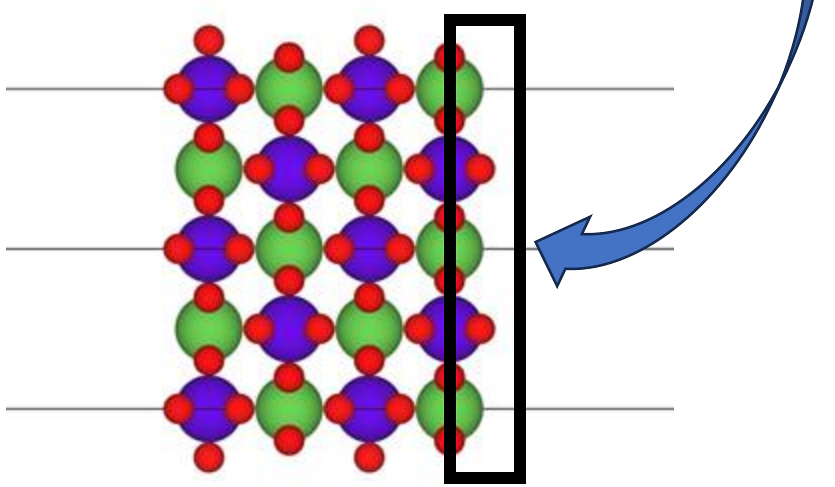
The Open Catalyst Project 2022

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)

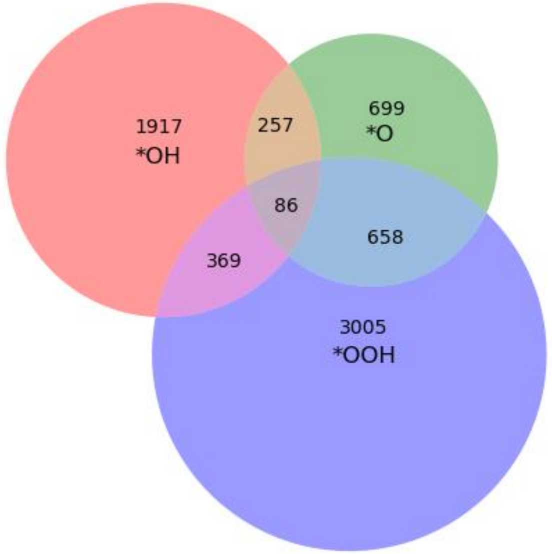
Carnegie Mellon University, Pittsburgh Pennsylvania 1900

Database scope

All data available at UH Dataverse Repository under:

Texas Data Repository

https://doi.org/10.18738/T8/APJFTM



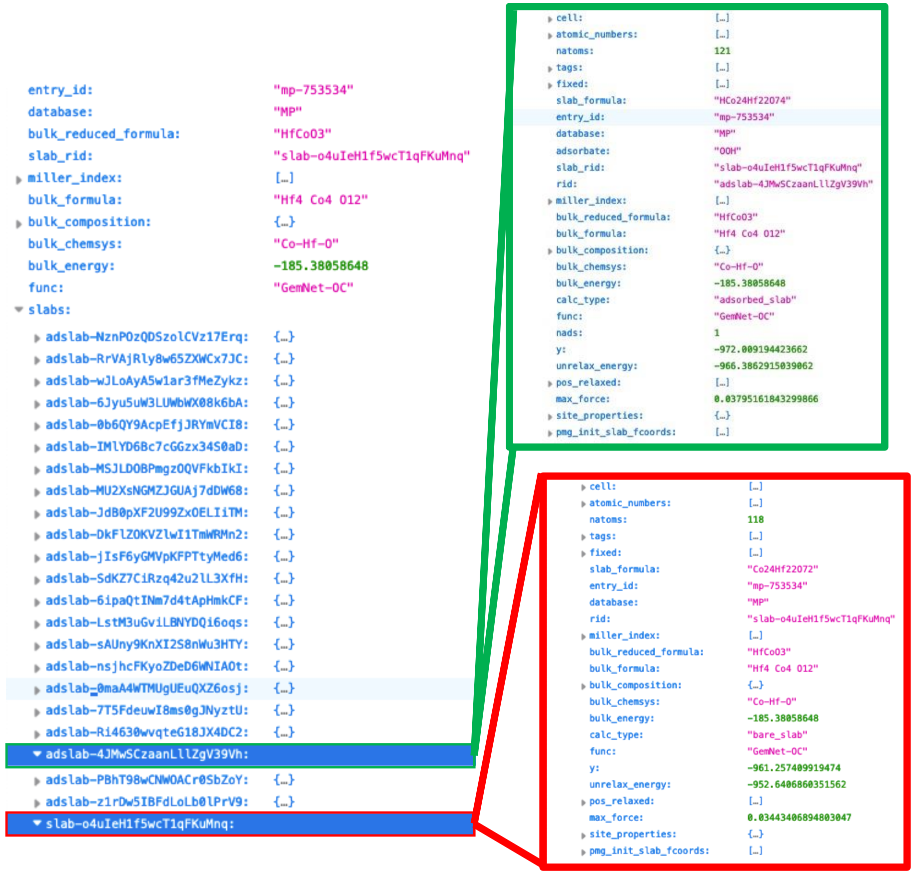
OC22 DFT dataset

# Predictions		6,068,572
# Materials		4,119
# Slab predictions		191,902
Ave. # slabs per material		47
# Adsorption predictions		5,876,670
Max Miller index		1
OH*	**O***	**OOH***
1,972,166	667,266	3,237,238

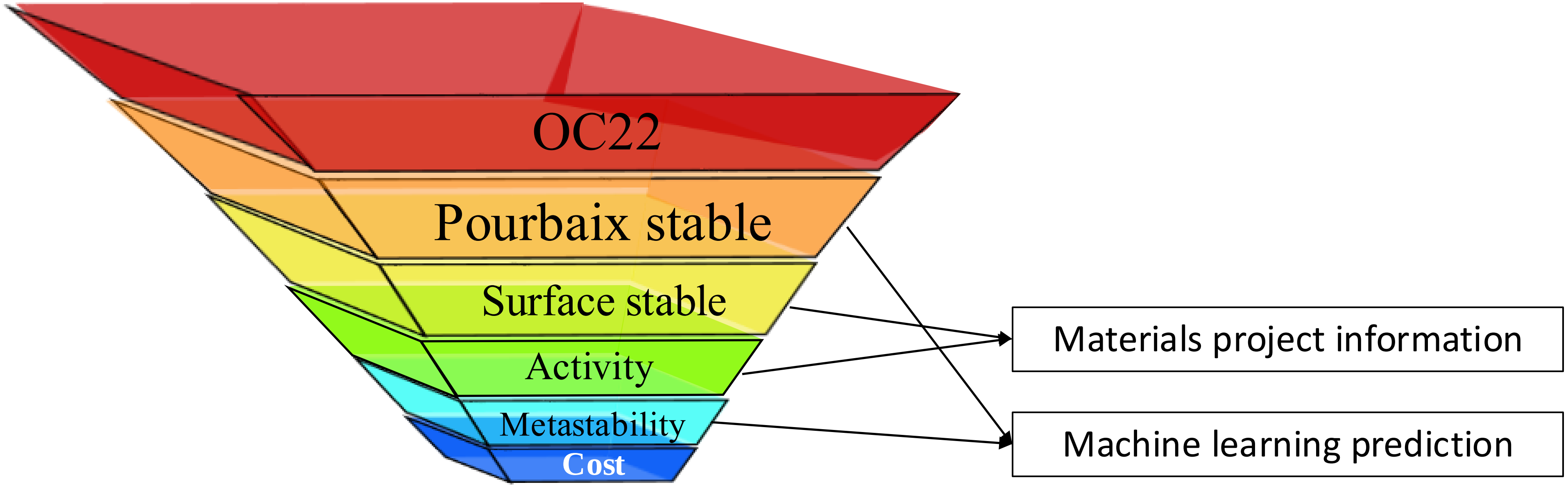
OC22 prediction dataset

We can substitute ΔG^{OOH^*} with:

$$\Delta G^{OOH^*} = \Delta G^{OH^*} + 3.26$$

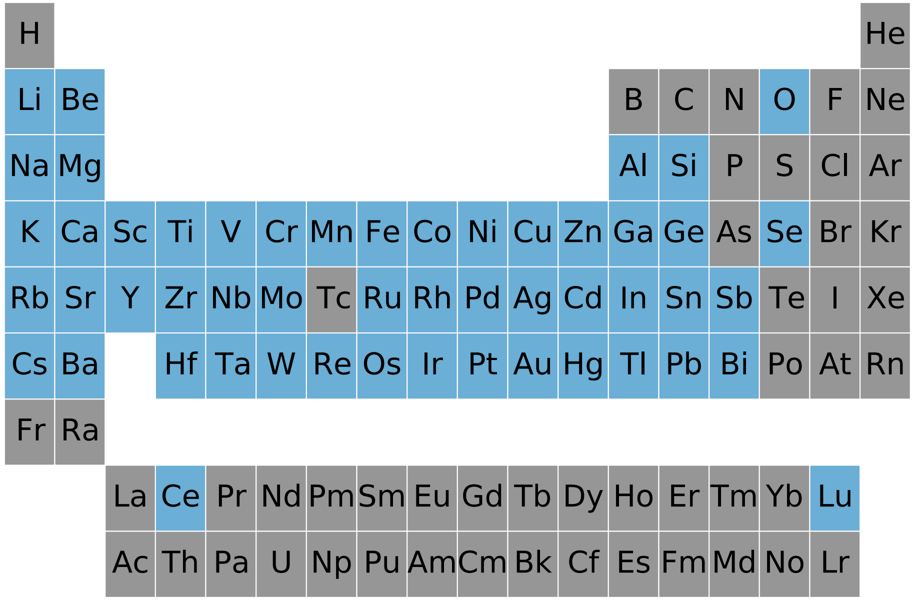


High throughput screening



OC22 dataset

OC22
(4,119)



- 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

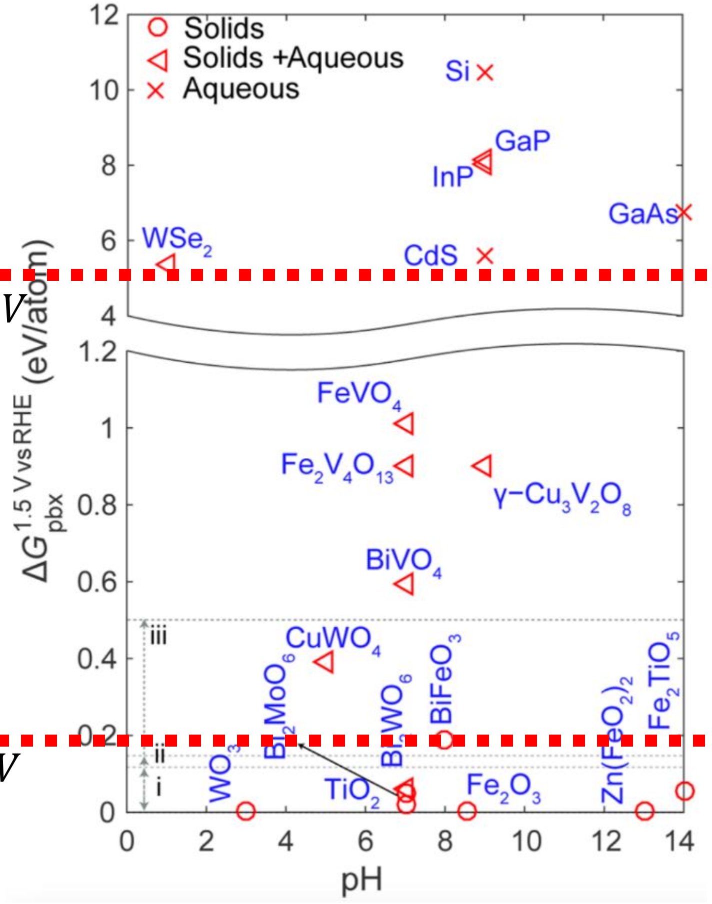
Pourbaix stability

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

OC22
(4,119)

Pourbaix stable (T, U, pH) 1,853

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



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

$$\mathbf{\Delta G_{PBX}(pH = 1, U = 1.8, T = 80^oC) < 0.5\ eV}$$

ΔG_{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

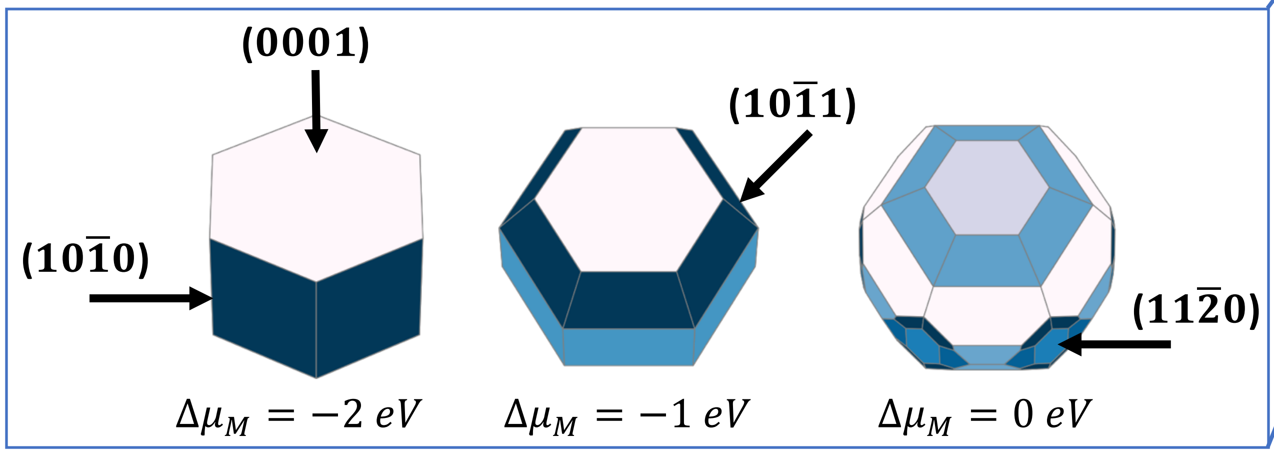
Surface stability

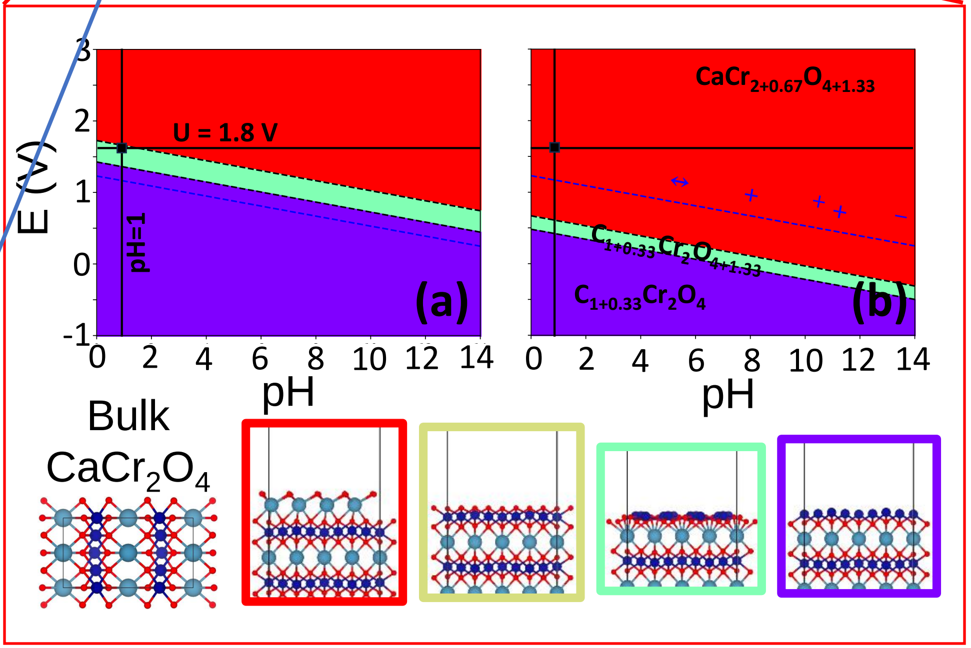
OC22
(4,119)

Pourbaix stable (T, U, pH) — 1,853

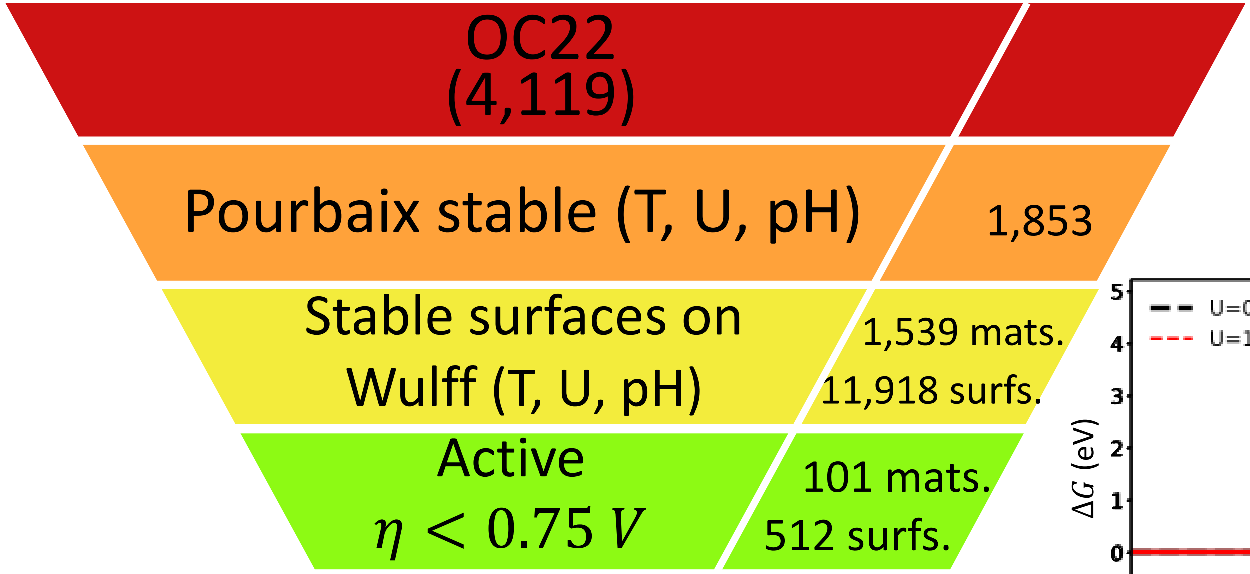
Stable surfaces on Wulff (T, U, pH) — 1,539 mats. 11,918 surfs.

$$\gamma(\mu_M, \mu_{O_2}) = \frac{E_{slab}^{A_{nx}B_{ny+k}O_{nz+j}} - nE_{bulk}^{A_xB_yO_z} + k\mu_B + j\frac{1}{2}\mu_{O_2}}{2A}$$

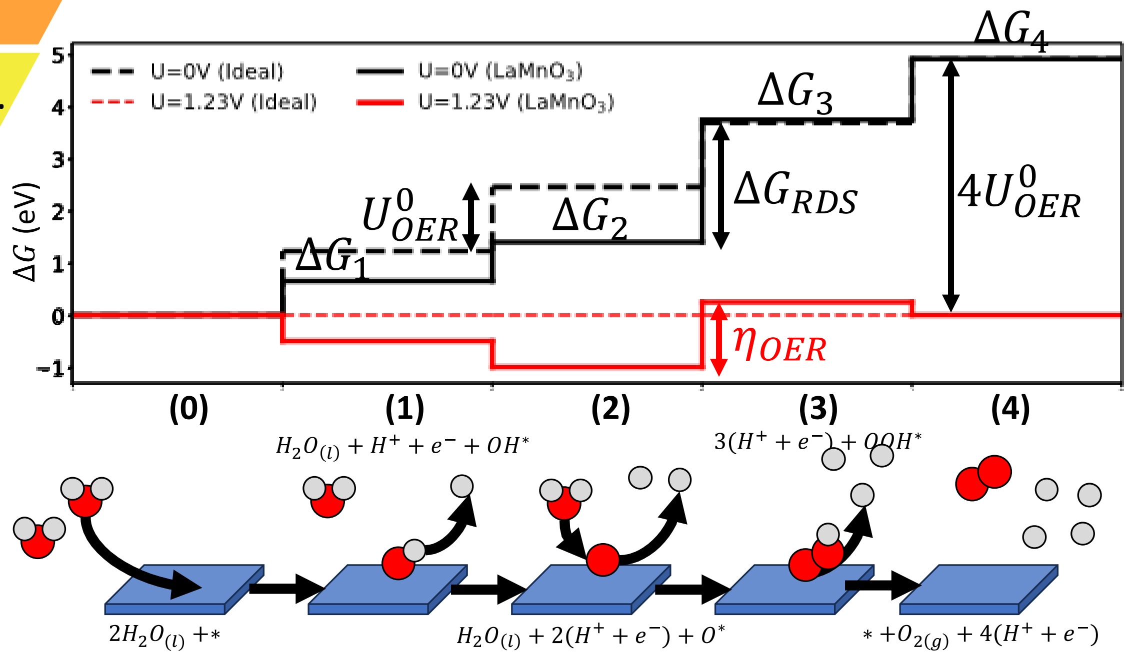




OER activity (overpotential)



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

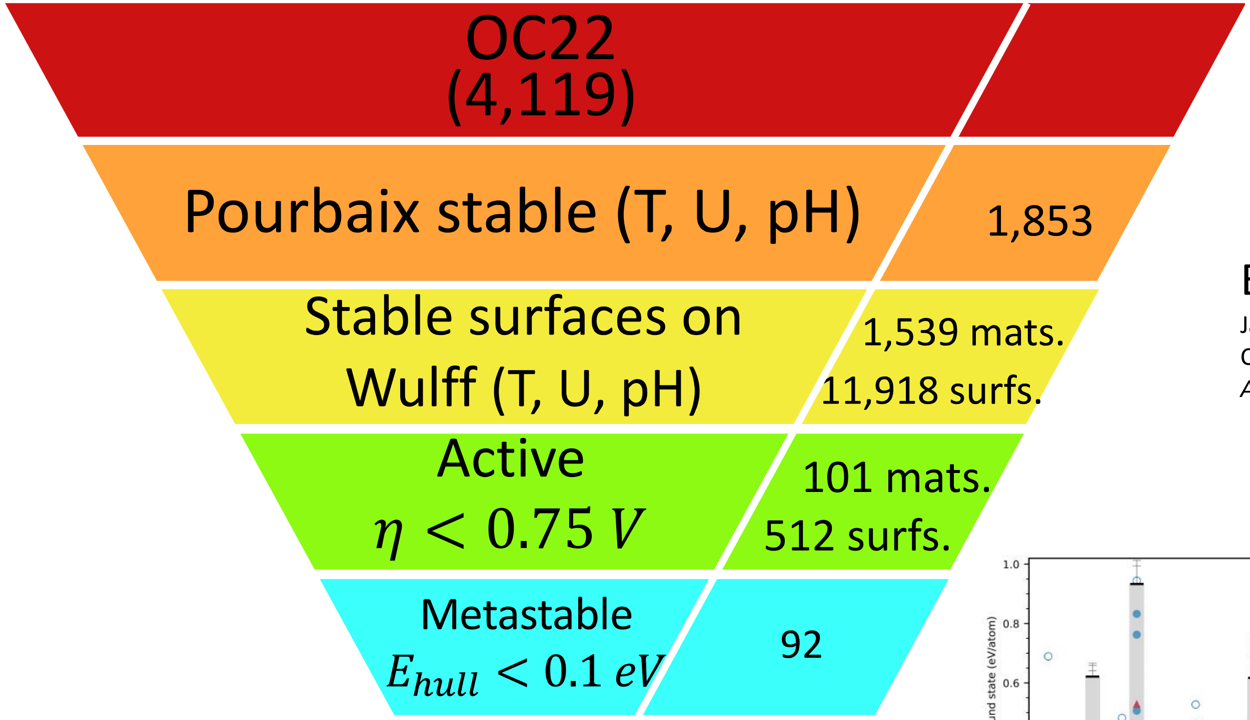


$$\Delta G_{RDS} = \max(\Delta G_1, \Delta G_2 - \Delta G_1, \Delta G_3 - \Delta G_2, \Delta G_4 - \Delta G_3,)$$

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

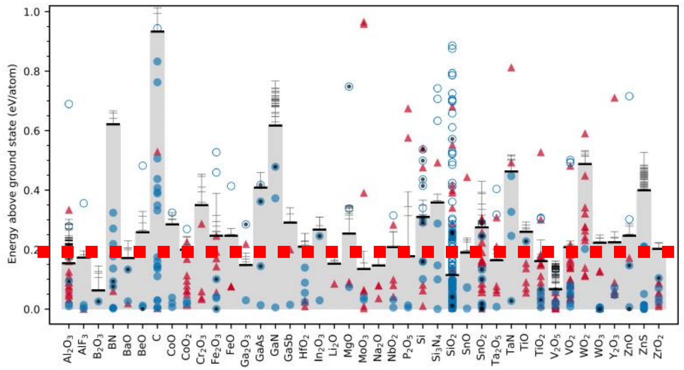
$$U^0_{OER} = 1.23\ V\ vs\ SHE$$

Metastability



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

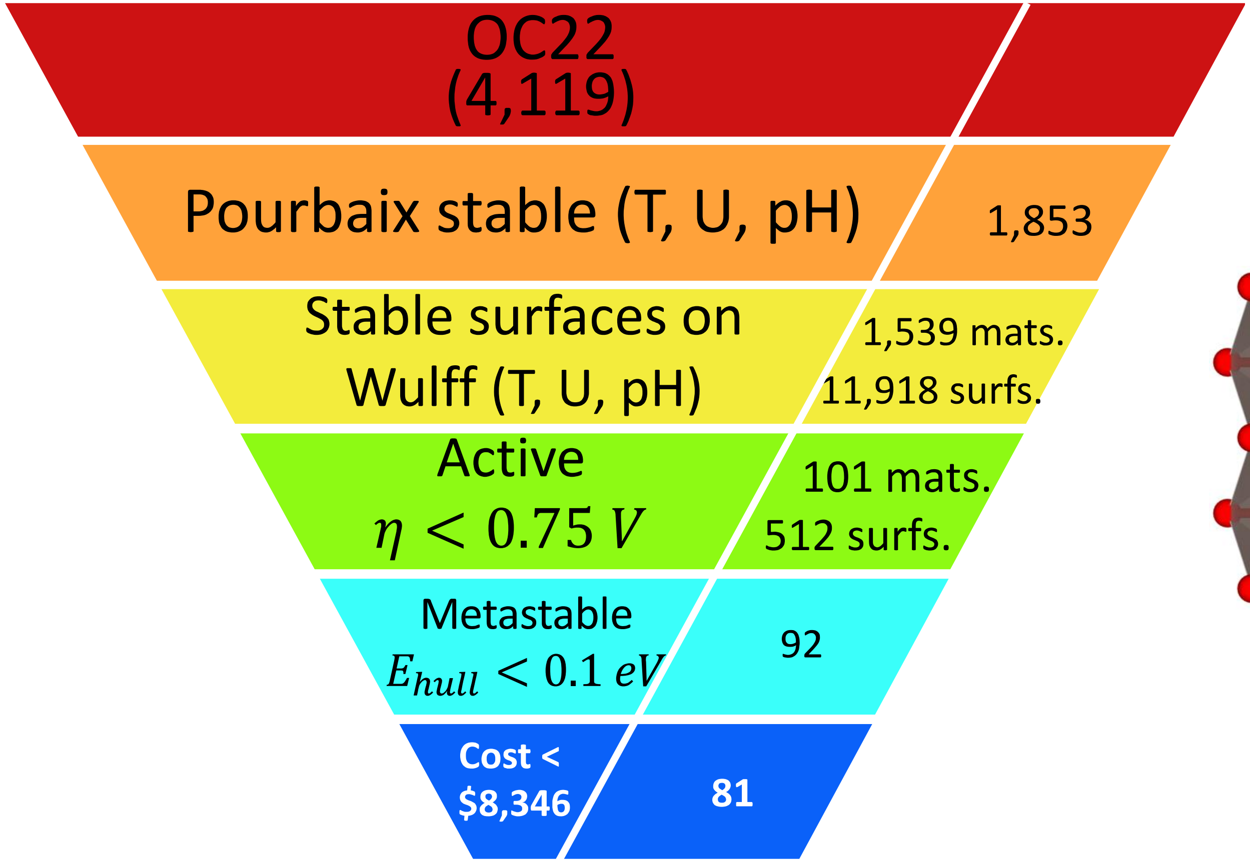


Aykol, M. *et al.* (2018). *Science Advances*, *4*(4), 1–8. doi.org/10.1126/sciadv.aaq0148

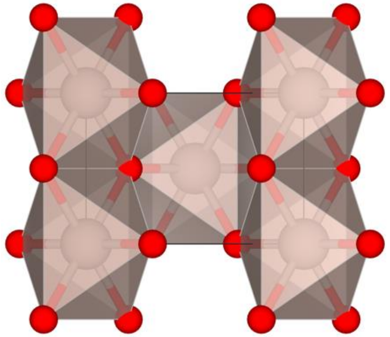
Metastability limits:

$E_{hull} < 0.2$ eV

Material cost



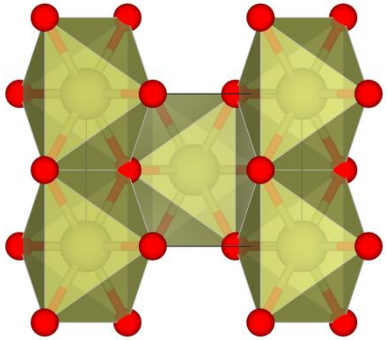
Cost < $18,315/kg



RuO_2

$18,315/kg

Ru: $24,113/kg

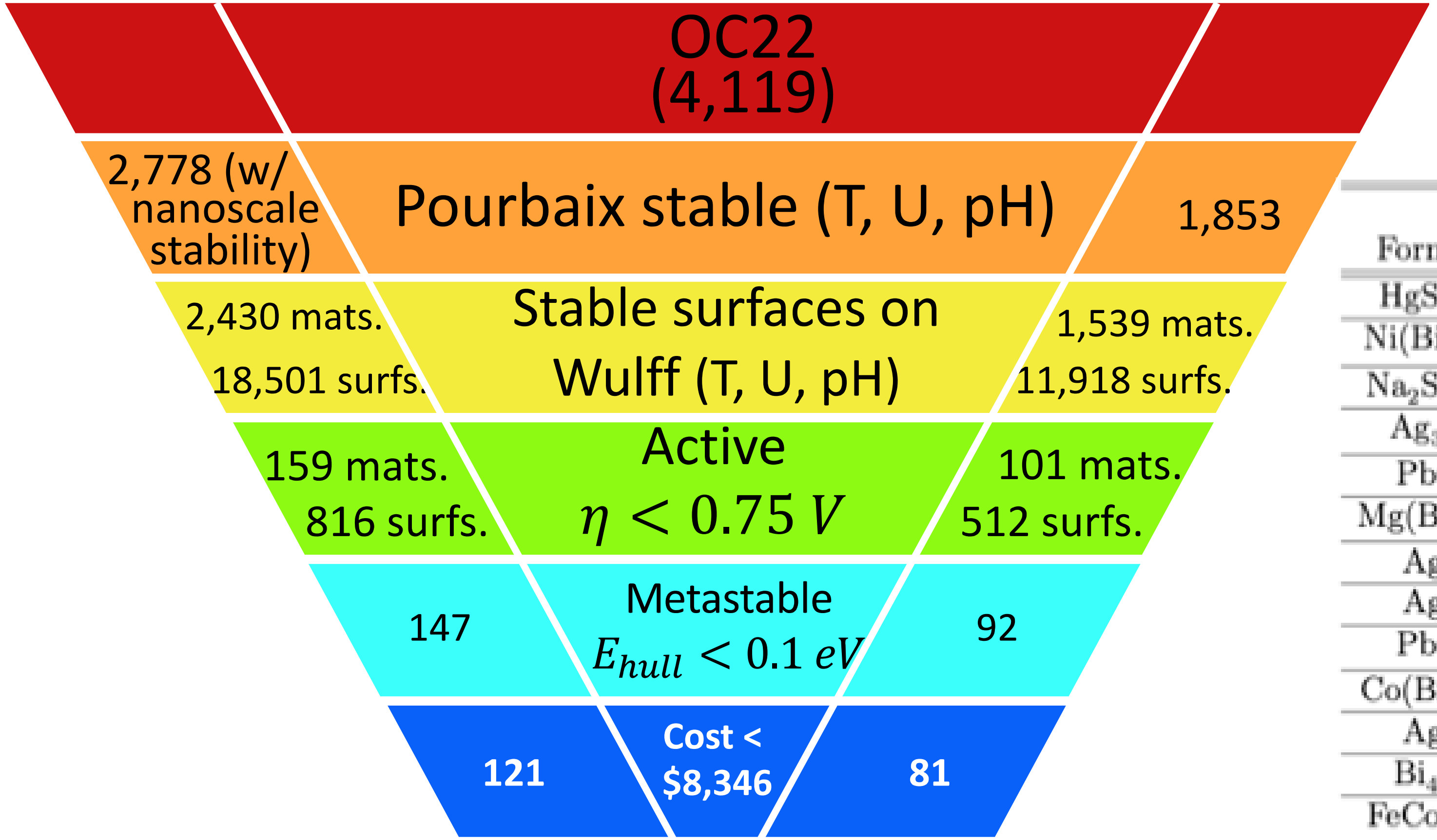


IrO_2

$155,727/kg

Ir: $181,651/kg

Final candidates

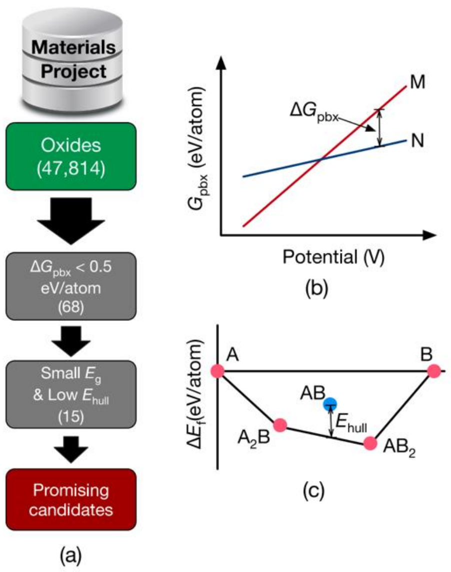


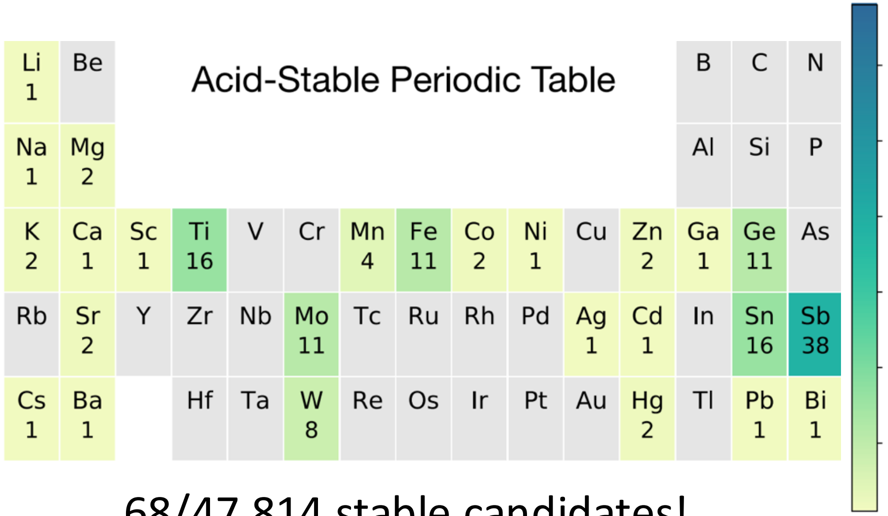
Formula	Space group	# facets	η (V)	E_{PBX} (eV)	E_{hull} (eV)	Cost ($/kg)
$HgSeO_4$	$Pmn2_1$	2	0.18	0.00	0.00	65.47
$Ni(BiO_3)_2$	$P4_2/mnm$	4	0.36	0.00	0.00	20.88
$Na_2Se_2O_7$	$P\bar{1}$	2	0.21	0.00	0.00	110.43
Ag_3O_4	$P2_1/c$	4	0.33	0.00	0.00	714.77
PbO_2	$P4_2/mnm$	2	0.33	0.00	0.00	2.41
$Mg(BiO_3)_2$	$P4_2/mnm$	3	0.52	0.00	0.00	20.41
AgO	$Cccm$	2	0.49	0.00	0.00	745.41
AgO	$C2/c$	2	0.51	0.01	0.01	745.41
PbO_2	$Pbcn$	2	0.56	0.01	0.01	2.41
$Co(BiO_3)_2$	$P4_2/mnm$	2	0.33	0.02	0.02	24.34
AgO	$P2_1/c$	3	0.50	0.04	0.04	745.41
Bi_4O_7	$P\bar{1}$	2	0.22	0.04	0.00	22.67
$FeCo_9O_{20}$	$P\bar{1}$	4	0.41	0.06	0.07	31.63

Limitations of Pourbaix stability

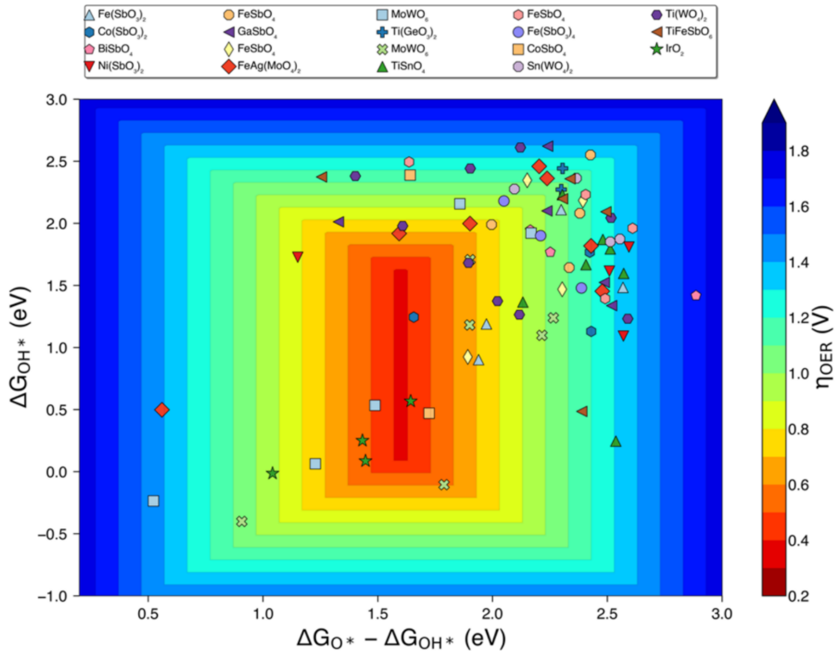
Gunasooriya, G. T. K. K., & Nørskov, J. K. (2020). Analysis of Acid-Stable and Active Oxides for the Oxygen Evolution Reaction. *ACS Energy Letters*, *5*(12), 3778–3787. https://doi.org/10.1021/acsenergylett.0c02030

Pourbaix stable (T, U, pH) 1,853





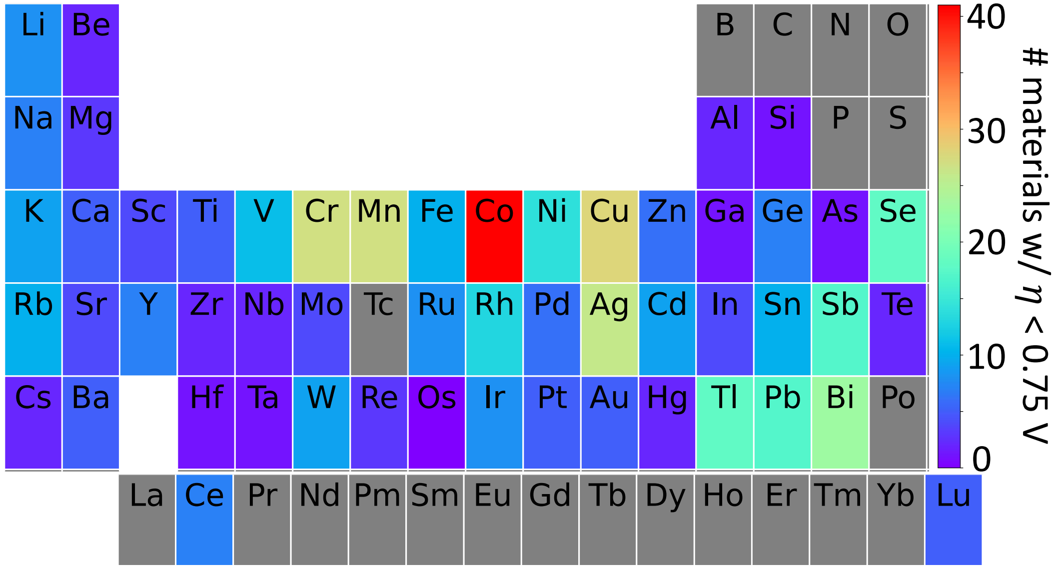
68/47,814 stable candidates!



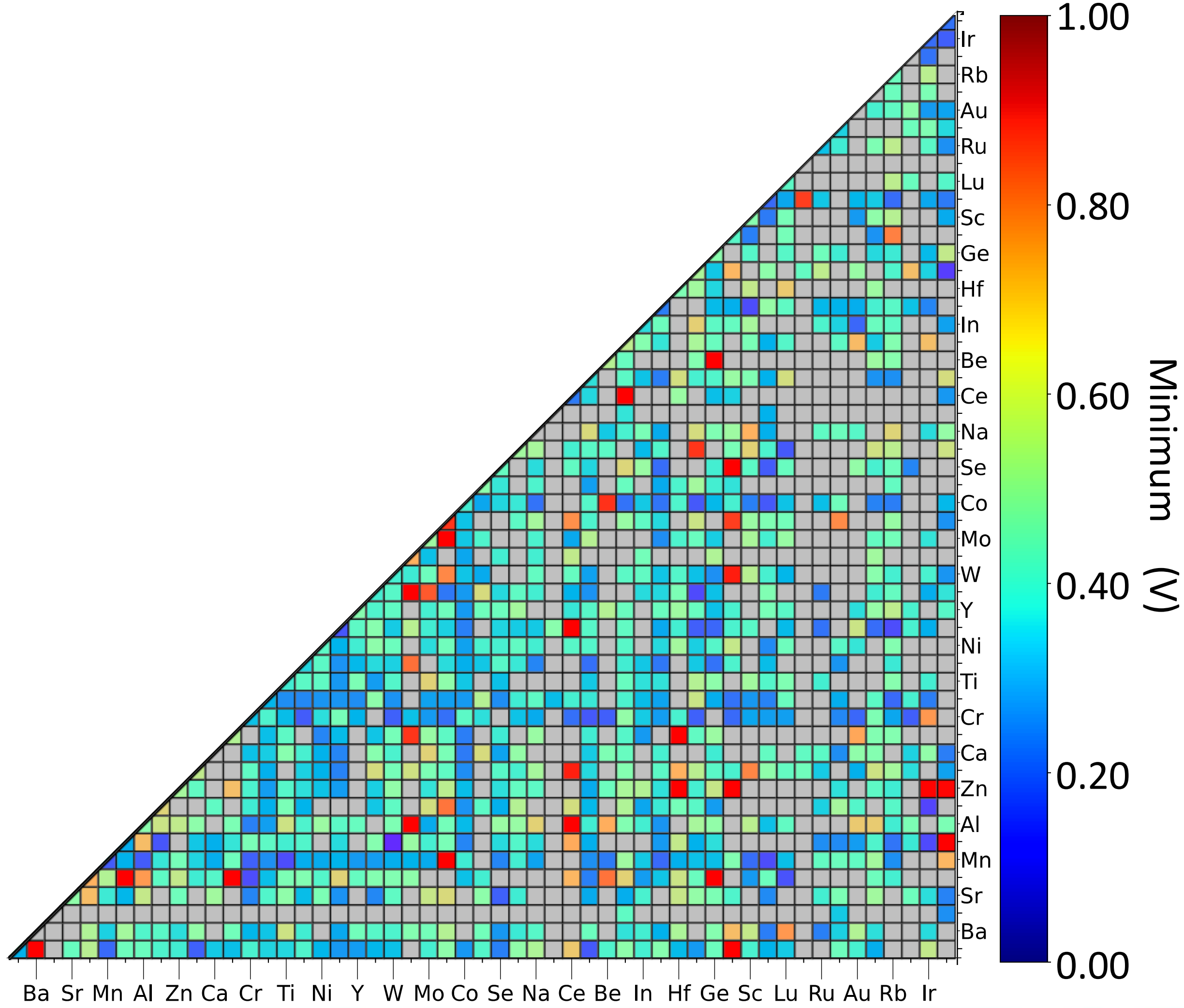
17 candidates w/ $\eta_{OER} < 0.8$ eV

Wang, Z., Zheng, Y. R., Chorkendorff, I., & Nørskov, J. K. (2020). Acid-Stable Oxides for Oxygen Electrocatalysis. *ACS Energy Letters*, *5*(9), 2905–2908. https://doi.org/10.1021/acsenergylett.0c01625

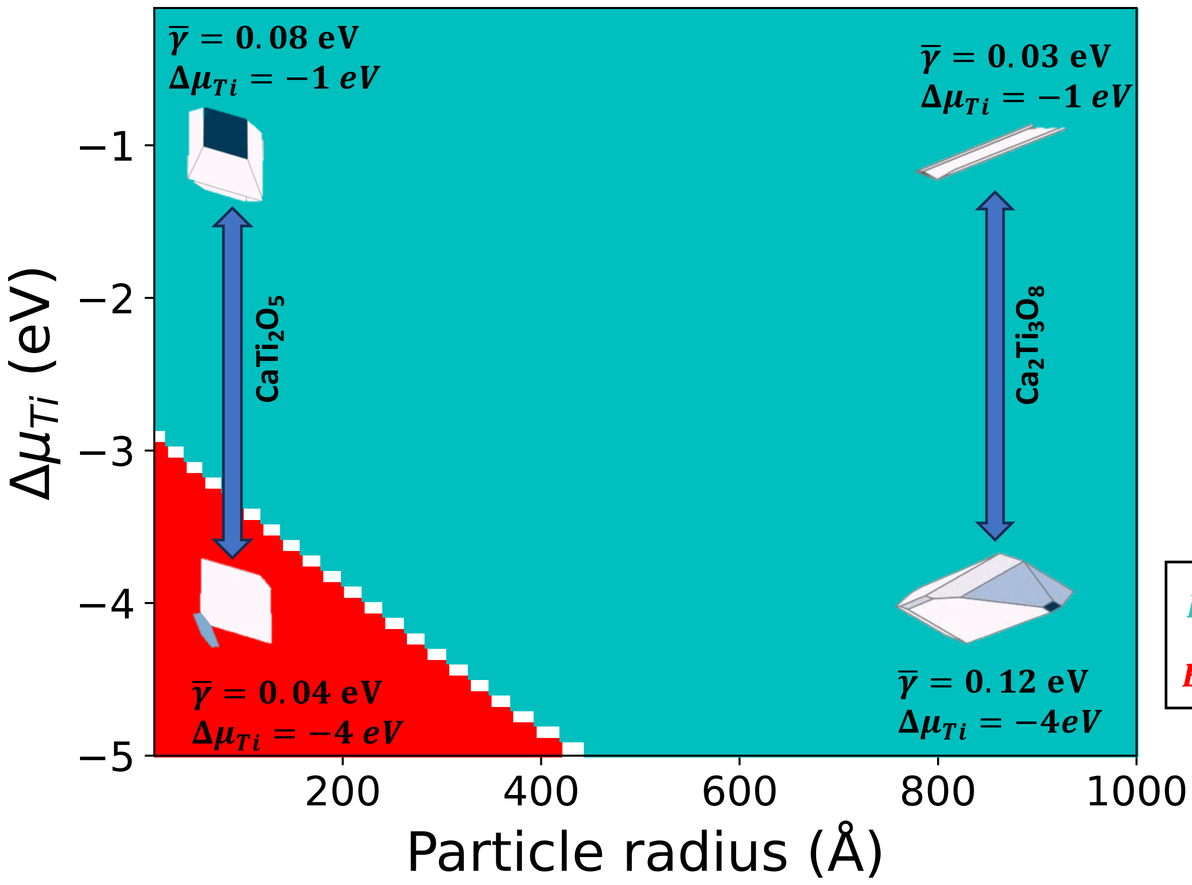
Overpotential assessment



Are there other oxides with low overpotential that are unstable? Is there a way to synthetically access these candidates?



Nanoscale stabilization



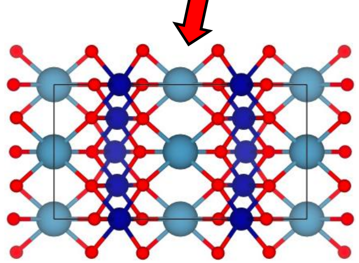
$E_{PBX}^{Ca_2Ti_3O_8} = -4.15\ eV$

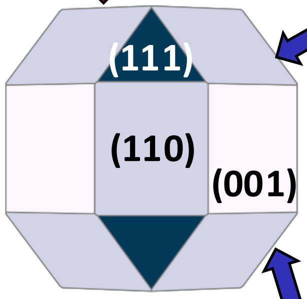
$E_{PBX}^{CaTi_2O_5} = -4.14\ eV$

Pourbaix formation energy of bulk per unit cell

Weighted surface energy

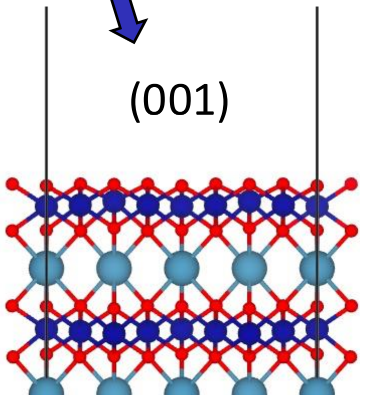
$$G_f^{NP} = E_V(pH, V, T)\left(\frac{4}{3}\pi r^3\right) + \bar{\gamma}(pH, V, T, \Delta\mu_M)(4\pi r^2)$$





Wulff shape (NP analog)

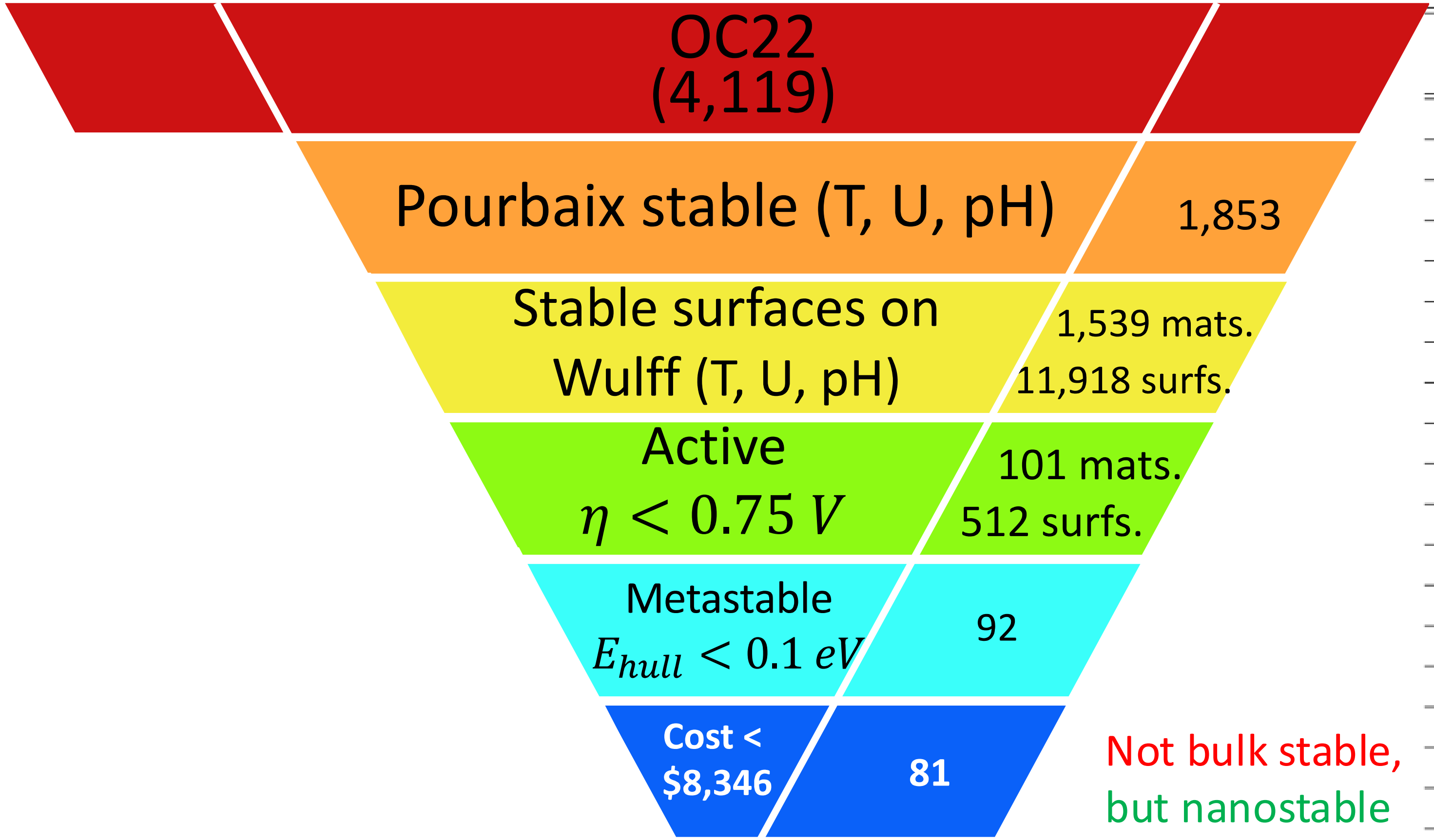
(001)



$$\bar{\gamma} = \sum_{hkl} \gamma_{hkl} f_{hkl}^A$$

f_{hkl}^A = fractional area of plane (hkl) on Wulff shape

Revised screening mechanism



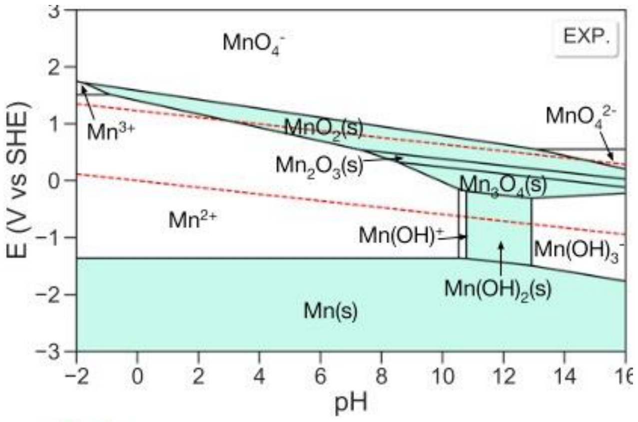
Not bulk stable, but nanostable

Formula	Space group	# facets	η (V)	E_{PBX} (eV)	E_{hull} (eV)	Cost ($/kg)
Cr_2WO_6	$P4_2/mnm$	2	0.25	0.52	0.00	20.20
$TlCuO_2$	$R\bar{3}m$	2	0.43	0.52	0.05	4091.65
$Y(FeO_2)_2$	$P\bar{1}$	2	0.53	0.54	0.01	11.23
$Sr_2Tl_2O_5$	$P2_1/c$	2	0.37	0.55	0.00	3694.92
$ZrCoO_3$	$P\bar{1}$	3	0.46	0.55	0.10	32.83
$TiVO_4$	$P2_1$	2	0.58	0.56	0.02	120.33
$HfFeO_3$	$Pnma$	2	0.53	0.56	0.06	569.53
Mn_4CuO_8	$C2/m$	3	0.39	0.56	0.07	3.51
$CoCu_2O_3$	$Pmmn$	3	0.42	0.57	0.07	18.94
$MnSe_2O_5$	$Pbcn$	2	0.50	0.58	0.00	79.93
$CrMoO_4$	$Cmmm$	2	0.42	0.59	0.00	21.89
KMn_2O_4	$P\bar{1}$	2	0.47	0.60	0.00	185.55
$Ba_2Tl_2O_5$	$Pnma$	2	0.34	0.60	0.00	3213.59
$LuMnO_3$	$Pnma$	3	0.54	0.61	0.05	4722.98
$TiMnO_3$	$R\bar{3}$	2	0.28	0.62	0.00	5.36
$KBiO_2$	$C2/c$	2	0.27	0.62	0.00	158.83
Na_5ReO_6	$C2/m$	2	0.30	0.62	0.00	1406.49
$CuTeO_4$	$Cmmm$	3	0.54	0.63	-5.71	177.66
$ScCrO_3$	$Pnma$	2	0.46	0.63	0.04	1077.47
Ta_2CrO_6	$P4_2/mnm$	2	0.56	0.64	0.01	109.26
$MnSnO_3$	$R\bar{3}$	2	0.43	0.65	0.00	18.71

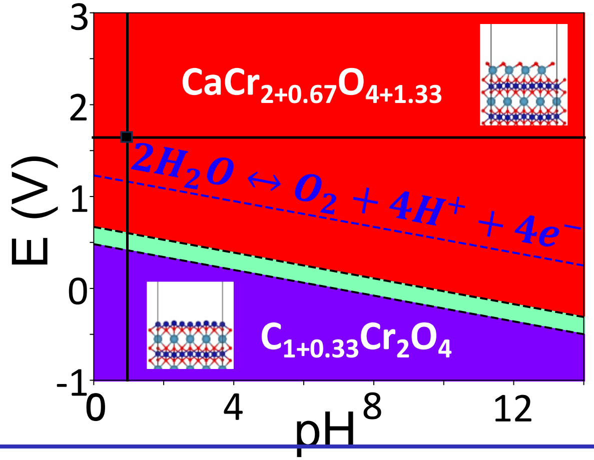
Alternate screening mechanisms

Temperature (^oC) Applied Potential (V)	60 1.8	60 1.2 to 2.0	80 1.8	80 1.2 to 2.0
Bulk	122[a]	99[e]	120[i]	99[m]
Bulk/Wulff	83[b]	62[f]	81[j]	62[n]
Bulk/Wulff/Nano	111[c]	83[g]	121[k]	84[o]
Bulk/Nano	168[d]	129[h]	181[l]	129[p]

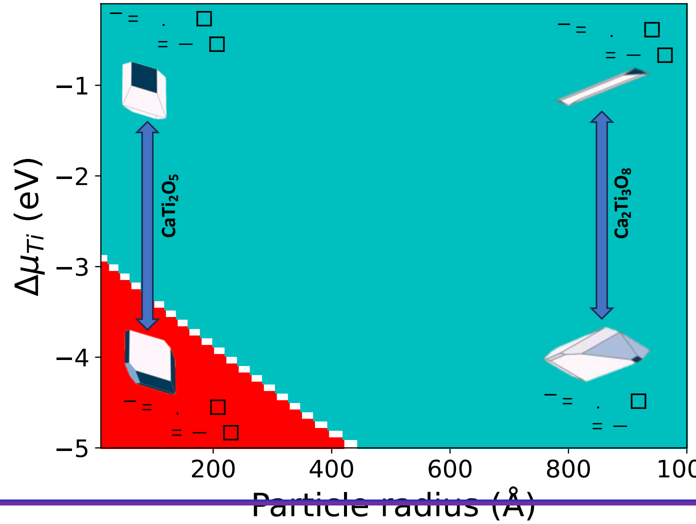
Bulk Pourbaix



Surface Pourbaix

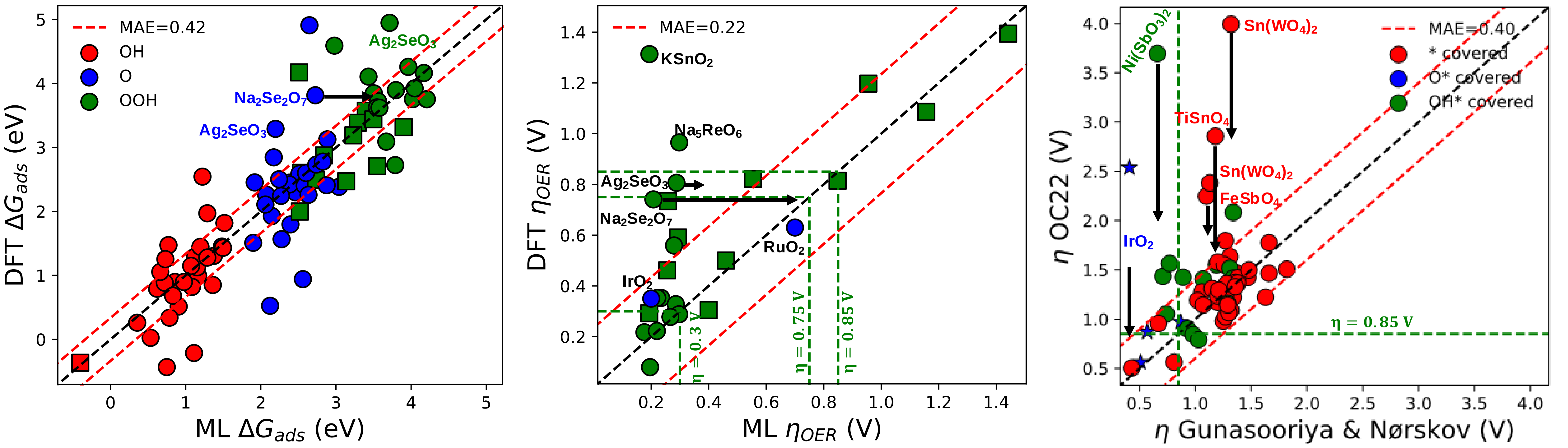


Nanoparticle PD



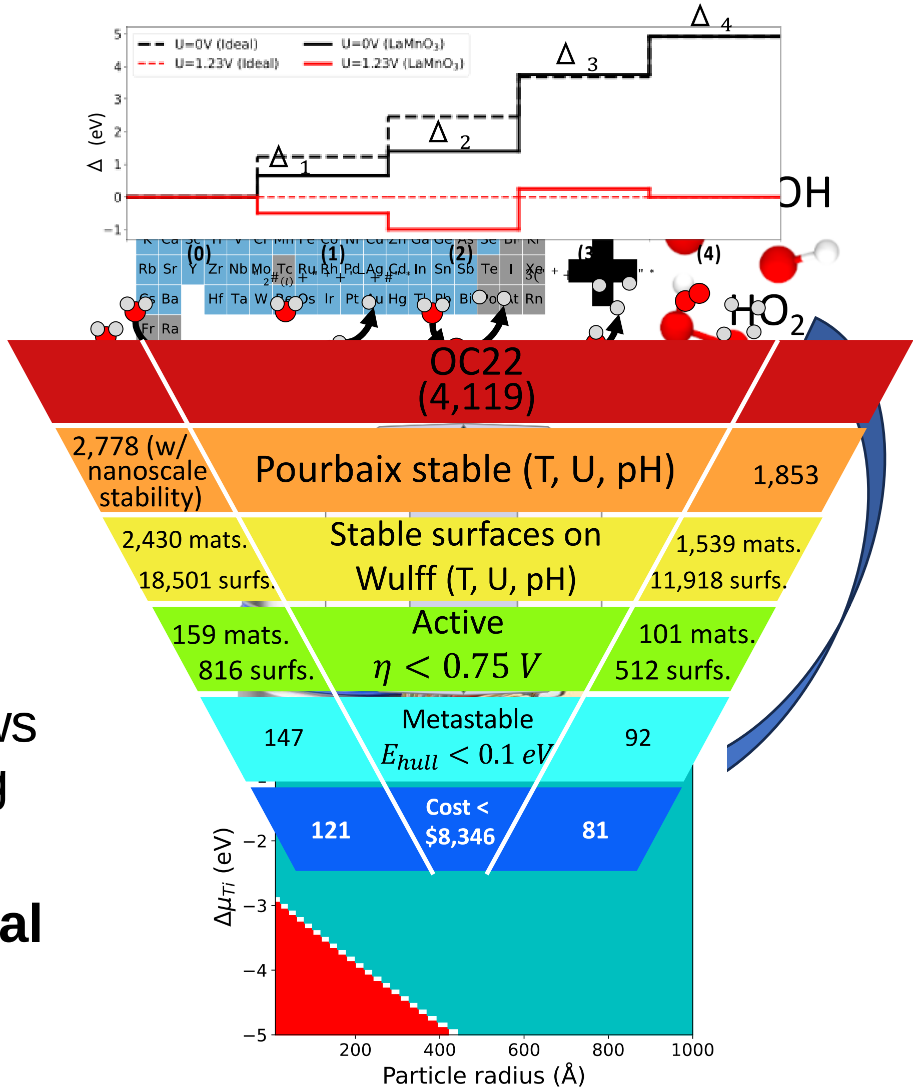
Validation

Gunasooriya, G. T. K. K., & Nørskov, J. K. (2020). Analysis of Acid-Stable and Active Oxides for the Oxygen Evolution Reaction. *ACS Energy Letters*, *5*(12), 3778–3787. https://doi.org/10.1021/acsenergylett.0c02030



Conclusion

- We have created a database of ML predicted **TOTAL DFT** energies for bare and adsorbed surfaces of oxides for OER
- Doing so allows us to perform complex surface analysis that typically requires enormous amounts of DFT calculations:
  - Prediction of **overpotential**
  - Prediction of **Wulff shapes**
  - Prediction of **nanoscale stability**
- The available of such analysis without the need of DFT allows us to construct complex screening frameworks for identifying oxides for OER
- **Identified 81 viable candidates for OER, with 40 additional candidates when considering nanoscale stabilization**



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Shell

UH ENERGY TRANSITION INSTITUTE

Supercomputing resources:

UNIVERSITY OF HOUSTON

Research Computing Data Core

Hewlett Packard Enterprise Data Science Institute

UH MODAL LAB

Multidisciplinary Modeling, Oilfield Data Analytics, and Well Logging Laboratory

The Computational Catalysis and Interface Chemistry Group

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