Supporting information

Volcano Relationships and a New Activity Descriptor of 2D Transition Metal-Fe Layered Double Hydroxides for Efficient Oxygen Evolution Reaction

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Keywords: Alkaline oxygen evolution reaction, layered double hydroxides, iron incorporation, adsorption capacitance, adsorption energy barrier

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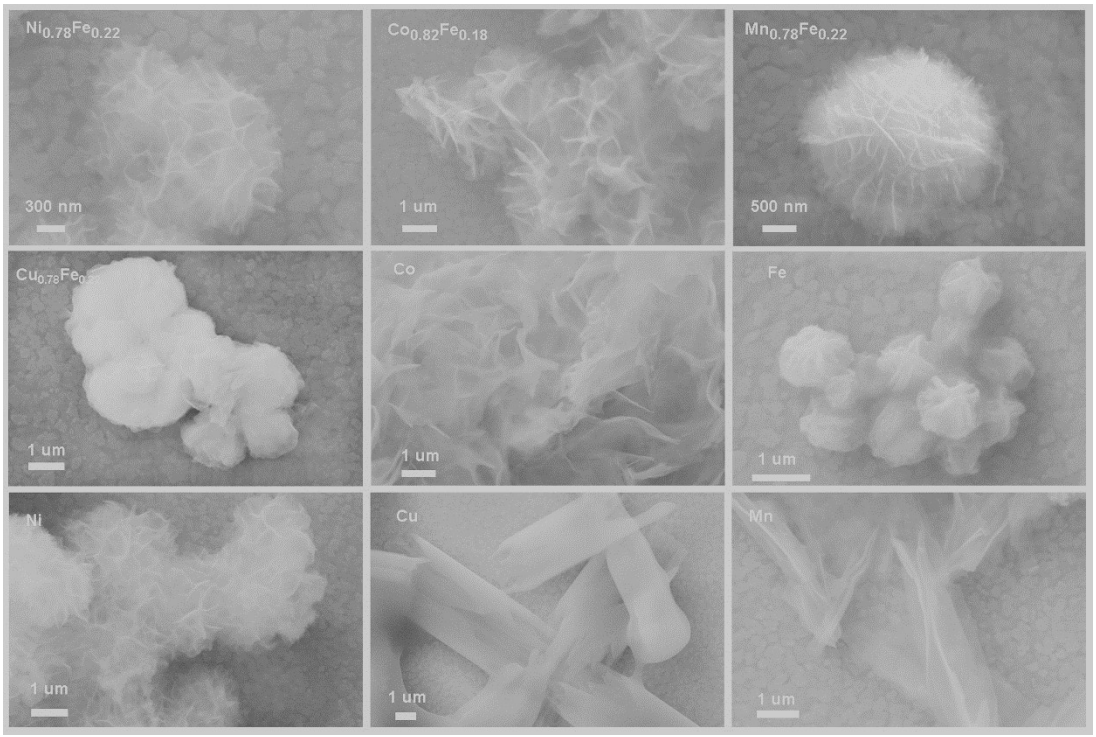

Figure S1. SEM images of different samples.

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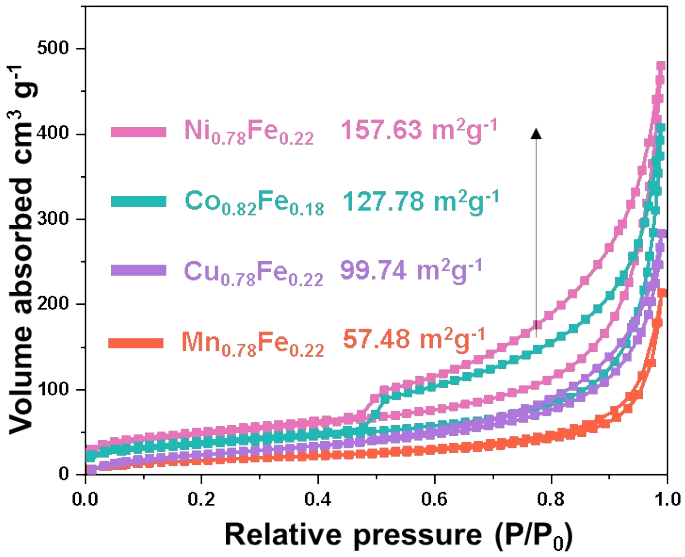

Figure S2. Nitrogen adsorption and desorption curves of $Ni_{0.82}Fe_{0.18}$, $Co_{0.82}Fe_{0.12}$, $Cu_{0.78}Fe_{0.22}$ and $Mn_{0.78}Fe_{0.22}$ samples.

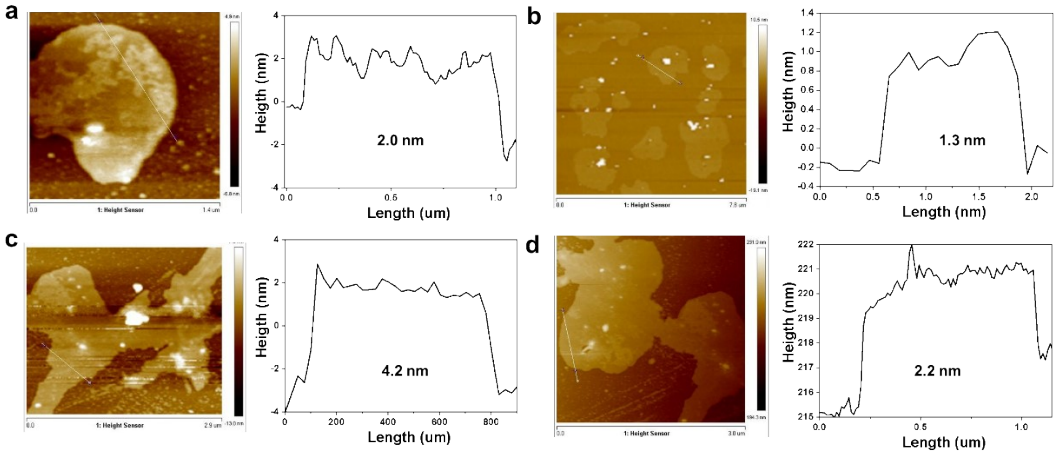

Figure S3. AFM images (a-d) of $Co_{0.82}Fe_{0.18}$, $Ni_{0.78}Fe_{0.22}$, $Cu_{0.78}Fe_{0.22}$, and $Mn_{0.78}Fe_{0.22}$ samples.

Figure S4. Optical images of different LDH samples.

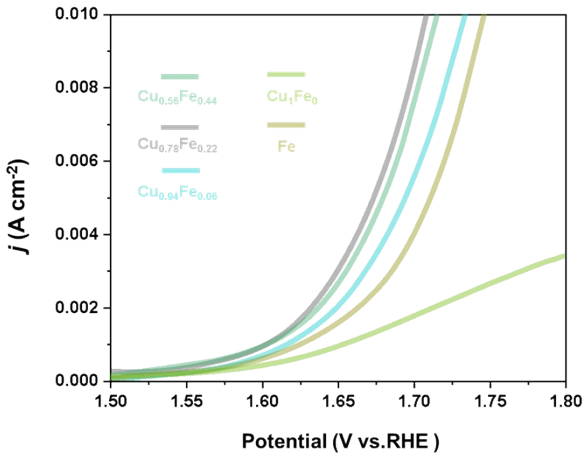

Figure S5. Polarization curves at 5 mV s^{-1} for $Cu_{1-x}Fe_x$-LDHs.

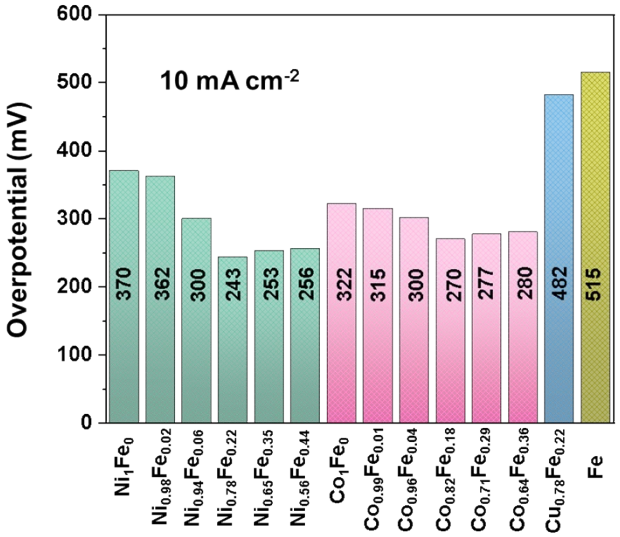

Figure S6. Overpotentials at 10 mA cm^{-2} of different LDH samples.

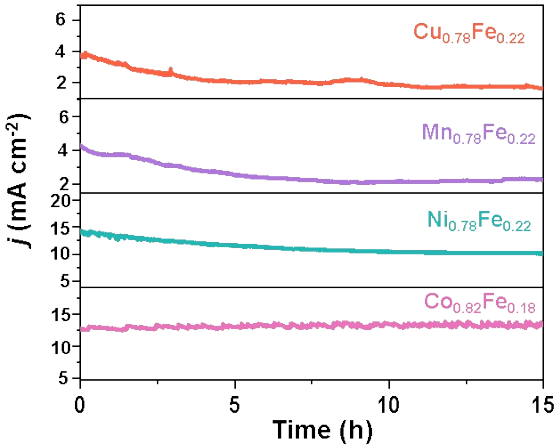

Figure S7. OER stability characterization with chronoamperometry test for 15 h.

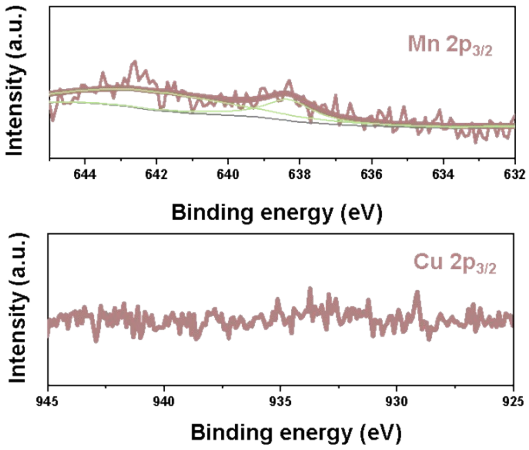

Figure S8. High-resolution XPS spectrums of Mn $2p_{3/2}$ and Cu $2p_{3/2}$ of $Mn_{0.78}Fe_{0.22}$ and $Cu_{0.78}Fe_{0.22}$ samples after OER reaction.

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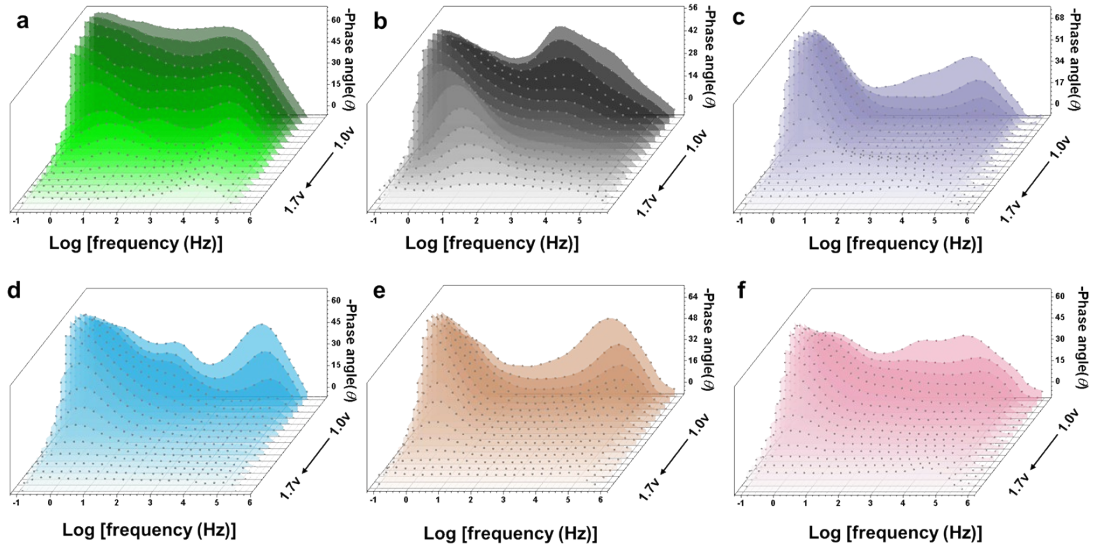

Figure S9. Bode plots of Co_1Fe_0, $Co_{0.99}Fe_{0.01}$, $Co_{0.96}Fe_{0.04}$, $Co_{0.82}Fe_{0.18}$, $Co_{0.71}Fe_{0.29}$ and $Co_{0.64}Fe_{0.36}$ samples.

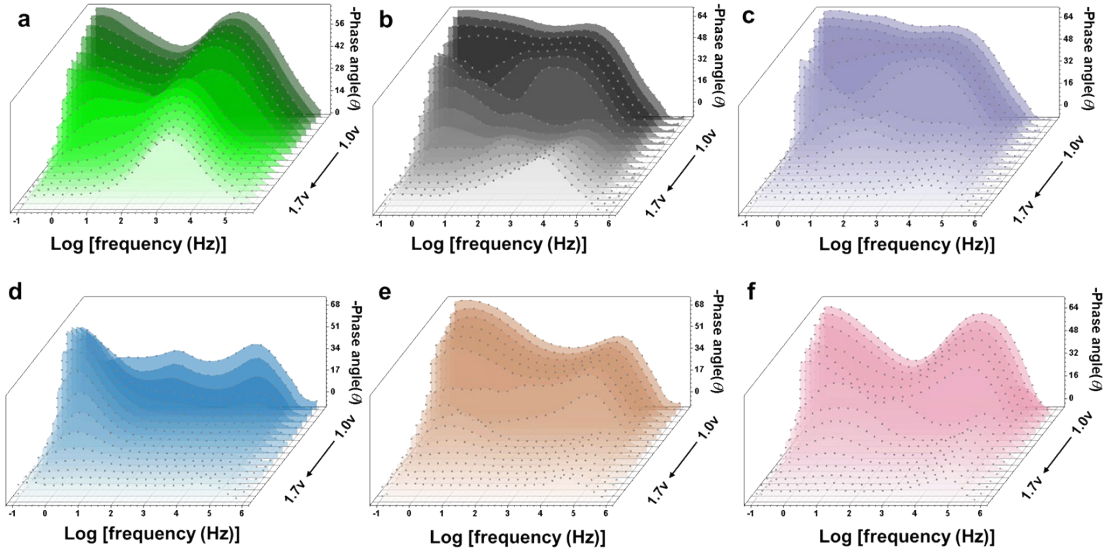

Figure S10. Bode plots of Ni_1Fe_0, $Ni_{0.98}Fe_{0.02}$, $Ni_{0.94}Fe_{0.06}$, $Ni_{0.78}Fe_{0.22}$, $Ni_{0.65}Fe_{0.35}$ and $Ni_{0.56}Fe_{0.44}$ samples.

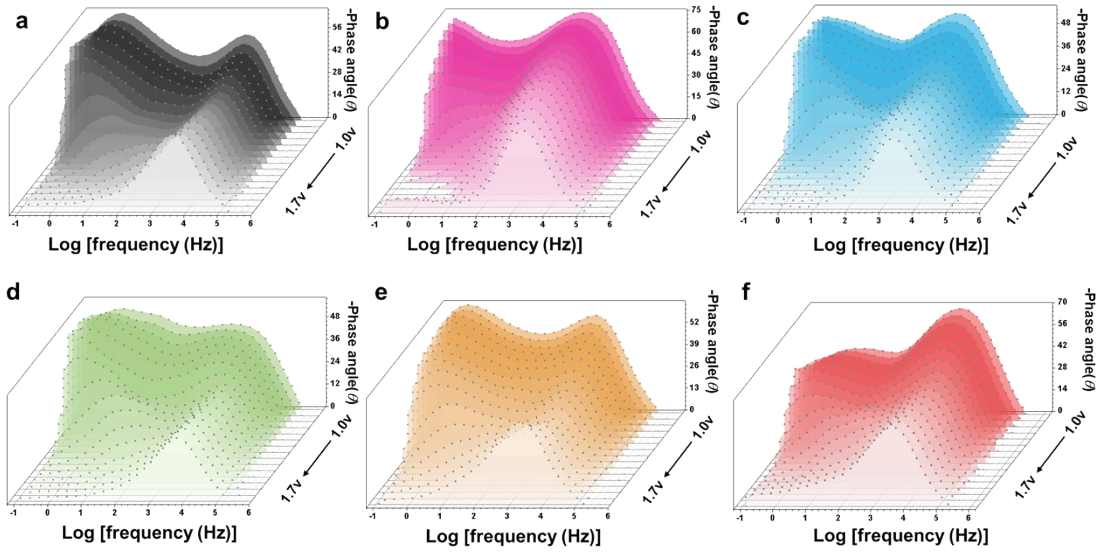

Figure S11. Bode plots of bare GC electrode, Fe, Mn, $Mn_{0.78}Fe_{0.22}$, Cu and $Cu_{0.78}Fe_{0.22}$ samples.

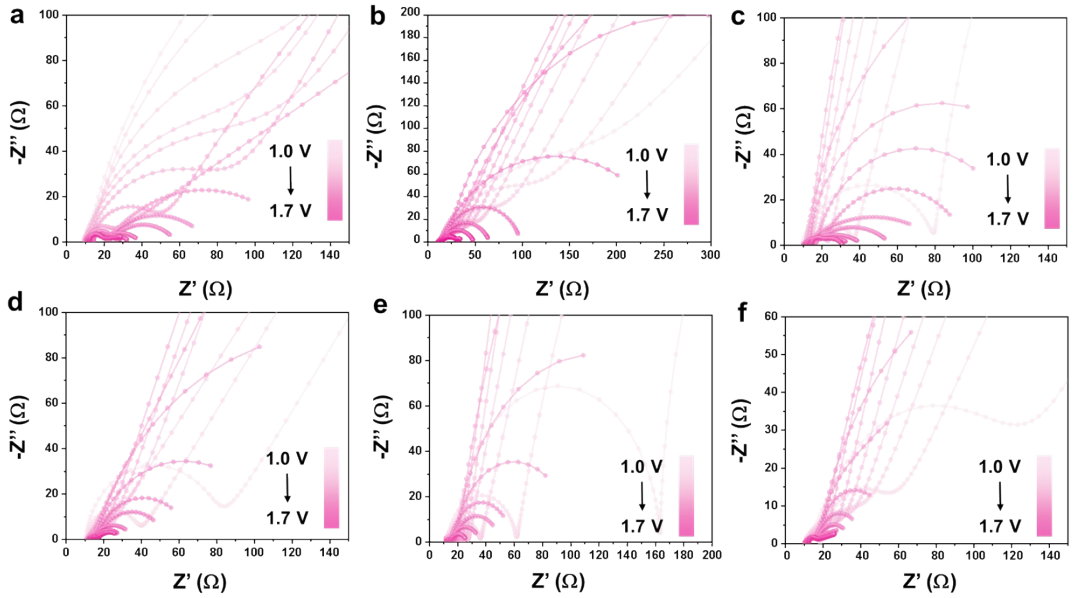

Figure S12. Nyquist plots (a-f) of Co_1Fe_0, $Co_{0.99}Fe_{0.01}$, $Co_{0.96}Fe_{0.04}$, $Co_{0.82}Fe_{0.18}$, $Co_{0.71}Fe_{0.29}$ and $Co_{0.64}Fe_{0.36}$ samples.

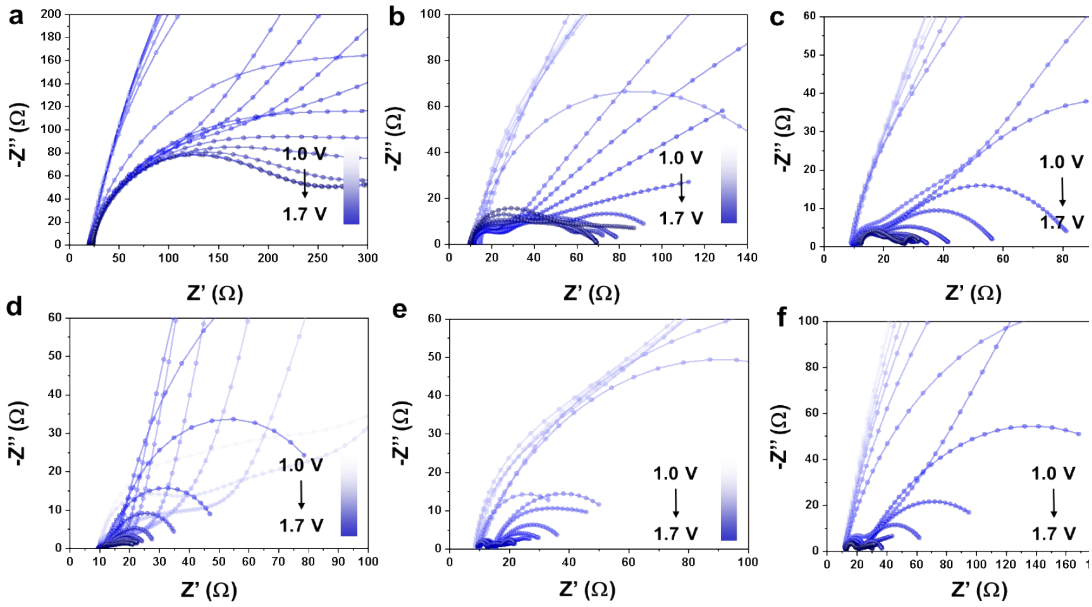

Figure S13. Nyquist plots (a-f) of Ni_1Fe_0, $Ni_{0.98}Fe_{0.02}$, $Ni_{0.94}Fe_{0.06}$, $Ni_{0.78}Fe_{0.22}$, $Ni_{0.65}Fe_{0.35}$ and $Ni_{0.56}Fe_{0.44}$ samples.

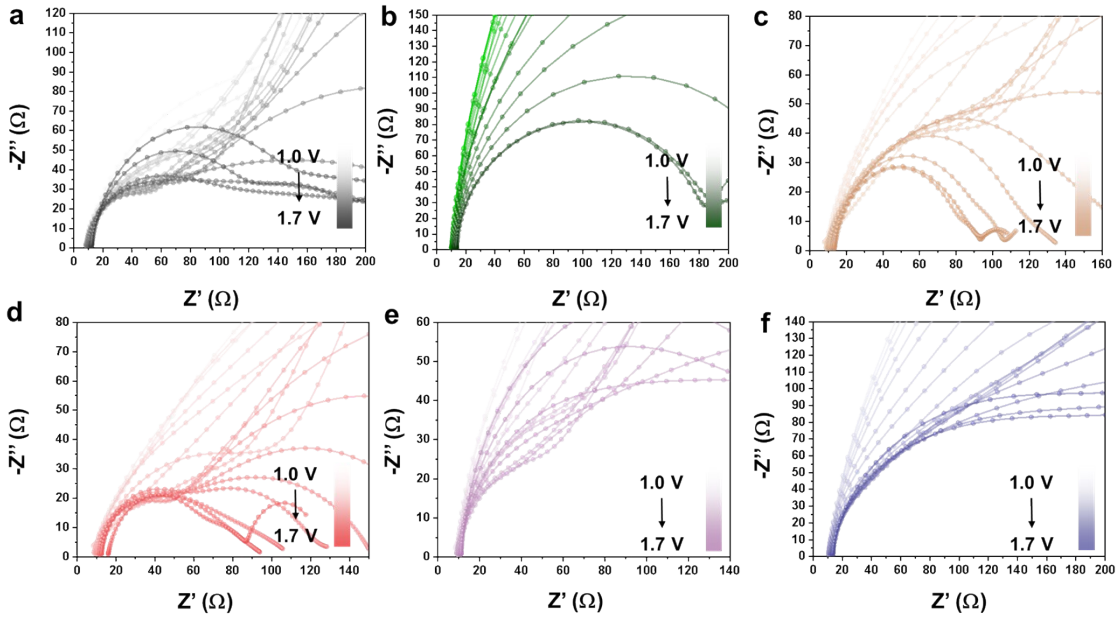

Figure S14. Nyquist plot of bare GC electrode, Fe, Mn, $Mn_{0.78}Fe_{0.22}$, Cu and $Cu_{0.78}Fe_{0.22}$ samples.

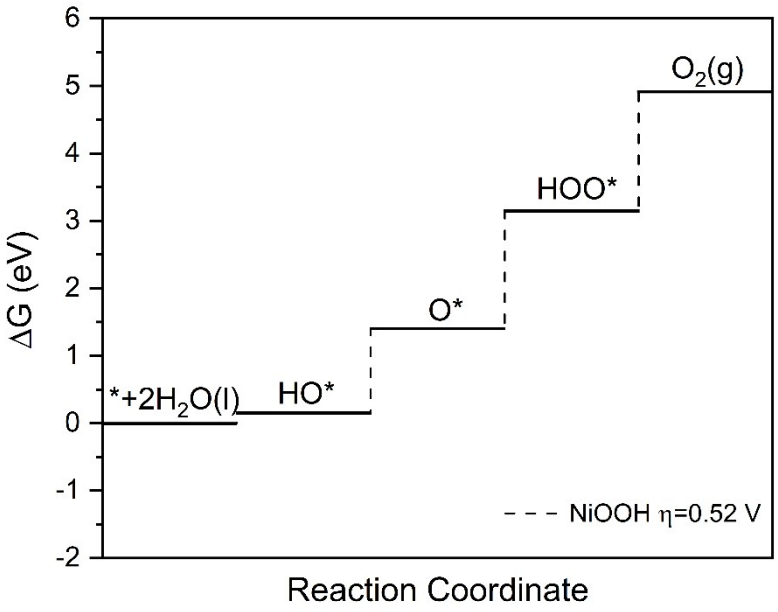

Figure S15 OER energy profile of NiOOH sample.

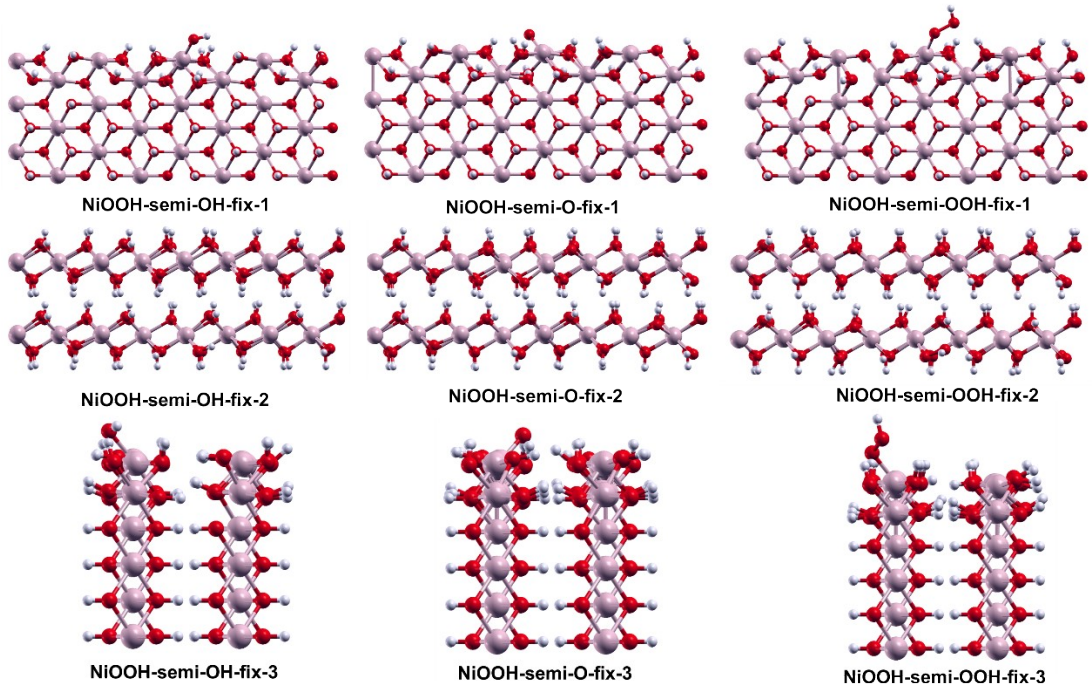

Figure S16 Top and side views of different oxygen evolution reaction steps.

Table S1. The elements contents of different samples calculated from ICP-OES experiment.

	Fe (wt)%	Ni, Co, Mn, Cu (wt) %	Total Metal (wt) %	Fe (at)%	M (at)%	Fe (at):M(at)	Fe/(Fe+M) (at)
Ni_1Fe_0	0.000	34.4970	34.4972	0.0000	0.5876	0.0000	0
$Ni_{0.98}Fe_{0.02}$	0.5901	39.9889	40.5791	0.0105	0.6812	0.0154	0.02
$Ni_{0.94}Fe_{0.06}$	2.1715	37.5607	39.7323	0.0387	0.6398	0.0606	0.06
$Ni_{0.78}Fe_{0.22}$	8.4555	30.5510	39.0065	0.1509	0.5204	0.2901	0.22
$Ni_{0.65}Fe_{0.35}$	13.5032	25.8717	39.3749	0.2411	0.4407	0.5470	0.35
$Ni_{0.56}Fe_{0.44}$	15.3287	20.5001	35.8289	0.2737	0.3492	0.7837	0.44
Co_1Fe_0	0.0000	45.2177	45.2177	0.0000	0.7677	0.0000	0.00
$Co_{0.99}Fe_{0.01}$	0.4818	40.5923	41.0742	0.0086	0.6891	0.0124	0.01
$Co_{0.96}Fe_{0.04}$	1.4496	40.0096	41.4593	0.0258	0.6792	0.0381	0.04
$Co_{0.82}Fe_{0.18}$	8.0123	37.8048	45.8172	0.1430	0.6418	0.2229	0.18
$Co_{0.71}Fe_{0.29}$	13.9608	35.7513	49.7121	0.2493	0.6069	0.4107	0.29
$Co_{0.64}Fe_{0.36}$	18.1508	33.5852	51.7360	0.3241	0.5702	0.5684	0.36
Mn	0.0000	32.7108	32.7108	0.0000	0.5958	0.0000	0
$Mn_{0.78}Fe_{0.22}$	7.0123	24.4874	31.4997	0.1252	0.4460	0.2807	0.22
Cu	0.0000	37.4368	37.4368	0.0000	0.5895	0.0000	0.00
$Cu_{0.78}Fe_{0.22}$	7.2690	29.1453	36.4143	0.1298	0.4589	0.2828	0.22
Fe	41.6372	0.0000	41.6372	0.7435	0.0000	0.0000	1

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Table S2. The calculated TOF (s^{-1}) values of different samples.

Co_1Fe_0 (S^{-1})	$Co_{0.99}Fe_{0.01}$ (S^{-1})	$Co_{0.96}Fe_{0.04}$ (S^{-1})	$Co_{0.82}Fe_{0.18}$ (S^{-1})	$Co_{0.71}Fe_{0.29}$ (S^{-1})	$Co_{0.64}Fe_{0.36}$ (S^{-1})
0.0352	0.06297	0.25567	0.45935	0.39473	0.39177
Ni_1Fe_0 (S^{-1})	$Ni_{0.98}Fe_{0.02}$ (S^{-1})	$Ni_{0.94}Fe_{0.06}$ (S^{-1})	$Ni_{0.78}Fe_{0.22}$ (S^{-1})	$Ni_{0.65}Fe_{0.35}$ (S^{-1})	$Ni_{0.56}Fe_{0.44}$ (S^{-1})
0.0115	0.01466	0.30566	0.53701	0.50396	0.49916

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Below are the tables of fitted values from different elements with the two equivalent circuit models:

Here, two constant phase elements (CPE) were used, which are CPE_{dl} and CPE_{ad} to represent the capacitance from double layer and intermediates adsorption. n is the constant phase exponent ($0 < n \leq 1$). For the circuit without R_i, equation (1) [1] was used:

$$C_{\varphi} = CPE_{ad}^{1/n} \left(\frac{1}{R_s} + 1/R_1\right) \quad (1)$$

and for the circuit with R_i, equation (2) [2] was employed:

$$C_{\varphi} = \frac{(R_i\, CPE_{ad})^{1/n}}{R_i} \quad (2)$$

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Table S3. EIS fitting data of sample Co_1Fe_0 in 1 KOH.

Potential (V vs RHE)	Rs (Ω)	CPE_{dl} (S·sec^n)	n_1	R_{ct} (Ω)	CPE_{ad} (S·sec^n)	n_2	Ri (Ω)	C (mF)
1.0365	11.50	6.06E-06	0.86	380.20	3.46E-05	0.71	-	0.00002
1.0865	13.53	5.15E-06	0.88	277.00	4.96E-05	0.71	-	0.00004
1.1365	12.97	4.23E-06	0.90	162.00	6.97E-05	0.72	-	0.00006
1.1865	12.01	4.05E-06	0.90	99.66	1.45E-04	0.65	-	0.00006
1.2365	9.18	1.62E-05	0.77	118.20	3.04E-04	0.64	-	0.00015
1.2865	11.67	1.50E-05	0.80	80.71	6.18E-04	0.64	-	0.00052
1.3365	13.32	2.80E-06	0.96	26.00	1.89E-03	0.75	-	0.00689
1.3865	10.28	1.09E-06	0.95	9.93	6.54E-03	0.77	-	0.03400
1.4365	11.81	1.32E-06	0.95	9.91	4.80E-03	0.73	-	0.05277
1.4865	12.15	3.48E-06	0.90	9.60	5.73E-03	0.75	52.71	0.05532
1.5365	14.43	1.26E-06	1.00	8.92	8.75E-03	0.81	35.23	0.09497
1.5865	10.58	1.67E-06	0.99	5.60	9.24E-03	0.86	24.91	0.10437
1.6365	12.12	1.55E-06	0.90	5.54	9.70E-03	0.93	16.24	0.12159
1.6865	10.88	5.29E-06	0.80	0.46	1.21E-02	0.93	21.46	0.15591

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Table S4. EIS fitting data of sample $Co_{0.99}Fe_{0.01}$ in 1 KOH.

Potential (V vs RHE)	Rs (Ω)	CPE_{dl} (S·sec^n)	n_1	R_{ct} (Ω)	CPE_{ad} (S·sec^n)	n_2	Ri (Ω)	C (mF)
1.0365	13.73	5.20E-05	0.85	192.20	0.0008	0.5423	-	0.00025
1.0865	14.25	5.79E-05	0.83	130.50	0.0010	0.5992	-	0.00072
1.1365	14.79	6.53E-05	0.89	71.20	0.0011	0.6348	-	0.00136
1.1865	14.06	7.32E-05	0.90	37.42	0.0014	0.6499	-	0.00214
1.2365	12.73	9.27E-05	0.84	33.02	0.0017	0.6673	-	0.00300
1.2865	14.45	3.23E-04	0.90	18.59	0.0023	0.6679	-	0.00459
1.3365	12.77	4.24E-04	0.70	18.30	0.0020	0.7073	-	0.00503
1.3865	14.19	4.80E-04	0.77	13.21	0.0028	0.7363	-	0.00981
1.4365	12.02	5.12E-04	0.71	12.12	0.0014	0.7437	346.9	0.01595
1.4865	12.34	5.97E-04	0.70	14.10	0.0019	0.8100	116.11	0.01904
1.5365	12.27	6.12E-04	0.74	6.40	0.0021	0.8361	79.95	0.02129
1.5865	12.79	6.77E-04	0.70	2.68	0.0027	0.8271	50.08	0.02592
1.6365	13.38	5.04E-04	0.78	2.69	0.0032	0.8810	32.81	0.03362
1.6865	12.73	2.43E-4	0.76	2.14	0.0029	0.9386	22.35	0.03516

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Table S5. EIS fitting data of sample $Co_{0.96}Fe_{0.04}$ in 1 KOH.

Potential (V vs RHE)	Rs (Ω)	CPE_{dl} (S·sec^n)	n_1	R_{ct} (Ω)	CPE_{ad} (S·sec^n)	n_2	Ri (Ω)	C (mF)
1.0365	13.58	3.01E-05	0.87	119.00	0.0016	0.57	-	0.00121
1.0865	11.02	1.21E-04	0.86	55.32	0.0022	0.56	-	0.00148
1.1365	10.50	8.68E-04	0.75	50.44	0.0021	0.63	-	0.00250
1.1865	10.14	1.12E-03	0.82	36.48	0.0028	0.68	-	0.00640
1.2365	9.85	1.45E-03	0.74	28.01	0.0031	0.73	-	0.01037
1.2865	10.11	1.78E-03	0.75	21.50	0.0035	0.74	-	0.01360
1.3365	10.15	1.96E-03	0.66	13.96	0.0046	0.78	-	0.02370
1.3865	10.98	3.52E-03	0.62	11.57	0.0050	0.78	-	0.02629
1.4365	9.96	1.01E-02	0.34	14.48	0.0056	0.84	-	0.04238
1.4865	9.73	1.78E-02	0.28	16.80	0.0050	0.88	-	0.04472
1.5365	12.27	3.12E-03	0.74	16.40	0.0033	0.99	79.95	0.04644
1.5865	9.84	4.11E-03	0.24	13.78	0.0044	0.94	41.64	0.05625
1.6365	9.83	7.74E-03	0.30	11.29	0.0048	0.90	17.36	0.05220
1.6865	9.34	6.13E-03	0.31	11.32	0.0053	0.92	11.24	0.05862

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Table S6. EIS fitting data of sample $Co_{0.82}Fe_{0.18}$ in 1 KOH.

Potential (V vs RHE)	Rs (Ω)	CPE_{dl} (S·sec^n)	n_1	R_{ct} (Ω)	CPE_{ad} (S·sec^n)	n_2	Ri (Ω)	C (mF)
1.0365	10.95	9.99E-07	0.94	69.46	0.0011	0.60	-	0.00074
1.0865	11.07	1.31E-06	0.93	27.61	0.0016	0.60	-	0.00122
1.1365	13.86	2.30E-03	0.53	18.64	0.0019	0.72	-	0.00545
1.1865	13.57	7.62E-04	0.74	34.46	0.0019	0.75	-	0.00720
1.2365	12.04	1.38E-03	0.66	28.07	0.0020	0.79	-	0.00965
1.2865	11.74	1.63E-03	0.69	19.08	0.0029	0.79	-	0.01468
1.3365	11.94	6.47E-03	0.65	9.90	0.0031	0.83	-	0.01980
1.3865	11.51	2.86E-03	0.73	9.01	0.0045	0.82	249.50	0.06554
1.4365	12.67	2.62E-03	0.78	5.28	0.0060	0.80	92.47	0.07437
1.4865	12.22	2.16E-03	0.83	3.17	0.0080	0.88	49.23	0.09994
1.5365	12.66	1.12E-03	0.84	2.00	0.0108	0.73	35.72	0.10830
1.5865	12.23	2.26E-03	0.64	2.76	0.0120	0.83	21.29	0.12950
1.6365	12.89	8.07E-04	0.71	2.43	0.0133	0.87	15.39	0.14935
1.6865	12.61	1.32E-04	0.85	2.17	0.0156	0.84	12.97	0.16258

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Table S7. EIS fitting data of sample $Co_{0.71}Fe_{0.29}$ in 1 KOH.

Potential (V vs RHE)	Rs (Ω)	CPE_{dl} (S·sec^n)	n_1	R_{ct} (Ω)	CPE_{ad} (S·sec^n)	n_2	Ri (Ω)	C (mF)
1.0365	19.25	5.16E-07	0.97	144.30	0.0029	0.82	-	0.02133
1.0865	13.08	1.40E-06	0.89	48.34	0.0038	0.79		0.02289
1.1365	11.79	5.09E-06	0.80	24.49	0.0047	0.79		0.02793
1.1865	12.79	8.39E-06	0.79	14.88	0.0054	0.81		0.03629
1.2365	11.68	2.03E-05	0.72	13.69	0.0058	0.85		0.04589
1.2865	12.71	2.57E-05	0.73	10.23	0.0064	0.87		0.05540
1.3365	9.45	1.55E-04	0.73	11.06	0.0070	0.88		0.06302
1.3865	12.80	9.64E-05	0.68	6.34	0.0068	0.90	153.00	0.09740
1.4365	10.61	1.08E-04	0.41	7.85	0.0077	0.91	87.92	0.10569
1.4865	12.66	4.31E-04	0.71	2.63	0.0092	0.89	43.81	0.11760
1.5365	12.38	2.41E-03	0.38	5.73	0.0108	0.90	33.56	0.13868
1.5865	12.02	1.14E-03	0.59	3.53	0.0124	0.90	20.55	0.15196
1.6365	11.21	6.34E-03	0.53	4.12	0.0152	0.92	11.38	0.18678
1.6865	10.80	4.16E-04	0.04	4.34	0.0193	0.89	8.63	0.22089

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Table S8. EIS fitting data of sample $Co_{0.64}Fe_{0.36}$ in 1 KOH.

Potential (V vs RHE)	Rs (Ω)	CPE_{dl} (S·sec^n)	n_1	R_{ct} (Ω)	CPE_{ad} (S·sec^n)	n_2	Ri (Ω)	C (mF)
1.0365	11.72	8.20E-06	0.82	69.70	0.0034	0.85	-	0.02494
1.0865	12.89	1.15E-05	0.86	22.17	0.0041	0.82		0.02789
1.1365	12.17	8.45E-05	0.79	8.49	0.0046	0.84	-	0.03210
1.1865	13.79	5.52E-03	0.79	5.40	0.0051	0.86	-	0.03904
1.2365	12.63	5.97E-03	0.86	3.80	0.0050	0.92	-	0.04820
1.2865	10.25	1.81E-04	0.70	3.32	0.0064	0.89	-	0.05446
1.3365	13.26	6.97E-04	0.92	3.10	0.0069	0.91	-	0.06745
1.3865	11.50	7.66E-04	0.90	3.93	0.0076	0.90	-	0.08090
1.4365	11.21	1.21E-03	0.77	2.08	0.0069	0.94	138.40	0.09852
1.4865	12.04	3.97E-03	0.63	1.38	0.0078	0.96	107.40	0.11104
1.5365	12.17	4.38E-03	0.61	0.96	0.0088	0.94	76.07	0.12164
1.5865	11.96	5.25E-03	0.40	1.31	0.0095	0.98	41.35	0.13359
1.6365	13.31	3.69E-04	0.51	1.25	0.0115	0.96	40.70	0.15901
1.6865	11.60	3.35E-04	1.00	2.33	0.0014	0.97	28.17	0.01874

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Table S9. EIS fitting data of sample Ni_1Fe_0 in 1 KOH.

Potential (V vs RHE)	Rs (Ω)	CPE_{dl} (S·sec^n)	n_1	R_{ct} (Ω)	CPE_{ad} (S·sec^n)	n_2	Ri (Ω)	C (mF)
1.0365	16.56	2.46E-06	0.83	2062.00	1.55E-05	0.75	-	1.32E-05
1.0865	16.51	2.50E-06	0.83	1961.00	1.68E-05	0.75	-	1.63E-05
1.1365	16.47	2.54E-06	0.83	1834.00	1.92E-05	0.76	-	2.22E-05
1.1865	16.49	2.48E-06	0.83	1608.00	2.44E-05	0.78		3.62E-05
1.2365	16.38	2.54E-06	0.83	1359.00	4.78E-05	0.78		9.38E-05
1.2865	16.56	2.29E-06	0.85	945.30	8.27E-05	0.81		2.47E-04
1.3365	16.21	5.30E-06	0.79	388.60	1.13E-04	0.85		5.39E-04
1.3865	16.75	3.00E-05	0.73	229.80	1.10E-04	0.66	12490.00	1.85E-03
1.4365	17.00	2.74E-05	0.76	235.10	1.65E-04	0.69	3709.00	1.89E-03
1.4865	17.02	2.43E-05	0.76	245.60	2.05E-04	0.86	1660.00	2.46E-03
1.5365	17.40	2.17E-05	0.75	268.50	3.79E-04	0.86	995.40	4.65E-03
1.5865	17.74	2.04E-05	0.74	279.70	5.07E-04	0.88	620.20	6.19E-03
1.6365	18.22	2.67E-05	0.75	244.80	6.10E-04	0.88	472.00	7.41E-03
1.6865	18.60	1.45E-05	0.75	236.10	7.12E-04	0.87	353.10	8.25E-03

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Table S10. EIS fitting data of sample $Ni_{0.98}Fe_{0.02}$ in 1 KOH.

Potential (V vs RHE)	Rs (Ω)	CPE_{dl} (S·sec^n)	n_1	R_{ct} (Ω)	CPE_{ad} (S·sec^n)	n_2	Ri (Ω)	C (mF)
1.0365	8.94	6.93E-06	0.83	570.60	1.32E-05	0.75	-	9.82E-06
1.0865	8.97	6.99E-06	0.83	561.30	1.34E-05	0.76	-	1.13E-05
1.1365	8.96	6.72E-06	0.83	505.70	1.48E-05	0.79	-	1.87E-05
1.1865	9.07	5.64E-06	0.85	355.20	2.21E-05	0.78	-	2.77E-05
1.2365	14.39	8.76E-06	0.83	792.90	1.36E-04	0.85	-	6.46E-04
1.2865	8.74	1.14E-05	0.79	577.30	2.29E-04	0.80	-	7.07E-04
1.3365	9.19	6.31E-06	0.86	152.50	4.58E-04	0.80	-	1.61E-03
1.3865	9.28	7.29E-06	0.86	14.00	1.92E-03	0.82	-	1.02E-02
1.4365	9.31	2.67E-05	0.75	13.80	5.81E-03	0.83	-	4.15E-02
1.4865	8.48	6.00E-04	0.77	35.36	4.17E-03	0.70	150.90	4.89E-02
1.5365	9.90	7.28E-03	0.72	1.42	5.37E-03	0.90	378.00	8.31E-02
1.5865	8.38	3.18E-03	0.29	1.64	6.38E-03	0.90	103.40	8.70E-02
1.6365	9.75	1.26E-03	0.70	6.90	7.55E-03	0.84	67.08	9.48E-02
1.6865	9.44	1.27E-03	0.80	8.10	7.46E-03	0.85	55.81	9.13E-02

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Table S11. EIS fitting data of sample $Ni_{0.94}Fe_{0.06}$ in 1 KOH.

Potential (V vs RHE)	Rs (Ω)	CPE_{dl} (S·sec^n)	n_1	R_{ct} (Ω)	CPE_{ad} (S·sec^n)	n_2	Ri (Ω)	C (mF)
1.0365	9.79	8.61E-06	0.81	644.70	1.09E-05	0.76	-	8.75E-06
1.0865	9.82	8.49E-06	0.81	616.10	2.14E-05	0.76	-	1.89E-05
1.1365	9.86	8.07E-06	0.81	543.20	4.32E-05	0.83	-	1.27E-04
1.1865	9.07	5.24E-06	0.85	237.90	7.52E-05	0.84	-	2.67E-04
1.2365	9.77	5.18E-06	0.85	48.14	9.77E-05	0.83	-	3.02E-04
1.2865	9.62	1.31E-05	0.77	99.40	2.92E-04	0.80	-	9.31E-04
1.3365	9.93	8.85E-05	0.81	38.20	7.29E-04	0.69	-	1.07E-03
1.3865	9.06	1.85E-05	0.69	41.70	3.95E-04	0.85	-	2.04E-03
1.4365	10.13	2.87E-05	0.74	14.24	6.37E-04	0.77	102.40	4.08E-03
1.4865	9.77	5.13E-05	0.887	8.82	8.68E-04	0.83	68.01	6.83E-03
1.5365	9.05	3.54E-05	0.74	1.03	8.45E-04	0.89	57.22	8.33E-03
1.5865	10.29	1.08E-04	0.70	4.67	1.17E-03	0.84	28.18	8.92E-03
1.6365	9.53	5.29E-04	0.76	1.55	1.21E-03	0.90	17.40	1.12E-02
1.6865	10.25	8.62E-05	0.70	4.25	1.38E-03	0.85	10.77	9.42E-03

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Table S12. EIS fitting data of sample $Ni_{0.78}Fe_{0.22}$ in 1 KOH.

Potential (V vs RHE)	Rs (Ω)	CPE_{dl} (S·sec^n)	n_1	R_{ct} (Ω)	CPE_{ad} (S·sec^n)	n_2	Ri (Ω)	C (mF)
1.0365	10.54	1.64E-06	0.80	42.02	1.56E-03	0.66	-	2.31E-03
1.0865	9.51	6.29E-05	0.60	68.93	2.20E-03	0.70	-	5.65E-03
1.1365	9.52	7.24E-04	0.71	57.78	2.17E-03	0.80	-	1.10E-02
1.1865	9.28	9.91E-04	0.53	32.87	2.62E-03	0.84	-	1.75E-02
1.2365	10.13	4.60E-04	0.62	46.79	3.17E-03	0.84	-	2.56E-02
1.2865	10.13	7.75E-04	0.62	10.95	4.27E-03	0.85	-	3.18E-02
1.3365	9.70	1.37E-03	0.66	8.86	4.92E-03	0.85	-	3.64E-02
1.3865	12.95	7.40E-03	0.74	8.60	6.08E-03	0.84	-	4.43E-02
1.4365	10.91	1.53E-03	0.55	3.91	5.75E-03	0.89	85.76	7.54E-02
1.4865	9.70	2.02E-03	0.58	2.59	7.42E-03	0.85	41.67	8.61E-02
1.5365	9.58	2.51E-03	0.65	2.72	9.80E-03	0.76	26.83	9.25E-02
1.5865	10.57	2.42E-02	0.54	6.16	1.15E-02	0.92	11.09	1.38E-01
1.6365	10.71	3.63E-02	0.62	3.14	1.43E-02	0.93	9.15	1.76E-01
1.6865	10.85	6.63E-03	0.62	3.47	1.79E-02	0.89	3.44	1.82E-01

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Table S13. EIS fitting data of sample $Ni_{0.65}Fe_{0.35}$ in 1 KOH.

Potential (V vs RHE)	Rs (Ω)	CPE_{dl} (S·sec^n)	n_1	R_{ct} (Ω)	CPE_{ad} (S·sec^n)	n_2	Ri (Ω)	C (mF)
1.0365	9.18	1.01E-05	0.74	209.80	4.15E-05	0.79	-	7.43E-05
1.0865	9.17	9.40E-06	0.75	198.60	4.37E-05	0.81	-	9.81E-05
1.1365	9.18	8.63E-06	0.75	187.00	4.68E-05	0.86	-	1.97E-04
1.1865	9.28	8.23E-06	0.76	178.80	5.37E-05	0.87	-	2.56E-04
1.2365	8.21	1.22E-05	0.72	195.00	6.71E-05	0.87	-	3.17E-04
1.2865	9.46	5.84E-06	0.79	124.60	1.48E-04	0.86	-	7.28E-04
1.3365	8.02	1.85E-05	0.70	161.50	1.15E-03	0.76	-	3.79E-03
1.3865	9.62	2.49E-06	0.89	33.54	2.96E-03	0.72	-	9.57E-03
1.4365	8.24	2.28E-05	0.74	12.01	3.18E-03	0.82	-	1.85E-02
1.4865	6.14	2.33E-05	0.79	28.00	3.75E-03	0.89	-	3.35E-02
1.5365	8.20	2.77E-05	0.67	18.76	5.56E-03	0.89	18.17	5.98E-02
1.5865	9.36	4.46E-05	0.62	10.85	7.48E-03	0.84	9.87	6.49E-02
1.6365	7.90	1.08E-4	0.67	8.32	7.95E-03	0.86	5.22	6.69E-02
1.6865	8.60	7.29E-05	0.63	4.45	8.42E-03	0.90	1.17	7.13E-02

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Table S14. EIS fitting data of sample $Ni_{0.56}Fe_{0.44}$ in 1 KOH.

Potential (V vs RHE)	Rs (Ω)	CPE_{dl} (S·sec^n)	n_1	R_{ct} (Ω)	CPE_{ad} (S·sec^n)	n_2	Ri (Ω)	C (mF)
1.0365	10.81	9.17E-07	0.85	1611.00	1.78E-05	0.76	-	1.64E-05
1.0865	11.79	9.44E-07	0.85	1416.00	1.95E-05	0.76	-	2.02E-05
1.1365	10.01	1.30E-06	0.83	1250.00	2.26E-05	0.76	-	2.18E-05
1.1865	11.58	1.26E-06	0.83	984.20	3.53E-05	0.72	-	2.31E-05
1.2365	11.49	1.44E-06	0.83	751.50	9.19E-05	0.73	-	1.02E-04
1.2865	11.19	2.36E-06	0.79	577.90	1.64E-04	0.79	-	4.45E-04
1.3365	11.07	4.47E-06	0.75	303.20	2.63E-04	0.76	-	5.63E-04
1.3865	8.86	4.33E-05	0.70	20.46	1.49E-03	0.80	-	6.47E-03
1.4365	12.06	2.74E-06	0.84	12.79	1.19E-03	0.81	125.50	1.08E-02
1.4865	12.45	1.92E-06	0.89	12.81	2.86E-03	0.75	92.52	2.65E-02
1.5365	10.66	1.12E-05	0.73	13.44	3.45E-03	0.82	43.19	3.21E-02
1.5865	11.79	2.97E-05	0.73	3.03	4.46E-03	0.84	24.49	4.19E-02
1.6365	10.04	1.08E-5	0.77	3.02	7.00E-03	0.83	5.22	5.01E-02
1.6865	10.00	7.29E-05	0.73	4.45	7.72E-03	0.87	1.17	5.40E-02

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Table S15. EIS fitting data of sample $Mn_{0.78}Fe_{0.22}$ in 1 KOH.

Potential (V vs RHE)	Rs (Ω)	CPE_{dl} (S·sec^n)	n_1	R_{ct} (Ω)	CPE_{ad} (S·sec^n)	n_2	Ri (Ω)	C (mF)
1.0365	9.59	1.27E-05	0.76	140.10	1.10E-05	0.60	-	1.49E-05
1.0865	9.41	1.64E-05	0.74	174.10	1.12E-05	0.61	-	1.92E-05
1.1365	9.30	1.97E-05	0.73	194.00	1.15E-05	0.64	-	3.17E-05
1.1865	9.26	2.15E-05	0.72	188.20	1.38E-05	0.66	-	5.46E-05
1.2365	9.20	2.20E-05	0.72	161.60	2.28E-04	0.62	-	7.27E-05
1.2865	9.00	2.99E-05	0.69	157.80	3.76E-04	0.66	-	2.53E-04
1.3365	9.31	2.73E-05	0.74	87.27	5.54E-04	0.61	-	2.51E-04
1.3865	9.11	3.87E-05	0.92	30.97	9.24E-04	0.46	-	3.41E-04
1.4365	9.23	3.57E-05	0.87	39.84	5.28E-04	0.57	675.50	3.50E-04
1.4865	9.93	2.11E-04	0.91	36.02	5.36E-04	0.57	351.10	7.55E-04
1.5365	10.69	2.48E-04	0.94	33.43	5.82E-04	0.57	218.10	8.62E-04
1.5865	10.62	3.06E-04	0.97	31.27	6.08E-04	0.84	152.90	4.44E-03
1.6365	10.29	3.17E-04	0.97	33.68	4.60E-04	0.74	107.10	3.23E-03
1.6865	10.45	2.94E-04	0.90	26.78	3.60E-04	0.70	90.76	1.64E-03

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Table S16. EIS fitting data of sample Mn in 1 KOH.

Potential (V vs RHE)	Rs (Ω)	CPE_{dl} (S·sec^n)	n_1	R_{ct} (Ω)	CPE_{ad} (S·sec^n)	n_2	Ri (Ω)	C (mF)
1.0365	9.96	1.10E-05	0.82	239.80	1.65E-04	0.60	-	3.20E-05
1.0865	9.90	1.24E-05	0.82	219.30	1.58E-04	0.61	-	3.16E-05
1.1365	9.85	1.48E-05	0.80	196.80	1.64E-04	0.62	-	4.40E-05
1.1865	9.87	1.58E-05	0.80	168.30	2.15E-04	0.62	-	6.02E-05
1.2365	9.80	2.04E-05	0.78	153.80	3.18E-04	0.61	-	1.05E-04
1.2865	9.71	2.40E-05	0.77	144.20	4.39E-04	0.64	-	2.72E-04
1.3365	9.81	2.27E-05	0.77	112.80	5.11E-04	0.66	-	4.35E-04
1.3865	9.04	2.21E-05	0.83	72.70	6.98E-04	0.54	-	1.14E-04
1.4365	9.09	2.53E-05	0.80	88.38	4.49E-04	0.61	965.00	4.70E-03
1.4865	9.42	2.46E-05	0.81	85.92	3.80E-04	0.61	662.20	2.27E-03
1.5365	9.74	3.15E-05	0.83	69.91	3.85E-04	0.76	366.00	1.18E-03
1.5865	9.39	3.44E-05	1.00	11.78	4.15E-04	0.88	252.10	4.30E-03
1.6365	9.34	3.46E-05	1.00	9.72	1.47E-04	0.75	155.20	9.09E-04
1.6865	9.60	2.54E-05	0.81	0.02	1.98E-04	0.08	164.80	1.50E-04

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Table S17. EIS fitting data of sample $Cu_{0.78}Fe_{0.22}$ in 1 KOH.

Potential (V vs RHE)	Rs (Ω)	CPE_{dl} (S·sec^n)	n_1	R_{ct} (Ω)	CPE_{ad} (S·sec^n)	n_2	Ri (Ω)	C (mF)
1.0365	10.55	1.23E-06	0.92	569.90	6.20E-05	0.49		4.07E-07
1.0865	10.55	1.36E-06	0.91	630.70	5.74E-05	0.52		7.88E-07
1.1365	10.41	1.53E-06	0.90	627.40	5.72E-05	0.54		1.39E-06
1.1865	10.35	1.88E-06	0.89	620.50	6.13E-05	0.56		2.53E-06
1.2365	10.22	2.14E-06	0.88	545.40	7.68E-05	0.55		3.06E-06
1.2865	10.11	2.64E-06	0.86	478.20	1.04E-04	0.54		4.64E-06
1.3365	10.18	2.42E-06	0.87	303.10	1.44E-04	0.51		3.36E-06
1.3865	10.54	1.32E-06	0.92	121.20	2.01E-04	0.45		1.32E-06
1.4365	10.52	1.70E-06	0.90	127.20	1.70E-04	0.50	5864.00	2.43E-03
1.4865	10.80	1.03E-06	0.95	84.95	1.42E-04	0.51	2885.00	8.59E-04
1.5365	11.06	9.09E-07	0.96	76.08	1.07E-04	0.54	1644.00	3.53E-04
1.5865	11.52	6.64E-07	0.99	61.04	9.83E-05	0.84	1007.00	9.03E-04
1.6365	11.89	5.99E-07	1.00	45.25	1.20E-04	0.50	719.60	1.49E-04
1.6865	11.51	2.52E-04	0.37	1.00	5.66E-07	1.00	677.10	8.09E-05

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Table S18. EIS fitting data of sample Cu in 1 KOH.

Potential (V vs RHE)	Rs (Ω)	CPE_{dl} (S·sec^n)	n_1	R_{ct} (Ω)	CPE_{ad} (S·sec^n)	n_2	Ri (Ω)	C (mF)
1.0365	8.89	7.19E-06	0.84	354.00	5.60E-05	0.70	-	2.83E-05
1.0865	8.91	7.09E-06	0.84	297.80	6.05E-05	0.69	-	2.98E-05
1.1365	8.92	7.44E-06	0.84	243.70	6.52E-05	0.69	-	3.19E-05
1.1865	7.16	8.87E-05	0.60	11.02	4.93E-14	0.80	--	5.08E-5
1.2365	8.85	8.96E-06	0.82	194.70	8.96E-05	0.67	-	3.69E-05
1.2865	8.84	9.98E-06	0.81	166.80	1.18E-04	0.65	-	4.09E-05
1.3365	8.85	1.16E-05	0.80	124.90	1.52E-04	0.65	-	5.74E-05
1.3865	8.89	1.02E-05	0.82	90.59	2.01E-04	0.64	-	7.69E-05
1.4365	9.14	3.04E-06	0.51	9740.00	9.58E-08	0.73	46790.00	1.92E-05
1.4865	9.68	3.87E-06	0.48	40100.00	5.27E-07	0.78	55430.00	2.79E-05
1.5365	9.17	8.40E-06	0.84	54.62	3.41E-04	0.62	1956.00	3.79E-04
1.5865	9.23	7.89E-06	0.85	41.05	2.19E-04	0.75	564.70	1.54E-03
1.6365	9.66	3.81E-06	0.91	33.48	6.37E-04	0.50	495.10	2.83E-03
1.6865	10.14	1.80E-06	0.98	18.33	8.30E-04	0.39	373.00	1.83E-03

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Table S19. EIS fitting data of sample Fe in 1 KOH.

Potential (V vs RHE)	Rs (Ω)	CPE_{dl} (S·sec^n)	n_1	R_{ct} (Ω)	CPE_{ad} (S·sec^n)	n_2	Ri (Ω)	C (mF)
1.0365	10.30	2.39E-06	0.92	2776.00	1.36E-05	0.71		4.96E-06
1.0865	10.32	2.30E-06	0.92	2480.00	1.38E-05	0.73		6.86E-06
1.1365	10.32	2.24E-06	0.93	2207.00	1.44E-05	0.73		8.55E-06
1.1865	10.40	2.11E-06	0.93	1922.00	1.60E-05	0.72		7.77E-06
1.2365	10.47	1.86E-06	0.95	1371.00	2.15E-05	0.67		4.88E-06
1.2865	10.48	1.80E-06	0.95	1207.00	3.14E-05	0.62		3.15E-06
1.3365	10.48	1.71E-06	0.95	914.90	4.64E-05	0.58		2.76E-06
1.3865	10.71	1.02E-06	1.00	186.20	9.09E-05	0.40		3.74E-08
1.4365	10.45	1.90E-06	0.95	730.50	5.29E-05	0.62	5881.00	3.73E-04
1.4865	10.32	2.42E-06	0.92	0.00	2.82E-04	0.18	6641000	4.20E+43
1.5365	10.71	1.93E-06	0.95	649.90	2.82E-05	0.73	1005.00	1.07E-04
1.5865	10.94	2.11E-06	0.94	618.40	8.55E-05	0.98	306.70	1.14E-03
1.6365	11.37	2.02E-06	0.95	403.80	5.59E-06	0.90	197.70	7.98E-03
1.6865	11.91	2.08E-06	0.95	293.00	5.28E-06	0.90	117.50	7.54E-03

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References:

1. R. Ge, Y. Wang, Z. Li, M. Xu, S.-M. Xu, H. Zhou, K. Ji, F. Chen, J. Zhou, H. Duan, Selective Electrooxidation of Biomass-Derived Alcohols to Aldehydes in a Neutral Medium: Promoted Water Dissociation over a Nickel-Oxide-Supported Ruthenium Single-Atom Catalyst, Angewandte Chemie International Edition 61 (19) (2022) e202200211.

2. F. Dionigi, J. Zhu, Z. Zeng, T. Merzdorf, H. Sarodnik, M. Gliech, L. Pan, W.-X. Li, J. Greeley, P. Strasser, Intrinsic Electrocatalytic Activity for Oxygen Evolution of Crystalline 3d-Transition Metal Layered Double Hydroxides, Angewandte Chemie International Edition 60 (26) (2021) 14446-14457.

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