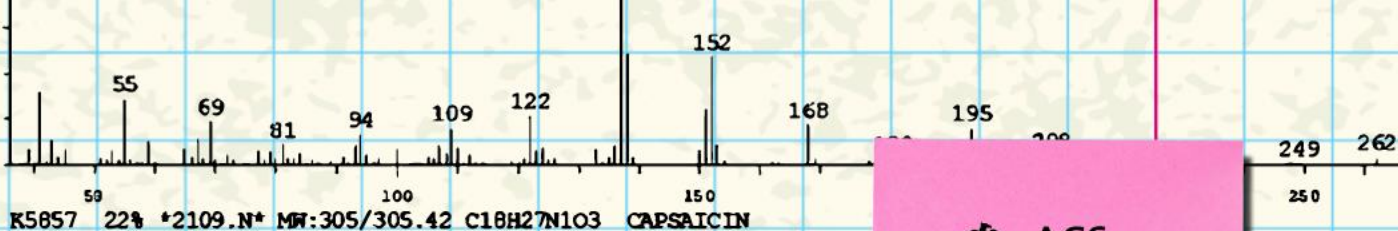


ACS SPRING 2024

MARCH 17-21
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Chemistry



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

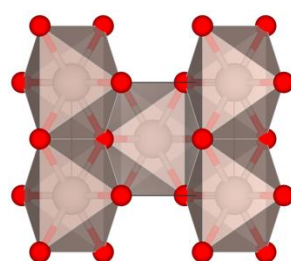
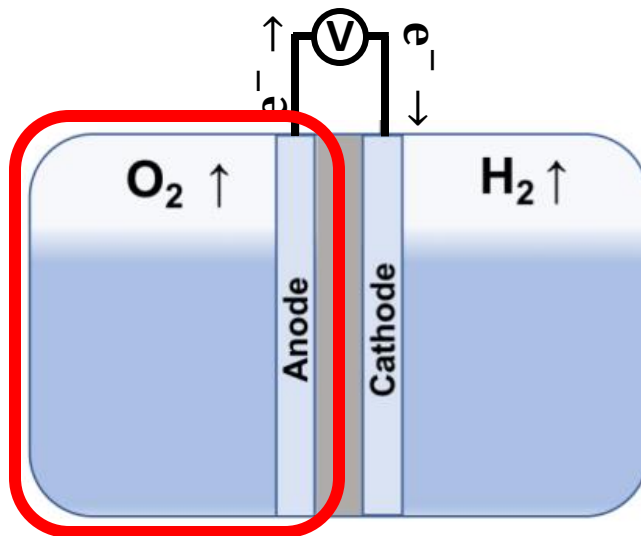
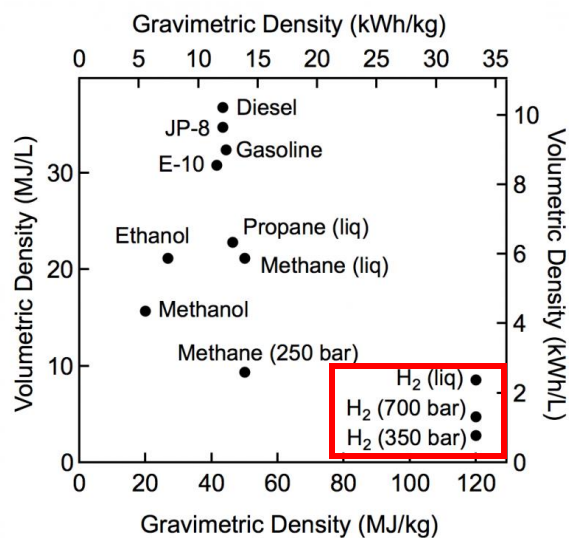
Richard Tran

3990961

rtran25@cougarnet.uh.edu



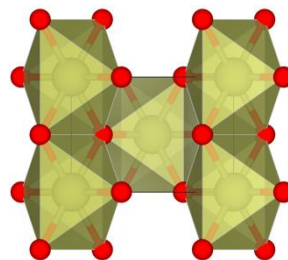
Harnessing the power of water



RuO₂

\$18,315/kg

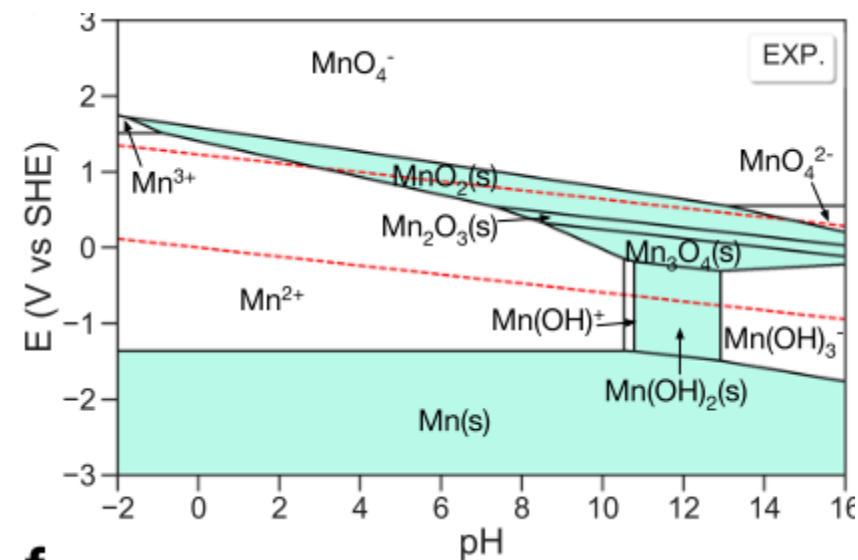
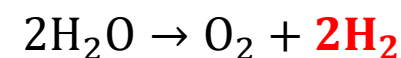
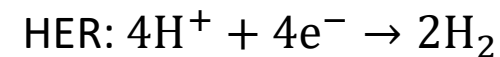
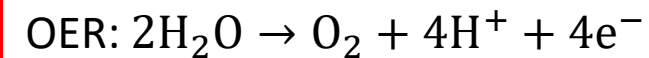
Ru: \$24,113/kg



IrO₂

\$155,727/kg

Ir: \$181,651/kg



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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>

Open Catalyst 2022 (OC22) Dataset

Contains:

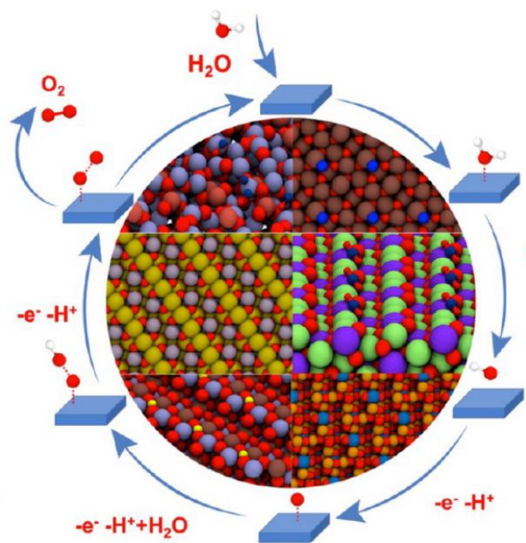
Adsorbate coverage

O, H, N, C, OH, OOH, H₂O, CO, O₂

Spin polarization

Vacancy defects

Binary oxides



Applications:



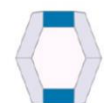
Water splitting, fuel cells



Batteries



H₂ production



Equilibrium nanoparticle shape

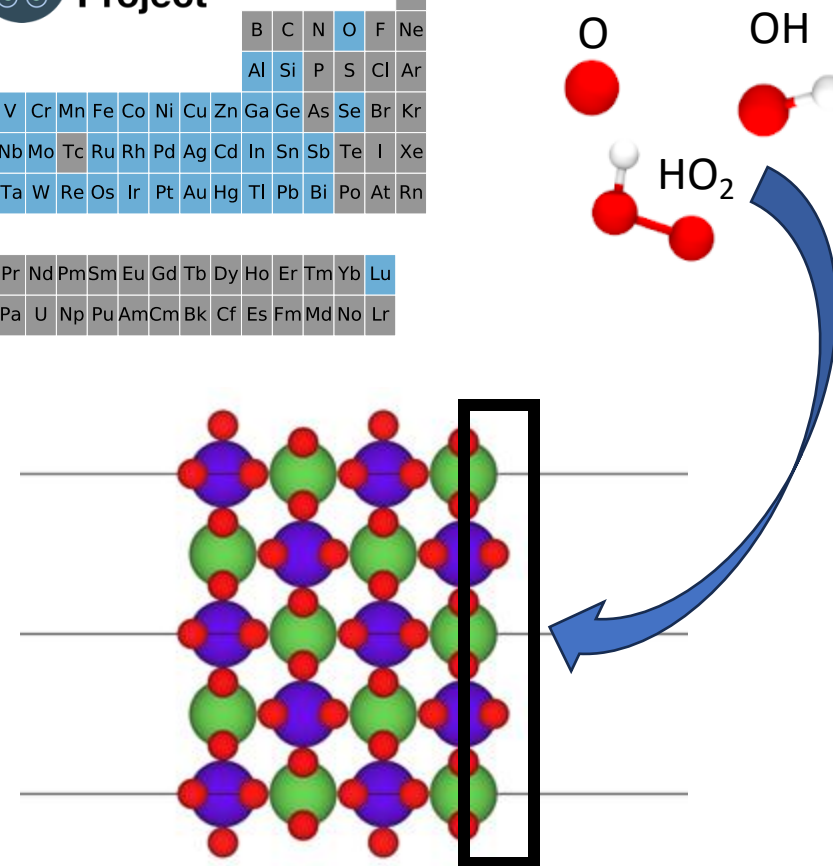
 **Meta AI**
Fundamental AI
Research (FAIR)





The Materials Project

H																	He
Li	Be											B	C	N	O	F	Ne
Na	Mg											Al	Si	P	S	Cl	Ar
K	Ca	Sc	Ti	V	Cr	Mn	Fe	Co	Ni	Cu	Zn	Ga	Ge	As	Se	Br	Kr
Rb	Sr	Y	Zr	Nb	Mo	Tc	Ru	Rh	Pd	Ag	Cd	In	Sn	Sb	Te	I	Xe
Cs	Ba		Hf	Ta	W	Re	Os	Ir	Pt	Au	Hg	Tl	Pb	Bi	Po	At	Rn
Fr	Ra																
		La	Ce	Pr	Nd	Pm	Sm	Eu	Gd	Tb	Dy	Ho	Er	Tm	Yb	Lu	
		Ac	Th	Pa	U	Np	Pu	Am	Cm	Bk	Cf	Es	Fm	Md	No	Lr	

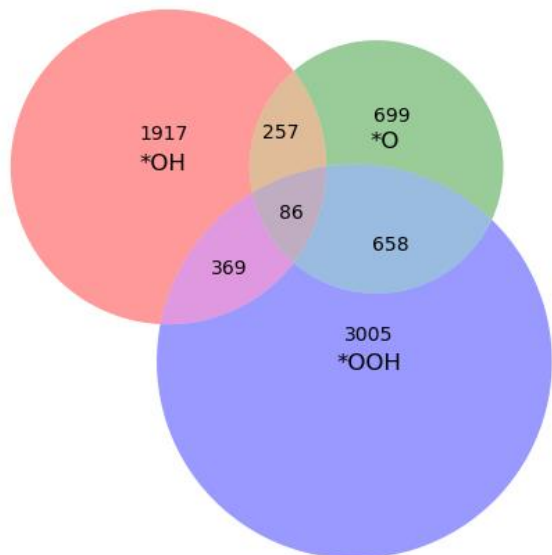


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Database scope



OC22 DFT dataset

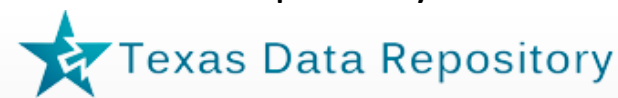
# Predictions		6,068,572
# Materials		4,119
# Slab predictions		191,902
Ave. # slabs per material		47
# Adsorption predictions		5,876,670
Max Miller index		1
OH*	O*	OOH*
1,972,166	667,266	3,237,238

OC22 prediction dataset

We can substitute ΔG^{OOH*} with:

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

All data available at UH
 Dataverse Repository under:



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

```
entry_id: "mp-753534"
database: "MP"
bulk_reduced_formula: "HfCoO3"
slab_id: "s_lab-o4uIeH1f5wcT1qFKuMnq"
> miller_index: [-]
bulk_formula: "Hf4 Co4 O12"
> bulk_composition: [-]
bulk_chemsys: "Co-Hf-O"
bulk_energy: -185.38058648
func: "GenNet-OC"
> slabs:
  > adslab-NznP0zQDSz0LCVz17Erq: [-]
  > adslab-RrVAjRly8w65ZXWcx73C: [-]
  > adslab-wJLoAyA5w1ar3fMeZykz: [-]
  > adslab-6JyuSuW3LUMbW88K6bA: [-]
  > adslab-0b6QY9AcPefJ3RYmVCIB: [-]
  > adslab-IMLYD6Bc7cGGzx34S0aD: [-]
  > adslab-MSJLD0BPmgz0QVFkbIKI: [-]
  > adslab-MU2XsNGMZJGUaj7dDW68: [-]
  > adslab-Jd80pXF2U99Zx0ELI1TM: [-]
  > adslab-DkFLZ0KVZlwI1TmWMn2: [-]
  > adslab-jIsF6yGMVpKFPPTtyMed6: [-]
  > adslab-SdKZ7CIRzq42u2L3XfH: [-]
  > adslab-61paQtINm7d4tApHmkCF: [-]
  > adslab-LstM3uGvILBNYDQ16oqs: [-]
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  > adslab-nsjhcFKyoZDe6WNIAT0t: [-]
  > adslab-0maA4WtMUGUEuQXZ6osj: [-]
  > adslab-775FdeuW8ms0gJNyztU: [-]
  > adslab-R14630wvqteG18JX4DC2: [-]
  > adslab-4JmWSzaanLl1ZgV39Vh: [-]
  > adslab-PBht98wCNWOCr05bZoY: [-]
  > adslab-z1Rdw5IBFdl0Lb01PrV9: [-]
  > slab-o4uIeH1f5wcT1qFKuMnq:
```

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> cell: [-]
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> natoms: 121
> tags: [-]
> fixed: [-]
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entry_id: "mp-753534"
database: "MP"
adsorbate: "OOH"
slab_id: "s_lab-o4uIeH1f5wcT1qFKuMnq"
rid: "ads_lab-4JmWSzaanLl1ZgV39Vh"
> miller_index: [-]
bulk_reduced_formula: "HfCoO3"
bulk_formula: "Hf4 Co4 O12"
> bulk_composition: [-]
bulk_chemsys: "Co-Hf-O"
bulk_energy: -185.38058648
calc_type: "adsorbed_slab"
func: "GenNet-OC"
nads: 1
y: -972.009194423662
unrelax_energy: -966.3862915039862
> pos_relaxed: [-]
max_force: 0.03795161843299866
> site_properties: [-]
pmg_init_slab_fcoords: [-]
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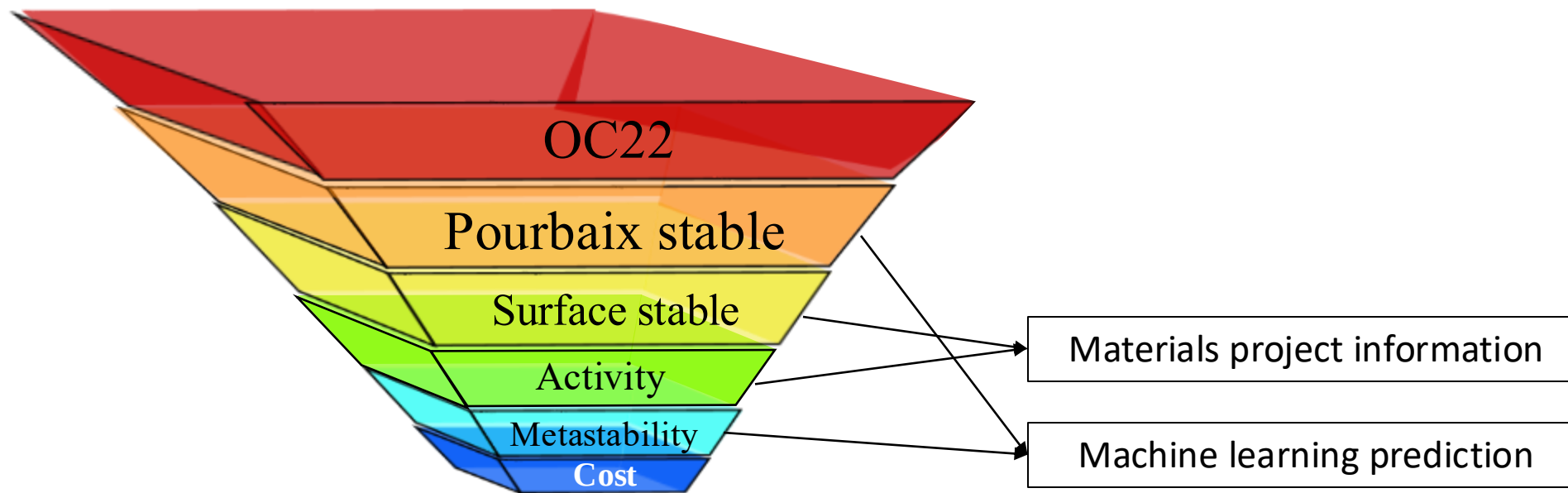
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bulk_formula: "Hf4 Co4 O12"
> bulk_composition: [-]
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bulk_energy: -185.38058648
calc_type: "bare_slab"
func: "GenNet-OC"
y: -961.257409919474
unrelax_energy: -952.6406860351562
> pos_relaxed: [-]
max_force: 0.03443406954803047
> site_properties: [-]
pmg_init_slab_fcoords: [-]
```



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High throughput screening



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OC22 dataset

OC22
(4,119)

H																	He
Li	Be											B	C	N	O	F	Ne
Na	Mg											Al	Si	P	S	Cl	Ar
K	Ca	Sc	Ti	V	Cr	Mn	Fe	Co	Ni	Cu	Zn	Ga	Ge	As	Se	Br	Kr
Rb	Sr	Y	Zr	Nb	Mo	Tc	Ru	Rh	Pd	Ag	Cd	In	Sn	Sb	Te	I	Xe
Cs	Ba		Hf	Ta	W	Re	Os	Ir	Pt	Au	Hg	Tl	Pb	Bi	Po	At	Rn
Fr	Ra																
		La	Ce	Pr	Nd	Pm	Sm	Eu	Gd	Tb	Dy	Ho	Er	Tm	Yb	Lu	
		Ac	Th	Pa	U	Np	Pu	Am	Cm	Bk	Cf	Es	Fm	Md	No	Lr	

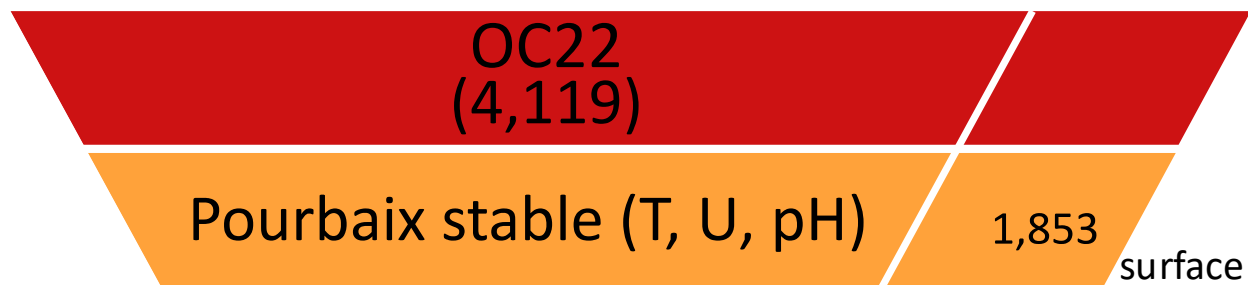
- Top 5 lowest E_{hull}
- Max # of atoms in bulk: 150
- 1720 bulks with U-values
- Unary bulks: 318
- Binary bulks: 4,414
- Total bulks: 4,732



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Pourbaix stability



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

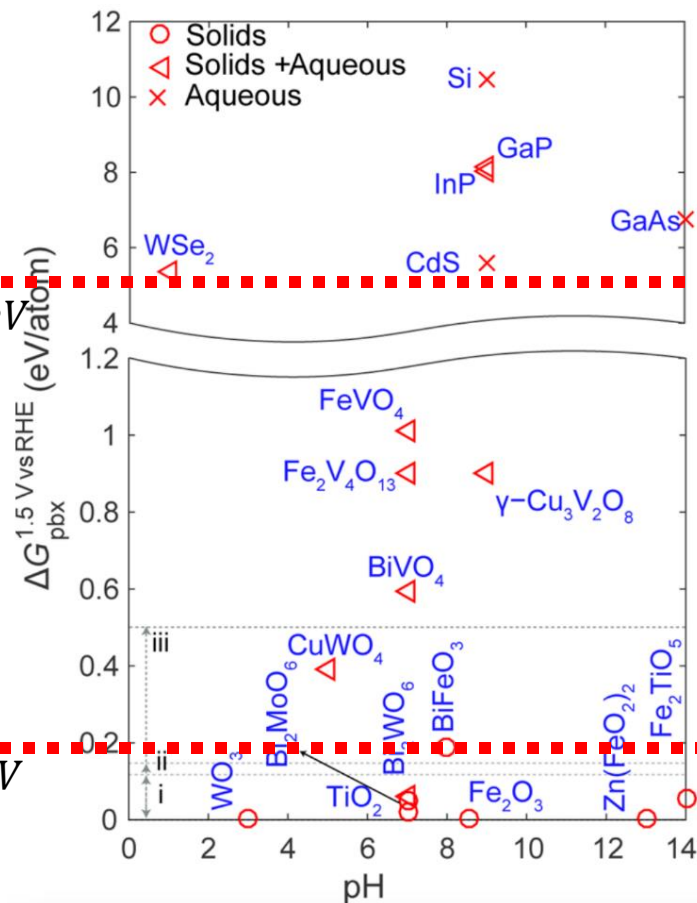
$$\Delta G_{PBX}(\text{pH} = 1, U = 1.8, T = 80^\circ\text{C}) < 0.5 \text{ eV}$$

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>

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>

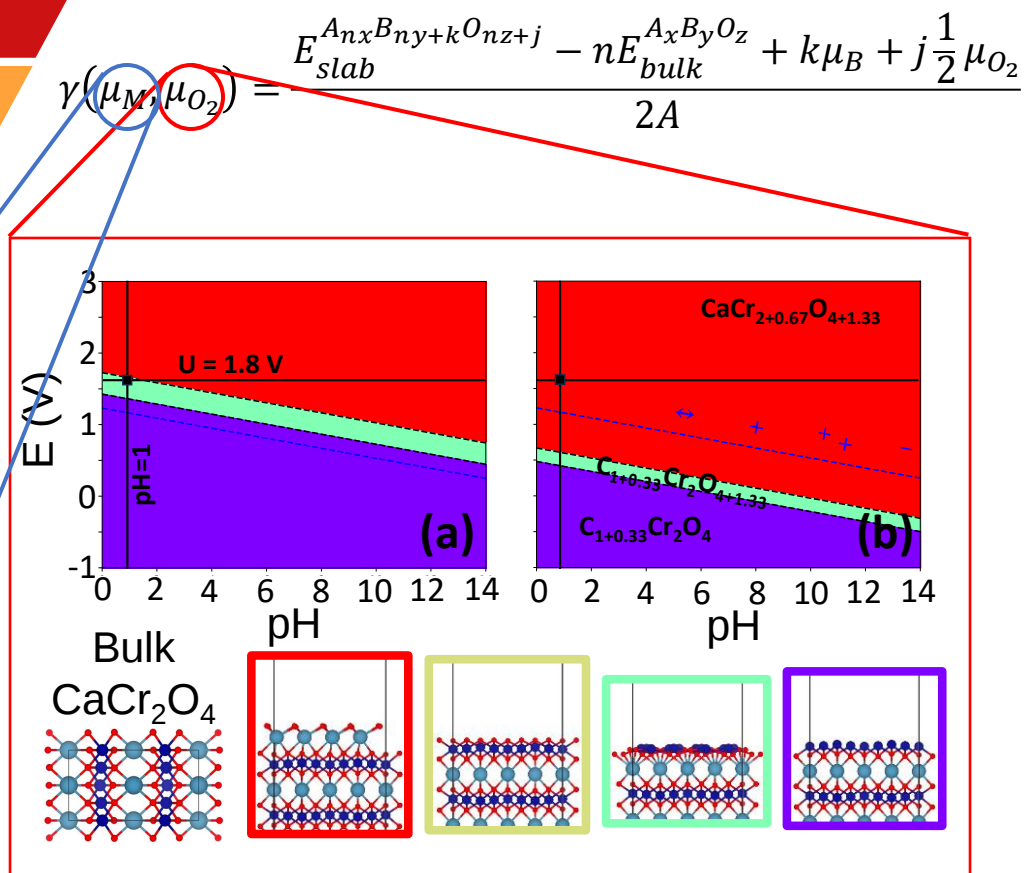
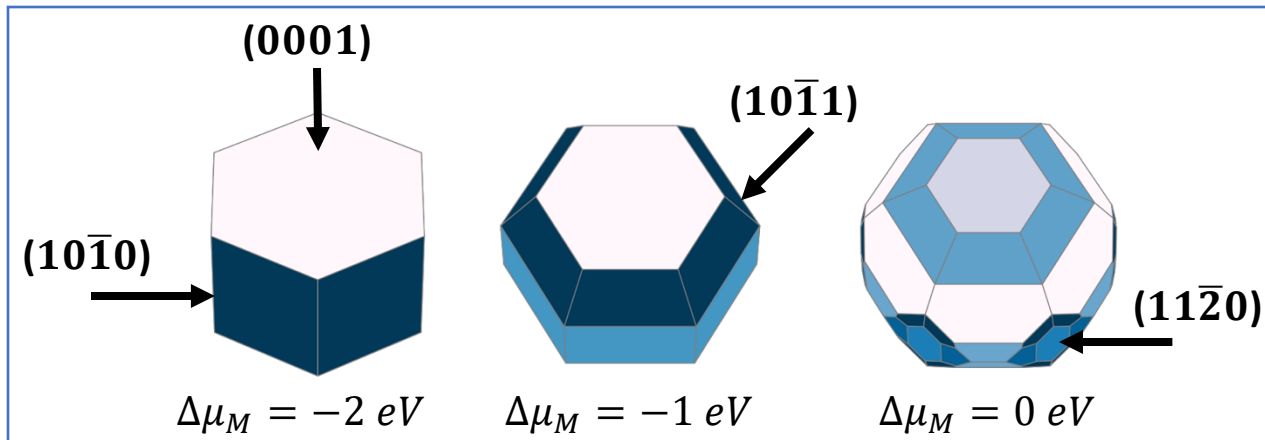
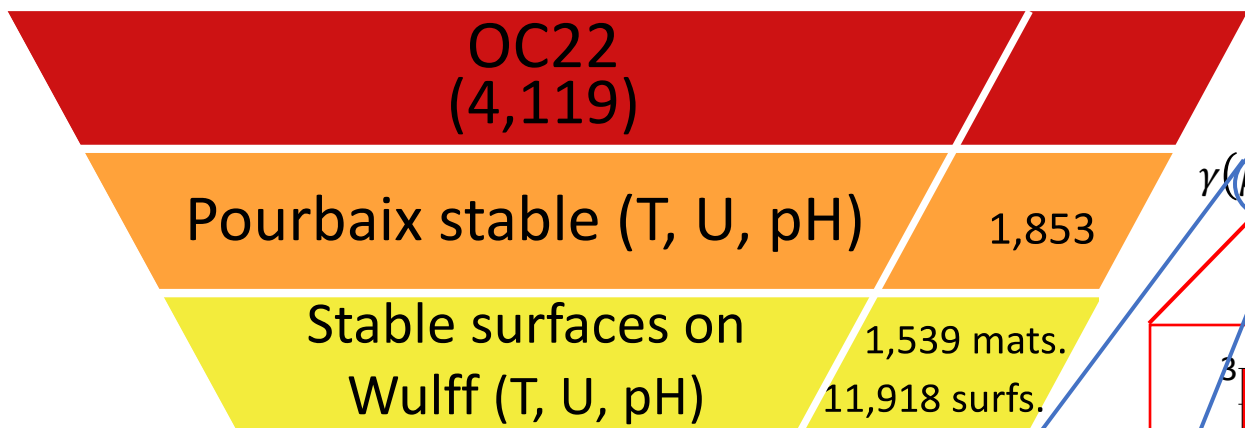
metastability
 $\Delta G_{PBX} < 0.2 \text{ eV}$



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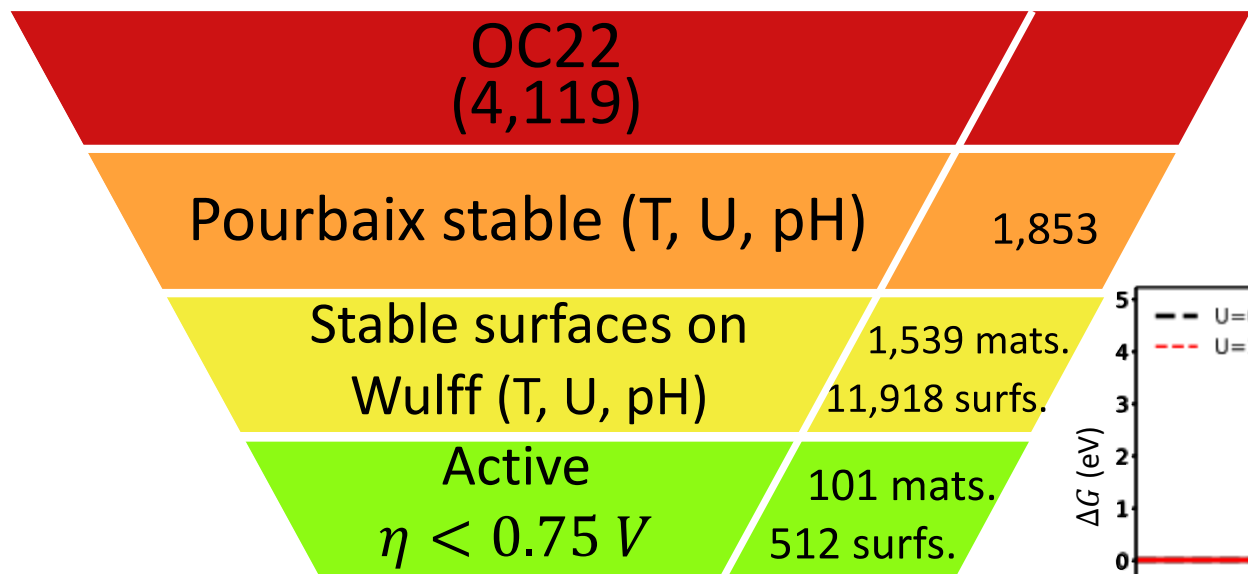
Surface stability



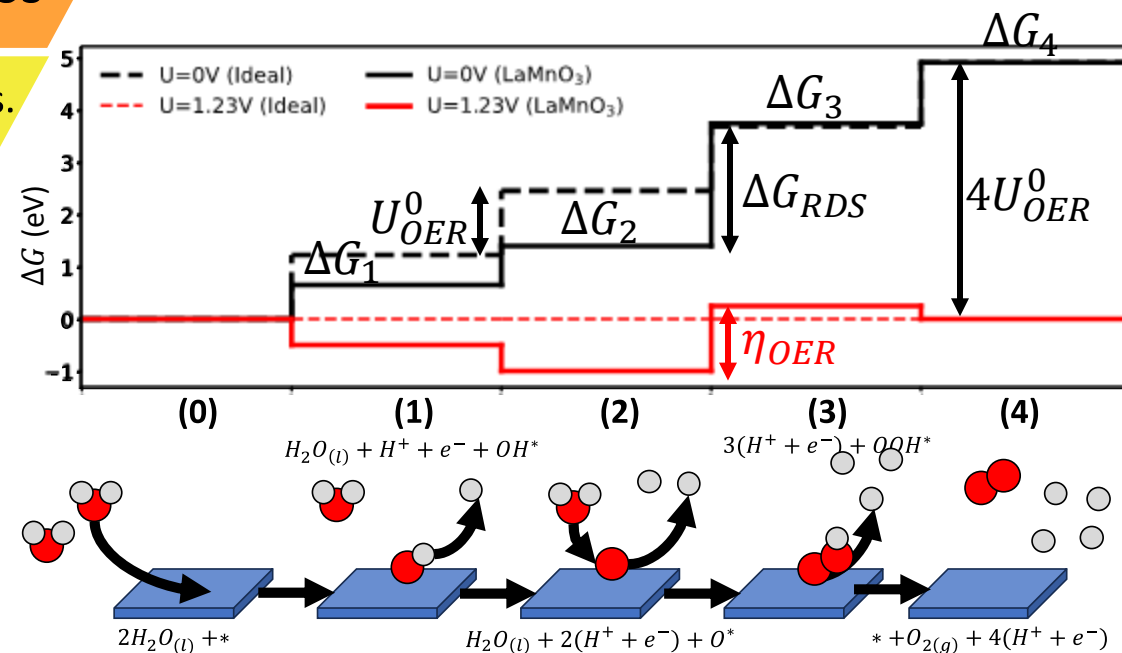
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OER activity (overpotential)



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>



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

$$\text{Overpotential: } \eta_{OER} = \Delta G_{RDS} - U_{OER}^0$$

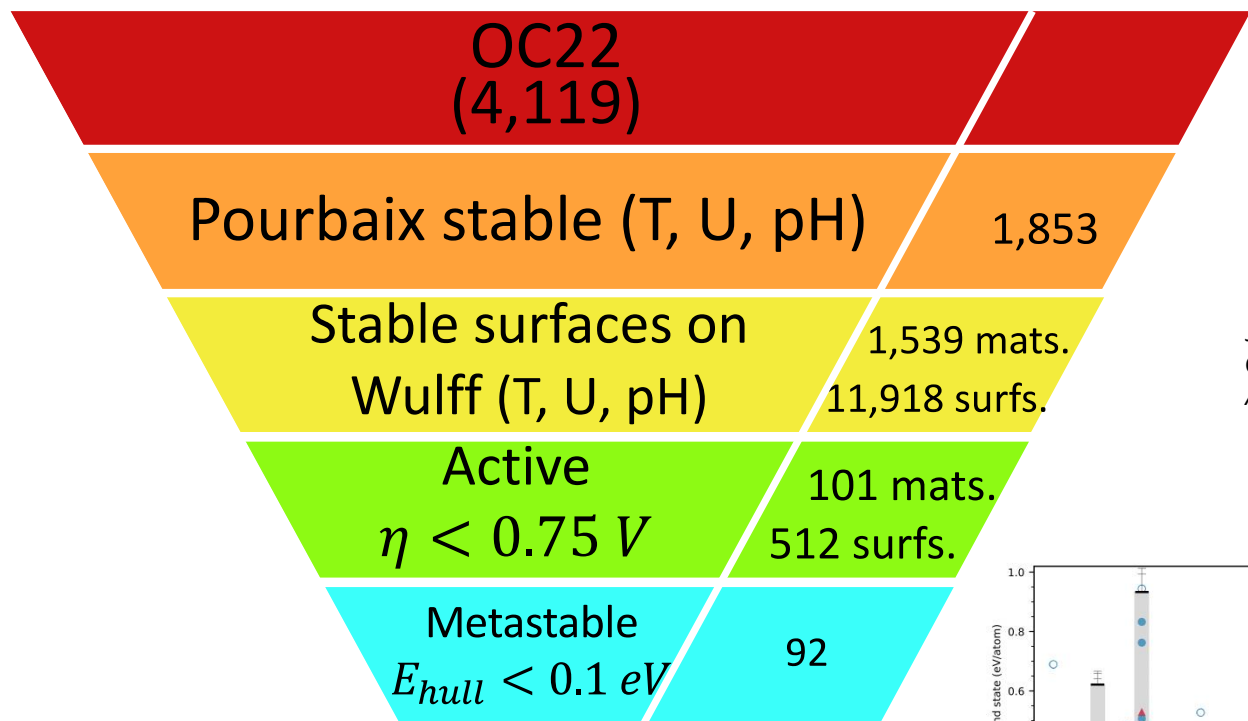
$$U_{OER}^0 = 1.23 V \text{ vs SHE}$$



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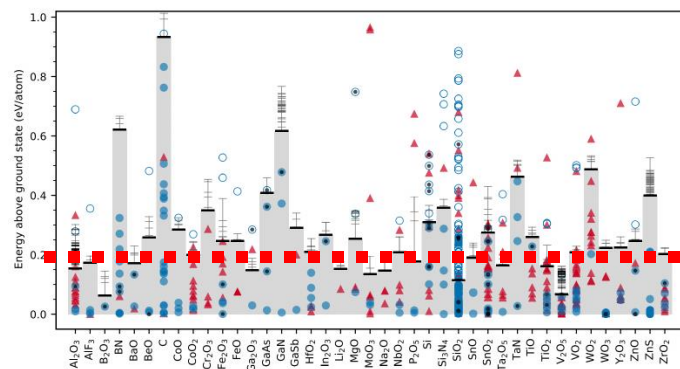
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Metastability



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.aag0148

Metastability limits:

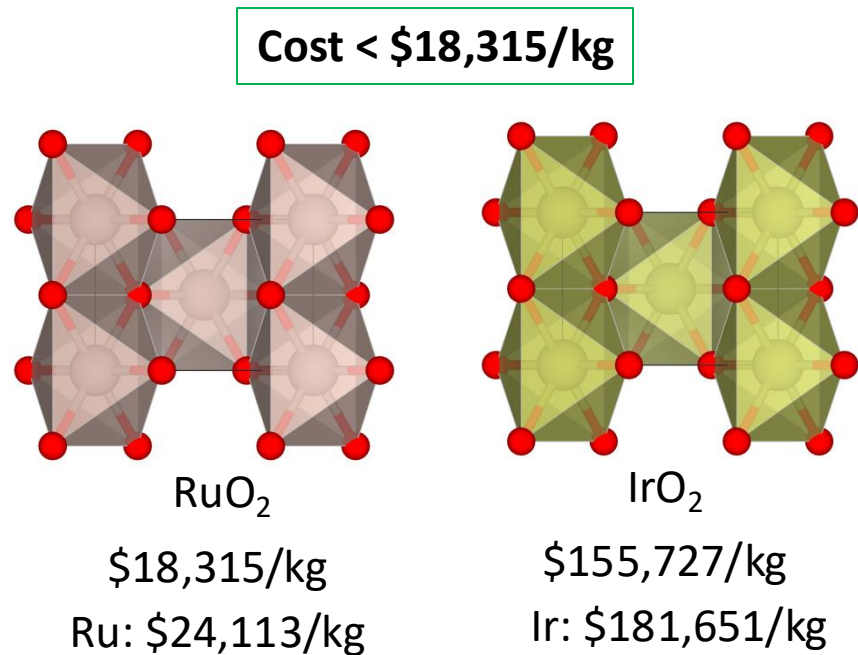
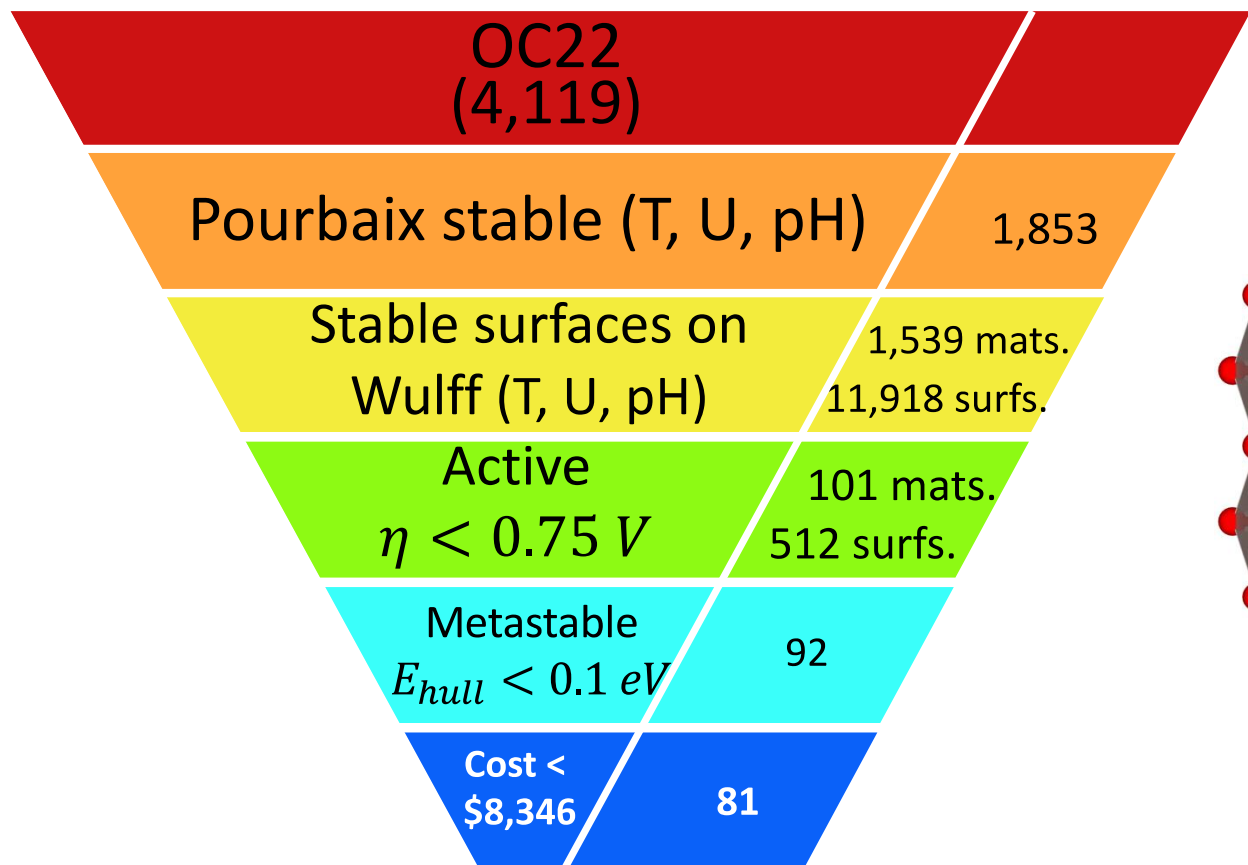
$$E_{hull} < 0.2 eV$$



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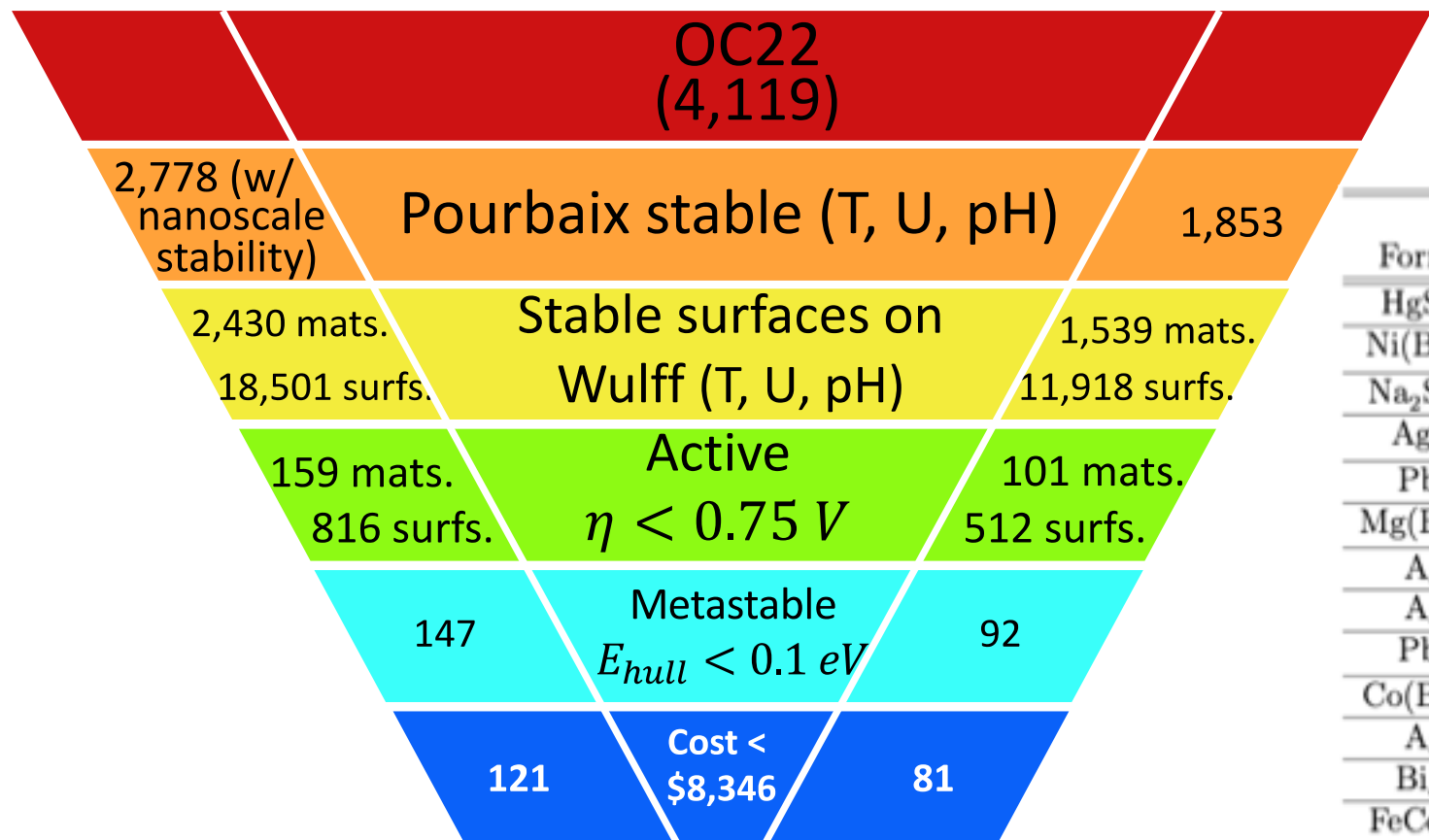
Material cost



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Final candidates



Formula	Space group	# facets	η (V)	E_{PBX} (eV)	E_{hull} (eV)	Cost (\$/kg)
HgSeO ₄	<i>Pmn2₁</i>	2	0.18	0.00	0.00	65.47
Ni(BiO ₃) ₂	<i>P4₂/mnm</i>	4	0.36	0.00	0.00	20.88
Na ₂ Se ₂ O ₇	<i>P1</i>	2	0.21	0.00	0.00	110.43
Ag ₃ O ₄	<i>P2₁/c</i>	4	0.33	0.00	0.00	714.77
PbO ₂	<i>P4₂/mnm</i>	2	0.33	0.00	0.00	2.41
Mg(BiO ₃) ₂	<i>P4₂/mnm</i>	3	0.52	0.00	0.00	20.41
AgO	<i>Cccm</i>	2	0.49	0.00	0.00	745.41
AgO	<i>C2/c</i>	2	0.51	0.01	0.01	745.41
PbO ₂	<i>Pbcn</i>	2	0.56	0.01	0.01	2.41
Co(BiO ₃) ₂	<i>P4₂/mnm</i>	2	0.33	0.02	0.02	24.34
AgO	<i>P2₁/c</i>	3	0.50	0.04	0.04	745.41
Bi ₄ O ₇	<i>P1</i>	2	0.22	0.04	0.00	22.67
FeCo ₉ O ₂₀	<i>P1</i>	4	0.41	0.06	0.07	31.63

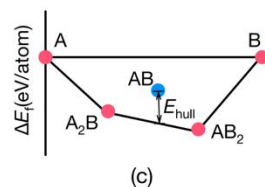


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A photograph of a single, vibrant red bell pepper resting on a dark wooden surface. The pepper is glossy and has a green stem. Overlaid on the left side of the pepper is a chemical structure of a pyrimidine derivative, specifically 2-isopropoxy-4-methylpyrimidine, which is a component of the 'M' in the MMR acronym.

1,853

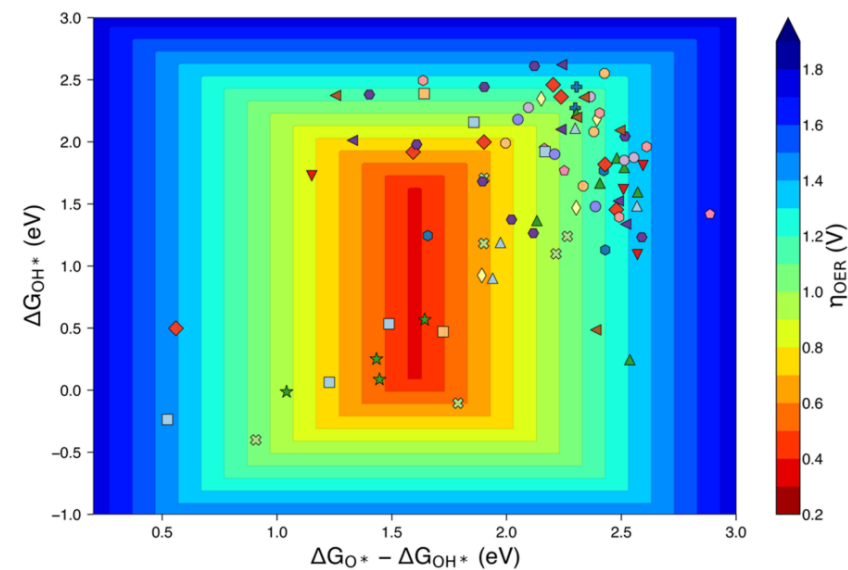


Li 1	Be	Acid-Stable Periodic Table										B	C	N
Na 1	Mg 2											Al	Si	P
K 2	Ca 1	Sc 1	Ti 16	V	Cr	Mn 4	Fe 11	Co 2	Ni 1	Cu	Zn 2	Ga 1	Ge 11	As
Rb	Sr 2	Y	Zr	Nb	Mo 11	Tc	Ru	Rh	Pd	Ag 1	Cd 1	In	Sn 16	Sb 38
Cs 1	Ba 1		Hf	Ta	W 8	Re	Os	Ir	Pt	Au	Hg 2	Tl	Pb 1	Bi 1

68/47,814 stable candidates!

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/acsenenergylett.0c01625>

 $\text{Fe}(\text{SbO}_3)_2$	 FeSbO_4	 MoWO_4	 FeSbO_4	 $\text{Ti}(\text{WO}_3)_2$
 $\text{Co}(\text{SbO}_3)_2$	 GaSbO_4	 $\text{Ti}(\text{GeO}_3)_2$	 $\text{Fe}(\text{SbO}_3)_4$	 TiFeSbO_6
 BiSbO_3	 FeSbO_4	 MoWO_4	 CoSbO_4	 IrO_2
 $\text{Ni}(\text{SbO}_3)_2$	 $\text{FeAg}(\text{MoO}_4)_2$	 TiSnO_3	 $\text{Sn}(\text{WO}_3)_2$	

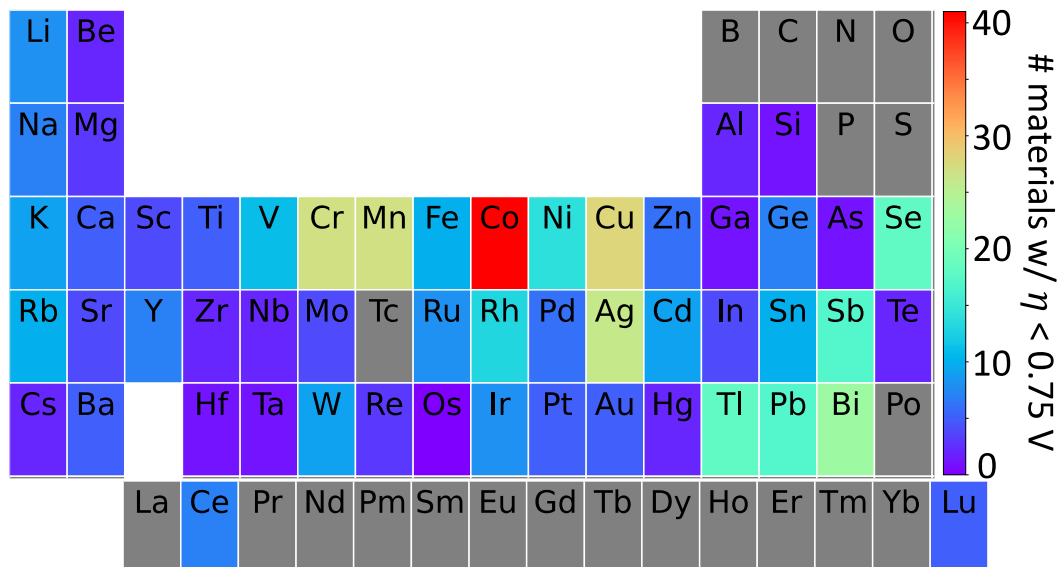


17 candidates w/ $\eta_{\text{OER}} < 0.8$ eV

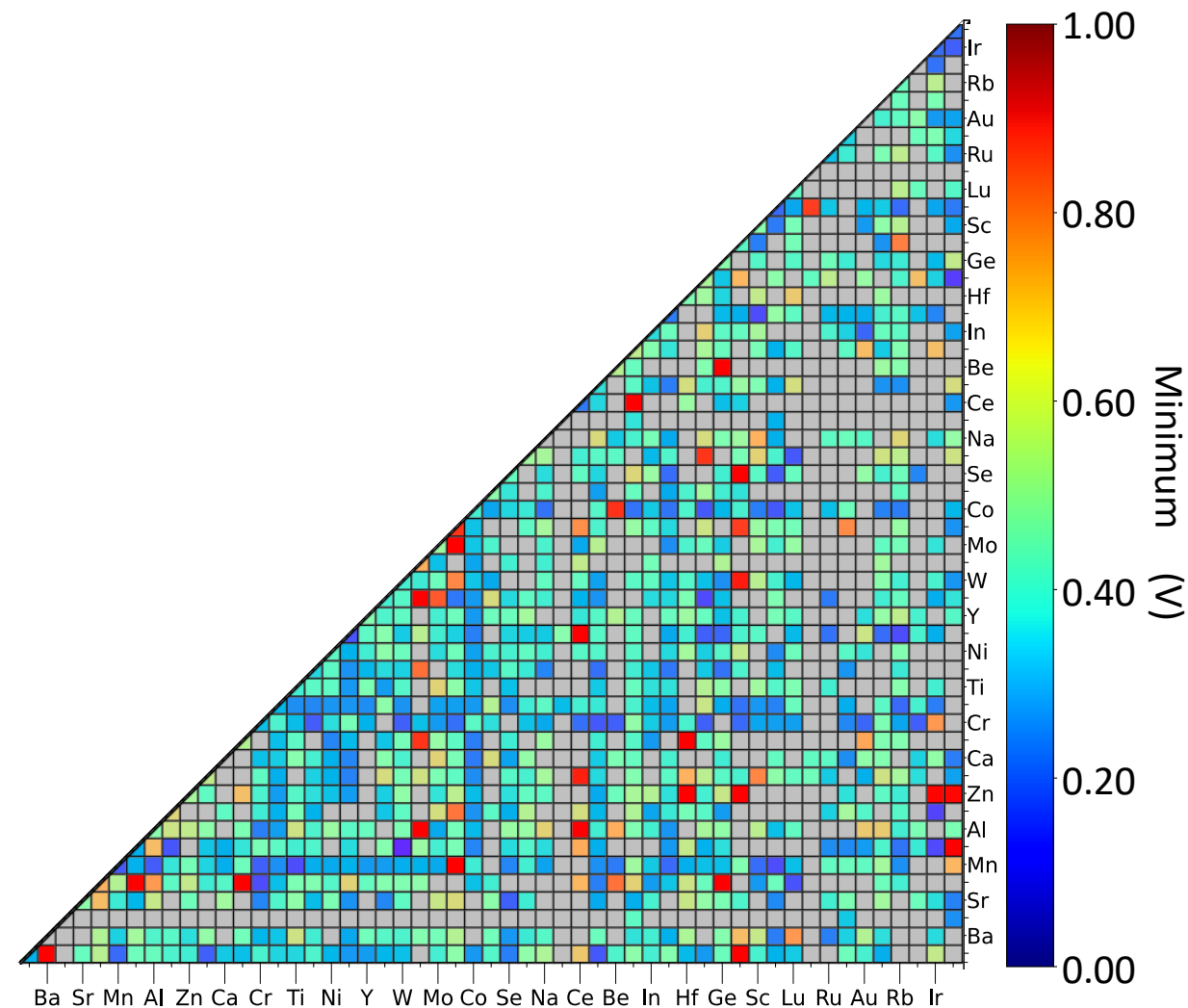
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Overpotential assessment



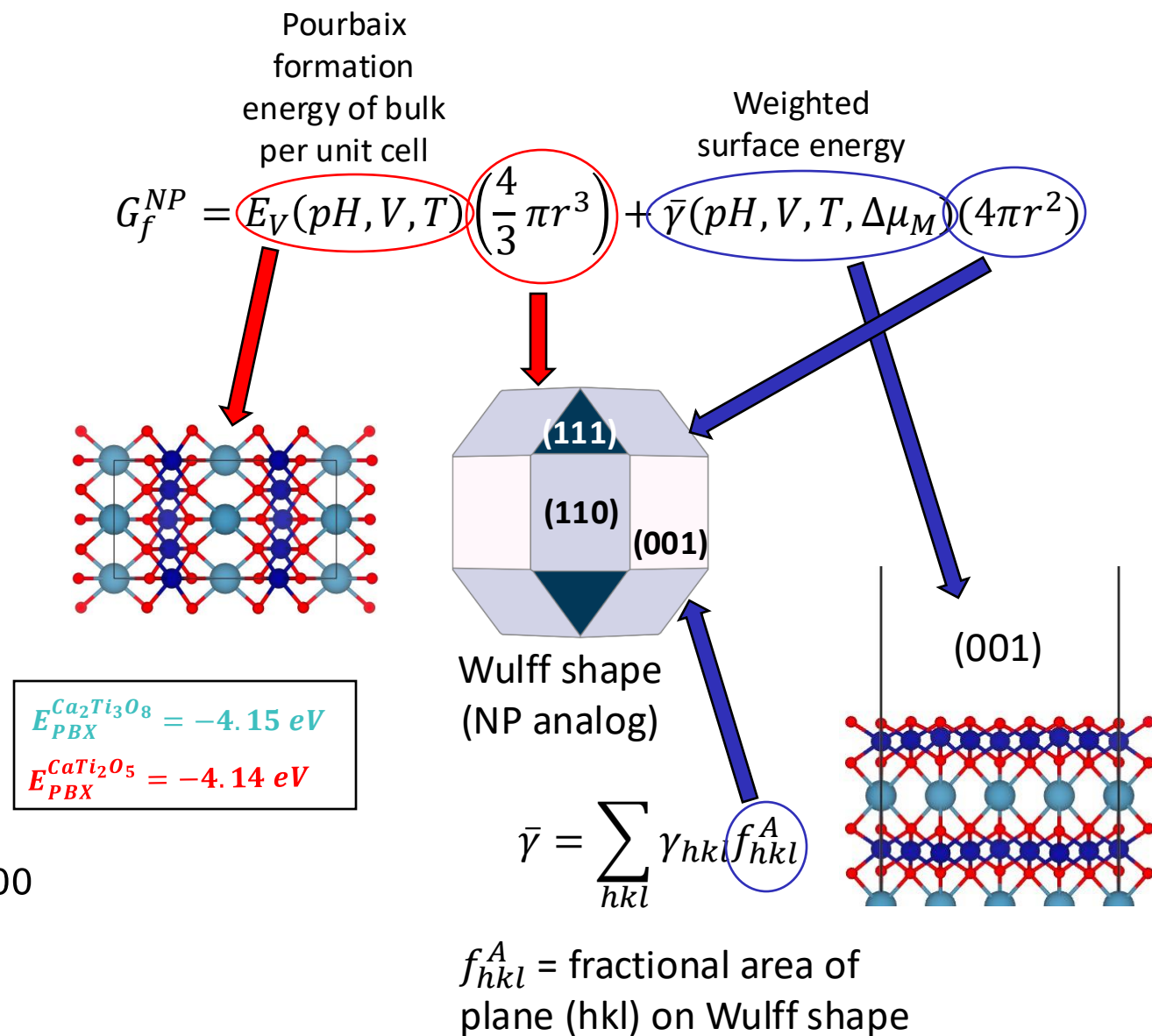
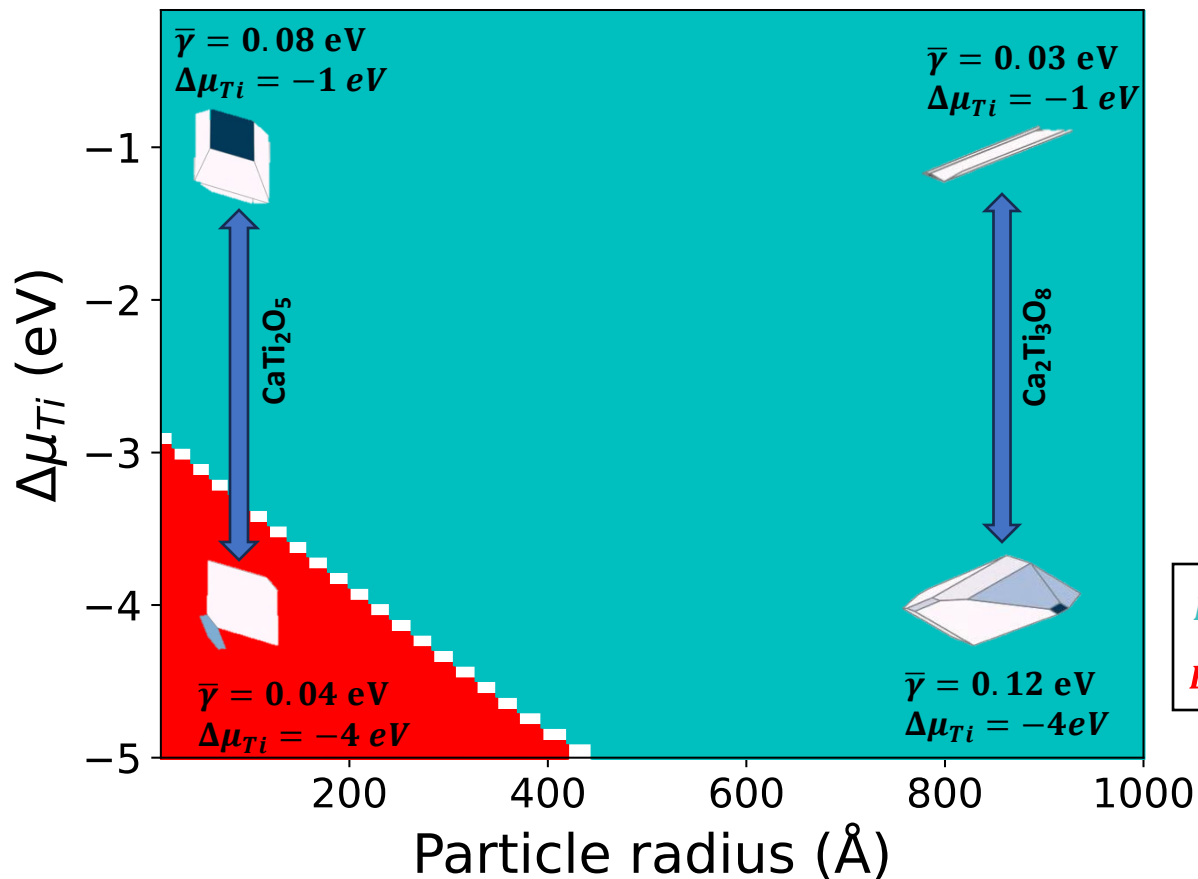
Are there other oxides with low overpotential that are unstable? Is there a way to synthetically access these candidates?



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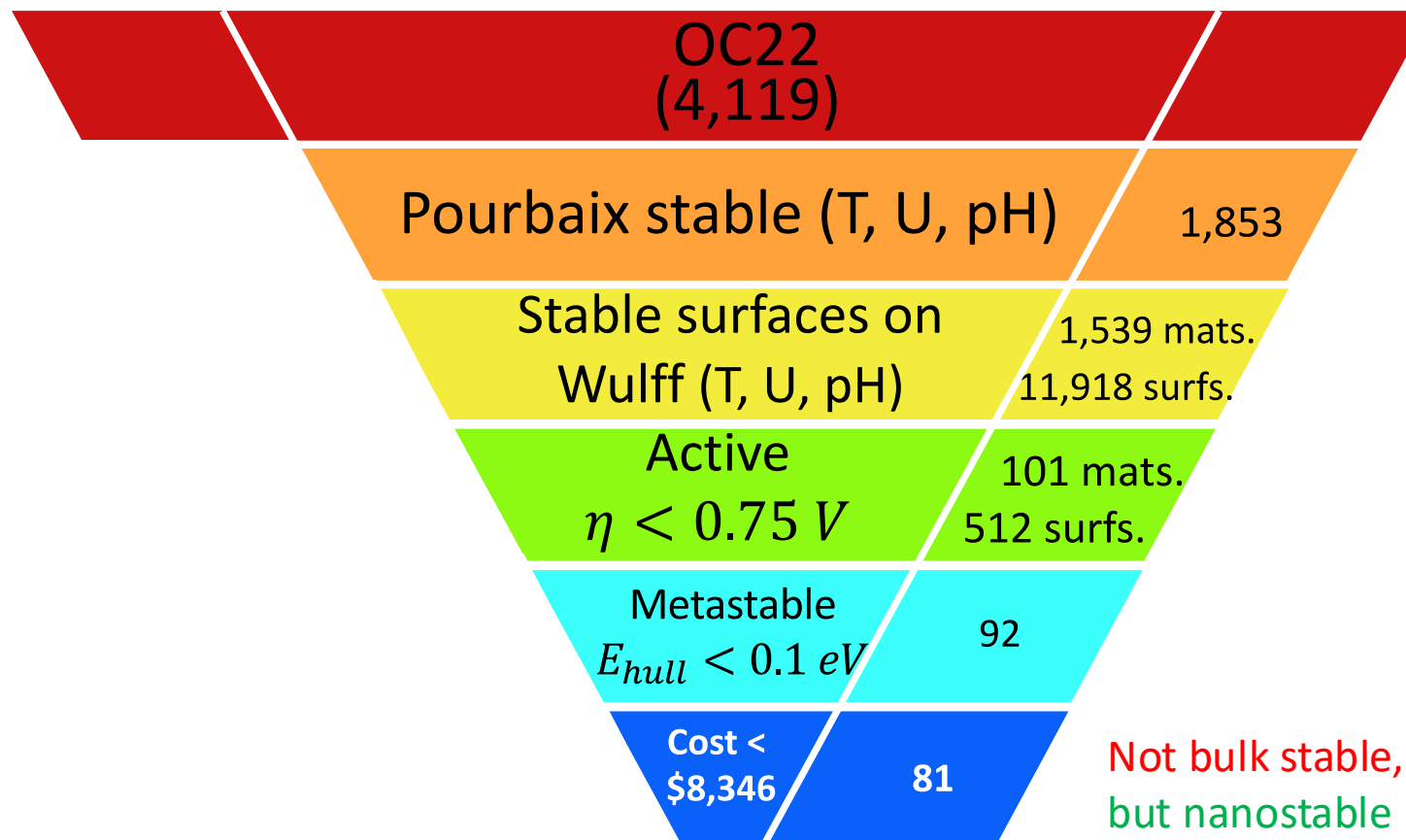
Nanoscale stabilization



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Revised screening mechanism



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
$\text{Y}(\text{FeO}_2)_2$	$P\bar{1}$	2	0.53	0.54	0.01	11.23
$\text{Sr}_2\text{Ti}_2\text{O}_5$	$P2_1/c$	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	$Pm\bar{m}n$	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
$\text{Ba}_2\text{Ti}_2\text{O}_5$	$Pnma$	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

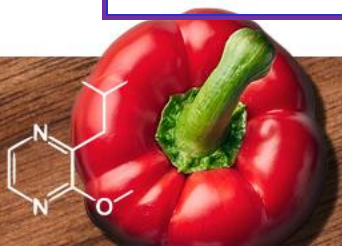
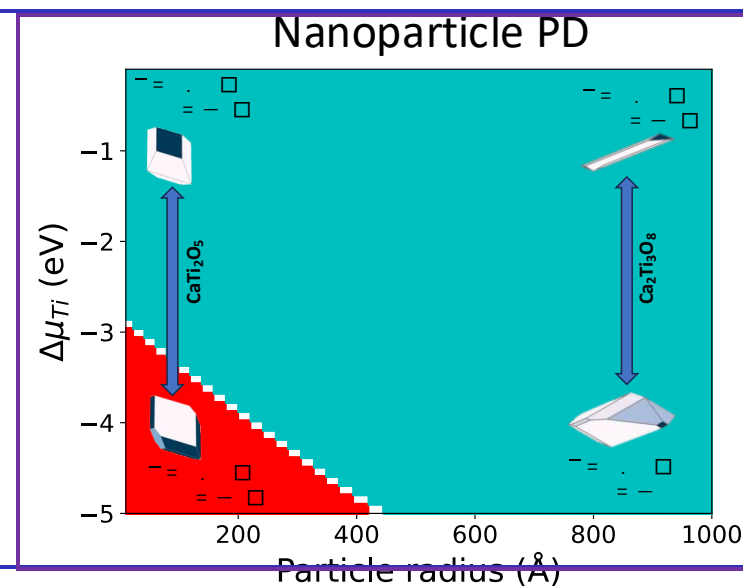
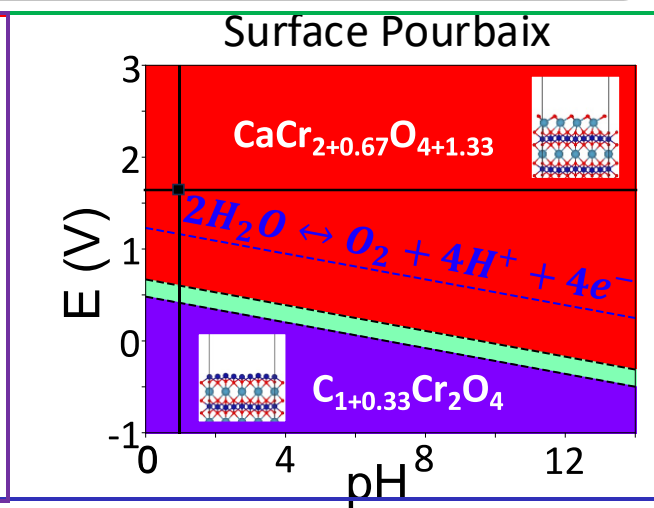
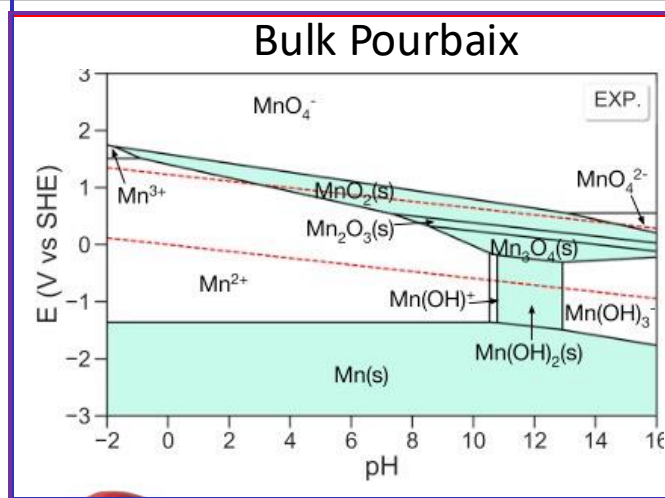


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Alternate screening mechanisms

Temperature ($^{\circ}\text{C}$)	60	60	80	80
Applied Potential (V)	1.8	1.2 to 2.0	1.8	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

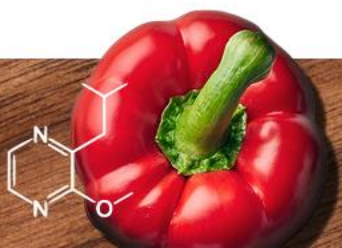
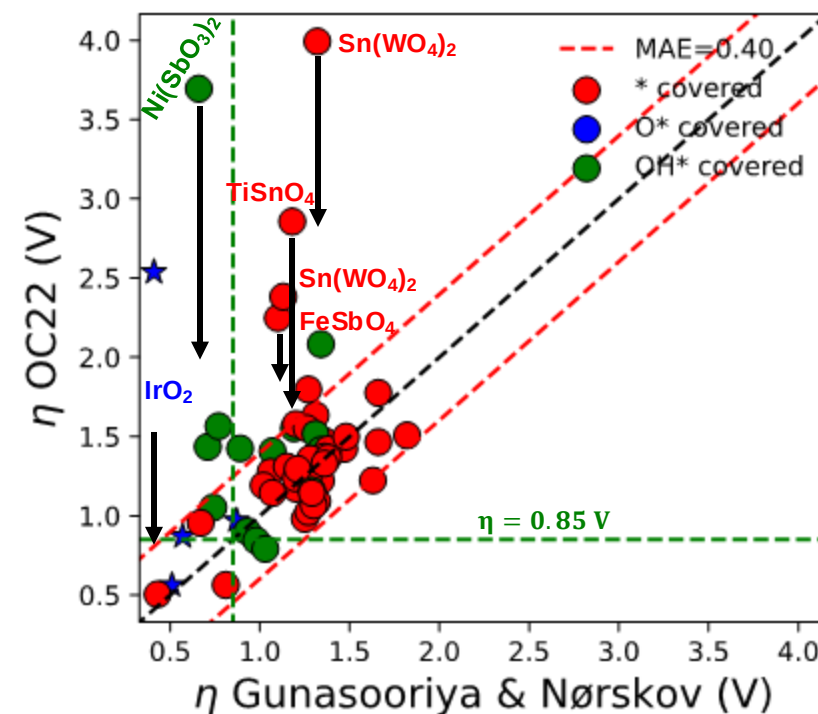
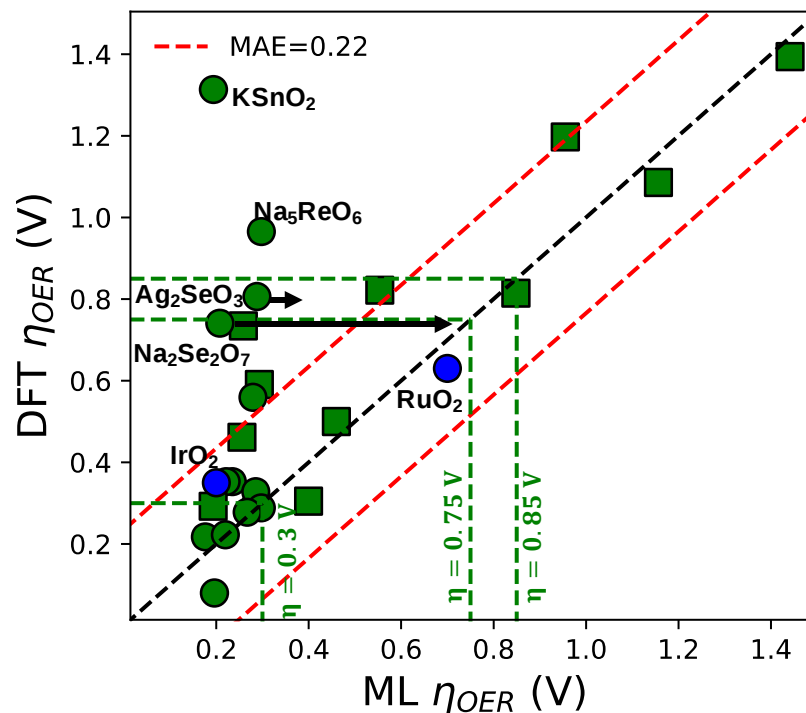
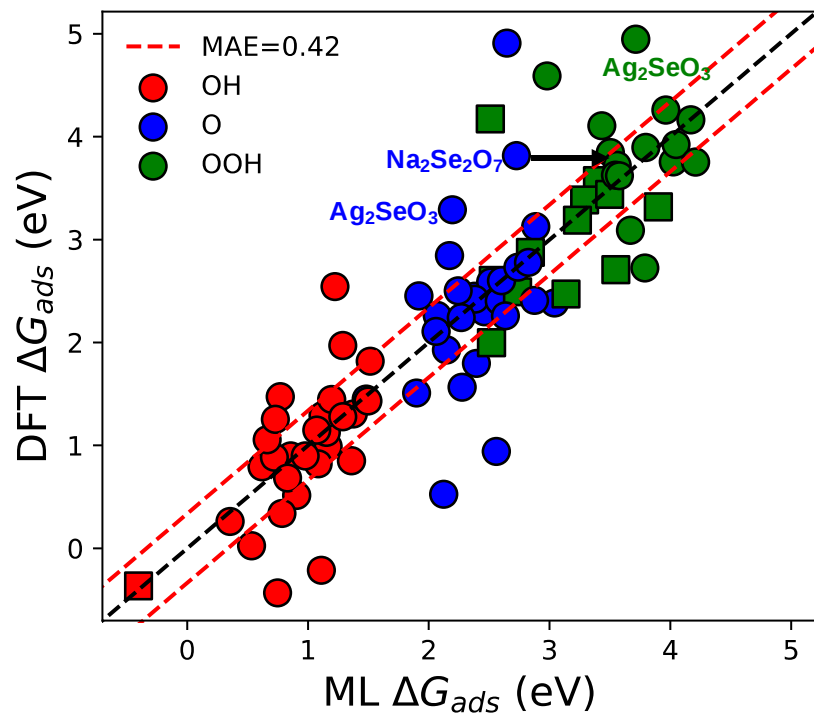


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Validation

Gunasekariya, 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/acseenergylett.0c02030>

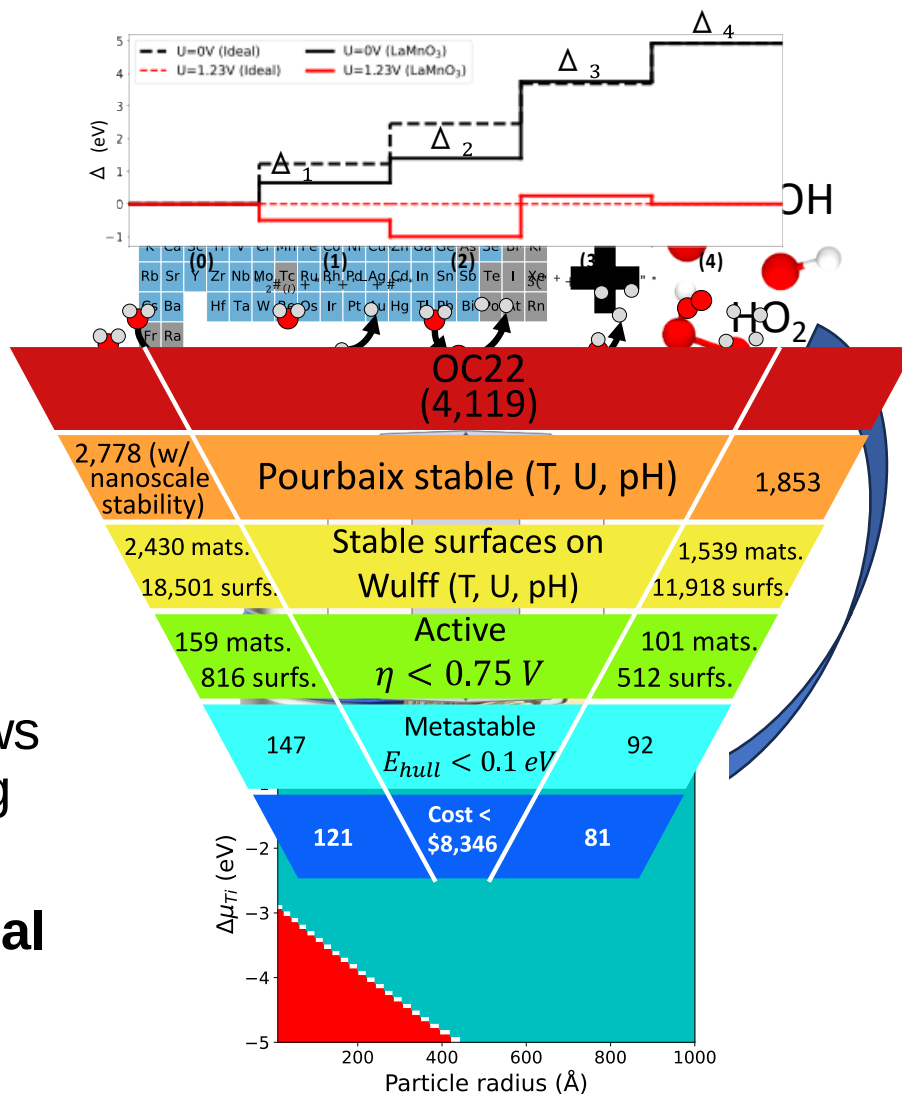


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Conclusion

- 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 availability 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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Research Computing Data Core
Hewlett Packard Enterprise Data Science Institute



UH MODAL LAB

Multidisciplinary Modeling, Oilfield Data Analytics, and Well Logging Laboratory



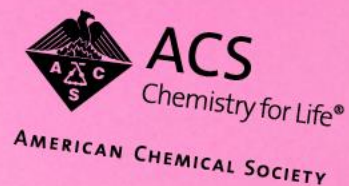
The Computational Catalysis and Interface Chemistry Group



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REPORT INSTANCES OF HARASSMENT

Contact ACS Secretary and
General Counsel Flint Lewis at
f_lewis@acs.org or call the
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