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Supporting Information

Directly Predicting Limiting Potentials from Easily Obtainable Physical Properties of Graphene-Supported Single-Atom Electrocatalysts by Machine Learning

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Data pretreatment. For the pretreatment of data, the data were divided into training data and test data randomly with a ratio around 0.85 for training data. Therefore, 104 input data were split into 88 training set and 16 test set for ORR/HER models. For OER model, the small data set for M-N-pyrrole systems affect the training performance, thus, the small data set (with 26 data) was amplified three times with random noises. Totally, 156 input data were produced for OER model, which were divided into 135 training data and 21 validation data.

DFT computations for catalysis performance. The changes of Gibbs free energy (ΔG) for each elementary step along the ORR/OER were evaluated using the computational hydrogen electrode (CHE) model developed by Nørskov and co-workers.[1] The computed electronic energy was converted into Gibbs free energy by adding zero-point energy. The energy of the triplet O_2 molecule cannot be accurately computed by the DFT method[2,3] thus, its free energy was computed relative to the free energies of $H_2O(l)$ and $H_2(g)$. The chemical potential of the H^+/e^- pair is equal to half of the gas-phase H_2 at standard hydrogen electrode (SHE) conditions.

Github website link. The three well-trained models were uploaded to Github: https://github.com/shirulin1026/ML_screening_ORROERHER, which is public now. The model files can be downloaded and employed for predictions through scikit-learn package. Training data and prediction data were also uploaded in the same folder on Github.

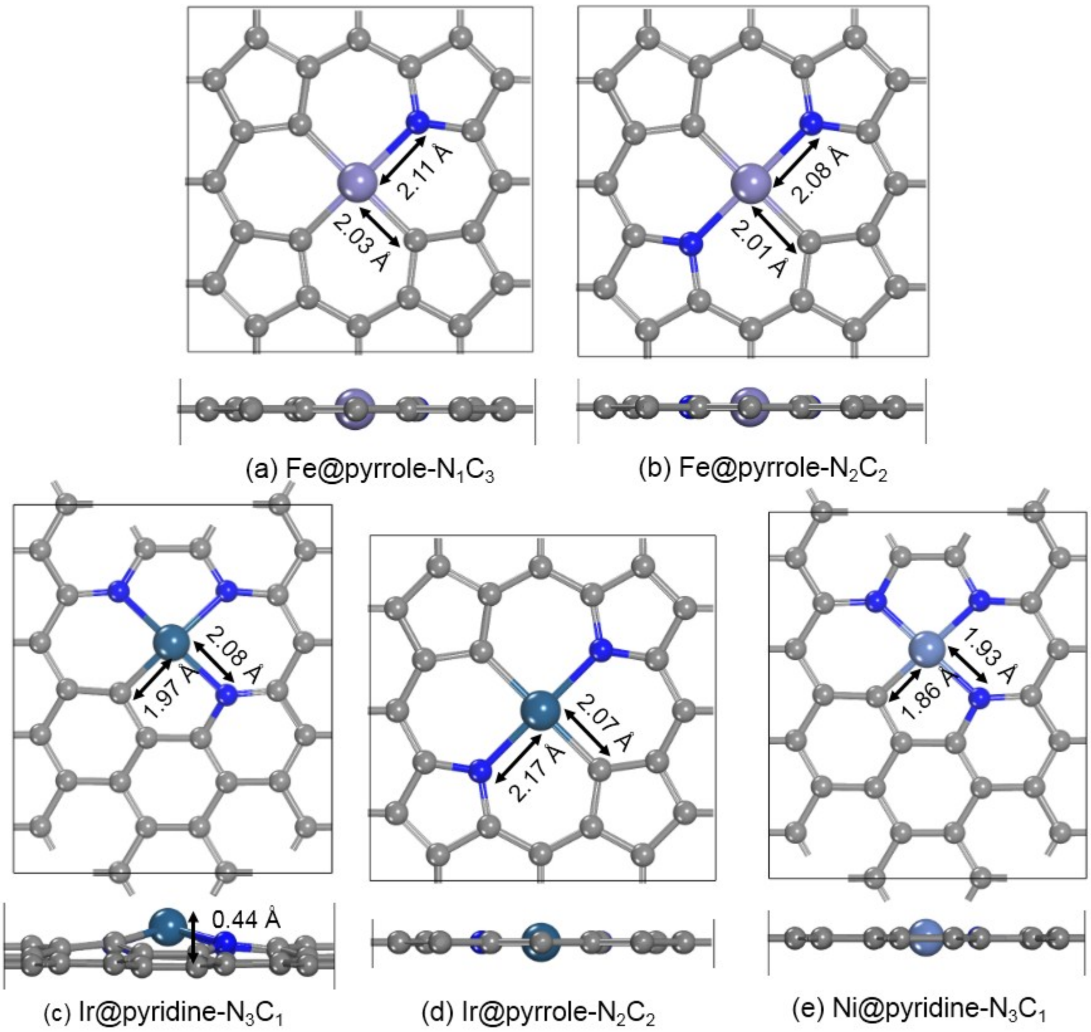

Figure S1. The geometries of ML-selected best SACs, (a, b) for ORR, (c, d) for OER, and (e) for HER.

Table S1. The structural compositions of 260 other graphene-supported SACs ($M@N_xC_y$) and their limiting potentials towards ORR/OER/HER predicted by ML models.

Num	Metal	Base	ORR	OER	HER
1	Sc	C_2N_1	-2.19	3.78	-2.16
2	Ti	C_2N_1	-1.85	3.25	-1.90
3	V	C_2N_1	-1.31	3.00	-1.35
4	Cr	C_2N_1	-0.98	3.05	-0.83
5	Mn	C_2N_1	-0.99	2.80	-0.96

Num	Metal	Base	ORR	OER	HER
6	Fe	C_2N_1	-0.53	3.14	-0.55
7	Co	C_2N_1	-0.24	3.05	-0.35
8	Ni	C_2N_1	-0.14	3.01	-0.17
9	Cu	C_2N_1	0.19	2.55	0.08
10	Zn	C_2N_1	0.10	2.33	0.01
11	Y	C_2N_1	-2.18	3.61	-2.03
12	Zr	C_2N_1	-1.77	3.17	-1.72
13	Nb	C_2N_1	-1.36	3.26	-1.42
14	Mo	C_2N_1	-0.76	3.48	-0.75
15	Tc	C_2N_1	-0.52	3.35	-0.41
16	Ru	C_2N_1	-0.17	2.83	-0.09
17	Rh	C_2N_1	-0.01	2.62	-0.04
18	Pd	C_2N_1	0.28	2.58	0.34
19	Cd	C_2N_1	0.19	2.27	0.20
20	Hf	C_2N_1	-1.86	3.39	-1.83
21	Ta	C_2N_1	-1.65	3.24	-1.66
22	W	C_2N_1	-0.93	3.16	-1.16
23	Re	C_2N_1	-0.99	3.07	-0.71
24	Os	C_2N_1	-0.32	3.42	-0.39
25	Ir	C_2N_1	-0.17	3.05	-0.11
26	Pt	C_2N_1	0.17	2.68	0.07
27	Sc	C_1N_2	-2.22	3.76	-2.15
28	Ti	C_1N_2	-1.80	3.20	-1.88
29	V	C_1N_2	-1.20	2.92	-1.27
30	Cr	C_1N_2	-0.65	2.82	-0.57
31	Mn	C_1N_2	-0.67	2.65	-0.77
32	Fe	C_1N_2	-0.34	2.76	-0.44
33	Co	C_1N_2	-0.13	2.70	-0.26
34	Ni	C_1N_2	-0.18	2.76	-0.18
35	Cu	C_1N_2	-0.07	2.53	-0.06
36	Zn	C_1N_2	-0.25	2.47	-0.22
37	Y	C_1N_2	-2.22	3.79	-2.02
38	Zr	C_1N_2	-1.75	3.18	-1.71
39	Nb	C_1N_2	-1.12	3.35	-1.20
40	Mo	C_1N_2	-0.45	3.02	-0.49
41	Tc	C_1N_2	-0.31	3.00	-0.27
42	Ru	C_1N_2	-0.08	2.53	-0.07
43	Rh	C_1N_2	-0.12	2.49	-0.09
44	Pd	C_1N_2	-0.05	2.72	-0.01
45	Cd	C_1N_2	-0.26	2.48	-0.10
46	Hf	C_1N_2	-1.85	3.39	-1.82
47	Ta	C_1N_2	-1.57	3.06	-1.60
48	W	C_1N_2	-0.68	3.29	-0.87

Num	Metal	Base	ORR	OER	HER
49	Re	C_1N_2	-0.59	2.72	-0.44
50	Os	C_1N_2	-0.18	2.88	-0.28
51	Ir	C_1N_2	-0.08	2.62	-0.06
52	Pt	C_1N_2	-0.04	2.59	0.02
53	Sc	N_3	-2.15	3.72	-2.17
54	Ti	N_3	-1.80	3.26	-1.86
55	V	N_3	-1.22	3.03	-1.29
56	Cr	N_3	-0.62	3.32	-0.48
57	Mn	N_3	-0.64	2.85	-0.62
58	Fe	N_3	-0.24	2.39	-0.34
59	Co	N_3	-0.05	1.93	-0.18
60	Ni	N_3	0.00	1.86	-0.08
61	Cu	N_3	0.19	1.89	0.11
62	Zn	N_3	0.03	2.03	-0.02
63	Y	N_3	-2.13	3.67	-2.02
64	Zr	N_3	-1.71	3.23	-1.67
65	Nb	N_3	-1.17	3.25	-1.24
66	Mo	N_3	-0.51	3.25	-0.51
67	Tc	N_3	-0.22	2.83	-0.26
68	Ru	N_3	-0.06	2.09	-0.04
69	Rh	N_3	-0.03	1.99	0.02
70	Pd	N_3	0.10	2.34	0.16
71	Cd	N_3	0.10	2.03	0.11
72	Hf	N_3	-1.83	3.25	-1.78
73	Ta	N_3	-1.57	3.13	-1.59
74	W	N_3	-0.75	3.36	-0.93
75	Re	N_3	-0.67	3.45	-0.45
76	Os	N_3	-0.16	2.56	-0.27
77	Ir	N_3	-0.07	2.16	-0.05
78	Pt	N_3	0.05	2.25	0.06
79	Sc	pyridine-C_3N_1	-2.16	3.73	-2.11
80	Ti	pyridine-C_3N_1	-1.57	3.17	-1.56
81	V	pyridine-C_3N_1	-1.12	3.02	-1.19
82	Cr	pyridine-C_3N_1	-0.60	3.13	-0.58
83	Mn	pyridine-C_3N_1	-0.48	2.73	-0.47
84	Fe	pyridine-C_3N_1	0.12	2.50	0.29
85	Co	pyridine-C_3N_1	0.31	2.04	0.36
86	Ni	pyridine-C_3N_1	0.41	2.06	0.49
87	Cu	pyridine-C_3N_1	0.50	2.13	0.56
88	Zn	pyridine-C_3N_1	0.34	2.16	0.39
89	Y	pyridine-C_3N_1	-2.17	3.81	-1.97
90	Zr	pyridine-C_3N_1	-1.57	3.23	-1.60
91	Nb	pyridine-C_3N_1	-1.03	3.25	-1.05

Num	Metal	Base	ORR	OER	HER
92	Mo	pyridine-C_3N_1	-0.43	3.06	-0.50
93	Tc	pyridine-C_3N_1	0.10	2.75	0.05
94	Ru	pyridine-C_3N_1	0.27	2.19	0.36
95	Rh	pyridine-C_3N_1	0.38	2.16	0.54
96	Pd	pyridine-C_3N_1	0.45	2.49	0.30
97	Cd	pyridine-C_3N_1	0.28	2.34	0.33
98	Hf	pyridine-C_3N_1	-1.56	3.29	-1.57
99	Ta	pyridine-C_3N_1	-1.32	3.09	-1.31
100	W	pyridine-C_3N_1	-0.82	3.32	-0.87
101	Re	pyridine-C_3N_1	-0.64	2.97	-0.48
102	Os	pyridine-C_3N_1	0.18	2.54	0.19
103	Ir	pyridine-C_3N_1	0.27	2.24	0.30
104	Pt	pyridine-C_3N_1	0.41	2.22	0.61
105	Sc	pyridine-C_2N_2	-2.20	3.73	-2.11
106	Ti	pyridine-C_2N_2	-1.56	3.17	-1.54
107	V	pyridine-C_2N_2	-0.98	3.02	-1.12
108	Cr	pyridine-C_2N_2	-0.15	3.13	-0.14
109	Mn	pyridine-C_2N_2	-0.11	2.73	-0.28
110	Fe	pyridine-C_2N_2	0.31	2.50	0.47
111	Co	pyridine-C_2N_2	0.38	2.04	0.44
112	Ni	pyridine-C_2N_2	0.23	2.06	0.20
113	Cu	pyridine-C_2N_2	0.04	2.13	0.05
114	Zn	pyridine-C_2N_2	-0.21	2.16	-0.29
115	Y	pyridine-C_2N_2	-2.20	3.81	-1.97
116	Zr	pyridine-C_2N_2	-1.59	3.23	-1.57
117	Nb	pyridine-C_2N_2	-0.83	3.25	-0.82
118	Mo	pyridine-C_2N_2	0.02	3.06	-0.01
119	Tc	pyridine-C_2N_2	0.30	2.75	0.24
120	Ru	pyridine-C_2N_2	0.26	2.19	0.38
121	Rh	pyridine-C_2N_2	0.07	2.16	0.18
122	Pd	pyridine-C_2N_2	-0.11	2.49	-0.37
123	Cd	pyridine-C_2N_2	-0.26	2.34	-0.36
124	Hf	pyridine-C_2N_2	-1.58	3.29	-1.59
125	Ta	pyridine-C_2N_2	-1.26	3.09	-1.24
126	W	pyridine-C_2N_2	-0.55	3.32	-0.61
127	Re	pyridine-C_2N_2	-0.16	2.97	0.01
128	Os	pyridine-C_2N_2	0.24	2.54	0.39
129	Ir	pyridine-C_2N_2	0.26	2.24	0.39
130	Pt	pyridine-C_2N_2	-0.04	2.22	0.25
131	Sc	pyridine-C_1N_3	-2.26	3.73	-2.11
132	Ti	pyridine-C_1N_3	-1.54	3.13	-1.52
133	V	pyridine-C_1N_3	-0.86	2.83	-1.03
134	Cr	pyridine-C_1N_3	0.28	2.60	0.25

Num	Metal	Base	ORR	OER	HER
135	Mn	pyridine-C_1N_3	0.30	2.46	-0.05
136	Fe	pyridine-C_1N_3	0.55	2.04	0.64
137	Co	pyridine-C_1N_3	0.47	1.66	0.54
138	Ni	pyridine-C_1N_3	0.03	1.94	-0.03
139	Cu	pyridine-C_1N_3	-0.44	2.32	-0.36
140	Zn	pyridine-C_1N_3	-0.81	2.65	-0.83
141	Y	pyridine-C_1N_3	-2.25	3.98	-1.97
142	Zr	pyridine-C_1N_3	-1.62	3.19	-1.54
143	Nb	pyridine-C_1N_3	-0.58	3.43	-0.58
144	Mo	pyridine-C_1N_3	0.44	2.51	0.41
145	Tc	pyridine-C_1N_3	0.55	2.41	0.46
146	Ru	pyridine-C_1N_3	0.25	1.69	0.38
147	Rh	pyridine-C_1N_3	-0.28	2.04	-0.12
148	Pd	pyridine-C_1N_3	-0.67	2.99	-0.97
149	Cd	pyridine-C_1N_3	-0.92	2.88	-0.93
150	Hf	pyridine-C_1N_3	-1.60	3.35	-1.59
151	Ta	pyridine-C_1N_3	-1.17	2.86	-1.18
152	W	pyridine-C_1N_3	-0.29	3.45	-0.32
153	Re	pyridine-C_1N_3	0.30	2.37	0.46
154	Os	pyridine-C_1N_3	0.30	1.96	0.60
155	Ir	pyridine-C_1N_3	0.25	1.64	0.47
156	Pt	pyridine-C_1N_3	-0.50	2.28	-0.07
157	Sc	pyrrole-C_4	-2.22	3.72	-2.11
158	Ti	pyrrole-C_4	-1.55	3.17	-1.54
159	V	pyrrole-C_4	-1.00	3.00	-1.11
160	Cr	pyrrole-C_4	-0.16	3.08	-0.18
161	Mn	pyrrole-C_4	-0.07	2.68	-0.24
162	Fe	pyrrole-C_4	0.36	2.40	0.46
163	Co	pyrrole-C_4	0.40	1.97	0.47
164	Ni	pyrrole-C_4	0.21	2.02	0.26
165	Cu	pyrrole-C_4	0.02	2.15	0.14
166	Zn	pyrrole-C_4	-0.26	2.27	-0.16
167	Y	pyrrole-C_4	-2.22	3.83	-1.97
168	Zr	pyrrole-C_4	-1.59	3.24	-1.57
169	Nb	pyrrole-C_4	-0.78	3.26	-0.81
170	Mo	pyrrole-C_4	-0.01	2.99	-0.08
171	Tc	pyrrole-C_4	0.34	2.71	0.27
172	Ru	pyrrole-C_4	0.26	2.14	0.36
173	Rh	pyrrole-C_4	0.03	2.14	0.25
174	Pd	pyrrole-C_4	-0.11	2.58	-0.30
175	Cd	pyrrole-C_4	-0.38	2.45	-0.23
176	Hf	pyrrole-C_4	-1.58	3.28	-1.58
177	Ta	pyrrole-C_4	-1.24	3.03	-1.25

Num	Metal	Base	ORR	OER	HER
178	W	pyrrole-C_4	-0.55	3.34	-0.59
179	Re	pyrrole-C_4	-0.18	2.92	-0.03
180	Os	pyrrole-C_4	0.25	2.42	0.40
181	Ir	pyrrole-C_4	0.27	2.13	0.38
182	Pt	pyrrole-C_4	-0.05	2.23	0.30
183	Sc	pyrrole-C_3N_1	-2.22	3.73	-2.11
184	Ti	pyrrole-C_3N_1	-1.55	3.13	-1.54
185	V	pyrrole-C_3N_1	-1.00	2.83	-1.11
186	Cr	pyrrole-C_3N_1	-0.16	2.60	-0.18
187	Mn	pyrrole-C_3N_1	-0.07	2.46	-0.24
188	Fe	pyrrole-C_3N_1	0.36	2.04	0.46
189	Co	pyrrole-C_3N_1	0.40	1.66	0.47
190	Ni	pyrrole-C_3N_1	0.21	1.94	0.26
191	Cu	pyrrole-C_3N_1	0.02	2.32	0.14
192	Zn	pyrrole-C_3N_1	-0.26	2.65	-0.16
193	Y	pyrrole-C_3N_1	-2.22	3.98	-1.97
194	Zr	pyrrole-C_3N_1	-1.59	3.19	-1.57
195	Nb	pyrrole-C_3N_1	-0.78	3.43	-0.81
196	Mo	pyrrole-C_3N_1	-0.01	2.51	-0.08
197	Tc	pyrrole-C_3N_1	0.34	2.41	0.27
198	Ru	pyrrole-C_3N_1	0.26	1.69	0.36
199	Rh	pyrrole-C_3N_1	0.03	2.04	0.25
200	Pd	pyrrole-C_3N_1	-0.11	2.99	-0.30
201	Cd	pyrrole-C_3N_1	-0.38	2.88	-0.23
202	Hf	pyrrole-C_3N_1	-1.58	3.35	-1.58
203	Ta	pyrrole-C_3N_1	-1.24	2.86	-1.25
204	W	pyrrole-C_3N_1	-0.55	3.45	-0.59
205	Re	pyrrole-C_3N_1	-0.18	2.37	-0.03
206	Os	pyrrole-C_3N_1	0.25	1.96	0.40
207	Ir	pyrrole-C_3N_1	0.27	1.64	0.38
208	Pt	pyrrole-C_3N_1	-0.05	2.28	0.30
209	Sc	pyrrole-C_2N_2	-2.26	3.73	-2.11
210	Ti	pyrrole-C_2N_2	-1.54	3.13	-1.52
211	V	pyrrole-C_2N_2	-0.86	2.83	-1.03
212	Cr	pyrrole-C_2N_2	0.28	2.60	0.25
213	Mn	pyrrole-C_2N_2	0.30	2.46	-0.05
214	Fe	pyrrole-C_2N_2	0.55	2.04	0.64
215	Co	pyrrole-C_2N_2	0.47	1.66	0.54
216	Ni	pyrrole-C_2N_2	0.03	1.94	-0.03
217	Cu	pyrrole-C_2N_2	-0.44	2.32	-0.36
218	Zn	pyrrole-C_2N_2	-0.81	2.65	-0.83
219	Y	pyrrole-C_2N_2	-2.25	3.98	-1.97
220	Zr	pyrrole-C_2N_2	-1.62	3.19	-1.54

Num	Metal	Base	ORR	OER	HER
221	Nb	pyrrole-C_2N_2	-0.58	3.43	-0.58
222	Mo	pyrrole-C_2N_2	0.44	2.51	0.41
223	Tc	pyrrole-C_2N_2	0.55	2.41	0.46
224	Ru	pyrrole-C_2N_2	0.25	1.69	0.38
225	Rh	pyrrole-C_2N_2	-0.28	2.04	-0.12
226	Pd	pyrrole-C_2N_2	-0.67	2.99	-0.97
227	Cd	pyrrole-C_2N_2	-0.92	2.88	-0.93
228	Hf	pyrrole-C_2N_2	-1.60	3.35	-1.59
229	Ta	pyrrole-C_2N_2	-1.17	2.86	-1.18
230	W	pyrrole-C_2N_2	-0.29	3.45	-0.32
231	Re	pyrrole-C_2N_2	0.30	2.37	0.46
232	Os	pyrrole-C_2N_2	0.30	1.96	0.60
233	Ir	pyrrole-C_2N_2	0.25	1.64	0.47
234	Pt	pyrrole-C_2N_2	-0.50	2.28	-0.07
235	Sc	pyrrole-C_1N_3	-2.26	3.73	-2.11
236	Ti	pyrrole-C_1N_3	-1.54	3.13	-1.52
237	V	pyrrole-C_1N_3	-0.86	2.83	-1.03
238	Cr	pyrrole-C_1N_3	0.28	2.60	0.25
239	Mn	pyrrole-C_1N_3	0.30	2.46	-0.05
240	Fe	pyrrole-C_1N_3	0.55	2.04	0.64
241	Co	pyrrole-C_1N_3	0.47	1.66	0.54
242	Ni	pyrrole-C_1N_3	0.03	1.94	-0.03
243	Cu	pyrrole-C_1N_3	-0.44	2.32	-0.36
244	Zn	pyrrole-C_1N_3	-0.81	2.65	-0.83
245	Y	pyrrole-C_1N_3	-2.25	3.98	-1.97
246	Zr	pyrrole-C_1N_3	-1.62	3.19	-1.54
247	Nb	pyrrole-C_1N_3	-0.58	3.43	-0.58
248	Mo	pyrrole-C_1N_3	0.44	2.51	0.41
249	Tc	pyrrole-C_1N_3	0.55	2.41	0.46
250	Ru	pyrrole-C_1N_3	0.25	1.69	0.38
251	Rh	pyrrole-C_1N_3	-0.28	2.04	-0.12
252	Pd	pyrrole-C_1N_3	-0.67	2.99	-0.97
253	Cd	pyrrole-C_1N_3	-0.92	2.88	-0.93
254	Hf	pyrrole-C_1N_3	-1.60	3.35	-1.59
255	Ta	pyrrole-C_1N_3	-1.17	2.86	-1.18
256	W	pyrrole-C_1N_3	-0.29	3.45	-0.32
257	Re	pyrrole-C_1N_3	0.30	2.37	0.46
258	Os	pyrrole-C_1N_3	0.30	1.96	0.60
259	Ir	pyrrole-C_1N_3	0.25	1.64	0.47
260	Pt	pyrrole-C_1N_3	-0.50	2.28	-0.07

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