Supporting information for

Same but Different: How Permutation Degeneracy Enhances Machine Learning in Single-Atom Catalyst Design

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Table S1. The E_f values of 164 M-N_xS_y.

Coordination	TM	E_f	Coordination	TM	E_f
N_4	Sc	-7.03	N_2S_2-sep	Y	-6.60
N_4	Ti	-5.76	N_2S_2-sep	Zr	-5.88
N_4	V	-4.79	N_2S_2-sep	Nb	3.89
N_4	Cr	-2.64	N_2S_2-sep	Mo	-2.85
N_4	Mn	-3.94	N_2S_2-sep	Tc	-13.03
N_4	Fe	-4.01	N_2S_2-sep	Ru	-2.67
N_4	Co	-4.65	N_2S_2-sep	Rh	-3.44
N_4	Ni	-4.43	N_2S_2-sep	Pd	-3.75
N_4	Cu	-3.44	N_2S_2-sep	Ag	-2.32
N_4	Y	-6.84	N_2S_2-sep	Hf	-6.07
N_4	Zr	-5.90	N_2S_2-sep	Ta	-4.35
N_4	Nb	-3.74	N_2S_2-sep	W	-2.58
N_4	Mo	-2.78	N_2S_2-sep	Re	-2.16
N_4	Tc	-13.52	N_2S_2-sep	Os	-2.41
N_4	Ru	-3.24	N_2S_2-opp	Sc	-6.55
N_4	Rh	-4.37	N_2S_2-opp	Ti	-4.94
N_4	Pd	-4.61	N_2S_2-opp	V	-3.48
N_4	Ag	-2.35	N_2S_2-opp	Cr	-4.36
N_4	Cd	-4.93	N_2S_2-opp	Mn	-3.36
N_4	Hf	-5.79	N_2S_2-opp	Fe	-4.27
N_4	Ta	-4.29	N_2S_2-opp	Co	-3.96
N_4	W	-2.91	N_2S_2-opp	Ni	-3.84
N_4	Re	-2.69	N_2S_2-opp	Cu	-2.73
N_4	Os	-2.35	N_2S_2-opp	Zn	-4.27
N_3S_1	Sc	-5.10	N_2S_2-opp	Y	-6.67
N_3S_1	Ti	-4.07	N_2S_2-opp	Zr	-4.75
N_3S_1	V	-3.34	N_2S_2-opp	Nb	2.92
N_3S_1	Cr	-3.02	N_2S_2-opp	Mo	-3.25
N_3S_1	Mn	-3.47	N_2S_2-opp	Tc	-13.46
N_3S_1	Fe	-2.96	N_2S_2-opp	Ru	-3.19
N_3S_1	Co	-3.13	N_2S_2-opp	Rh	-3.91
N_3S_1	Ni	-4.36	N_2S_2-opp	Pd	-4.19
N_3S_1	Cu	-3.38	N_2S_2-opp	Ag	-1.55
N_3S_1	Y	-6.65	N_2S_2-opp	Cd	-3.76
N_3S_1	Zr	-6.31	N_2S_2-opp	Hf	-5.20
N_3S_1	Nb	-4.74	N_2S_2-opp	Ta	-3.20
N_3S_1	Mo	-3.33	N_2S_2-opp	W	-2.05
N_3S_1	Tc	-11.99	N_2S_2-opp	Re	-2.65
N_3S_1	Ru	-1.65	N_2S_2-opp	Os	-3.09
N_3S_1	Rh	-2.64	N_1S_3	Sc	-5.03
N_3S_1	Pd	-2.92	N_1S_3	Ti	-4.15
N_3S_1	Ag	-2.62	N_1S_3	V	-3.00
N_3S_1	Cd	-5.02	N_1S_3	Cr	-2.88
N_3S_1	Hf	-5.75	N_1S_3	Mn	-3.12
N_3S_1	Ta	-2.41	N_1S_3	Fe	-2.68
N_3S_1	W	-2.87	N_1S_3	Co	-2.81
N_3S_1	Re	-1.00	N_1S_3	Ni	-2.64

N_3S_1	Os	-0.71	**N_1S_3**	Cu	-1.63
N_2S_2-adj	Sc	-5.02	**N_1S_3**	Y	-5.47
N_2S_2-adj	Ti	-3.88	**N_1S_3**	Zr	-4.93
N_2S_2-adj	V	-2.89	**N_1S_3**	Nb	7.44
N_2S_2-adj	Cr	-2.79	**N_1S_3**	Mo	-1.97
N_2S_2-adj	Mn	-3.04	**N_1S_3**	Tc	-12.28
N_2S_2-adj	Fe	-2.52	**N_1S_3**	Ru	-1.98
N_2S_2-adj	Co	-2.56	**N_1S_3**	Rh	-2.70
N_2S_2-adj	Ni	-2.31	**N_1S_3**	Pd	-2.95
N_2S_2-adj	Cu	-1.27	**N_1S_3**	Ag	-1.42
N_2S_2-adj	Zn	-2.86	**N_1S_3**	Hf	-4.74
N_2S_2-adj	Y	-5.16	**N_1S_3**	Ta	-3.14
N_2S_2-adj	Zr	-4.19	**N_1S_3**	W	-1.63
N_2S_2-adj	Nb	5.16	**N_1S_3**	Re	-1.39
N_2S_2-adj	Mo	-1.72	**N_1S_3**	Os	-1.26
N_2S_2-adj	Tc	-12.01	**S_4**	Sc	-5.21
N_2S_2-adj	Ru	-1.75	**S_4**	Ti	-3.65
N_2S_2-adj	Rh	-2.45	**S_4**	V	-3.03
N_2S_2-adj	Pd	-2.68	**S_4**	Cr	-3.00
N_2S_2-adj	Ag	-1.07	**S_4**	Mn	-3.20
N_2S_2-adj	Cd	-3.18	**S_4**	Fe	-2.76
N_2S_2-adj	Hf	-4.45	**S_4**	Co	-3.07
N_2S_2-adj	Ta	-2.38	**S_4**	Ni	-3.01
N_2S_2-adj	W	-1.44	**S_4**	Y	-5.06
N_2S_2-adj	Re	-1.21	**S_4**	Zr	-4.70
N_2S_2-adj	Os	-1.00	**S_4**	Nb	10.56
N_2S_2-sep	Sc	-6.42	**S_4**	Mo	-2.10
N_2S_2-sep	Ti	-5.80	**S_4**	Tc	-12.59
N_2S_2-sep	V	-4.27	**S_4**	Ru	-1.40
N_2S_2-sep	Cr	-3.97	**S_4**	Rh	-3.09
N_2S_2-sep	Mn	-4.45	**S_4**	Hf	-4.61
N_2S_2-sep	Fe	-3.68	**S_4**	Ta	-2.18
N_2S_2-sep	Co	-3.77	**S_4**	W	-1.24
N_2S_2-sep	Ni	-3.52	**S_4**	Re	-1.81
N_2S_2-sep	Cu	-2.89	**S_4**	Os	-2.20

Table S2. The U_{diss} values of 164 M-N_xS_y.

Coordination	TM	U_{diss}	Coordination	TM	U_{diss}
N_4	Sc	0.03	**N_2S_2-sep**	Y	-0.21
N_4	Ti	2.32	**N_2S_2-sep**	Zr	0.69
N_4	V	3.37	**N_2S_2-sep**	Nb	2.27
N_4	Cr	3.91	**N_2S_2-sep**	Mo	3.42
N_4	Mn	3.39	**N_2S_2-sep**	Tc	Nan
N_4	Fe	3.78	**N_2S_2-sep**	Ru	5.10
N_4	Co	3.28	**N_2S_2-sep**	Rh	4.28
N_4	Ni	2.63	**N_2S_2-sep**	Pd	3.54
N_4	Cu	2.39	**N_2S_2-sep**	Ag	3.63

N_4	Y	-0.21	**N_2S_2-sep**	Hf	0.94
N_4	Zr	0.69	**N_2S_2-sep**	Ta	3.35
N_4	Nb	2.27	**N_2S_2-sep**	W	4.42
N_4	Mo	3.42	**N_2S_2-sep**	Re	4.45
N_4	Tc	Nan	**N_2S_2-sep**	Os	2.24
N_4	Ru	5.10	**N_2S_2-opp**	Sc	0.03
N_4	Rh	4.28	**N_2S_2-opp**	Ti	2.32
N_4	Pd	3.54	**N_2S_2-opp**	V	3.37
N_4	Ag	3.63	**N_2S_2-opp**	Cr	3.91
N_4	Cd	Nan	**N_2S_2-opp**	Mn	3.39
N_4	Hf	0.94	**N_2S_2-opp**	Fe	3.78
N_4	Ta	3.35	**N_2S_2-opp**	Co	3.28
N_4	W	4.42	**N_2S_2-opp**	Ni	2.63
N_4	Re	4.45	**N_2S_2-opp**	Cu	2.39
N_4	Os	2.24	**N_2S_2-opp**	Zn	-0.13
N_3S_1	Sc	0.03	**N_2S_2-opp**	Y	-0.21
N_3S_1	Ti	2.32	**N_2S_2-opp**	Zr	0.69
N_3S_1	V	3.37	**N_2S_2-opp**	Nb	2.27
N_3S_1	Cr	3.91	**N_2S_2-opp**	Mo	3.42
N_3S_1	Mn	3.39	**N_2S_2-opp**	Tc	Nan
N_3S_1	Fe	3.78	**N_2S_2-opp**	Ru	5.10
N_3S_1	Co	3.28	**N_2S_2-opp**	Rh	4.28
N_3S_1	Ni	2.63	**N_2S_2-opp**	Pd	3.54
N_3S_1	Cu	2.39	**N_2S_2-opp**	Ag	3.63
N_3S_1	Y	-0.21	**N_2S_2-opp**	Cd	Nan
N_3S_1	Zr	0.69	**N_2S_2-opp**	Hf	0.94
N_3S_1	Nb	2.27	**N_2S_2-opp**	Ta	3.35
N_3S_1	Mo	3.42	**N_2S_2-opp**	W	4.42
N_3S_1	Tc	Nan	**N_2S_2-opp**	Re	4.45
N_3S_1	Ru	5.10	**N_2S_2-opp**	Os	2.24
N_3S_1	Rh	4.28	**N_1S_3**	Sc	0.03
N_3S_1	Pd	3.54	**N_1S_3**	Ti	2.32
N_3S_1	Ag	3.63	**N_1S_3**	V	3.37
N_3S_1	Cd	Nan	**N_1S_3**	Cr	3.91
N_3S_1	Hf	0.94	**N_1S_3**	Mn	3.39
N_3S_1	Ta	3.35	**N_1S_3**	Fe	3.78
N_3S_1	W	4.42	**N_1S_3**	Co	3.28
N_3S_1	Re	4.45	**N_1S_3**	Ni	2.63
N_3S_1	Os	2.24	**N_1S_3**	Cu	2.39
N_2S_2-adj	Sc	0.03	**N_1S_3**	Y	-0.21
N_2S_2-adj	Ti	2.32	**N_1S_3**	Zr	0.69
N_2S_2-adj	V	3.37	**N_1S_3**	Nb	2.27
N_2S_2-adj	Cr	3.91	**N_1S_3**	Mo	3.42
N_2S_2-adj	Mn	3.39	**N_1S_3**	Tc	Nan
N_2S_2-adj	Fe	3.78	**N_1S_3**	Ru	5.10
N_2S_2-adj	Co	3.28	**N_1S_3**	Rh	4.28
N_2S_2-adj	Ni	2.63	**N_1S_3**	Pd	3.54
N_2S_2-adj	Cu	2.39	**N_1S_3**	Ag	3.63
N_2S_2-adj	Zn	-0.13	**N_1S_3**	Hf	0.94

N_2S_2-adj	Y	-0.21	**N_1S_3**	Ta	3.35
N_2S_2-adj	Zr	0.69	**N_1S_3**	W	4.42
N_2S_2-adj	Nb	2.27	**N_1S_3**	Re	4.45
N_2S_2-adj	Mo	3.42	**N_1S_3**	Os	2.24
N_2S_2-adj	Tc	Nan	**S_4**	Sc	0.03
N_2S_2-adj	Ru	5.10	**S_4**	Ti	2.32
N_2S_2-adj	Rh	4.28	**S_4**	V	3.37
N_2S_2-adj	Pd	3.54	**S_4**	Cr	3.91
N_2S_2-adj	Ag	3.63	**S_4**	Mn	3.39
N_2S_2-adj	Cd	Nan	**S_4**	Fe	3.78
N_2S_2-adj	Hf	0.94	**S_4**	Co	3.28
N_2S_2-adj	Ta	3.35	**S_4**	Ni	2.63
N_2S_2-adj	W	4.42	**S_4**	Y	-0.21
N_2S_2-adj	Re	4.45	**S_4**	Zr	0.69
N_2S_2-adj	Os	2.24	**S_4**	Nb	2.27
N_2S_2-sep	Sc	0.03	**S_4**	Mo	3.42
N_2S_2-sep	Ti	2.32	**S_4**	Tc	Nan
N_2S_2-sep	V	3.37	**S_4**	Ru	5.10
N_2S_2-sep	Cr	3.91	**S_4**	Rh	4.28
N_2S_2-sep	Mn	3.39	**S_4**	Hf	0.94
N_2S_2-sep	Fe	3.78	**S_4**	Ta	3.35
N_2S_2-sep	Co	3.28	**S_4**	W	4.42
N_2S_2-sep	Ni	2.63	**S_4**	Re	4.45
N_2S_2-sep	Cu	2.39	**S_4**	Os	2.24

Table S3. The 11 configurations are excluded according DFT calculations.

TM-N_xS_y	***OH, *O, *OOH**
Cu-S_4	**O**
Zn-N_4	**O**
Zn-N_3S_1	**OH**
Zn-N_2S_2-sep	**OH**
Zn-N_1S	**OH**
Zn-S_4	**OH**
Pd-S_4	**OOH**
Ag-S_4	**O**
Cd-N_2S_2-sep	**OH**
Cd-N_1S	**OH**
Cd-S_4	**OH**

Table S4. The ΔG_{*OH}, ΔG_{*O}, and ΔG_{*OOH} values of 164 TM-N_xS_y

Coordination	TM	ΔG_{*OH}	ΔG_{*O}	ΔG_{*OOH}	Coordination	TM	ΔG_{*OH}	ΔG_{*O}	ΔG_{*OOH}
N_4	Sc	-1.45	0.23	1.62	N_2S_2-sep	Y	-1.21	1.23	2.00
N_4	Ti	-1.51	-1.27	1.27	N_2S_2-sep	Zr	-1.32	-0.44	2.06
N_4	V	-1.57	-1.11	1.66	N_2S_2-sep	Nb	-1.78	-1.42	1.46
N_4	Cr	-0.09	0.14	2.86	N_2S_2-sep	Mo	-1.36	-1.23	1.70
N_4	Mn	-1.87	-1.51	0.88	N_2S_2-sep	Tc	-0.79	-0.72	2.19
N_4	Fe	-0.40	0.81	2.51	N_2S_2-sep	Ru	0.03	0.77	3.00
N_4	Co	1.08	2.61	3.90	N_2S_2-sep	Rh	0.94	2.58	3.75
N_4	Ni	1.67	4.00	4.54	N_2S_2-sep	Pd	2.04	4.38	4.67
N_4	Cu	1.83	2.12	4.52	N_2S_2-sep	Ag	1.45	1.69	4.54
N_4	Y	-1.70	0.14	1.53	N_2S_2-sep	Hf	-1.30	-0.17	2.10
N_4	Zr	-2.21	-1.70	0.97	N_2S_2-sep	Ta	-2.00	-1.38	1.34
N_4	Nb	-2.51	-2.73	0.69	N_2S_2-sep	W	-1.77	-1.90	1.52
N_4	Mo	-2.41	-2.69	0.52	N_2S_2-sep	Re	-1.27	-1.25	1.83
N_4	Tc	-1.48	-1.70	0.90	N_2S_2-sep	Os	-0.41	0.26	2.32
N_4	Ru	-0.11	0.74	2.76	N_2S_2-opp	Sc	-1.14	0.98	2.46
N_4	Rh	1.05	2.64	3.86	N_2S_2-opp	Ti	-0.60	0.19	2.67
N_4	Pd	2.02	4.40	4.88	N_2S_2-opp	V	-0.55	0.21	2.47
N_4	Ag	1.52	1.83	4.76	N_2S_2-opp	Cr	-0.13	0.36	3.00
N_4	Cd	1.51	4.24	4.17	N_2S_2-opp	Mn	0.70	1.22	3.65
N_4	Hf	-2.69	-1.97	0.64	N_2S_2-opp	Fe	0.69	1.85	3.69
N_4	Ta	-2.86	-2.57	0.45	N_2S_2-opp	Co	1.00	2.49	3.88
N_4	W	-2.44	-2.78	0.97	N_2S_2-opp	Ni	1.67	3.87	4.43
N_4	Re	-1.29	-1.47	1.25	N_2S_2-opp	Cu	1.42	1.71	4.33
N_4	Os	0.40	0.69	2.88	N_2S_2-opp	Zn	0.84	1.97	4.01
N_3S_1	Sc	-1.27	0.56	1.75	N_2S_2-opp	Y	-1.25	1.12	1.93
N_3S_1	Ti	-0.96	0.05	2.29	N_2S_2-opp	Zr	-1.14	-0.03	2.00
N_3S_1	V	-0.72	-0.10	2.47	N_2S_2-opp	Nb	-1.21	-1.08	1.75
N_3S_1	Cr	-0.17	0.25	2.79	N_2S_2-opp	Mo	-1.20	-1.18	2.02
N_3S_1	Mn	0.49	1.02	3.43	N_2S_2-opp	Tc	-1.02	-0.90	2.04
N_3S_1	Fe	0.58	1.73	3.51	N_2S_2-opp	Ru	-0.14	0.55	2.72
N_3S_1	Co	1.08	2.70	3.94	N_2S_2-opp	Rh	0.97	2.58	3.76
N_3S_1	Ni	1.67	3.31	4.46	N_2S_2-opp	Pd	2.07	4.38	4.68
N_3S_1	Cu	2.00	2.37	4.69	N_2S_2-opp	Ag	2.11	1.94	4.58
N_3S_1	Y	-1.50	0.51	1.70	N_2S_2-opp	Cd	1.79	2.09	4.29
N_3S_1	Zr	-1.20	-0.28	1.88	N_2S_2-opp	Hf	-1.06	0.00	2.06
N_3S_1	Nb	-2.15	-1.90	1.18	N_2S_2-opp	Ta	-1.59	-1.21	1.51
N_3S_1	Mo	-1.39	-1.46	1.89	N_2S_2-opp	W	-1.33	-1.53	2.00
N_3S_1	Tc	-1.53	-1.54	1.26	N_2S_2-opp	Re	-1.50	-1.40	1.71
N_3S_1	Ru	-0.10	0.56	2.80	N_2S_2-opp	Os	0.30	0.27	3.07
N_3S_1	Rh	1.01	2.61	3.86	N_1S_3	Sc	-0.94	-0.09	2.13
N_3S_1	Pd	2.07	4.43	4.76	N_1S_3	Ti	-0.67	0.06	2.37
N_3S_1	Ag	1.87	2.15	4.47	N_1S_3	V	-1.03	-0.62	2.06
N_3S_1	Cd	2.01	1.60	4.40	N_1S_3	Cr	-0.14	0.21	2.90
N_3S_1	Hf	-2.78	-2.06	0.51	N_1S_3	Mn	1.76	3.34	3.55
N_3S_1	Ta	-2.11	-2.22	1.27	N_1S_3	Fe	0.71	1.90	3.57
N_3S_1	W	-1.75	-1.71	1.40	N_1S_3	Co	1.02	2.47	3.95
N_3S_1	Re	-1.51	-1.50	-0.71	N_1S_3	Ni	1.68	3.89	4.47

N_3S_1	Os	-1.62	-2.19	1.04	**N_1S_3**	Cu	1.81	1.75	4.56
N_2S_2-adj	Sc	-1.10	0.81	2.08	**N_1S_3**	Y	-0.59	0.49	2.81
N_2S_2-adj	Ti	-1.40	-0.57	2.09	**N_1S_3**	Zr	-0.98	0.01	2.62
N_2S_2-adj	V	-1.00	-0.77	2.24	**N_1S_3**	Nb	-2.08	-1.50	-1.74
N_2S_2-adj	Cr	-0.03	0.24	2.88	**N_1S_3**	Mo	-0.85	0.50	2.30
N_2S_2-adj	Mn	0.83	1.32	3.71	**N_1S_3**	Tc	-0.74	1.24	2.31
N_2S_2-adj	Fe	0.67	2.01	3.62	**N_1S_3**	Ru	0.08	0.56	2.72
N_2S_2-adj	Co	1.03	2.51	3.96	**N_1S_3**	Rh	1.06	2.64	3.83
N_2S_2-adj	Ni	1.63	3.85	4.51	**N_1S_3**	Pd	2.02	4.32	4.70
N_2S_2-adj	Cu	1.51	1.72	4.46	**N_1S_3**	Ag	1.80	1.41	4.68
N_2S_2-adj	Zn	1.15	0.84	4.24	**N_1S_3**	Hf	-1.02	-0.03	2.09
N_2S_2-adj	Y	-1.26	1.12	1.71	**N_1S_3**	Ta	-1.70	-0.82	1.74
N_2S_2-adj	Zr	-2.10	-1.01	1.39	**N_1S_3**	W	-1.55	-1.59	1.67
N_2S_2-adj	Nb	-2.12	-1.76	1.01	**N_1S_3**	Re	-1.49	-1.31	1.76
N_2S_2-adj	Mo	-1.22	-1.30	1.93	**N_1S_3**	Os	-0.79	-1.32	3.03
N_2S_2-adj	Tc	-0.86	-0.88	2.07	**S_4**	Sc	1.26	2.08	4.84
N_2S_2-adj	Ru	0.04	0.73	2.93	**S_4**	Ti	-0.46	0.13	2.63
N_2S_2-adj	Rh	1.01	2.69	3.81	**S_4**	V	-1.22	-0.60	2.03
N_2S_2-adj	Pd	1.99	2.12	4.78	**S_4**	Cr	-0.24	0.12	2.85
N_2S_2-adj	Ag	1.70	1.95	4.48	**S_4**	Mn	1.00	1.98	4.05
N_2S_2-adj	Cd	1.71	1.38	4.55	**S_4**	Fe	0.34	1.43	3.20
N_2S_2-adj	Hf	-1.30	-0.22	1.73	**S_4**	Co	1.10	2.50	3.97
N_2S_2-adj	Ta	-1.66	-1.17	1.37	**S_4**	Ni	1.81	4.01	4.58
N_2S_2-adj	W	-1.51	-1.57	1.67	**S_4**	Y	-1.15	-0.22	2.06
N_2S_2-adj	Re	-1.43	-1.36	1.71	**S_4**	Zr	-0.84	-0.01	2.34
N_2S_2-adj	Os	-0.47	-0.10	1.96	**S_4**	Nb	-1.20	0.02	1.88
N_2S_2-sep	Sc	-1.08	1.02	2.10	**S_4**	Mo	-1.51	-0.10	1.81
N_2S_2-sep	Ti	-0.69	0.18	2.58	**S_4**	Tc	-1.09	-0.88	2.04
N_2S_2-sep	V	-0.88	-0.55	2.13	**S_4**	Ru	-2.07	-1.51	0.91
N_2S_2-sep	Cr	-0.10	0.27	2.74	**S_4**	Rh	1.16	2.69	4.01
N_2S_2-sep	Mn	0.85	1.32	3.71	**S_4**	Hf	-0.97	0.45	2.17
N_2S_2-sep	Fe	0.69	1.88	3.56	**S_4**	Ta	-1.63	-0.09	1.57
N_2S_2-sep	Co	1.01	2.45	3.92	**S_4**	W	-1.38	-1.13	2.14
N_2S_2-sep	Ni	1.63	3.79	4.38	**S_4**	Re	-1.44	-1.18	1.80
N_2S_2-sep	Cu	1.70	1.76	4.68	**S_4**	Os	-0.23	-0.23	2.78

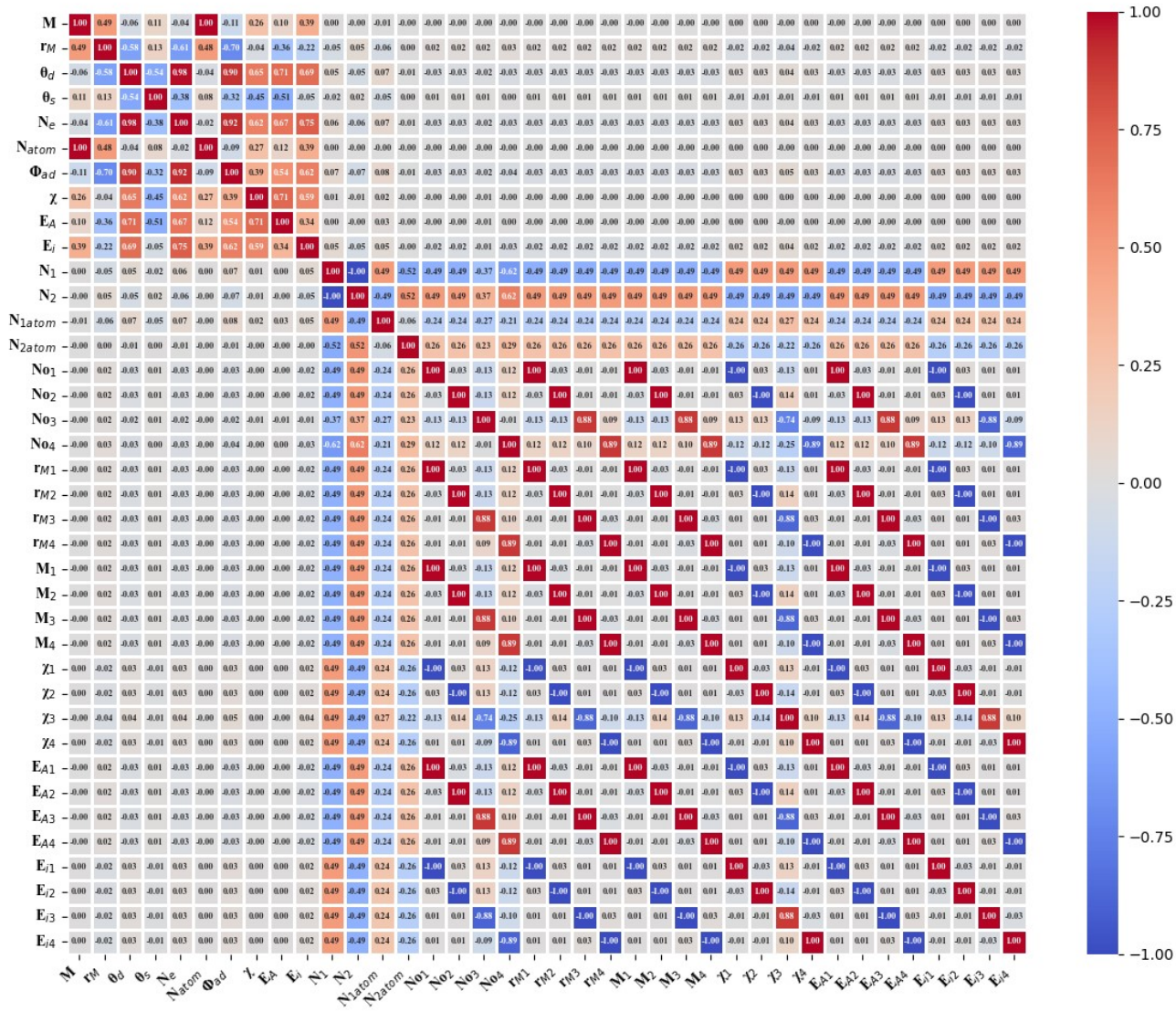


Figure S1. Heat map of Pearson correlation coefficients for different features before feature selection.

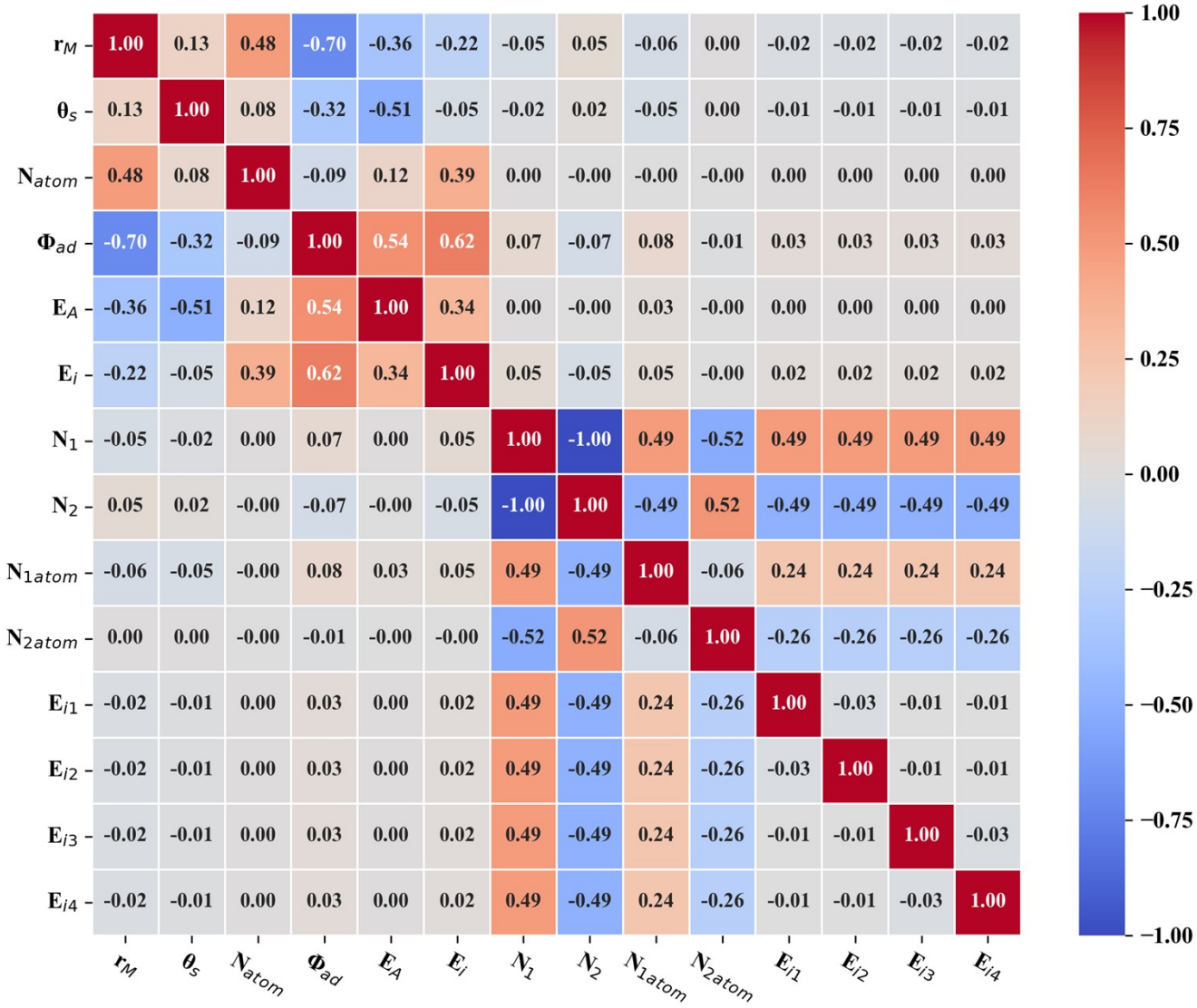


Figure S2. Heat map of Pearson correlation coefficients for different features after feature selection.

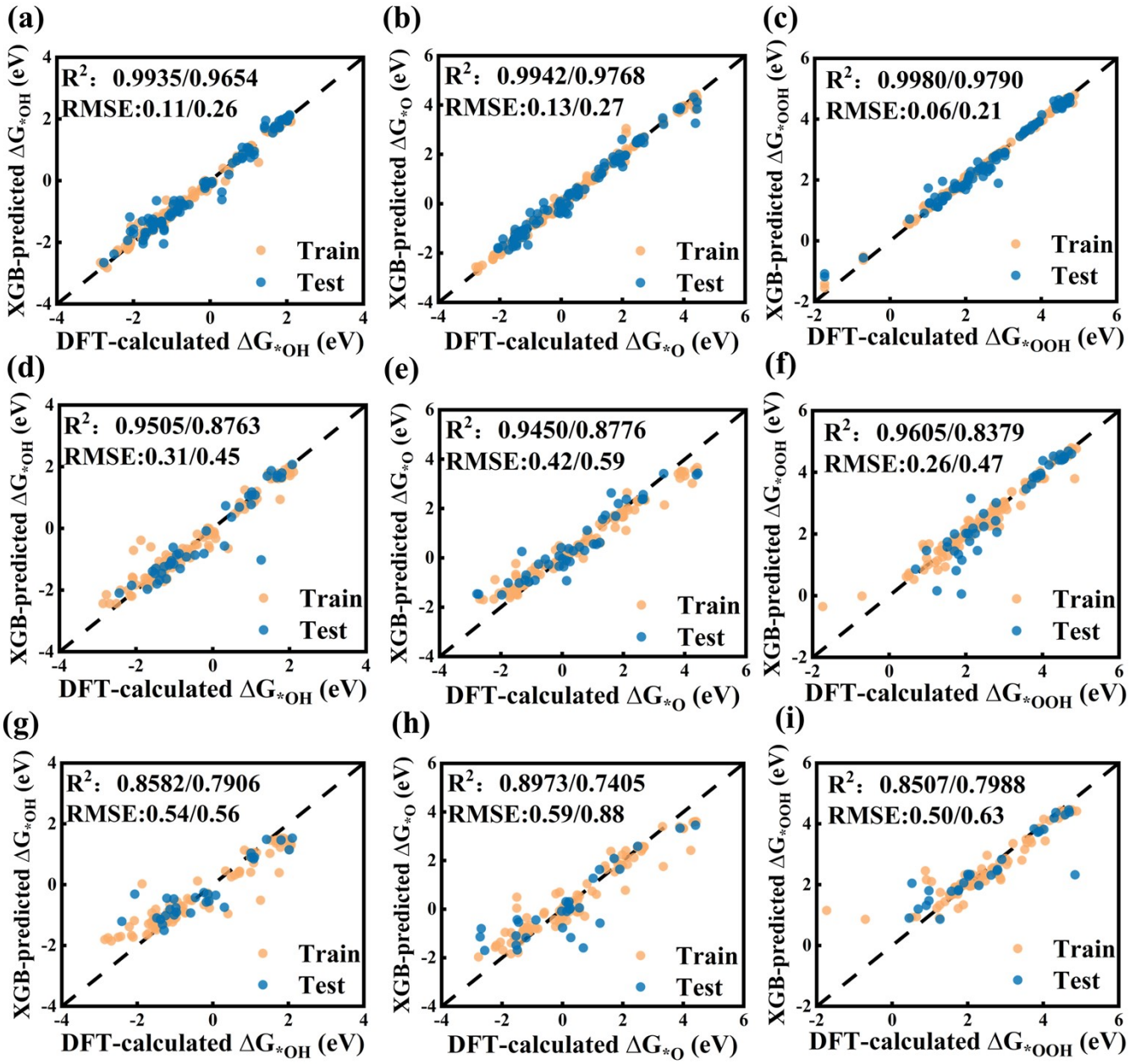


Figure S3. Comparison of DFT-calculated (a) ΔG_{OH^*}, (b) ΔG_{O^*}, and (c) ΔG_{OOH^*} values with those predicted by the best performing XGB models in the dataset 3. Comparison of DFT-calculated (d) ΔG_{OH^*}, (e) ΔG_{O^*}, and (f) ΔG_{OOH^*} values with those predicted by the best performing XGB models in the dataset 2. Comparison of DFT-calculated (g) ΔG_{OH^*}, (h) ΔG_{O^*}, and (i) ΔG_{OOH^*} values with those predicted by the best performing XGB models in the dataset 1.

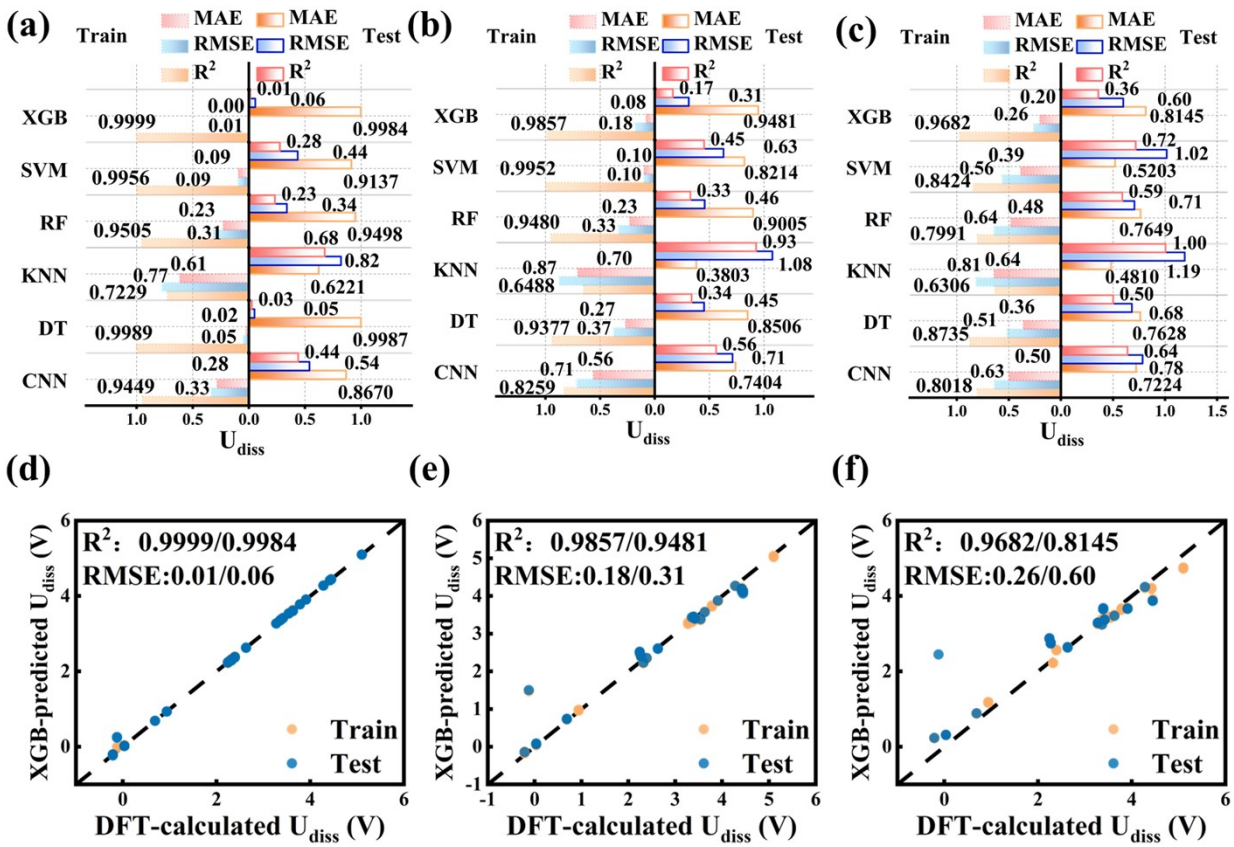
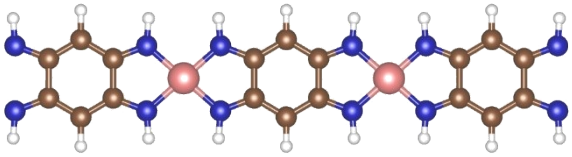
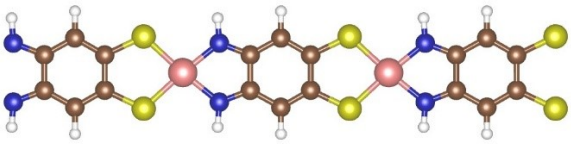
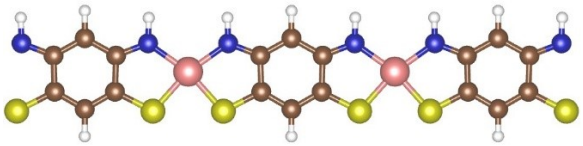
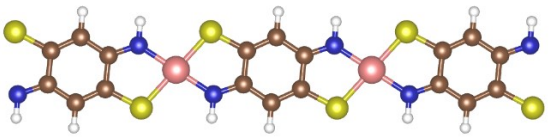
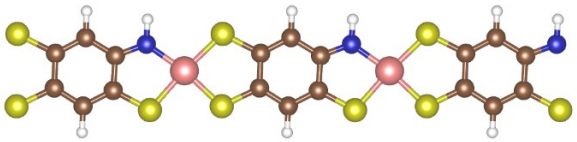
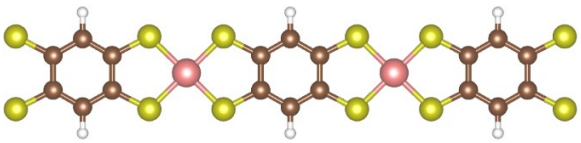
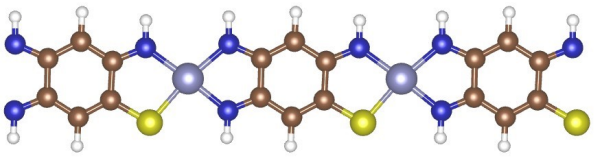
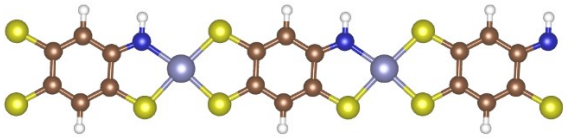
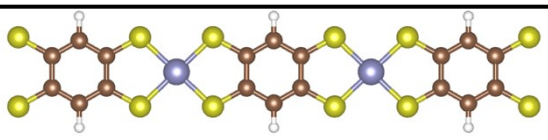
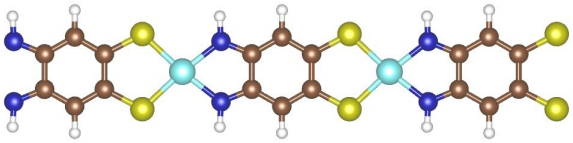
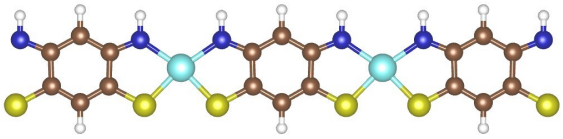
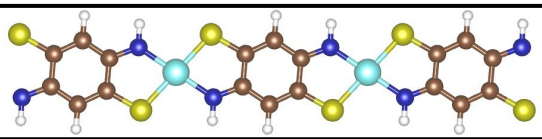
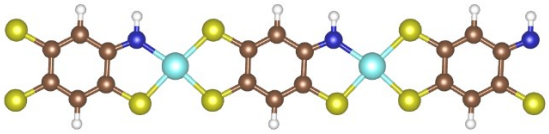


Figure S4. RMSE, MAE, and the R^2 score of each model for U_{diss} in (a) the dataset 3, (b) the dataset 2, and (c) thedataset 1. Comparison of DFT-calculated U_{diss} values with those predicted by the best performing XGB models in (d) the third dataset, (e) the second dataset, and (f) the first dataset.

Table S5 The optimized structures and overpotentials of 13 excellent catalysts.

$M\text{-}N_xS_y$	Structures	Overpotential
$Co\text{-}N_4$		0.30 V
$Co\text{-}N_2S_2\text{-adj}$		0.25 V
$Co\text{-}N_2S_2\text{-sep}$		0.24 V
$Co\text{-}N_2S_2\text{-opp}$		0.26V
$Co\text{-}N_1S_3$		0.25 V
$Co\text{-}S_4$		0.24 V
$Rh\text{-}N_3S_1$		0.37 V
$Rh\text{-}N_1S_3$		0.35 V
$Rh\text{-}S_4$		0.31 V
$Ir\text{-}N_2S_2\text{-adj}$		0.31 V

Ir-N_2S_2-sep		0.25 V
Ir-N_2S_2-opp		0.37 V
Ir-N_3S_1		0.35 V

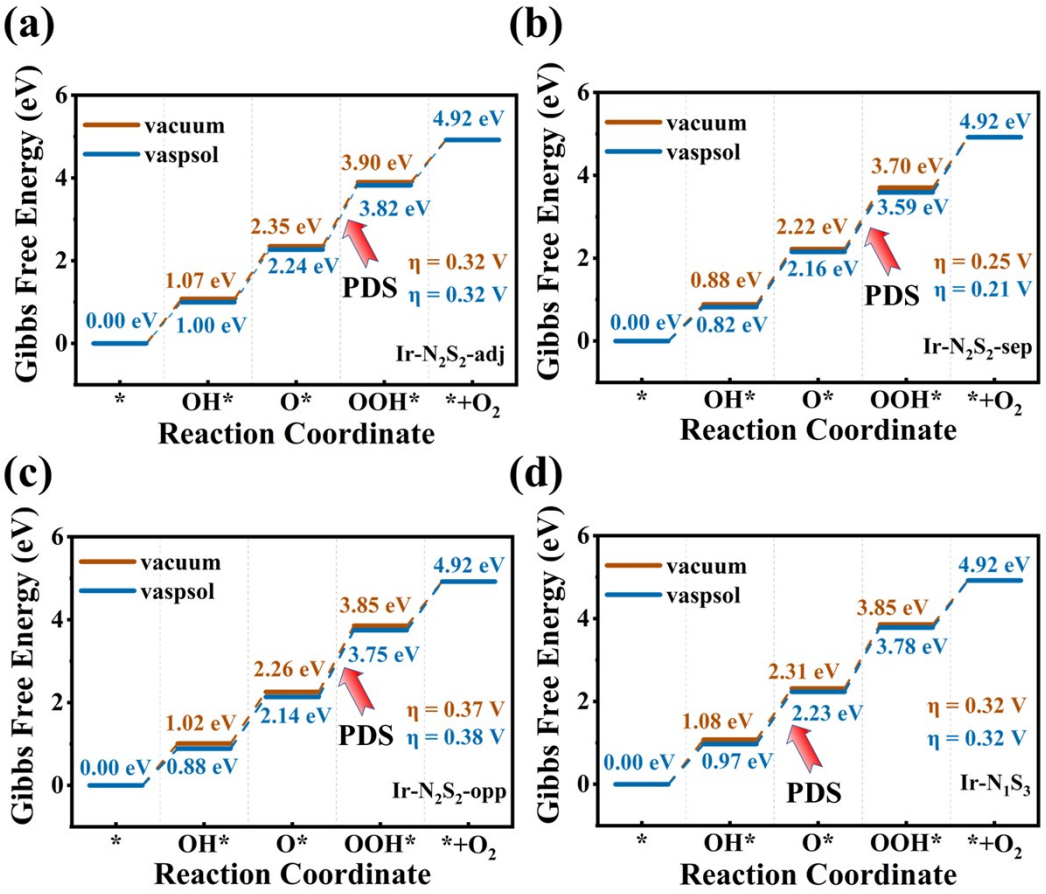


Figure S5. Gibbs free energy change diagrams of (a)Ir-N_2S_2-adj, (b)Ir-N_2S_2-sep, (c)Ir-N_2S_2-opp, and (d)Ir-N_1S_3, along the AEM pathway under vacuum and implicit solvation.

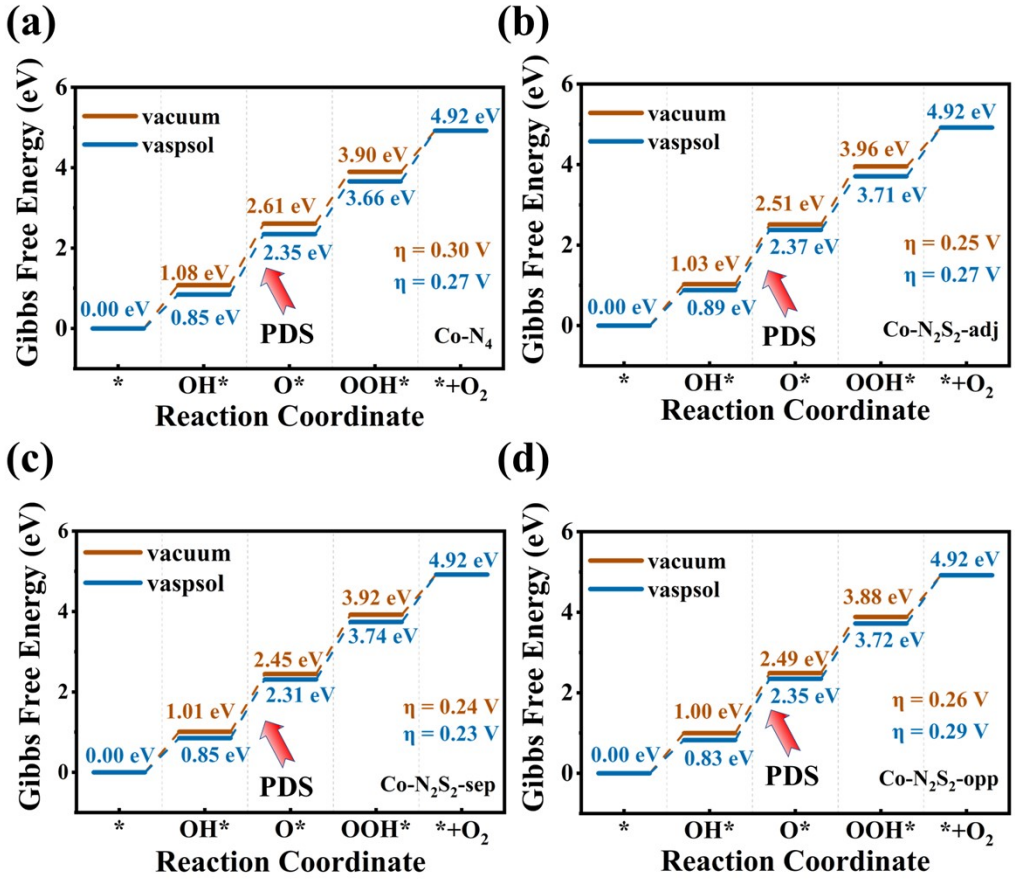


Figure S6. Gibbs free energy change diagrams of (a)Co-N_4, (b)Co-N_2S_2-adj, (c)Co-N_2S_2-sep, and (d)Co-N_2S_2-opp, along the AEM pathway under vacuum and implicit solvation.

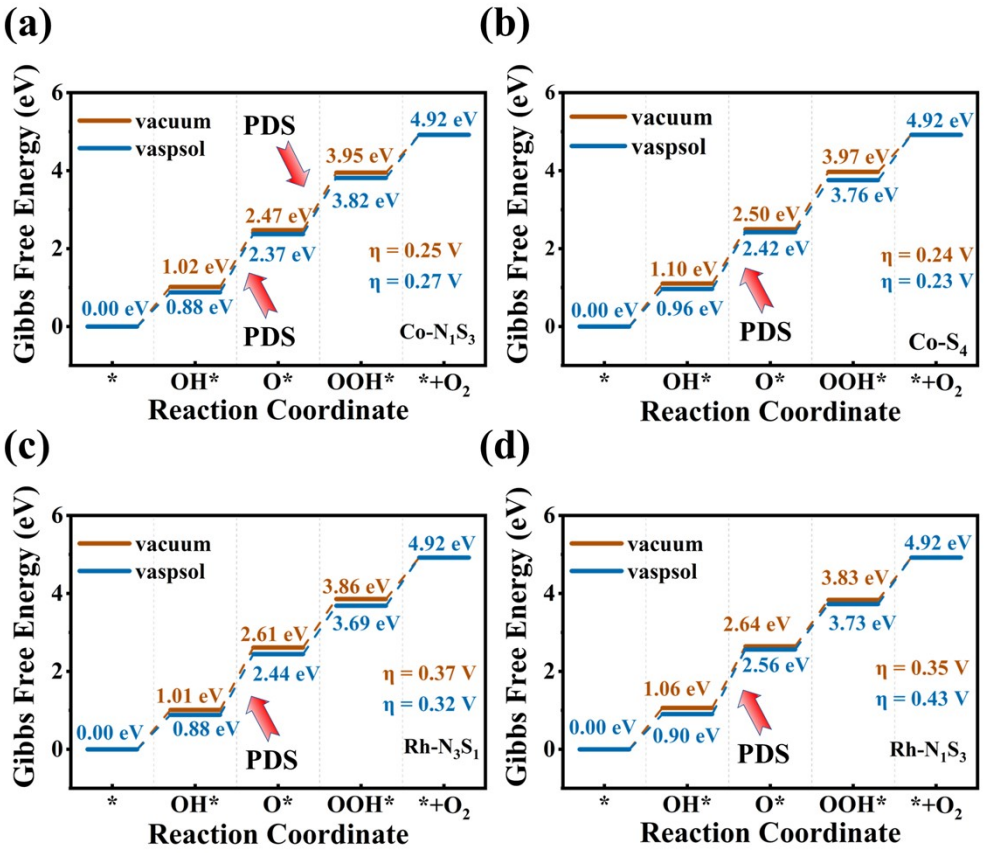


Figure S7. Gibbs free energy change diagrams of (a)Co-N_1S_3, (b)Co-S_4, (c)Rh-N_3S_1, and (d)Rh-N_1S_3, along the AEM pathway under vacuum and implicit solvation.

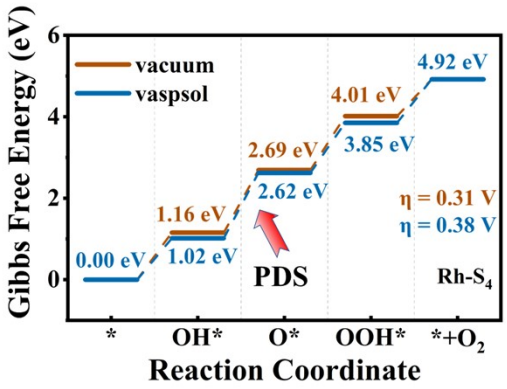


Figure S8. Gibbs free energy change diagrams of Rh-S_4, along the AEM pathway under vacuum and implicit solvation.

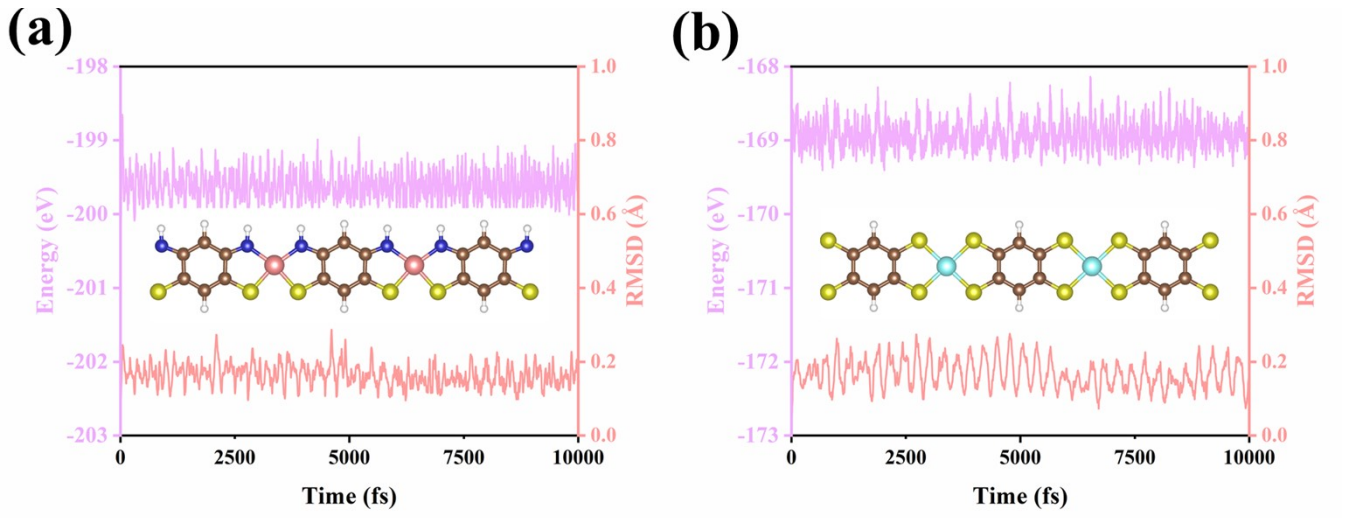


Figure S9. Ab initio molecular dynamics (AIMD) simulations and root-mean-square deviations (RMSDs) of (a)Co-N_2S_2-ortho and (b)Co-S_4.