Supplementary Information of Application of machine learning to discover new intermetallic catalysts for the hydrogen evolution and the oxygen reduction reactions

Carmen Martínez-Alonso[1,2,3], Valentin Vassilev-Galindo[1], Benjamin M. Comer[3], Frank Abild-Pedersen[3], Kirsten T. Winther[3], and Javier LLorca*[1,4]

[1]IMDEA Materials Institute, C/Eric Kandel 2, 28906 - Getafe, Madrid, Spain.
[2]Department of Inorganic Chemistry, Complutense University of Madrid, 28040 Madrid, Spain.
[3]SUNCAT Center for Interface Science and Catalysis, Department of Chemical Engineering, Stanford University, Stanford, California 94305, United States; SUNCAT Center for Interface Science and Catalysis, SLAC National Accelerator Laboratory, Menlo Park, California 94025, United States.
[4]Department of Materials Science, Polytechnic University of Madrid, E. T. S. de Ingenieros de Caminos, 28040 Madrid, Spain.

May 20, 2024

*To whom correspondence should be addressed: javier.llorca@upm.es , javier.llorca@imdea.org

Minimum and maximum values of the descriptors

Descriptor	max value	min value
Unit cell volume	358.93 Å^3	10.49 Å^3
Weighted atomic radius (WAR)	180 pm	127.5 pm
Generalized coordination number (GCN)	5.25	0.5
Weighted electronegativity (WEN)	2.4 (Pauling)	1.23 (Pauling)
Weighted first ionization energy (WIE)	9.39 eV	5.86 eV
Outer electrons of A (S_A)	12	2
Outer electrons of B (S_B)	12	1
Ψ	106.73	3.41
Biaxial strain	-8.00%	8.00%

Table S 1: Minimum and maximum values of the descriptors.

Database details

Type	H		O		OH		System
	No Strain	Strain	No Strain	Strain	No Strain	Strain	
Pure metals	72	125	70	126	68	107	24
AB bcc(101)	220	190	193	182	218	152	58
AB fcc(101)	14	0	14	0	0	0	8
A_2B hcp(0001)	21	0	19	0	0	0	6
A_3B fcc(111)	111	105	114	110	103	125	26
A_3B hcp(0001)	36	30	36	27	31	4	8

Table S 2: Number of entries in the databases.

Hyperparameter values

Hyperparameter	E_{ads}^{H}	$E_{\text{ads}}^{O/OH}$
n_estimators	1000	1000
max_features	9	10
min_samples_split	4	3
min_samples_leaf	1	1
max_depth	600	300

Table S 3: Optimized hyperparameters for the Random Forest Regressor model in the prediction of the adsorption energy of hydrogen, oxygen, and hydroxyl.

Calculation of equilibrium lattice parameters

Stoichiometry	Material	Calculated by DFT		Materials Project	
		a (Å)	c (Å)	a (Å)	c (Å)
A	Pt	3.96	3.96	3.94	3.94
	Ag	4.09	4.09	4.10	4.10
	Au	4.16	4.16	4.17	4.17
	Ir	3.87	3.87	3.85	3.85
	Pd	3.94	3.94	3.92	3.92
	Co	2.49	4.04	2.47	4.03
AB	AlFe	2.86	2.86	2.85	2.85
	AlNi	2.89	2.89	2.86	2.86
	AlRu	3.00	3.00	2.98	2.98
	CuPd	3.02	3.02	2.98	2.98
	DyIn	3.74	3.74	3.74	3.74
	FeRh	3.00	3.00	2.99	2.99
	HfRu	3.23	3.23	3.23	3.23
	MgSc	3.58	3.58	3.60	3.60
	NdZn	3.67	3.67	3.67	3.67
	RuV	3.00	3.00	3.00	3.00
	TiRe	3.12	3.12	3.11	3.11
A_2B	Fe_2Zr	5.22	16.66	5.02	16.43
	Os_2Y	5.21	8.98	5.28	8.93
	Re_2Sm	5.24	8.98	5.45	8.93
	Ru_2Sm	5.15	8.80	5.27	9.06
A_3B	Al_3Li	4.02	4.02	4.00	4.00
	Co_3Ti	3.60	3.60	3.59	3.59
	In_3Sc	4.53	4.53	4.50	4.50
	In_3Y	4.65	4.65	4.63	4.63
	Ir_3Ti	3.87	3.87	3.86	3.86
	Ir_3Zr	3.98	3.98	3.96	3.96
	Pd_3Y	4.13	4.13	4.10	4.10
	Pt_3In	4.05	4.05	4.02	4.02
	Zn_3Mn	3.82	3.82	3.81	3.81
	Zn_3Nb	3.94	3.94	3.91	3.91
	Zr_3Al	4.38	4.38	4.38	4.38

Table S 4: Equilibrium lattice parameters (a and c) in Å. Displayed values calculated by DFT and obtained from Materials Project Database for all the different stoichiometries.

Feature distributions

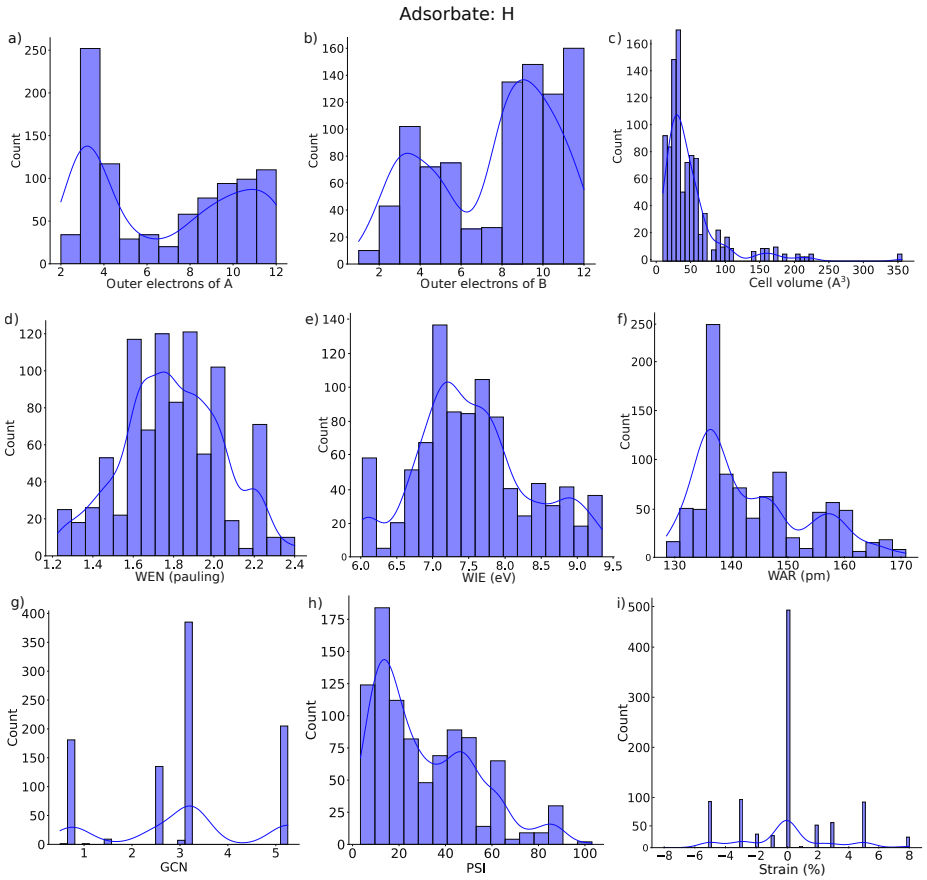

Figure S 1: **Feature distributions:** Hydrogen adsorption database.

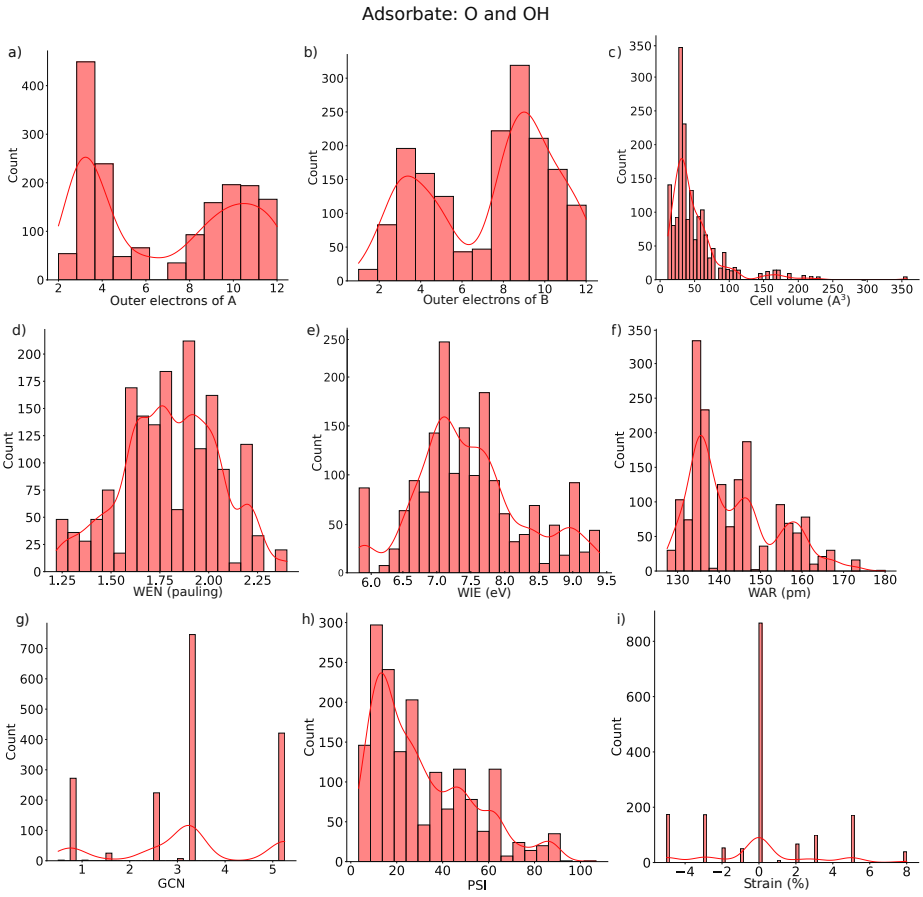

Figure S 2: **Feature distributions:** Oxygen and hydroxyl adsorption database.

Learning curves

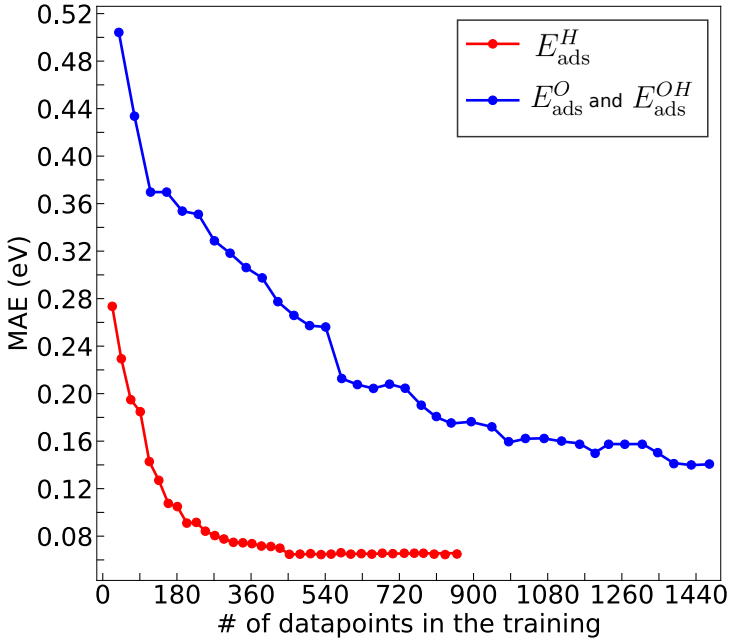

Figure S 3: **Learning curves:** for the hydrogen (red) and the oxygen/hydroxyl (blue) machine learning models.

Verification parity plots

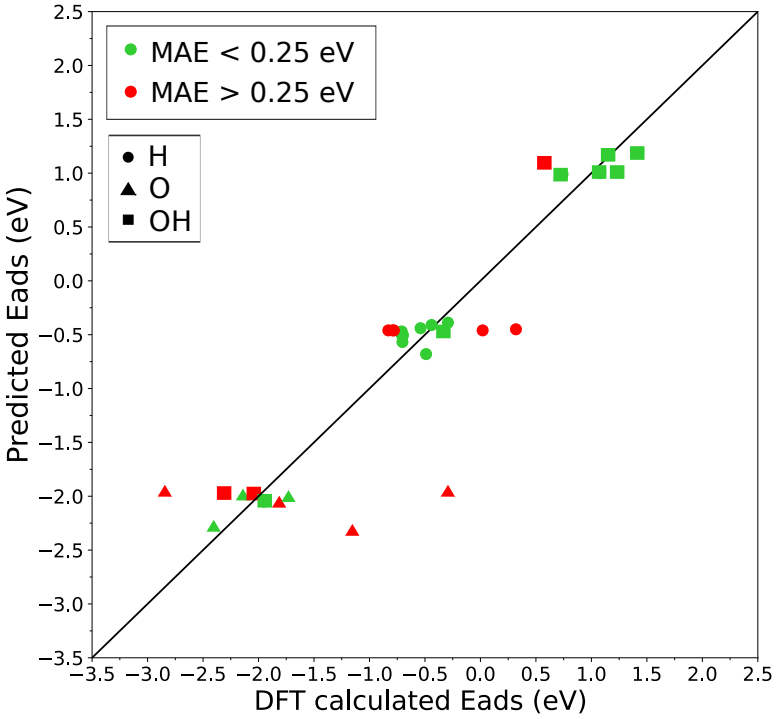

Figure S 4: **Parity plot for the 30 verification candidates:** Green signs represent the candidates that obtained accurate predictions by the RF model (MAE <0.25 eV). Red signs represent the candidates that obtained high errors (MAE >0.25 eV) by the RF model. Dots, triangles, and squares, illustrate the adsorption energies for H, O, and OH, respectively.

Optimum adsorption energies

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Material	Strain	Adsorption site	E^H_{ads} (eV)
Pt	0	FCC	-0.49
Ce3Ga	5	fccAAA	-0.49
Ce3In	5	fccAAA	-0.49
Ce3Sn	1	fccAAB	-0.49
Ir3W	-3	hcpAAA	-0.52
Nd3In	5	hcpAAA	-0.47
Ni3Fe	0	hcpAAB	-0.50
Ni3Pt	1	hcpAAA	-0.49
Pd3Ce	-1	fccAAB	-0.49
Pd3Dy	1	hcpAAA	-0.49
Pd3Fe	-5	fccAAA	-0.49
Pd3Nd	-3	hcpAAA	-0.49
Pd3Pr	-5	hcpAAA	-0.49
Pd3Sc	-3	fccAAA	-0.49
Pd3Sm	-5	hcpAAA	-0.47
Pd3Y	-3	fccAAA	-0.48
Pr3In	5	fccAAB	-0.48
Pt3Co	0	fccAAB	-0.49
Pt3Dy	0	hcpAAA	-0.49
Pt3Sc	-3	fccAAB	-0.49
Pt3Sm	-5	fccAAB	-0.49
Pt3Sn	-5	fccAAA	-0.50
Pt3Y	0	hcpAAA	-0.48
Rh3Mo	0	fccAAA	-0.46
Sm3In	3	hcpAAA	-0.47
Sn3Nd	5	fccAAB	-0.47
Sn3Sm	3	fccAAB	-0.49
Zn3Ti	-5	fccAAB	-0.48

Table S 5: Optimum hydrogen adsorption energies (in comparison with Pt at zero strain). The adsorption sites and the strains are indicated in the table.

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Material	Strain	Adsorption site	E^{O}_{ads} (eV)	Strain	Adsorption site	E^{OH}_{ads} (eV)
Pt	0	FCC	-1.79	0	FCC	1.19
Ag3In	5	fccAAB	-1.79	-5	hcpAAA	1.15
ZnAu	3	threefoldAAB	-1.78	0	longbridgeB	1.36
Cu3Pt	-5	fccAAB	-1.79	3	ontopB	1.19
Ir3Sc	-1	ontopB	-1.94	-1	fccAAA	1.19
Ni3Pt	-5	hcpAAA	-1.89	3	ontopA	1.16
Pd3Dy	-1	fccAAB	-1.81	0	fccAAA	1.19
Pd3Sn	-5	fccAAB	-1.84	3	fccAAA	1.13
Pd3Sc	1	hcpAAA	-1.77	-5	fccAAA	1.00
Pt3Co	5	hcpAAA	-1.76	-1	hcpAAA	1.2
Pt3Dy	-1	hcpAAA	-1.79	1	ontopA	1.18
Pt3Mn	3	hcpAAA	-1.81	1	hcpAAA	1.17
Pt3Sc	-5	fccAAB	-1.84	-5	fccAAA	1.19
Pt3Sn	-5	fccAAB	-1.87	1	fccAAA	1.18
Pt3Y	5	hcpAAA	-1.75	0	fccAAA	1.06

Table S 6: Optimum oxygen and hydroxyl adsorption energies (in comparison with Pt at zero strain). The adsorption sites and the applied strain are indicated in the table.

Variation of E_{ads} with strain

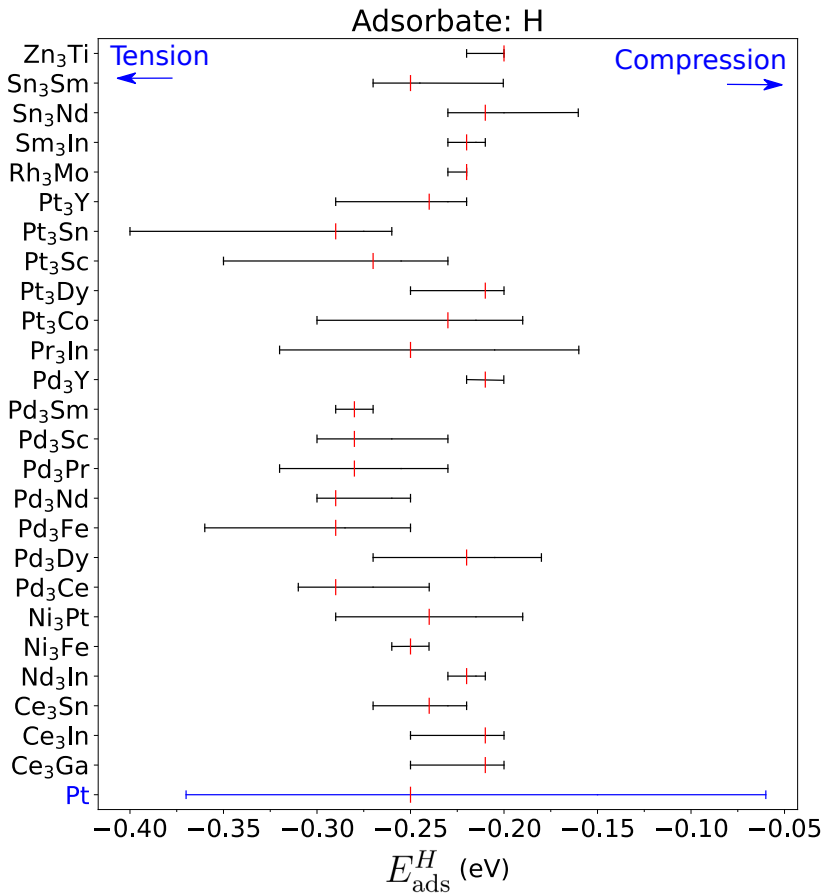

Figure S 5: **Variation of E^H_{ads}:** From -5% compression to 5% tension for the 25 candidates with the most similar energies to pure platinum. The red vertical lines represent the E^H_{ads} at zero strain.

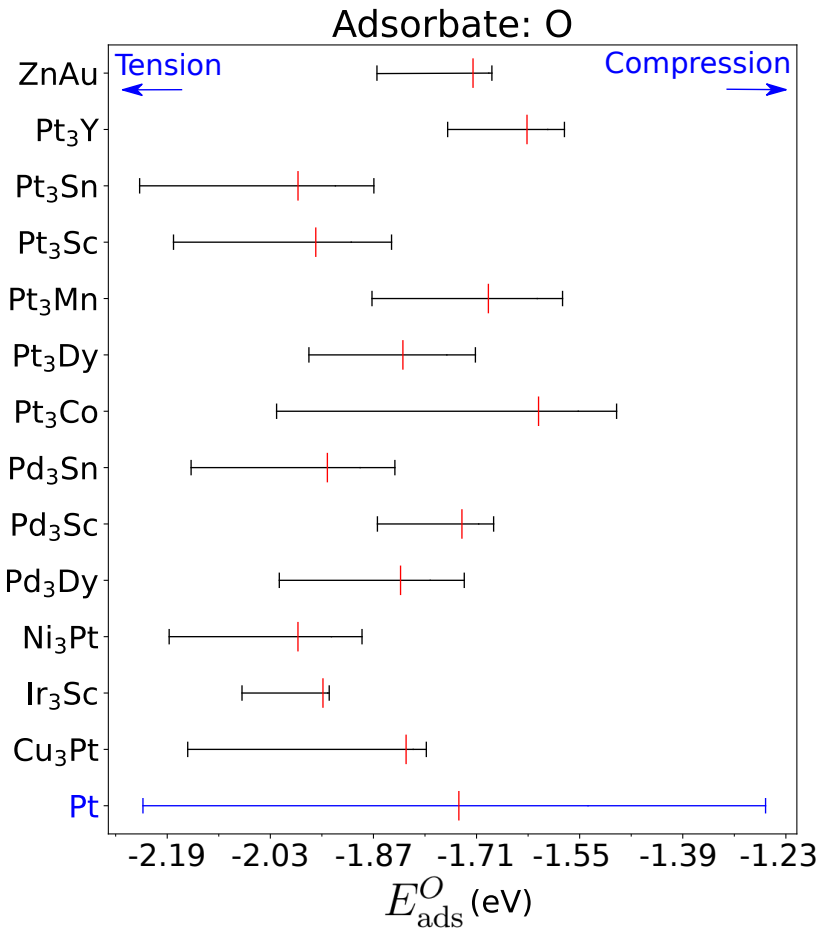

Figure S 6: **Variation of E^O_{ads}:** From -5% compression to 5% tension for the 13 candidates with the most similar energies to pure platinum. The red vertical lines represent the E^O_{ads} at zero strain.

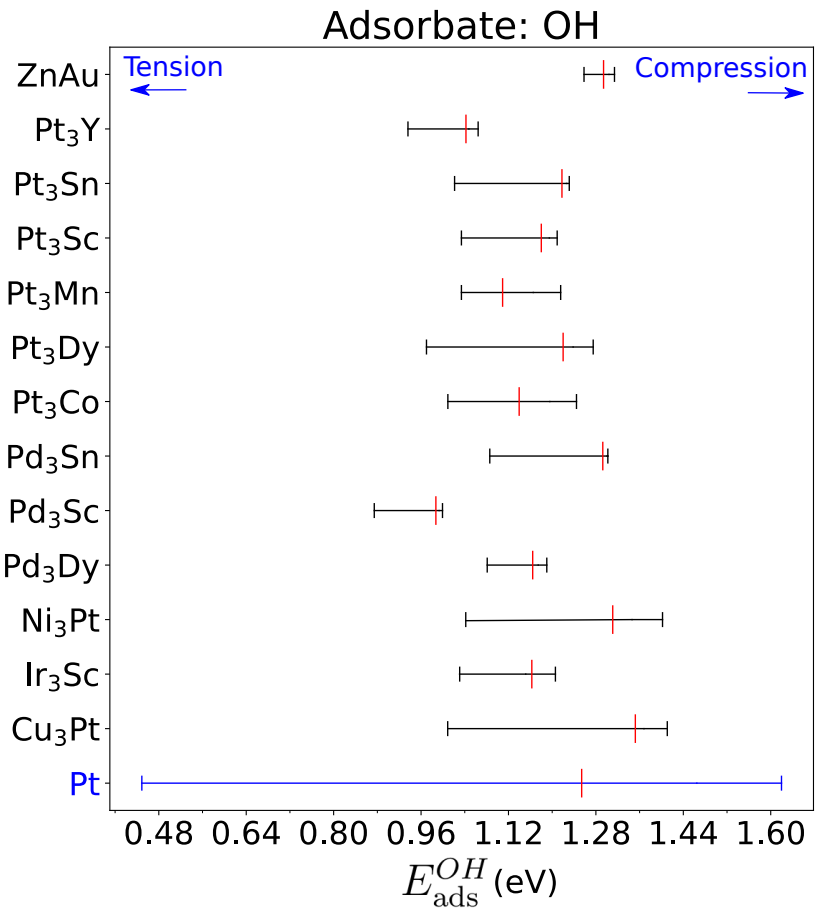

Figure S 7: **Variation of E_{ads}^{OH}:** From -5% compression to 5% tension for the 13 candidates with the most similar energies to pure platinum. The red vertical lines represent the E_{ads}^{OH} at zero strain.