

Supplementary Information: Benchmarking Machine-Learned Potentials for Adsorption on Pt and IrO₂ Surfaces Using OC20 and OMat24

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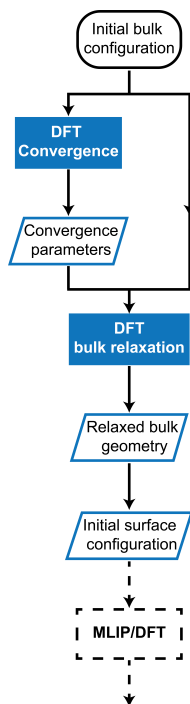


Figure S1: Both Pt and IrO₂ slab bulk configurations were relaxed using DFT; the lattice parameter of the resulting relaxed bulk geometry was used to design subsequent surface configurations for adsorption calculations

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Table S1: The adsorption energies of common adsorbates like CO, NO, O, and H on Pt(111) and Pt(110) calculated using UMA-OC20, obtained from experiments (Silbaugh and Campbell), and computed using spin-polarized DFT without vdW corrections. Here, both the DFT flavors and the geometries are different

Surf ace	Adso rbate	Cove rage	UMA-OC20		Exp. E _{ads} (eV)	Diff. OC20- Exp. (eV)	DFT		Diff. OC20-DFT	
			E _{ads} (eV)	Site			E _{ads} (eV)	Site	E _{ads} (eV)	Norm (Å)
Pt (111)	CO	1/4	-1.493	fcc	-1.244	-0.249	-1.420	fcc	-0.073	0.085
	NO	1/4	-1.741	bridge, fcc	-1.182	-0.559	-1.538	fcc	-0.203	0.094
	O ₂ → 2O	1/9	-3.092	fcc	-2.156	-0.936	-2.025	fcc	-1.067*	0.179
	H ₂ → 2H	1/4	-0.681	fcc	-0.746	0.065	-0.718	top**	0.037	0.066
Pt (110)	CO	1/4	-1.858	short bridge	-1.244	-0.614	-1.829	short bridge	-0.029	0.074
	NO	1/4	-2.290	short bridge	-1.171	-1.119	-2.039	short bridge	-0.251	0.102
	O ₂ → 2O	1/4	-3.412	short bridge	-2.28	-1.132	-2.174	short bridge	-1.238*	0.113

*Gas phase references, E_{gas}, for each adsorbate was computed as a linear combination of N₂, H₂O, CO, and H₂ resulting in the atomic energies of O₂ to be -14.408 eV (correct value from DFT is -10.545)

** The fcc site is shown to be the most stable site for *H on Pt(111). Adding vdW corrections to DFT changes the site from top to fcc.

Table S2: The adsorption energies of common adsorbates like CO, NO, O, and H on Pt(111) and Pt(110) calculated from MLIPs UMA-OC20 and non-spin polarized DFT with no vdW corrections. Here, the DFT flavors are the same but the geometries are different

Surf ace	Adso rbate	Cove rage	UMA-OC20		DFT	DFT	Diff. OC20-DFT	
			E _{ads} (eV)	Site		E _{ads} (eV)	E _{ads} (eV)	Norm (Å)
Pt(111)	CO	1/4	-1.493	fcc	-1.420	fcc	-0.073	0.085
	NO	1/4	-1.741	bridge, fcc	-1.844	fcc	0.103	0.094
	O ₂ → 2O	1/9	-3.092	fcc	-3.050	fcc	-0.042	0.158
	H ₂ → 2H	1/4	-0.646	top*	-0.718	top*	0.072	0.066
Pt(110)	CO	1/4	-1.858	short bridge	-1.829	short bridge	-0.029	0.074
	NO	1/4	-2.290	short bridge	-2.338	short bridge	0.048	0.08
	O ₂ → 2O	1/4	-3.412	short bridge	-3.298	short bridge	-0.114	0.106

Table S3: The adsorption energies of common adsorbates like CO, NO, O, and H on Pt(111) and Pt(110) calculated from single-point MLIPs UMA-OC20 and non-spin polarized DFT with no vdW corrections, on the relaxed structure obtained from the non-spin polarized DFT with no vdW corrections. Here, both the DFT flavors and the geometries are the same. All Euclidean Norm of coordinate difference is 0 and hence, is not displayed

Surface	Adsorbate	Coverage	UMA-OC20		DFT		Diff.
			E _{ads} (eV)	Site	E _{ads} (eV)	Site	OC20-DFT (eV)
Pt(111)	CO	1/4	-1.490	fcc	-1.420	fcc	-0.070
	NO	1/4	-1.739	bridge, fcc	-1.844	fcc	0.105
	O ₂ → 2O	1/9	-3.070	fcc	-3.050	fcc	-0.020
	H ₂ → 2H	1/4	-0.644	top*	-0.718	top*	0.074
Pt(110)	CO	1/4	-1.866	shortbridge	-1.829	shortbridge	-0.037
	NO	1/4	-2.299	shortbridge	-2.338	shortbridge	0.039
	O ₂ → 2O	1/4	-3.414	shortbridge	-3.298	shortbridge	-0.116

Table S4: The adsorption energies of common adsorbates like CO, NO, O, and H on Pt(231) calculated from single-point MLIPs UMA-OC20 and non-spin polarized DFT with no vdW corrections, on structures with the adsorbate 2 Å from three top sites corresponding to the three Pt atoms exposed to vacuum. Here, both the DFT flavors and the geometries are the same. All Norm is 0 and hence, is not displayed

Surface	Adsorbate	Coverage	Site	E _{ads}		Diff.
				UMA-OC20 (eV)	DFT (eV)	OC20-DFT (eV)
Pt(231)	CO	1/4	site1	-1.782	-1.669	-0.113
			site2	0.149	0.210	-0.061
			site3	-0.060	-0.007	-0.053
	NO	1/4	site1	-1.794	-1.842	0.048
			site2	-0.261	-0.339	0.078
			site3	-0.664	-0.726	0.062
	O	1/4	site1	-0.575	-0.470	-0.105
			site2	2.265	2.278	-0.013
			site3	2.147	2.123	0.024
	H	1/4	site1	0.498	0.546	-0.048
			site2	2.249	2.244	0.005
			site3	2.162	2.185	-0.023

Table S5: The adsorption energies of *NO and *O on Pt(111) and Pt(110) calculated from spin polarized and non-spin polarized DFT, along with MLIP

Technique	Spin state	Slab	Adsorbate (X)	$E_{\text{ads}}(\text{X})$ (eV)	$E(\text{slab}+*\text{X})$ (eV)	$E(\text{slab})$ (eV)	$E(\text{X}_{\text{ref gas}})$ (eV)
DFT	Spin	Pt(111)- 2 $\times$ 2	NO	-1.538	-126.287	-111.864	-12.885
	No spin			-1.844	-126.287	-111.864	-12.579
MLIP				-1.741	-118.118	-104.675	-11.702
DFT	Spin	Pt(111)- 3 $\times$ 3	O*	-1.013	-257.926	-251.641	-5.273
	No spin			-1.525	-257.871	-251.641	-4.705
MLIP				-1.546	-241.301	-235.519	-4.236
DFT	Spin	Pt(110)- 2 $\times$ 2	NO	-2.039	-121.861	-106.937	-12.885
	No spin			-2.338	-121.855	-106.937	-12.579
MLIP				-2.290	-113.617	-99.625	-11.702
DFT	Spin		O*	-1.087	-113.297	-106.937	-5.273
	No spin			-1.649	-113.292	-106.937	-4.705
MLIP				-1.706	-105.567	-99.625	-4.236

* The adsorption energy for O is for a coverage of 1 O in the slab, which is half of the value depicted in other tables which consider 2 O in the slab. The reference gas column for O depicts $0.5 \cdot E(\text{O}_2 \text{ gas})$

Table S6: The adsorption energies of *NO and *O on Pt(111) and Pt(110) calculated from spin polarized and non-spin polarized DFT. Most of the difference between them arise from the reference gas calculations. We obtain the same adsorption energies for *CO and *H on Pt(111) and Pt(110) from both spin-polarized and non-spin polarized DFT

Spin state	Slab	Adsorbate (X)	$E_{\text{ads}}(\text{X})$ (eV)	$E(\text{slab}+*\text{X})$ (eV)	$E(\text{slab})$ (eV)	$E(\text{X}_{\text{ref gas}})$ (eV)	Norm Å
Spin	Pt(111)- 2 $\times$ 2	NO	-1.538	-126.287	-111.864	-12.885	0.022
No spin			-1.844	-126.287	-111.864	-12.579	
Difference			0.306	0	0	-0.306	
Spin	Pt(111)- 3 $\times$ 3	O*	-1.013	-257.926	-251.641	-5.273	0.046
No spin			-1.525	-257.871	-251.641	-4.705	
Difference			0.512	-0.055	0	-0.567	
Spin	Pt(110)- 2 $\times$ 2	NO	-2.039	-121.861	-106.937	-12.885	0.044
No spin			-2.338	-121.855	-106.937	-12.579	
Difference			0.3	-0.006	0	-0.306	
Spin		O*	-1.087	-113.297	-106.937	-5.273	0.021
No spin			-1.649	-113.292	-106.937	-4.705	
Difference			0.562	-0.006	0	-0.567	

* The adsorption energy for O is for a coverage of 1 O in the slab, which is half of the value depicted in other tables which consider 2 O in the slab. The reference gas column for O depicts $0.5 \cdot E(\text{O}_2 \text{ gas})$

Table S7: The adsorption energies of adsorbates in the water splitting reaction on IrO₂(110) calculated from UMA-OMat24 MLIP and spin polarized DFT with vdW corrections. Here, the DFT flavors are the same. The SP and DFT geometries are the same but the rest are different from them

Surf	Adso	UMA-OMat24 E _{ads}			DFT	Difference					
ace	rbate	R1	SP	R2	E _{ads}	R1-DFT		SP-DFT		R2-DFT	
		(eV)	(eV)	(eV)	(eV)	E _{ads} (eV)	Norm (Å)	E _{ads} (eV)	Norm (Å)	E _{ads} (eV)	Norm (Å)
	H, -	-0.848	-1.809	-1.819	-1.746	0.898*	9.125	-0.063	0	-0.073	0.244
	H, H	-3.745	-3.744	-3.745	-3.726	-0.019	11.107	-0.018	0	-0.019	0.202
IrO ₂	O, -	2.383	2.427	2.383	2.491	-0.108	6.817	-0.064	0	-0.108	0.52
(110)O,	O	4.945	5.007	4.928	5.181	-0.236	5.499	-0.174	0	-0.253	0.56
	H, O	0.542	0.730	0.681	0.849	-0.307	16.554	-0.119	0	-0.168	0.519
	OH, H	-0.471	-0.467	-0.471	-0.451	-0.02	11.111	-0.016	0	-0.02	0.26

* The high error arises from *H configuration not changing from the initial position during the first MLIP relaxation

Table S8: The relative computational time for Pt(110) and Pt(110) calculated from UMA-OC20 MLIP and DFT. DFT computations used VASP 5.4.4 with 64 MPI processes, each spawning 4 OpenMP threads, for a total of 256 cores with 1 CPU compute node in perlmutter. We set NCORE to 8 to organize the 64 MPI ranks into groups of 8. No other parallelization was added for VASP. MLIP computations used uma-s-1p1 using OC20 with 1 GPU compute node in perlmutter.

Surface	Adsorbate	MLIP time (s)	DFT time (s)		Enhancement	
			spin	no spin	spin	no spin
Pt(111)	CO fcc	3.9	17699	17699	4538	4538
	NO fcc	2.0	37766	16384	18883	8192
	H fcc	1.3	5059	5059	3891	3891
	H top	1.9	5334	5334	2807	2807
	O fcc	2.3	65249	23352	28369	10153
Pt(110)	CO sb	2.1	7437	7437	3541	3541
	NO sb	2.6	17856	8124	6868	3125
	O sb	2.2	10429	3544	4740	1611

Table S9: The relative computational time for IrO₂(110) calculated from UMA-OMat24 MLIP and spin polarized DFT with vdW corrections. DFT computations used VASP 5.4.4 with 64 MPI processes, each spawning 4 OpenMP threads, for a total of 256 cores with 1 CPU compute node in perlmutter. We set NCORE to 8 to organize the 64 MPI ranks into groups of 8. No other parallelization was added for VASP. MLIP computations used uma-s-1p1 using OC20 with 1 GPU compute node in perlmutter.

Surface	Adsorbate	MLIP time (s)	DFT time (s)	Enhancement
IrO ₂ (110)	H, -	26.2	9673	369
	H, H	8.8	16301	1852
	O, -	17.7	35273	1992
	O, O	9.9	48335	4882
	H, O	14.9	51766	3474
	OH, H	11.3	17864	1580