Electronic Supplementary Information (ESI)

Accelerating High-Throughput Screening of Hydrogen Peroxide Production via DFT and Machine Learning

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Contents

Fig. S3. Illustration of the reactions and the excellent catalyst during ORR reaction.

Fig. S4. Partial density of states (PDOS) of d orbitals for 21 $2e^-$ ORR catalysts screened out by the contour map. The Fermi level (E_F) is set to 0 eV.

Fig. S5. Band center of metal atom (ε_d) versus Gibbs free energy of O^* (a) and OOH^* (g) on different SACs. The values of ε_d and ΔG_{*O} on SACs with different N coordination (b-f).

Supporting Information Note 1. Additional computational details for SISSO

Table S15. The full list of primary features compiled from the literature before feature elimination.

Table S16. Atomic primary features obtained from the literature.

Fig. S6. Pearson correlation coeffcient between every pair of primary features from Table S15.

Table S17. The full list of primary features compiled from the literature after feature elimination.

Table S18. The features of electronic bands.

Fig. S7. (a) Importance of the selected features for target property of ΔG_{*OOH}. (b) Training RMSE from 5-fold cross validation (averaged over the 5 folds) for all tested SISSO hyperparameters. (d) Validation of the models constructed by the selected features for target property of ΔG_{*OOH}, shown by the values of the ML prediction and density functional theory (DFT) calculations.

Table S1. Computed formation energies (E_{bind}) of metal atoms at each substrate.

Metal	Formation energy (E_{bind}, eV)				
	MC4	**MC3N**	**MC2N2**	**MCN3**	**MN4**
Sc	-1.35	-3.40	-4.63	-4.57	-4.23
Ti	-1.91	-3.35	-4.18	-3.53	-2.98
V	-1.29	-2.47	-3.19	-2.70	-2.36
Cr	-1.04	-2.21	-2.92	-2.74	-2.69
Mn	-1.31	-2.76	-3.64	-3.13	-2.78
Fe	-0.95	-2.33	-3.14	-2.70	-2.39
Co	-1.02	-2.37	-3.26	-2.95	-2.58
Ni	-1.37	-3.00	-4.03	-3.54	-2.90
Cu	-1.41	-2.90	-3.42	-2.37	-1.72
Zn	-1.31	-2.09	-2.49	-2.33	-2.32
Y	-1.13	-3.12	-4.39	-4.35	-4.07
Zr	-1.85	-3.30	-4.16	-3.42	-2.65
Nb	-1.33	-2.48	-2.76	-1.89	-1.09
Mo	-0.55	-1.67	-1.84	-0.79	0.09
Tc	-0.85	-1.62	-1.70	-0.55	-0.03
Ru	-1.04	-1.52	-1.53	-0.82	-0.36
Rh	-1.04	-2.12	-2.37	-1.96	-1.66
Pd	-0.69	-2.09	-3.12	-2.67	-2.06
Ag	0.09	-1.38	-2.00	-0.94	0.50
Cd	0.34	-0.42	-0.71	-0.80	-0.66
Lu	-1.11	-3.17	-4.46	-4.40	-4.10
Hf	-1.69	-3.13	-3.97	-3.24	-2.52
Ta	-1.02	-2.14	-2.47	-1.55	-0.82
W	-0.11	-0.99	-1.22	-0.19	0.76
Re	-0.26	1.07	-0.95	0.27	1.01
Os	-0.54	-0.94	-0.86	-0.05	0.53
Ir	-1.01	-1.96	-2.24	-1.59	-1.11
Pt	-1.55	-2.73	-3.56	-2.88	-2.19
Au	-1.23	-2.45	-2.95	-1.74	-0.09
Hg	-0.07	-0.58	-0.55	0.20	0.12

Table. S2. The calculated electronic energy (E_{elect}, eV), zero-point energy (ZPE, eV), entropy (S, eV/K) and G (eV) at 298 K for reaction intermediates in ORR reaction.

	E_{elect}	**ZPE**	**-TS**	**G**	**S (eV/K)**
H_2 (g)	-6.80	0.27	-0.40	-6.93	0.001354 (ref.1)
H_2O (g)	-14.22	0.57	-0.58	-14.23	0.001957 (ref.1)
O*	-	0.07	-0.07	-	0.000247
OH*	-	0.31	-0.15	-	0.00051
OOH*	-	0.44	-0.21	-	0.000707

Ref. 1. NIST Chemistry WebBook, NIST Standard Reference Database Number 69, (Eds: P.J. Linstrom, W.G. Mallard), National Institute of Standards and Technology, Gaithersburg MD, p. 20899 (retrieved 2019).

Table. S3. Computed Gibbs free energies of O* (ΔG_{*O}) on catalyst surfaces. All free energies are computed relative to $H_2O(l)$ and $H_2(g)$.

Metal	ΔG_{*O} (eV)				
	MC4	**MC3N**	**MC2N2**	**MCN3**	**MN4**
Sc	0.46	1.46	1.70	0.97	0.13
Ti	0.39	-0.27	-0.94	-1.86	-2.44
V	-1.03	-1.65	-1.66	-2.04	-2.23
Cr	-0.86	-1.39	-1.37	7.83	-0.41
Mn	-0.44	-0.10	0.18	0.16	0.75
Fe	0.01	0.30	0.83	0.88	1.12
Co	0.94	1.18	1.56	1.71	2.46
Ni	2.31	2.86	3.39	3.51	4.00
Cu	4.04	4.19	4.23	0.99	3.82
Zn	3.99	3.41	3.39	3.09	3.07
Y	0.39	1.53	1.83	1.25	0.59
Zr	0.72	0.10	-0.48	-1.48	-2.34
Nb	-0.83	-1.51	-2.02	-2.60	-2.91
Mo	-1.48	-1.57	-1.98	-2.69	-
Tc	-0.89	-1.34	-1.87	-2.45	-1.80
Ru	-0.34	-0.78	-0.88	-0.40	0.39
Rh	0.66	0.94	1.32	1.85	2.42
Pd	2.55	2.86	3.75	3.97	4.43
Ag	-	4.39	4.39	4.31	-
Cd	-	3.14	2.73	2.60	2.58
Lu	0.93	1.78	1.78	1.17	0.52
Hf	0.56	-0.12	-0.75	-1.71	-2.53
Ta	-1.27	-1.97	-2.52	-3.10	-3.16
W	-2.00	-2.14	-2.60	-2.86	-
Re	-1.51	-	-2.30	-	-
Os	4.30	1.62	-0.55	-0.57	-
Ir	-0.28	0.17	0.96	1.53	2.09
Pt	2.07	2.57	3.53	3.71	4.22
Au	4.31	1.42	4.64	4.67	4.42
Hg	4.48	3.57	2.84	-	-

Table. S4. Computed Gibbs free energies of OH* (ΔG_{*OH}) on catalyst surfaces. All free energies are computed relative to $H_2O(l)$ and $H_2(g)$.

Metal	ΔG_{*OH} (eV)				
	MC4	**MC3N**	**MC2N2**	**MCN3**	**MN4**
Sc	-0.69	-0.68	-0.94	-1.40	-2.05
Ti	-1.06	-1.65	-1.92	-2.38	-2.31
V	-1.47	-1.58	-1.75	-2.02	-1.57
Cr	-1.07	-1.25	-1.15	-0.86	-0.24
Mn	-0.40	-0.32	-0.06	-0.22	0.30
Fe	-0.09	-0.02	0.16	0.02	0.39
Co	0.08	0.29	0.55	0.63	0.84
Ni	0.95	1.27	1.50	1.44	1.87
Cu	2.07	2.13	1.81	1.64	1.71
Zn	1.75	1.08	0.86	0.66	0.57
Y	-0.71	-0.67	-0.80	-1.25	-1.89
Zr	-0.95	-1.59	-1.92	-2.52	-2.79
Nb	-1.56	-1.75	-2.21	-2.47	-2.39
Mo	-1.41	-1.53	-1.81	-2.00	-
Tc	-0.86	-1.12	-1.36	-1.49	-0.95
Ru	-0.32	-1.02	-0.57	-0.70	-0.17
Rh	0.33	0.36	0.24	0.68	0.87
Pd	1.08	1.09	1.78	1.84	2.27
Ag	-	2.16	1.97	1.93	-
Cd	-	0.54	0.27	0.09	0.07
Lu	-0.91	-0.81	-1.01	-1.47	-2.16
Hf	-1.33	-1.99	-2.37	-2.96	-3.19
Ta	-2.11	-2.33	-2.71	-2.71	-2.65
W	-1.73	-1.92	-2.29	-2.22	-
Re	-1.37	-	-1.78	-	-
Os	1.24	1.18	-1.17	-0.64	-
Ir	-0.34	0.05	0.24	0.67	0.92
Pt	1.11	1.07	1.78	1.79	2.32
Au	2.58	2.33	2.14	2.14	1.97
Hg	2.24	1.39	0.84	-	-

Table. S5. Computed Gibbs free energies of OOH* (ΔG_{*OOH}) on catalyst surfaces. All free energies are computed relative to $H_2O(l)$ and $H_2(g)$.

Metal	ΔG_{*OOH} (eV)				
	MC4	**MC3N**	**MC2N2**	**MCN3**	**MN4**
Sc	2.37	2.44	2.27	1.82	1.21
Ti	2.01	1.52	1.35	0.93	-
V	1.66	-	-	-	-
Cr	2.10	-	-	-	-
Mn	-	-	-	-	-
Fe	2.69	2.83	-	3.41	3.29
Co	2.96	3.26	3.57	3.59	3.76
Ni	3.82	4.25	4.43	4.39	4.68
Cu	4.81	4.83	4.68	4.58	4.59
Zn	4.52	4.16	4.01	3.88	3.80
Y	2.24	2.48	2.41	1.99	1.35
Zr	2.08	1.57	1.35	0.77	0.49
Nb	1.52	1.40	-	-	-
Mo	-	-	-	-	-
Tc	-	-	-	-	-
Ru	2.58	1.93	-	-	-
Rh	2.81	3.35	-	3.73	3.82
Pd	3.86	3.94	4.69	4.73	4.84
Ag	-	4.76	4.78	4.72	-
Cd	-	3.94	3.34	3.41	3.35
Lu	2.24	2.36	2.22	1.78	1.11
Hf	1.77	1.23	0.96	0.39	0.16
Ta	1.09	0.95	-	-	-
W	-	-	-	-	-
Re	-	-	-	-	-
Os	-	-	-	-	-
Ir	-	2.98	-	3.71	3.86
Pt	4.05	4.00	4.72	4.70	4.78
Au	4.89	4.86	4.93	4.81	4.75
Hg	4.76	4.42	4.06	-	-

Table. S6. The free energy changes of each elementary step (ΔG_1, ΔG_2, ΔG_3 and ΔG_4) and the overpotential of ORR (η_{ORR}) for MC4.

MC4	ΔG_1 (eV)	ΔG_2 (eV)	ΔG_3 (eV)	ΔG_4 (eV)	η_{ORR} (eV)
Sc	-2.55	-1.91	-1.15	0.69	1.92
Ti	-2.91	-1.61	-1.45	1.06	2.29
V	-3.26	-2.69	-0.44	1.47	2.70
Cr	-2.82	-2.96	-0.22	1.07	2.30
Mn	-	-	-	-	-
Fe	-2.23	-2.68	-0.11	0.09	1.32
Co	-1.96	-2.02	-0.85	-0.08	1.15
Ni	-1.10	-1.51	-1.37	-0.95	0.28
Cu	-0.11	-0.78	-1.97	-2.07	1.12
Zn	-0.40	-0.54	-2.23	-1.75	0.83
Y	-2.68	-1.85	-1.10	0.71	1.94
Zr	-2.84	-1.36	-1.67	0.95	2.18
Nb	-3.40	-2.36	-0.73	1.56	2.79
Mo	-	-	-	-	-
Tc	-	-	-	-	-
Ru	-2.34	-2.92	0.03	0.32	1.55
Rh	-2.11	-2.15	-0.33	-0.33	0.90
Pd	-1.06	-1.31	-1.47	-1.08	0.17
Cd	-	-	-	-	-
Lu	-2.68	-1.31	-1.85	0.91	2.14
Hf	-3.15	-1.21	-1.89	1.33	2.56
Ta	-3.83	-2.35	-0.84	2.11	3.34
W	-	-	-	-	-
Re	-	-	-	-	-
Os	-	-	-	-	-
Ir	-	-	-	-	-
Au	-0.03	-0.57	-1.73	-2.58	1.20
Hg	-0.16	-0.28	-2.24	-2.24	1.07
Pt	-0.87	-1.98	-0.96	-1.11	0.36
Ag	-	-	-	-	-

Table. S7. The free energy changes of each elementary step (ΔG_1, ΔG_2, ΔG_3 and ΔG_4) and the overpotential of ORR (η_{ORR}) for MC3N.

MC3N	ΔG_1 (eV)	ΔG_2 (eV)	ΔG_3 (eV)	ΔG_4 (eV)	η_{ORR} (eV)
Sc	-2.48	-0.98	-2.14	0.68	1.91
Ti	-3.40	-1.80	-1.37	1.65	2.88
V	-	-	-	-	-
Cr	-	-	-	-	-
Mn	-	-	-	-	-
Fe	-2.09	-2.53	-0.32	0.02	1.25
Co	-1.66	-2.07	-0.89	-0.29	0.94
Ni	-0.67	-1.40	-1.58	-1.27	0.56
Cu	-0.09	-0.64	-2.05	-2.13	1.14
Zn	-0.76	-0.75	-2.33	-1.08	0.47
Y	-2.44	-0.95	-2.20	0.67	1.90
Zr	-3.35	-1.47	-1.68	1.59	2.82
Nb	-3.52	-2.91	-0.24	1.75	2.98
Mo	-	-	-	-	-
Tc	-	-	-	-	-
Ru	-2.99	-2.72	-0.24	1.02	2.25
Rh	-1.57	-2.40	-0.59	-0.36	0.87
Pd	-0.98	-1.08	-1.77	-1.09	0.25
Cd	-0.98	-0.79	-2.60	-0.54	0.69
Lu	-2.56	-0.59	-2.58	0.81	2.04
Hf	-3.69	-1.35	-1.87	1.99	3.22
Ta	-3.97	-2.93	-0.35	2.33	3.56
W	-	-	-	-	-
Re	-	-	-	-	-
Os	-	-	-	-	-
Ir	-1.94	-2.81	-0.12	-0.05	1.18
Au	-0.06	-3.44	0.91	-2.33	2.14
Hg	-0.50	-0.85	-2.18	-1.39	0.73
Pt	-0.92	-1.43	-1.50	-1.07	0.31
Ag	-0.16	-0.38	-2.22	-2.16	1.07

Table. S8. The free energy changes of each elementary step (ΔG_1, ΔG_2, ΔG_3 and ΔG_4) and the overpotential of ORR (η_{ORR}) for MC2N2.

MC2N2	ΔG_1 (eV)	ΔG_2 (eV)	ΔG_3 (eV)	ΔG_4 (eV)	η_{ORR} (eV)
Sc	-2.65	-0.57	-2.64	0.94	2.17
Ti	-3.57	-2.29	-0.98	1.92	3.15
V	-	-	-	-	-
Cr	-	-	-	-	-
Mn	-	-	-	-	-
Fe	-	-	-	-	-
Co	-1.35	-2.01	-1.02	-0.55	0.68
Ni	-0.49	-1.04	-1.89	-1.50	0.74
Cu	-0.24	-0.44	-2.43	-1.81	0.99
Zn	-0.91	-0.61	-2.54	-0.86	0.37
Y	-2.51	-0.58	-2.63	0.80	2.03
Zr	-3.57	-1.83	-1.44	1.92	3.15
Nb	-	-	-	-	-
Mo	-	-	-	-	-
Tc	-	-	-	-	-
Ru	-	-	-	-	-
Rh	-	-	-	-	-
Pd	-0.23	-0.94	-1.97	-1.78	1.00
Cd	-1.58	-0.61	-2.46	-0.27	0.96
Lu	-2.70	-0.45	-2.78	1.01	2.24
Hf	-3.96	-1.71	-1.62	2.37	3.60
Ta	-	-	-	-	-
W	-	-	-	-	-
Re	-	-	-	-	-
Os	-	-	-	-	-
Ir	-	-	-	-	-
Au	0.01	-0.29	-2.50	-2.14	1.24
Hg	-0.86	-1.22	-1.99	-0.84	0.39
Pt	-0.20	-1.19	-1.75	-1.78	1.03
Ag	-0.14	-0.38	-2.42	-1.97	1.09

Table. S9. The free energy changes of each elementary step (ΔG_1, ΔG_2, ΔG_3 and ΔG_4) and the overpotential of ORR (η_{ORR}) for MCN3.

MCN3	ΔG_1 (eV)	ΔG_2 (eV)	ΔG_3 (eV)	ΔG_4 (eV)	η_{ORR} (eV)
Sc	-3.10	-0.85	-2.37	1.40	2.63
Ti	-3.99	-2.79	-0.51	2.38	3.61
V	-	-	-	-	-
Cr	-	-	-	-	-
Mn	-	-	-	-	-
Fe	-1.51	-2.53	-0.87	-0.02	1.21
Co	-1.33	-1.88	-1.08	-0.63	0.60
Ni	-0.53	-0.87	-2.08	-1.44	0.70
Cu	-0.34	-3.59	0.64	-1.64	1.87
Zn	-1.04	-0.78	-2.43	-0.66	0.57
Y	-2.93	-0.74	-2.49	1.25	2.48
Zr	-4.15	-2.25	-1.05	2.52	3.75
Nb	-	-	-	-	-
Mo	-	-	-	-	-
Tc	-	-	-	-	-
Ru	-	-	-	-	-
Rh	-1.19	-1.88	-1.18	-0.68	0.55
Pd	-0.19	-0.76	-2.13	-1.84	1.04
Cd	-1.51	-0.82	-2.50	-0.09	1.14
Lu	-3.14	-0.61	-2.65	1.47	2.70
Hf	-4.53	-2.10	-1.26	2.96	4.19
Ta	-	-	-	-	-
W	-	-	-	-	-
Re	-	-	-	-	-
Os	-	-	-	-	-
Ir	-1.21	-2.18	-0.86	-0.67	0.56
Au	-0.11	-0.14	-2.52	-2.14	1.12
Hg	-	-	-	-	-
Pt	-0.22	-0.99	-1.92	-1.79	1.01
Ag	-0.20	-0.41	-2.38	-1.93	1.03

Table. S10. The free energy changes of each elementary step (ΔG_1, ΔG_2, ΔG_3 and ΔG_4) and the overpotential of ORR (η_{ORR}) for MN4.

MN4	ΔG_1 (eV)	ΔG_2 (eV)	ΔG_3 (eV)	ΔG_4 (eV)	η_{ORR} (eV)
Sc	-3.71	-1.07	-2.18	2.05	3.28
Ti	-	-	-	-	-
V	-	-	-	-	-
Cr	-	-	-	-	-
Mn	-	-	-	-	-
Fe	-1.63	-2.16	-0.74	-0.39	0.84
Co	-1.16	-1.30	-1.62	-0.84	0.39
Ni	-0.24	-0.68	-2.13	-1.87	0.99
Cu	-0.33	-0.77	-2.11	-1.71	0.90
Zn	-1.12	-0.73	-2.50	-0.57	0.66
Y	-3.57	-0.76	-2.49	1.89	3.12
Zr	-4.43	-2.83	-0.45	2.79	4.02
Nb	-	-	-	-	-
Mo	-	-	-	-	-
Tc	-	-	-	-	-
Ru	-	-	-	-	-
Rh	-1.10	-1.39	-1.55	-0.87	0.36
Pd	-0.08	-0.41	-2.16	-2.27	1.15
Cd	-1.57	-0.77	-2.51	-0.07	1.16
Lu	-3.81	-0.59	-2.68	2.16	3.39
Hf	-4.76	-2.69	-0.66	3.19	4.42
Ta	-	-	-	-	-
W	-	-	-	-	-
Re	-	-	-	-	-
Os	-	-	-	-	-
Ir	-1.06	-1.77	-1.18	-0.92	0.31
Au	-0.17	-0.33	-2.45	-1.97	1.06
Hg	-	-	-	-	-
Pt	-0.14	-0.56	-1.90	-2.32	1.09
Ag	-	-	-	-	-

Table. S11. Overview of recent literature works aimed at synthesis of catalysts in ORR reaction.

Metal	Ref	Conclusion
Co	*J. Am. Chem. Soc.* **2022,** 144, 14505-14516	CoN4-4e$^-$
Ni	*Angew. Chem. Int. Edit.* **2022,** 61, e202206544	NiN4-2e$^-$
Cu	*Nat. Sustain.* **2021,** 4, 233-241	CuC3N4-2e$^-$
Zn	*Angew. Chem. Int. Edit.* **2019,** 58, 7035-7039	ZnN4-4e$^-$
Rh	*RSC. Adv.* **2021,** 11, 13906-13911	RhNC-4e$^-$
Pd	*ChemElectroChem* **2019,** 6, 4757-4764	PdN4-2e$^-$
Ag	*Angew. Chem. Int. Edit.* **2022,** 61, e202212397	AgCN-2e$^-$
Pt	*Angew. Chem. Int. Edit.* **2019,** 58, 1163-1167	PtC4-4e$^-$
Au	*J. Mater. Chem. A* **2022,** 10, 8371-8377	AuCN-2e$^-$

Table. S12. Band center of metal atoms (ε_d) for O* adsorbed on catalyst surfaces. The Fermi level is set to be 0 eV.

Metal	Band center of the metal atom (ε_d, eV)				
	MC4	**MC3N**	**MC2N2**	**MCN3**	**MN4**
Sc	2.82	3.81	4.06	4.18	4.11
Ti	2.87	2.73	2.64	1.98	1.26
V	1.25	0.51	-0.10	0.48	0.39
Cr	0.39	-0.17	-0.73	-2.79	-0.85
Mn	-1.32	-1.04	-0.95	-1.53	-1.43
Fe	-1.40	-1.64	-1.65	-1.61	-1.41
Co	-2.26	-1.94	-2.05	-2.29	-2.50
Ni	-2.56	-2.50	-2.70	-2.53	-2.63
Cu	-3.34	-3.33	-3.47	-2.40	-2.61
Zn	-6.25	-6.59	-5.69	-5.07	-5.25
Y	3.09	3.48	3.78	3.69	3.53
Zr	3.09	3.00	3.04	2.58	2.10
Nb	1.99	1.33	0.81	0.50	0.37
Mo	-0.58	0.15	-0.58	-0.89	-
Tc	-1.20	-0.92	-1.12	-1.76	-2.43
Ru	-1.78	-2.11	-2.64	-2.39	-2.12
Rh	-3.10	-3.22	-2.84	-2.89	-2.72
Pd	-3.71	-3.76	-3.77	-3.57	-3.49
Ag	-	-4.72	-4.62	-5.31	-
Cd	-	-7.56	-7.06	-6.70	-6.79
Lu	3.22	3.44	3.47	3.15	2.91
Hf	3.04	3.00	2.98	2.61	2.28
Ta	2.02	1.47	1.16	0.70	0.43
W	-0.09	-0.52	-0.33	-0.26	-
Re	-1.14	-	-0.90	-	-
Os	-0.94	-1.47	-1.20	-1.48	-
Ir	-3.10	-3.57	-3.40	-3.05	-2.93
Pt	-4.45	-3.94	-3.83	-3.65	-3.98
Au	-4.63	-4.68	-4.40	-5.14	-5.54
Hg	-6.66	-6.87	-6.12	-	-

Table. S13. Band center of metal atoms (ε_d) for OOH* adsorbed on catalyst surfaces. The Fermi level is set to be 0 eV.

Metal	Band center of the metal atom (ε_d, eV)				
	MC4	**MC3N**	**MC2N2**	**MCN3**	**MN4**
Sc	3.14	3.47	3.89	3.34	2.67
Ti	2.61	1.96	1.46	0.90	-
V	0.53	-	-	-	-
Cr	0.09	-	-	-	-
Mn	-	-	-	-	-
Fe	-0.65	-1.13	-	-1.10	-1.02
Co	-1.61	-1.50	-1.30	-1.52	-1.38
Ni	-1.89	-2.18	-1.72	-2.01	-2.38
Cu	-2.77	-2.92	-3.34	-3.62	-3.00
Zn	-6.45	-6.69	-5.90	-5.16	-5.30
Y	3.52	3.82	4.15	3.71	3.23
Zr	3.21	2.66	2.28	1.77	1.25
Nb	1.24	0.99	-	-	-
Mo	-	-	-	-	-
Tc	-	-	-	-	-
Ru	-0.89	-1.56	-	-	-
Rh	-2.64	-2.36	-	-1.83	-2.11
Pd	-3.13	-3.21	-2.88	-2.94	-3.14
Ag	-	-4.61	-4.85	-5.49	-
Cd	-	-7.66	-7.16	-6.93	-7.12
Lu	3.21	3.34	3.85	3.52	3.10
Hf	2.96	2.75	2.44	1.96	1.38
Ta	1.34	1.08	-	-	-
W	-	-	-	-	-
Re	-	-	-	-	-
Os	-	-	-	-	-
Ir	-	-2.96	-	-2.27	-2.19
Pt	-3.04	-3.12	-2.79	-2.56	-2.94
Au	-3.91	-4.23	-4.50	-5.06	-5.64
Hg	-7.04	-6.96	-5.97	-	-

Table. S14. Computed bader charge of O* on catalyst surfaces.

Metal	O*				
	MC4	**MC3N**	**MC2N2**	**MCN3**	**MN4**
Sc	-0.07	-0.04	-0.07	-0.04	0.04
Ti	-0.09	-0.19	-0.22	-0.29	-0.46
V	-0.39	-0.39	-0.41	-0.45	-0.37
Cr	-0.35	-0.28	-0.35	-0.22	-0.26
Mn	-0.22	-0.09	-0.13	-0.09	-0.10
Fe	0.00	-0.06	-0.05	-0.16	-0.18
Co	-0.14	-0.20	-0.27	-0.30	-0.33
Ni	-0.15	-0.27	-0.29	-0.25	-0.24
Cu	-0.16	-0.13	-0.10	-0.01	-0.13
Zn	-0.04	-0.10	-0.08	-0.08	-0.12
Y	-0.23	-0.11	-0.14	-0.09	-0.06
Zr	-0.32	-0.30	-0.30	-0.36	-0.51
Nb	-0.54	-0.55	-0.64	-0.69	-0.64
Mo	-0.61	-0.56	-0.59	-0.70	-
Tc	-0.56	-0.52	-0.57	-0.45	-0.47
Ru	-0.60	-0.55	-0.51	-0.55	-0.46
Rh	-0.51	-0.43	-0.40	-0.59	-0.59
Pd	-0.24	-0.30	-0.37	-0.34	-0.21
Ag	-	-0.11	-0.10	-0.08	-
Cd	-	-0.09	-0.08	-0.09	-0.17
Lu	-0.15	-0.13	-0.11	-0.08	-0.05
Hf	-0.20	-0.25	-0.25	-0.45	-0.50
Ta	-0.55	-0.55	-0.70	-0.74	-0.68
W	-0.64	-0.75	-0.60	-0.61	-
Re	-0.68	-	-0.49	-	-
Os	0.00	-0.01	0.00	-0.13	-
Ir	-0.49	-0.51	-0.53	-0.59	-0.65
Pt	-0.37	-0.37	-0.44	-0.43	-0.47
Au	-0.18	-0.08	-0.12	-0.11	-0.09
Hg	-0.08	-0.05	0.04	-	-

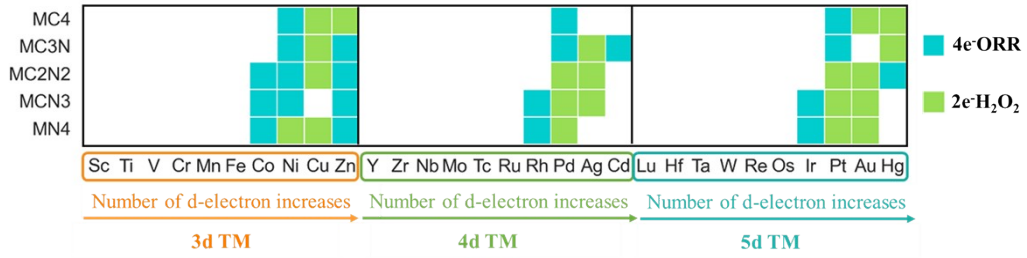

Fig. S1. The classified diagram of the excellent and high-performance $2e^-$ and $4e^-$ ORR catalysts screened out by the volcano map.

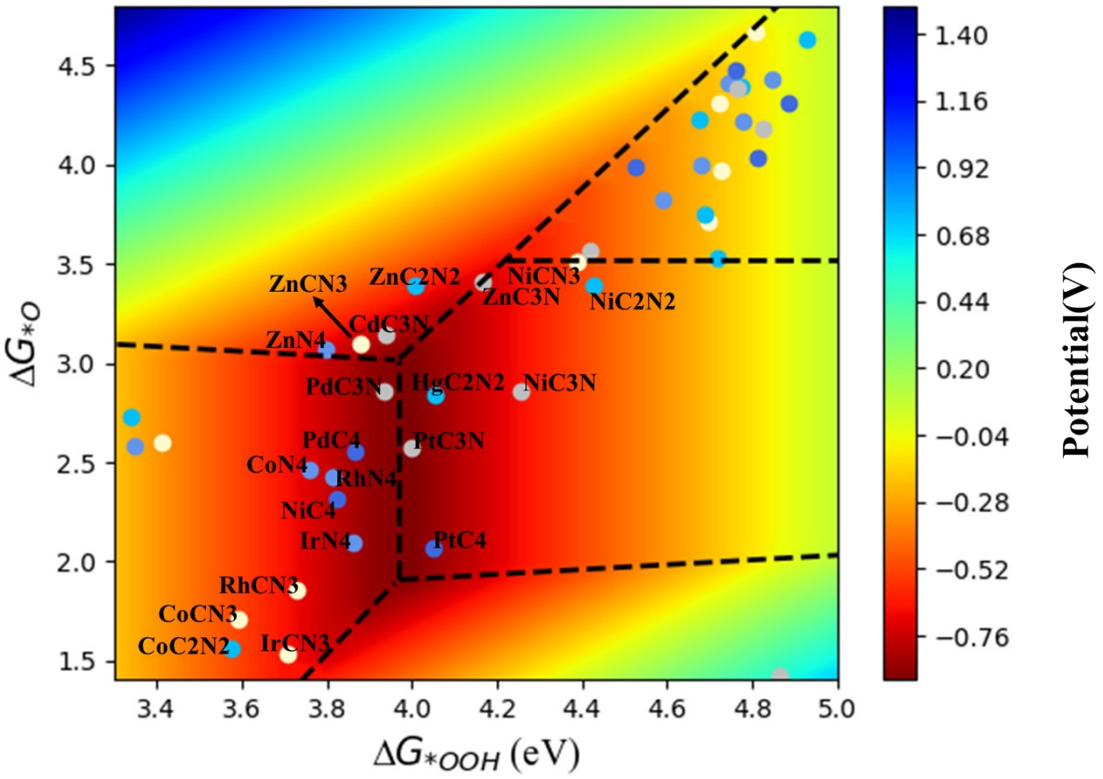

Fig. S2. The close-up view of the 4e- ORR Contour maps.

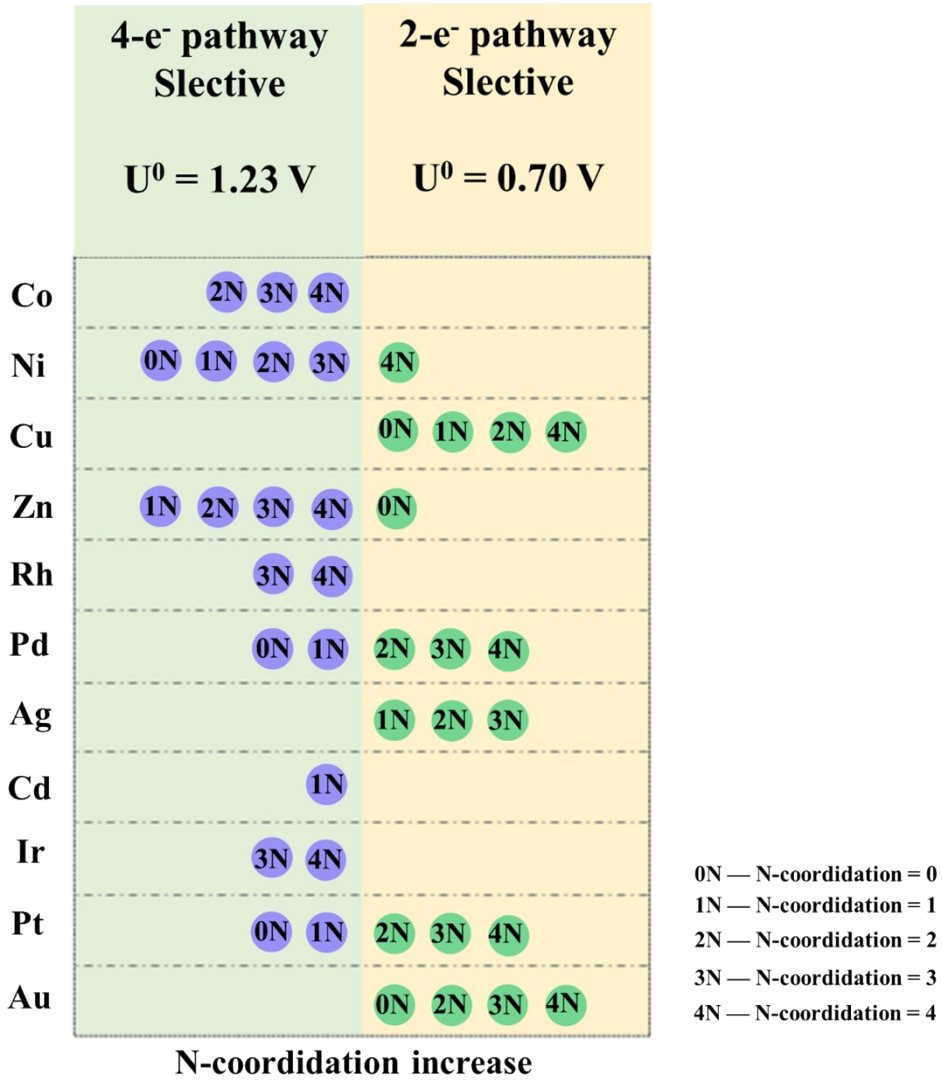

Fig. S3. Illustration of the reactions and the excellent catalyst during ORR reaction.

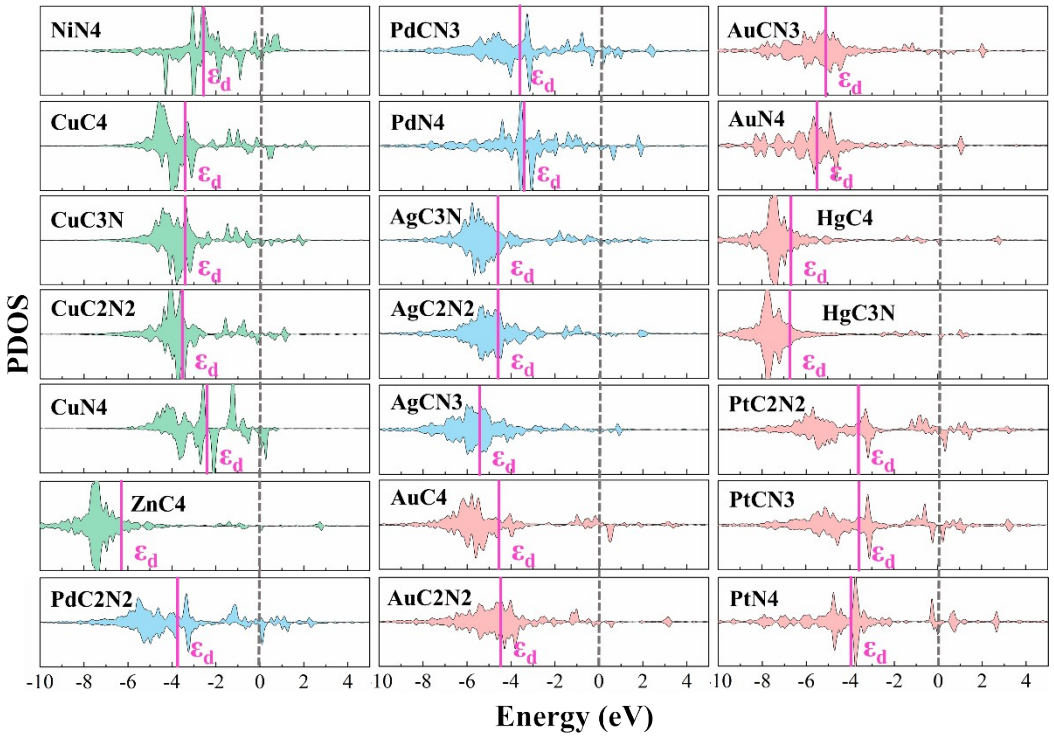

Fig. S4. Partial density of states (PDOS) of d orbitals for 21 $2e^-$ ORR catalysts screened out by the contour map. The Fermi level (E_F) is set to 0 eV.

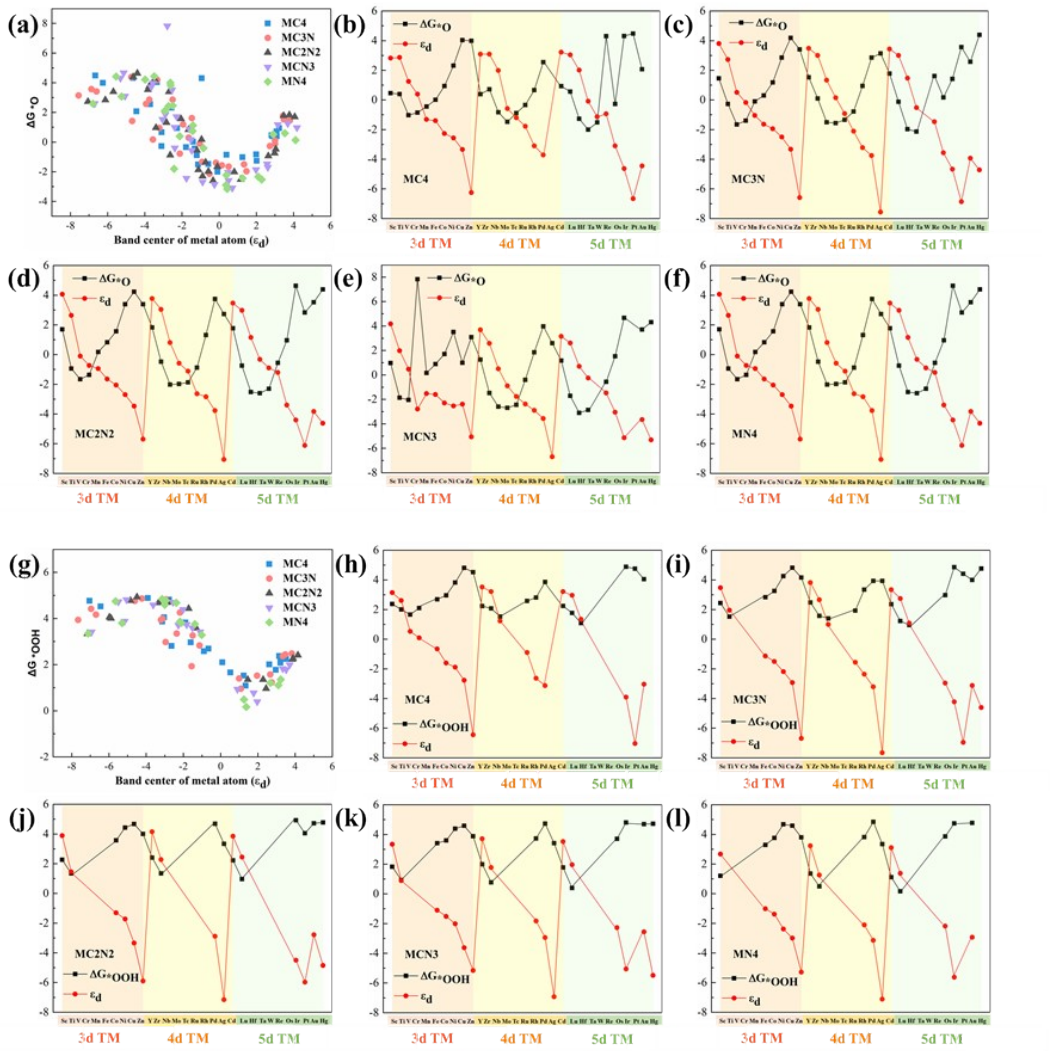

Fig. S5. Band center of metal atom (ε_d) versus Gibbs free energy of O^* (a) and OOH^* (g) on different SACs. The values of ε_d and ΔG_{*O} on SACs with different N coordination (b-f). The values of ε_d and ΔG_{*OOH} on SACs with different N coordination (h-l). Detailed ε_d values of ΔG_{*O} and ΔG_{*OOH} are given in Tables S12 and S13.

Supporting Information Note 1. Additional computational details for SISSO

Primary Features

In this work, our search for optimal descriptors in SISSO began with an extensive list of 18 fundamental quantities, which are potential factors determining the reactivity of oxide surfaces and the adsorption enthalpies. These primary features were collected from general physicochemical considerations. These features, which are categorized into geometric and electronic properties, can be characterized as describing the entire surface, the local adsorption site, or the specific cus metal atom to which the adsorbates coordinate. (ref.2) Although SISSO is designed to handle correlated features, it is preferable to filter out obvious correlations in advance to alleviate the dramatic increase in computational cost of the SISSO method with the increase in the number of primary features. Therefore, we evaluated the Pearson correlation coefficient between each pair of primary features in our database.

Table S15 provides a list of 18 basic quantities obtained from the literature that are important factors in determining the reactivity and adsorption energy of SACs surfaces (Detailed values of these quantities are given in Table S16). Next, the Pearson correlation coefficient between each pair of primary features was calculated as shown in Fig. S6, and 0.88 was selected as the standard to screen out the features with low correlation (details as follows).

- R_M^{atom} and R_M^{CRC} show a very high correlation (0.90). We removed R_M^{CRC} due to the R_M^{atom} is more versatile.

- N_M^{LPE} and N_M^{men} are all highly correlated (0.88). We selected N_M^{men}.

After removing above three primary features, we got 16 primary features used for the SISSO descriptor identification in Table S17. The correlation coefficients between each primary feature and calculated DFT adsorption energy of O is also calculated.

Compressed Sensing.

SISSO is a compressed sensing-based method for describing and predicting an attribute of the key characteristics (here adsorption energy). (ref.3-5) The resulting predictive model is represented in the form of analytical formulas (constructed by applying the following set of algebraic/functional operators), built from simple physical properties.

$$\hat{H}^{(m)} \equiv \{I, +, -, \times, /, log, exp, ^{-1}, ^{2}, ^{3}, \sqrt{}, \sqrt[3]{}\}$$

The superscript m indicates that the dimensional analysis was performed to ensure that only physically meaningful combinations were retained. The operator is applied iteratively to the generated feature space, and N is the number of iterations and a hyperparameter of the method (denoted as gradient, ΦN). The size of the subspace is controlled by the sure independence screening (SIS) parameters. First, a large feature space is pre-screened using SIS, and then a smaller subspace of top-ranked candidate features is selected based on the correlation of features with attributes and the correlation of features with the remaining error of the selected features. In general, a larger SIS value indicates a higher probability of finding the best descriptor. For very small feature spaces, a further constraint on the SIS value is that the resulting feature subspace is smaller than the total feature space. Then, a sparsification operator (SO, here the ℓ_0 constraint) is used to find the best sparse solution. M is another hyperparameter denoted as the dimensions of the descriptor. The final M linear combinations of features needed are also referred to as descriptors.

Ref. 2 W. Xu, M. Andersen and K. Reuter, *ACS Catal*, 2021, **11**, 734-742.

Ref. 3. Rossmeisl, J.; Qu, Z.-W.; Zhu, H.; Kroes, G.-J.; N_rskov, J. K. J. *Electroanal. Chem*. **2007**, *607*, 83-89.

Ref. 4. Nϕrskov, J. K.; Rossmeisl, J.; Logadottir, A.; Lindqvist, L.; Kitchin, J. R.; Bligaard, T.; Jonsson, H. *J. Phys. Chem. B* **2004**, *108*, 17886-17892.

Ref. 5. Man, I. C.; Su, H.-Y.; Calle-Vallejo, F.; Hansen, H. A.; Martinez, J. I.; Inoglu, N. G.; Kitchin, J.; Jaramillo, T. F.; N_rskov, J. K.; Rossmeisl, J. *ChemCatChem* **2011**, *3*, 1159-1165.

Table. S15. The full list of primary features compiled from the literature before feature elimination.

Symbol	Physical meaning	Symbol	Physical meaning
N_M^{LPE}	The number of lone pair electrons of the central metal atom	R_M^{CRC}	Covalent radius of the central metal atom by Cordero et al.
N_M^{men}	Mendeleev number of the central metal atom	R_M^{RS}	*s* orbital radius of the central metal atom
N_M^{group}	Group of the central metal atom	R_M^{RD}	*d* orbital radius of the central metal atom
N_M^{atom}	Atomic number of the central metal atom	R_M^{atom}	Atomic radius of the central metal atom
E_M^{ion}	Ion-energy of the central metal atom	R_M^{vdw}	Van der Waals radius of the central metal atom
P_M^{EA}	Electron affinity of the central metal atom	N_{C1}^{men}	Mendeleev number of the first coordinating atom
χ_M	Pauling electronegativity of the central metal atom	N_{C2}^{men}	Mendeleev number of the second coordinating atom
C_M^{TC}	Thermal conductivity of the central metal atom	N_{C3}^{men}	Mendeleev number of the third coordinating atom
V_M^{atom}	Volume of the central metal atom	N_{C4}^{men}	Mendeleev number of the fourth coordinating atom

Table. S16. Atomic primary features obtained from the literature (ref.5-8).

Metal	N_M^{men}	N_M^{group}	N_M^{atom}	E_M^{ion}	P_M^{EA}	χ_M	C_M^{TC}	V_M^{atom}	R_M^{RD}	R_M^{atom}	R_M^{vdw}
Sc	11	3	21	6.56	0.19	1.36	16.00	15.00	1.12	3.48	4.87
Ti	43	4	22	6.83	0.08	1.54	21.90	10.64	1.00	3.32	4.65
V	46	5	23	6.75	0.52	1.63	30.70	8.32	0.91	3.23	4.57
Cr	49	6	24	6.77	0.67	1.66	94.00	7.23	0.86	2.64	4.63
Mn	52	7	25	7.43	-0.50	1.55	7.80	7.35	0.77	3.04	4.63
Fe	55	8	26	7.90	0.15	1.83	80.00	7.09	0.72	2.95	4.61
Co	58	9	27	7.88	0.66	1.88	100.00	6.67	2.49	2.87	4.53
Ni	61	10	28	7.64	1.16	1.91	91.00	6.59	0.64	2.81	4.53
Cu	64	11	29	7.73	1.23	1.90	400.00	7.11	0.61	2.74	4.50
Zn	69	12	30	9.39	-0.60	1.65	116.00	9.16	0.57	2.68	4.51
Y	12	3	39	6.22	0.31	1.22	17.20	19.88	1.77	4.00	5.20
Zr	44	4	40	6.63	0.43	1.33	14.02	14.02	1.59	3.89	4.76
Nb	47	5	41	6.76	0.92	1.60	54.00	10.83	1.49	3.74	4.84
Mo	50	6	42	7.09	0.75	2.16	139.00	9.38	1.38	3.59	4.63
Tc	53	7	43	7.12	0.55	1.90	51.00	8.63	1.26	3.46	4.61
Ru	56	8	44	7.36	1.05	2.20	120.00	8.17	1.21	3.36	4.65
Rh	59	9	45	7.46	1.14	2.28	150.00	8.28	1.14	3.27	4.61
Pd	62	10	46	8.34	0.56	2.20	72.00	8.56	1.10	3.19	4.06
Ag	65	11	47	7.58	1.30	1.93	430.00	10.27	1.03	3.12	4.78
Cd	70	12	48	8.99	-0.70	1.69	97.00	13.00	0.98	3.04	4.70
Lu	41	18	71	5.43	0.24	1.27	16.00	17.78	0.47	4.10	4.97
Hf	45	4	72	6.83	0.18	1.30	23.00	13.44	1.65	3.93	4.78
Ta	48	5	73	7.89	0.32	1.50	57.00	10.85	1.55	3.78	4.85
W	51	6	74	7.86	0.82	2.36	174.00	9.47	1.47	3.65	4.70
Re	54	7	75	7.83	0.06	1.90	48.00	8.86	1.40	3.55	4.68
Os	57	8	76	8.44	1.10	2.20	88.00	8.42	1.33	3.49	4.55
Ir	60	9	77	8.97	1.57	2.20	150.00	8.52	1.28	3.40	4.33
Pt	63	10	78	8.96	2.13	2.28	72.00	9.09	1.25	3.34	4.38
Au	66	11	79	9.23	2.31	2.54	320.00	10.21	1.20	3.29	4.63
Hg	71	12	80	10.44	0	2.00	8.30	14.09	1.15	3.23	4.67

Ref. 5. Winter, M. Accessed 29th May 2020. https://www.webelements.com/.

Ref. 6. Tran, K.; Ulissi, Z. W. *Nat. Catal.* **2018**, *1*, 696-703.

Ref. 7. Clementi, E.; Raimondi, D.; Reinhardt, W. P. *J. Chem. Phys.* **1967**, *47*, 1300-

1307.

Ref. 8. Ruban, A.; Hammer, B.; Stoltze, P.; Skriver, H. L.; Nϕrskov, J. K. *J. Mol. Catal. A-Chem.* **1997**, *115*, 421-429.

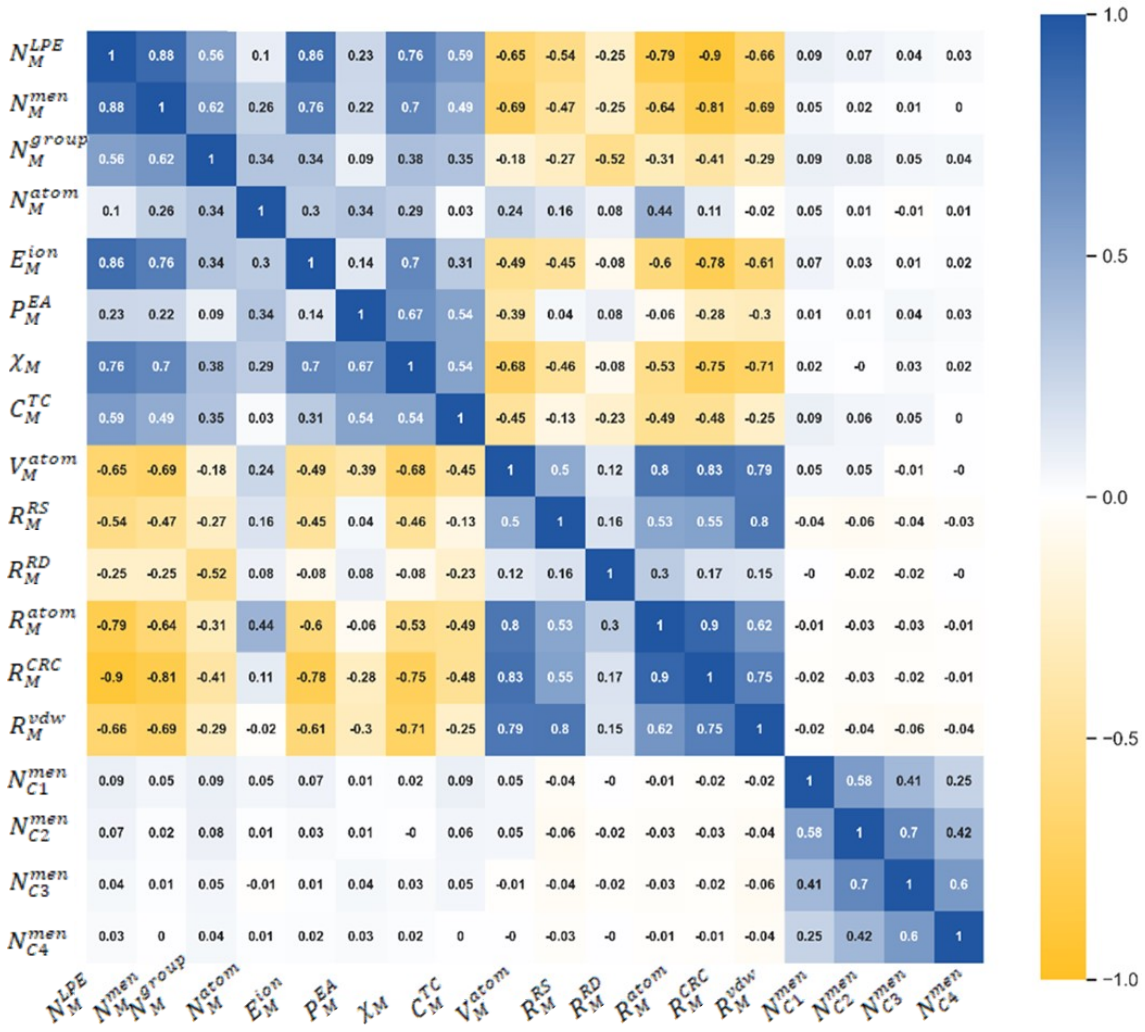

Fig. S6. Pearson correlation coefficient between every pair of primary features from Table. S15.

Table. S17. The full list of primary features compiled from the literature after feature elimination.

Symbol	Physical meaning	Symbol	Physical meaning
N_M^{men}	Mendeleev number of the central metal atom	R_M^{RD}	*d* orbital radius of the central metal atom
N_M^{group}	Group of the central metal atom	R_M^{RS}	*s* orbital radius of the central metal atom
N_M^{atom}	Atomic number of the central metal atom	R_M^{atom}	Atomic radius of the central metal atom
E_M^{ion}	Ion-energy of the central metal atom	R_M^{vdw}	Van der Waals radius of the central metal atom
P_M^{EA}	Electron affinity of the central metal atom	N_{C1}^{men}	Mendeleev number of the first coordinating atom
χ_M	Pauling electronegativity of the central metal atom	N_{C2}^{men}	Mendeleev number of the second coordinating atom
C_M^{TC}	Thermal conductivity of the central metal atom	N_{C3}^{men}	Mendeleev number of the third coordinating atom
V_M^{atom}	Volume of the central metal atom	N_{C4}^{men}	Mendeleev number of the fourth coordinating atom

Table. S18. The features of electronic bands.

Symbol	Physical meaning	Symbol	Physical meaning
ε_d	Center of the d-band	K_d	Kurtosis of the d-band
S_d	Skewness of the d-band	f_d	Filling of the d-band
W_d	Width of the d-band		

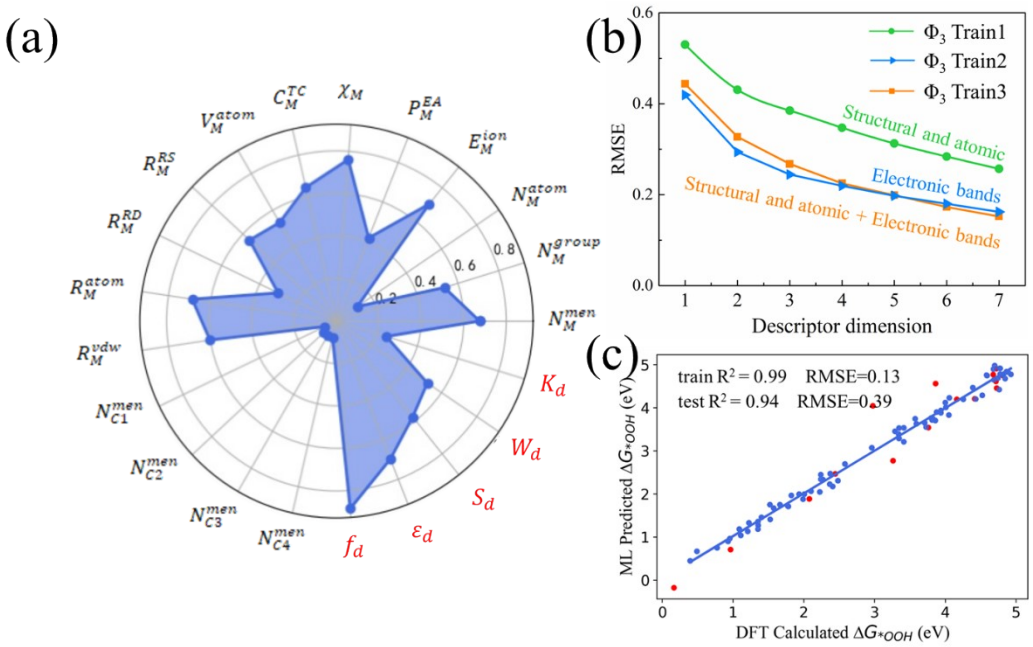

Fig. S7 (a) Importance of the selected features for target property of ΔG_{*OOH}. (b) Training RMSE from 5-fold cross validation (averaged over the 5 folds) for all tested SISSO hyperparameters. (d) Validation of the models constructed by the selected features for target property of ΔG_{*OOH}, shown by the values of the ML prediction and density functional theory (DFT) calculations. The red points in each diagram correspond to the structures newly calculated by DFT calculations, which are treated as validation data. The blue points in each diagram correspond to the data that have been put into SISSO to get the descriptors, which are treated as training data.