Supplement Information

Optimized Ni, Co, Mn Oxides Anchored on Graphite Plates for Highly Efficient Overall Water Splitting

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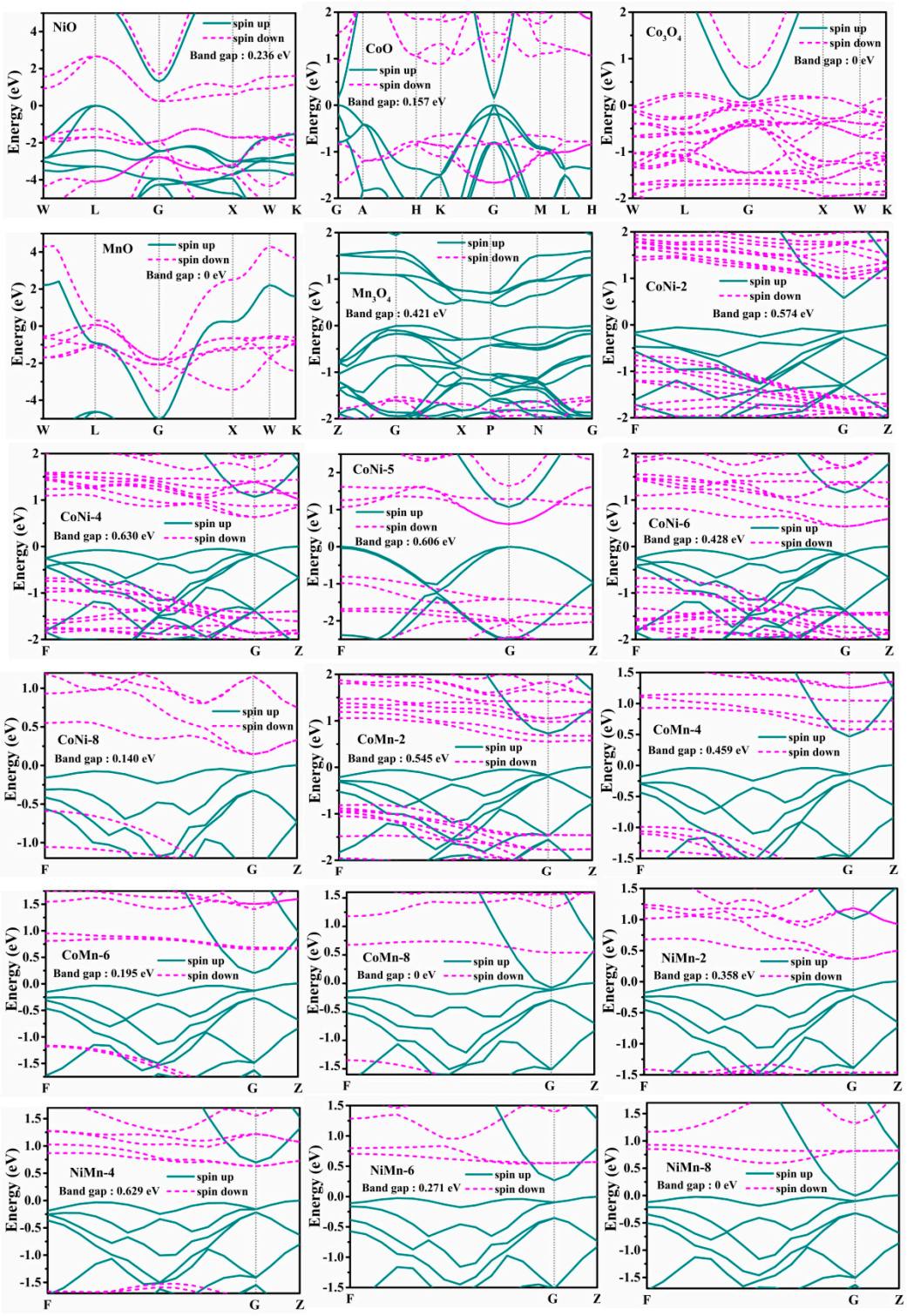


Figure S1 Band structure of Co_3O_4, Mn_3O_4, $Co_xNi_{1-x}O$, $Ni_xMn_{1-x}O$, $Co_{1-x}Mn_xO$ (x = 0, 0.2, 0.4, 0.6, 0.8).

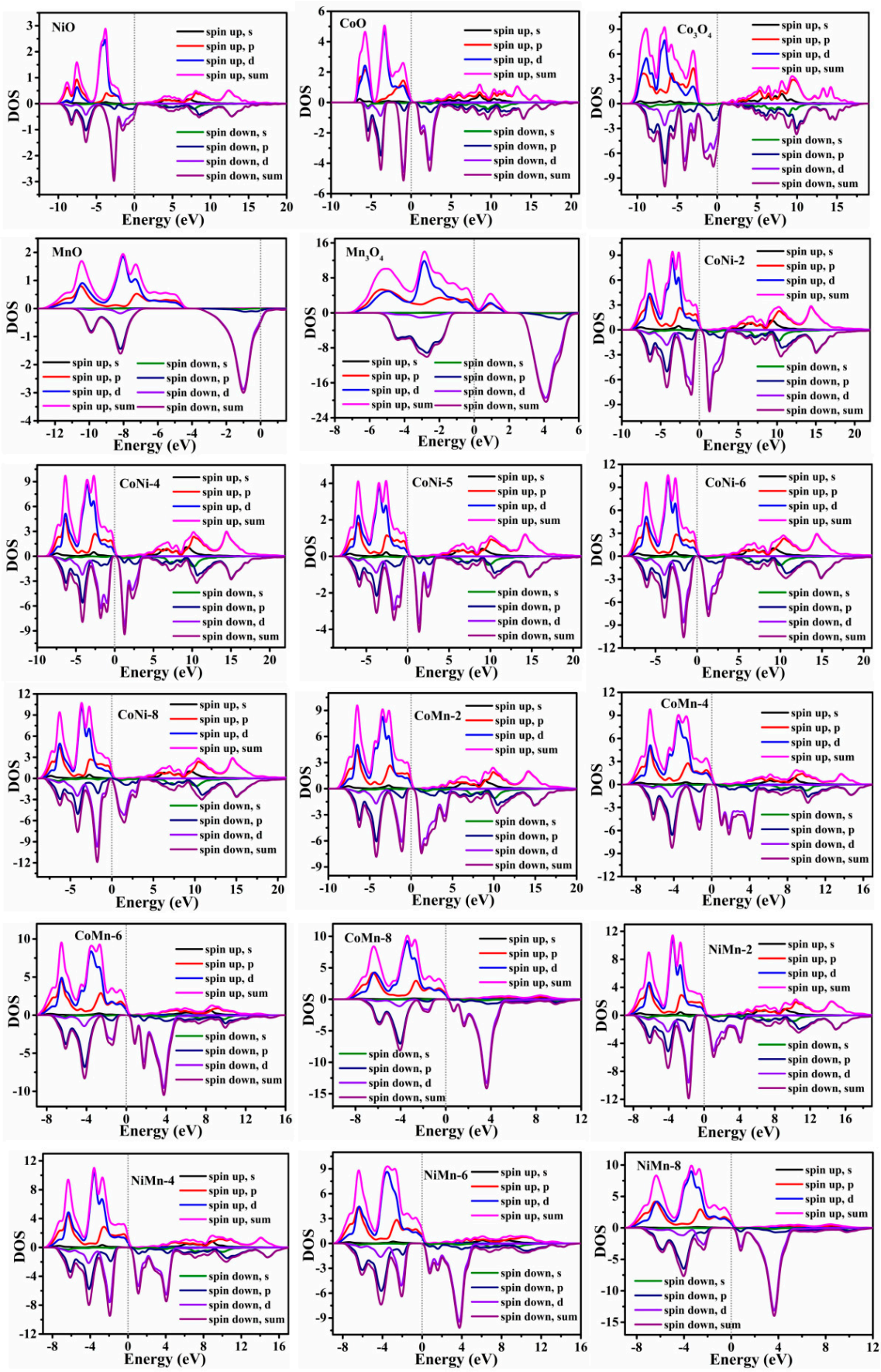


Figure S2 DOS of Co_3O_4, Mn_3O_4, $Co_xNi_{1-x}O$, $Ni_xMn_{1-x}O$, $Co_{1-x}Mn_xO$ (x = 0, 0.2, 0.4, 0.6, 0.8).

Table S1 Band gap of Co_3O_4, Mn_3O_4, $Co_xNi_{1-x}O$, $Ni_xMn_{1-x}O$, $Co_{1-x}Mn_xO$ (x = 0, 0.2, 0.4, 0.6, 0.8).

No.	Materials	Band gap
1	NiMn-8	0
2	CoMn-8	0
3	Co_3O_4	0
4	MnO	0
5	CoNi-8	0.140
6	CoO	0.157
7	CoMn-6	0.195
8	NiO	0.236
9	NiMn-6	0.271
10	NiMn-2	0.358
11	Mn_3O_4	0.421
12	CoNi-6	0.428
13	CoMn-4	0.459
14	CoMn-2	0.545
15	CoNi-2	0.574
16	CoNi-5	0.606
17	NiMn-4	0.629
18	CoNi-4	0.630

Table S2 d-band center values of Co_3O_4, Mn_3O_4, $Co_xNi_{1-x}O$, $Ni_xMn_{1-x}O$, $Co_{1-x}Mn_xO$ (x = 0, 0.2, 0.4, 0.6, 0.8).

No.	Materials	d-band center (eV)		
		Spin up	Spin down	Average
1	NiO	4.6097	2.8516	3.7307
2	CoO	3.6863	0.3398	2.0131
3	Co_3O_4	7.0021	2.9789	4.9905
4	MnO	8.4377	1.5909	5.0143
5	Mn_3O_4	2.9809	3.6133	3.2971
6	CoNi-2	4.0919	0.2828	2.1873
7	CoNi-4	4.0253	0.5215	2.2734
8	CoNi-5	3.8768	0.5166	2.1967
9	CoNi-6	3.9178	0.7105	2.3141
10	CoNi-8	3.9659	1.0698	2.5178
11	CoMn-2	4.1097	0.5593	2.3345
12	CoMn-4	4.0775	1.1458	2.6116
13	CoMn-6	4.0495	1.7190	2.8842
14	CoMn-8	4.0092	2.2855	3.1473
15	NiMn-2	3.9081	0.4547	2.1814
16	NiMn-4	3.9358	0.3805	2.1582
17	NiMn-6	3.9752	1.1857	2.5804
18	NiMn-8	3.9792	2.0114	2.9953

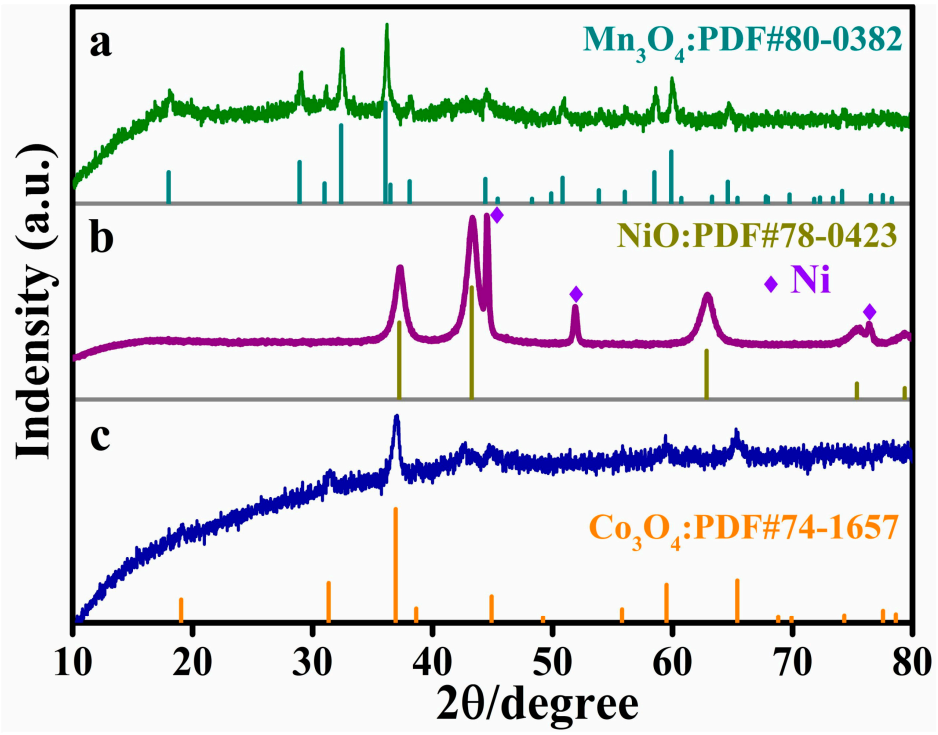


Figure S3 (a) XRD pattern of heated $Mn(CH_3COO)_2 \cdot 4H_2O$, (b) XRD pattern of heated $Ni(CH_3COO)_2 \cdot 4H_2O$, (c) XRD pattern of heated $Co(CH_3COO)_2 \cdot 4H_2O$.

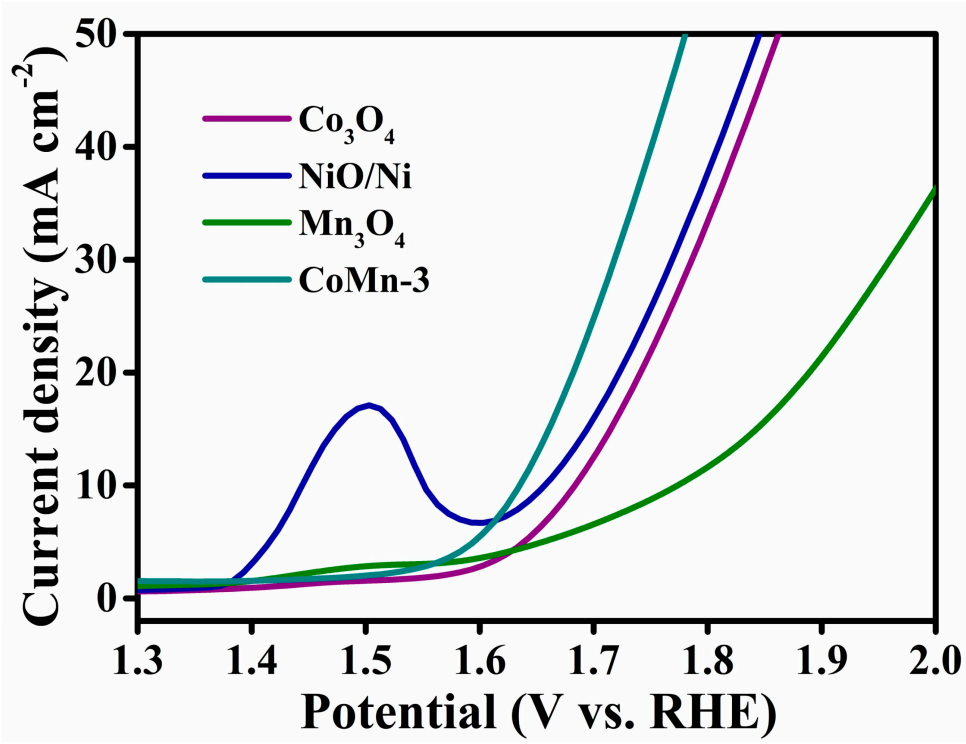


Figure S4 LSV of CoMn-3, Ni/NiO, Co_3O_4 and Mn_3O_4 without IR compensation. Notice that Ni/NiO, Co_3O_4 and Mn_3O_4 were deposed on graphite plates by immersing the graphite plates in 1.0 mol/L $Ni(CH_3COO)_2 \cdot 4H_2O$, $Co(CH_3COO)_2 \cdot 4H_2O$, and $Mn(CH_3COO)_2 \cdot 4H_2O$ aqueous solution, then dried, and heated at 300 °C for 2 h.

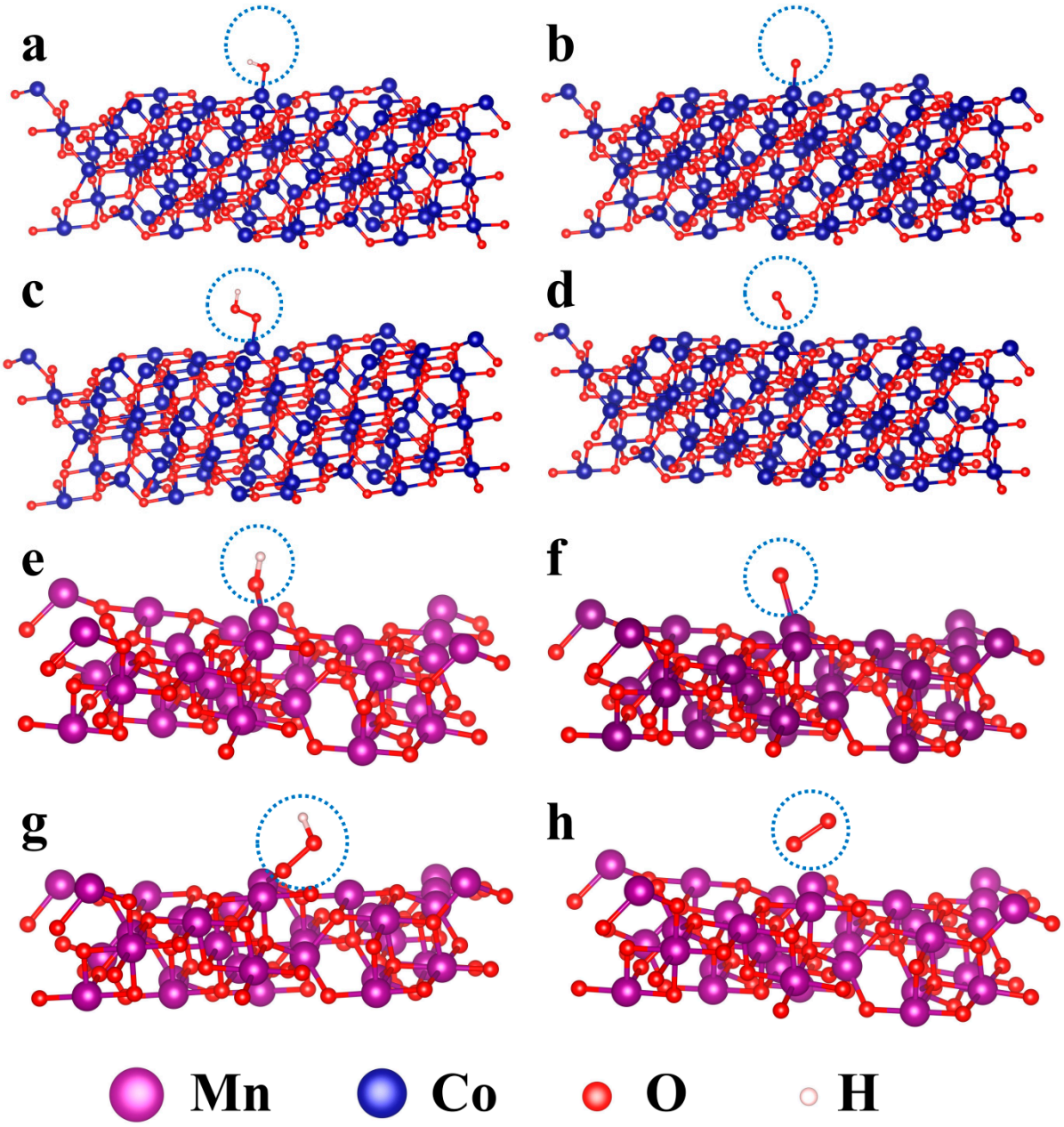


Figure S5 (a-h) The OER process on Co_3O_4. (a) based-OH^* radical, (b) based-O^* radical, (c) based-OOH^* radical, (d) based + O_2. The O_2 is desorb and released. (e-f) The OER process on Mn_3O_4. (e) based-OH^* radical, (f) based-O^* radical, (g) based-OOH^* radical, (h) based + O_2. The O_2 is desorb and released.

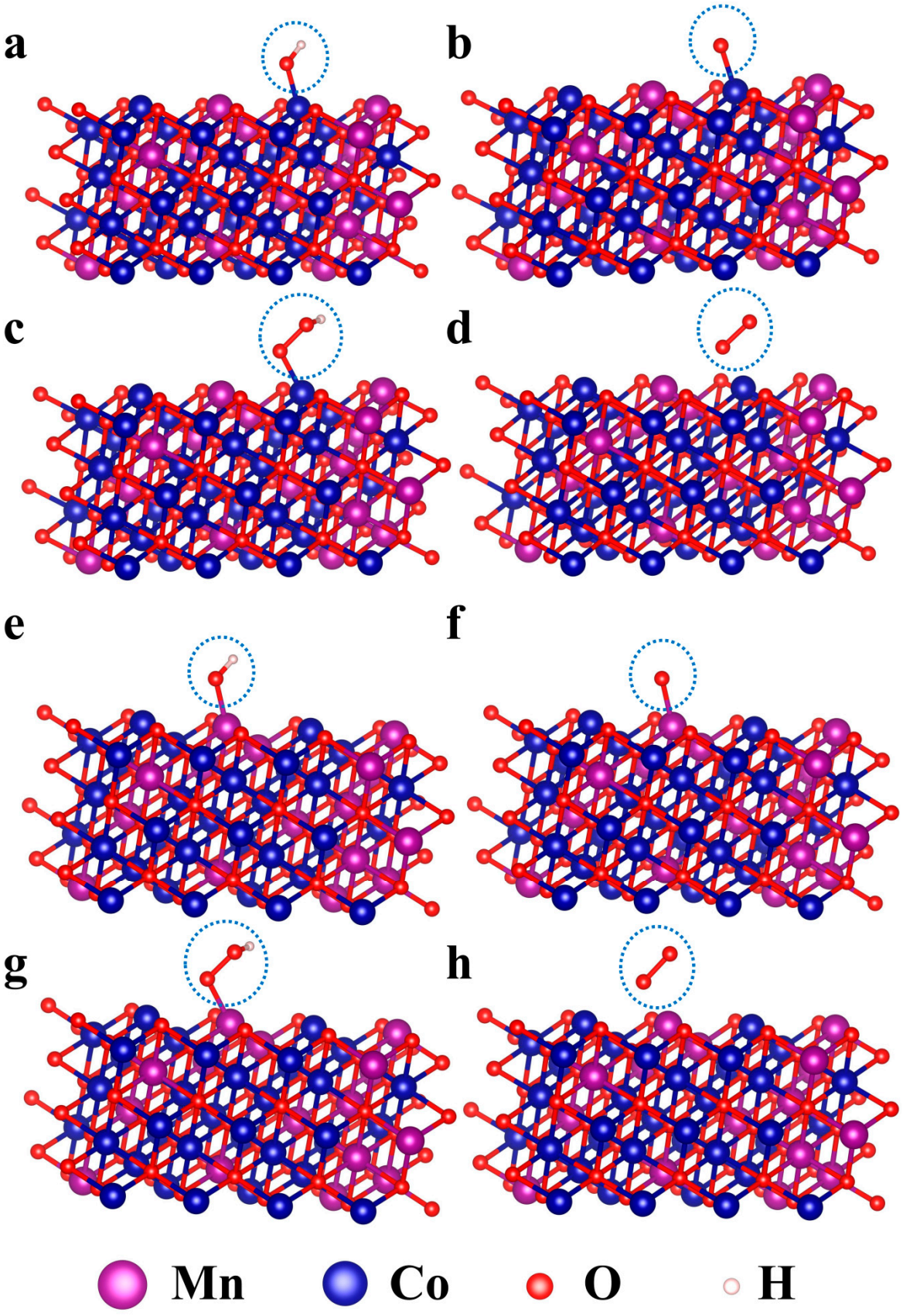


Figure S6 The OER process of H_2O on CoMn-3. (a-d) OER process on Co site, (e-h) OER process on Mn site. (a, e) based-OH^* radical, (b, f) based-O^* radical, (c, g) based-OOH^* radical, (d, h) based + O_2. The O_2 is desorb and released.

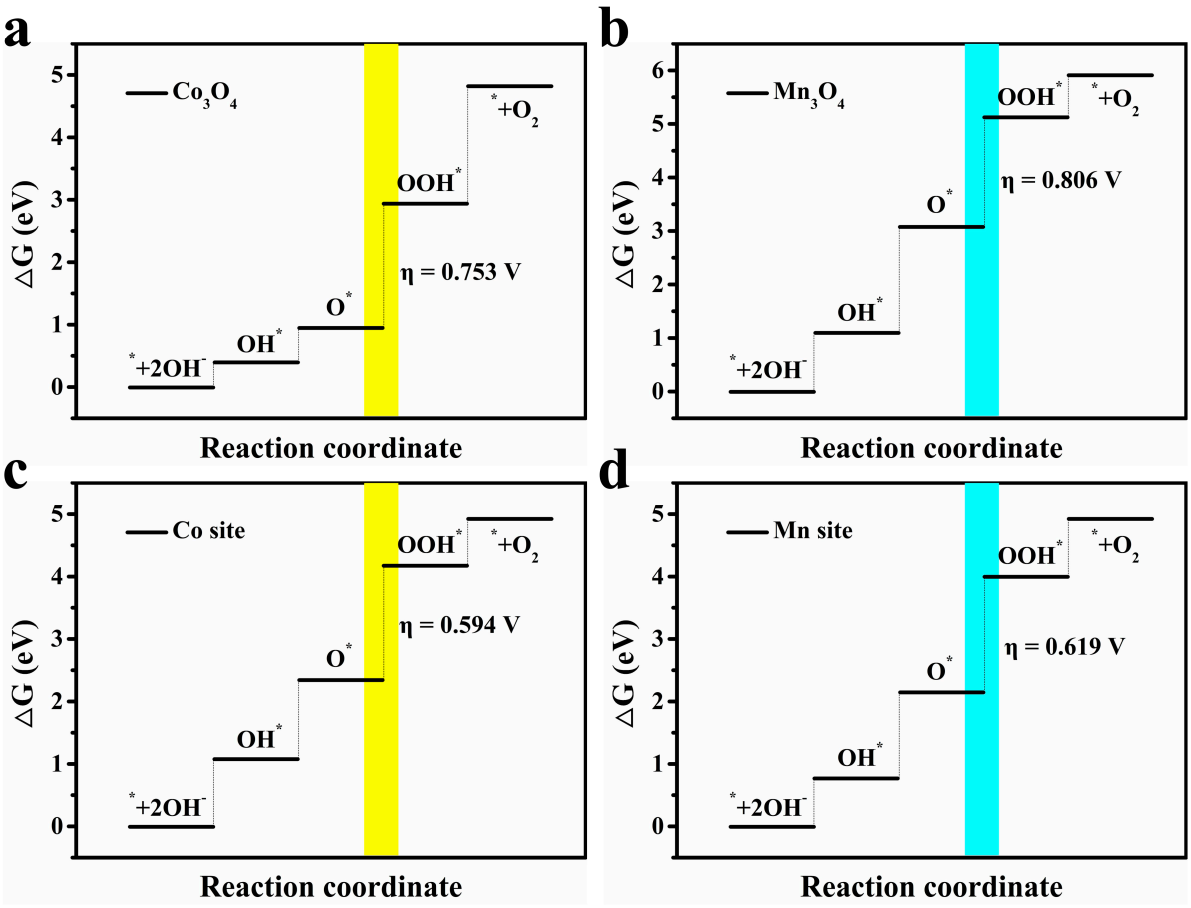


Figure S7 Theoretical calculated Gibbs free energy of four elementary reactions of OER. (a) on Co_3O_4, (b) on Mn_3O_4, (c) on Co site of CoMn-3, (d) on Mn site of CoMn-3. The overpotential was calculated based on:

$\eta = |\max\{\Delta G_1, \Delta G_2, \Delta G_3, \Delta G_4\}/e^-| - 1.23$ (V).

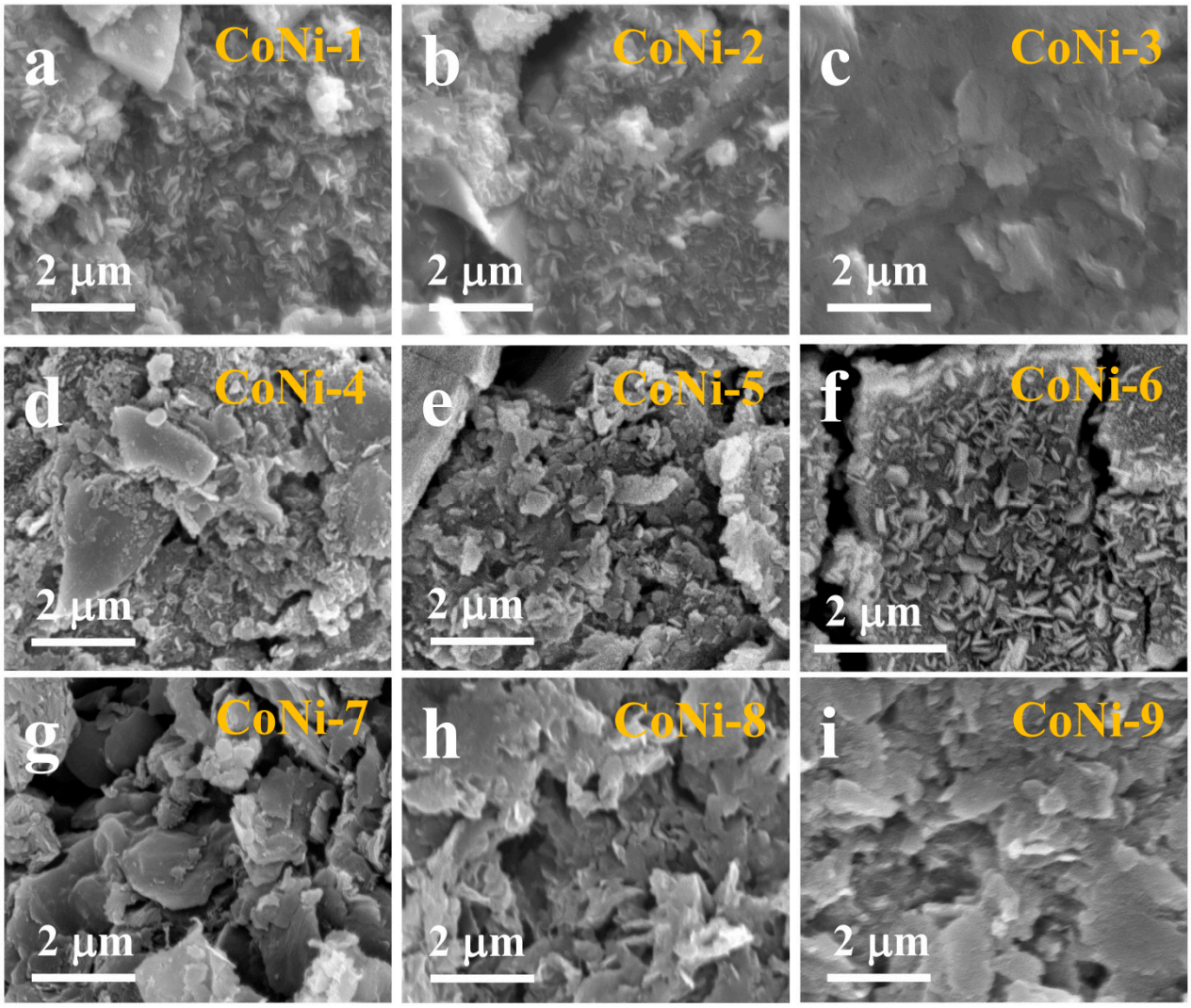


Figure S8 SEM images of CoNi-*n*, *n*=1~9.

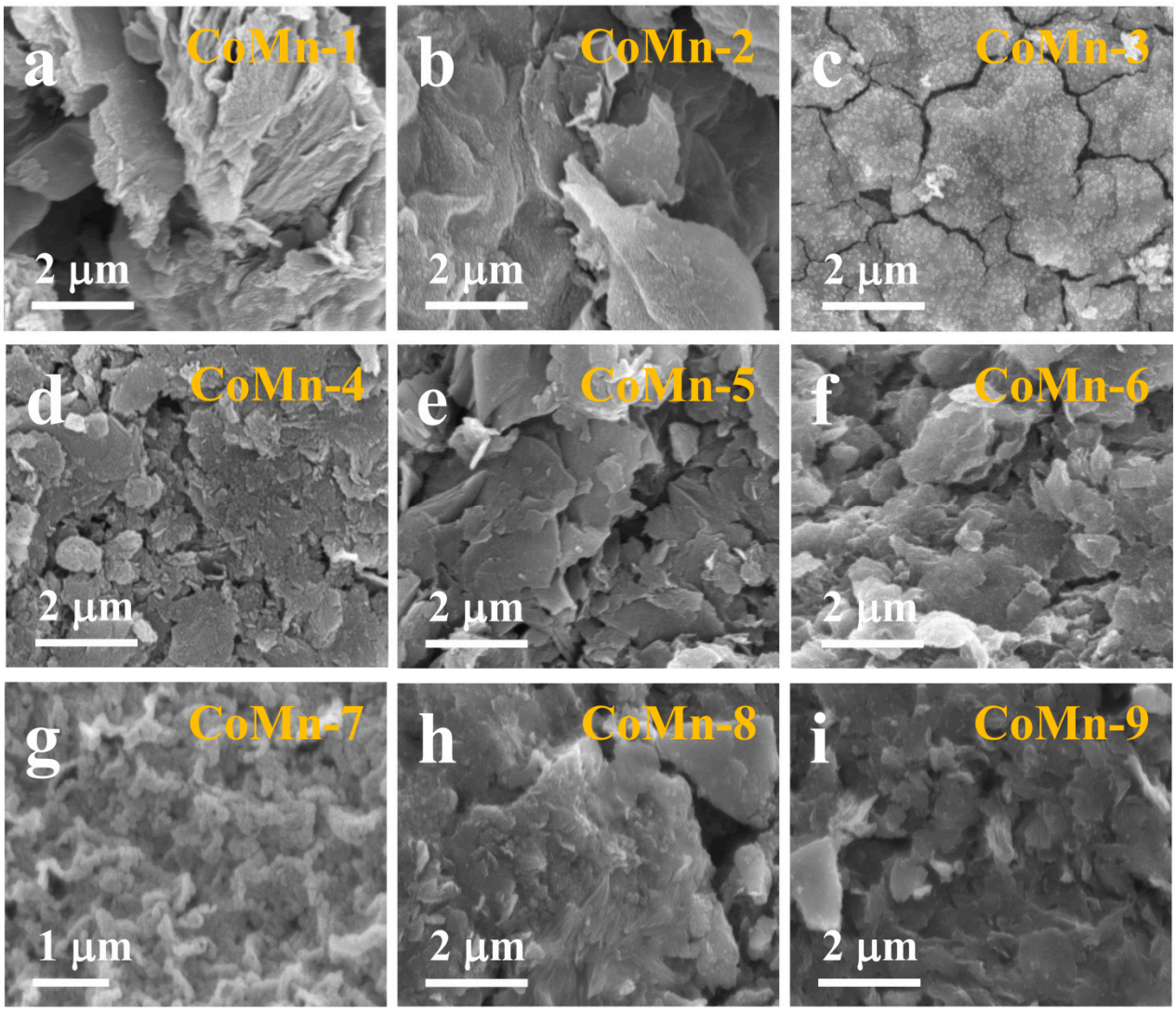


Figure S9 SEM images of CoMn-*n*, *n*=1~9.

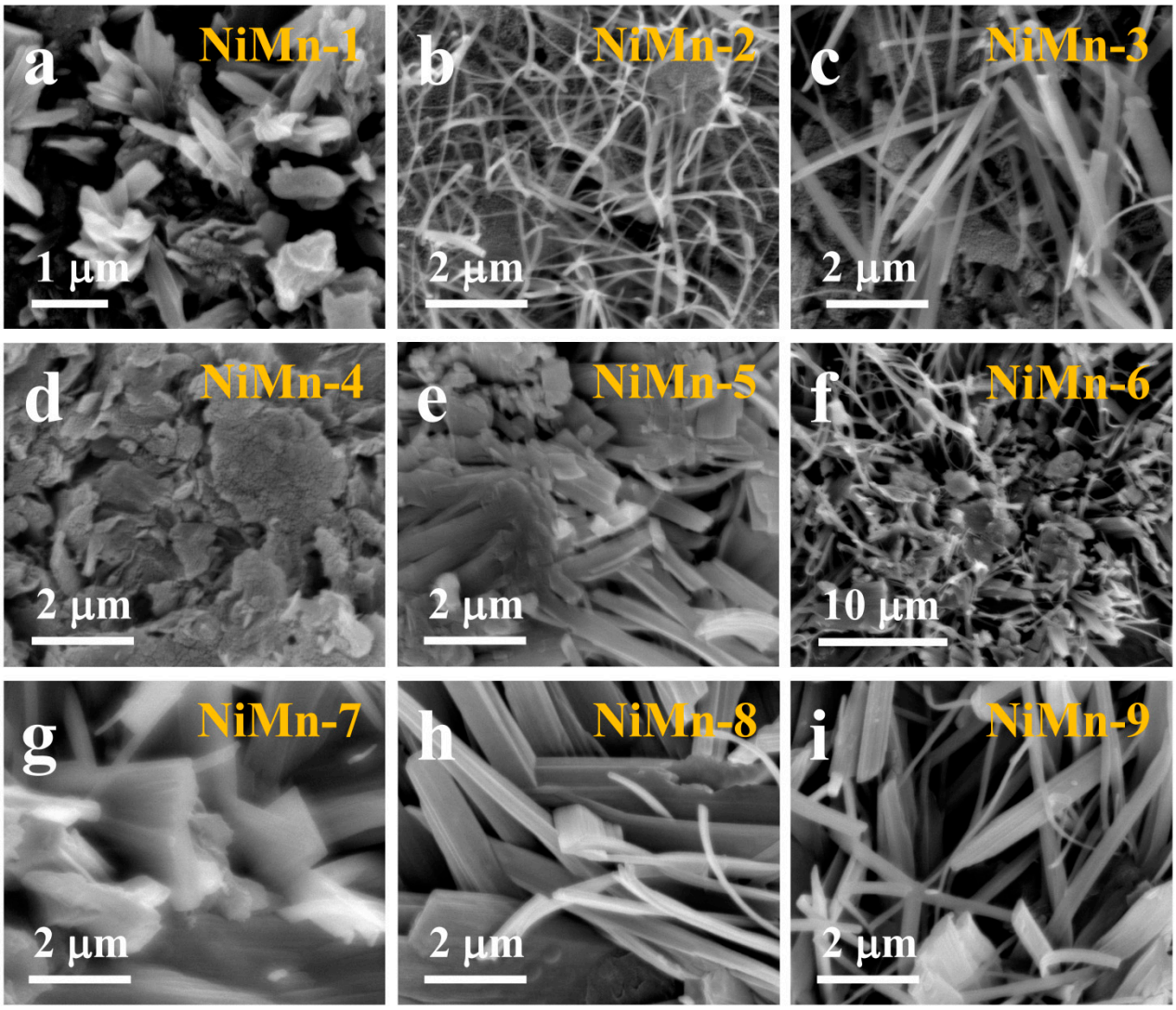


Figure S10 SEM images of NiMn-*n*, *n*=1~9.

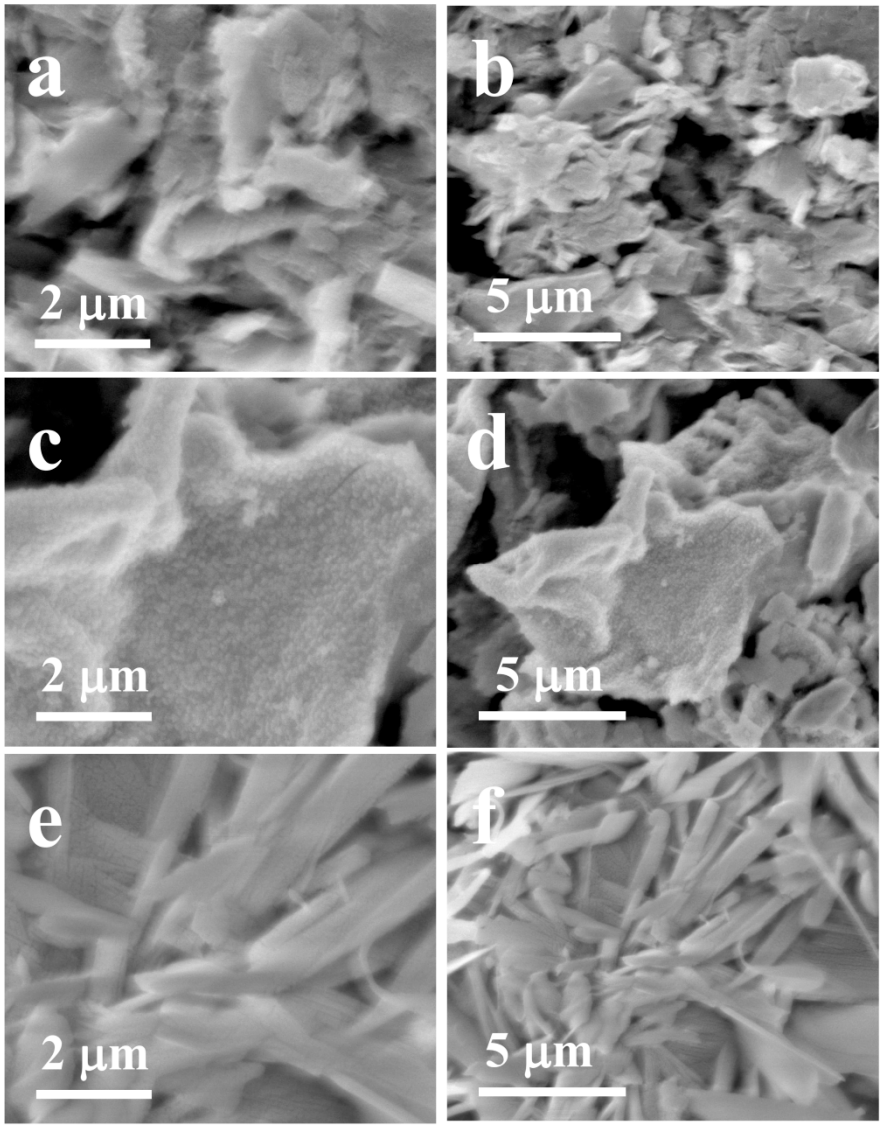


Figure S11 (a, b) SEM images Ni/NiO, (c, d) SEM images Co_3O_4, (e, f) SEM images Mn_3O_4.

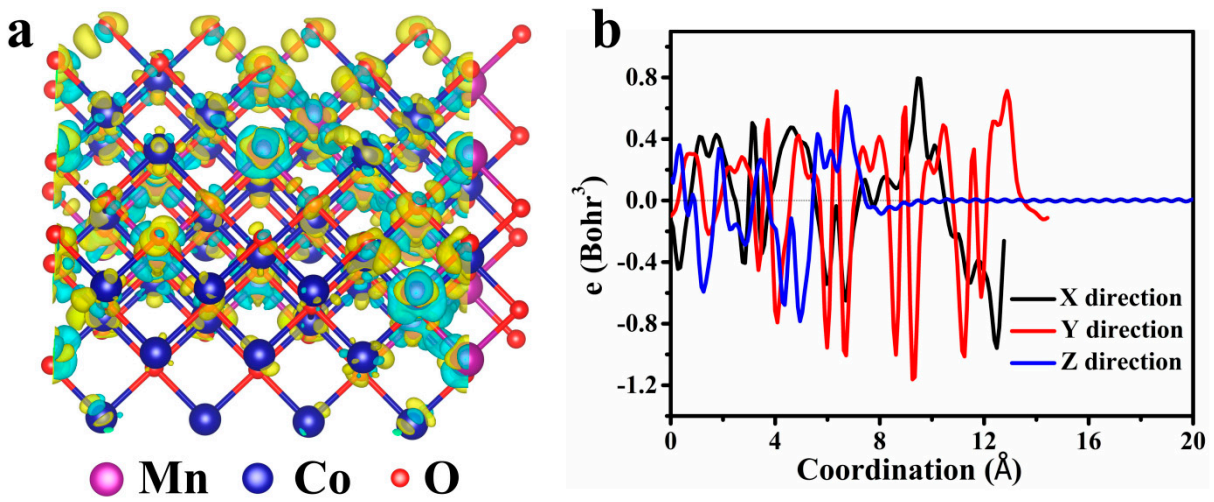


Figure S12 (a) Charge density difference of CoMn-3, (b) electron density fluctuation in X, Y, Z directions.

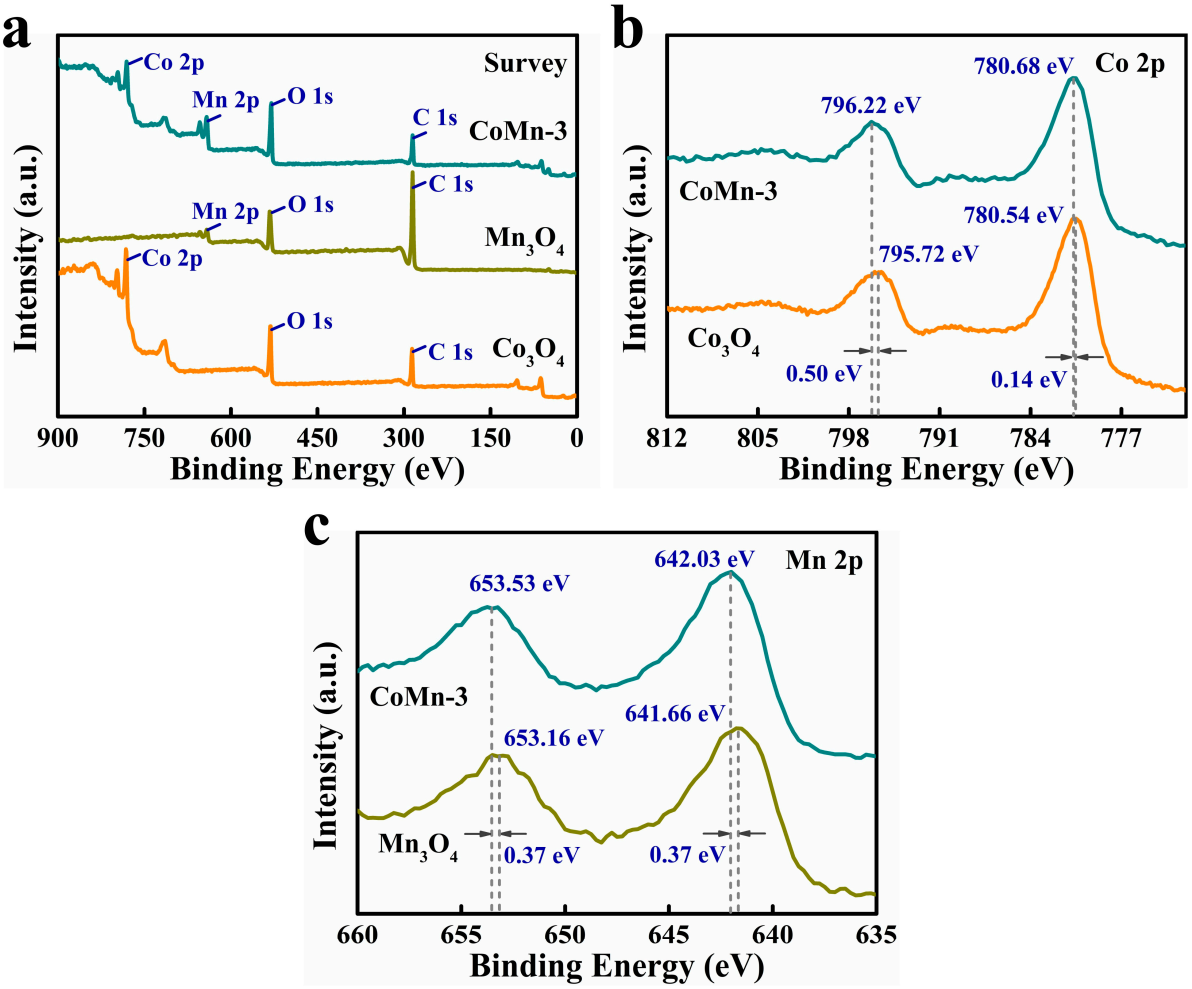


Figure S13 (a) XPS survey of Co_3O_4, Mn_3O_4 and CoMn-3. (b) Co 2p spectra of Co_3O_4 and CoMn-3, (c) Mn 2p spectra of Mn_3O_4 and CoMn-3.

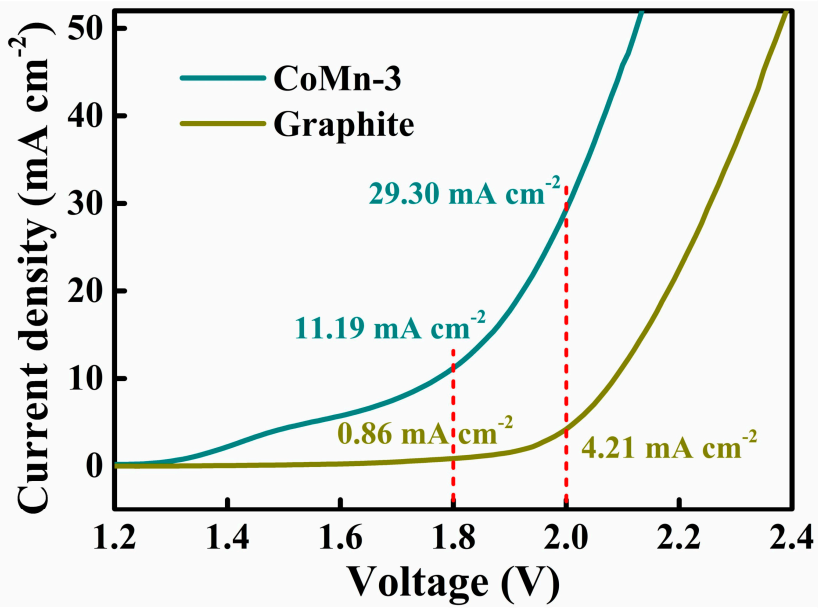


Figure S14 Compare the LSV curve of CoMn-3 with graphite for the overall water splitting

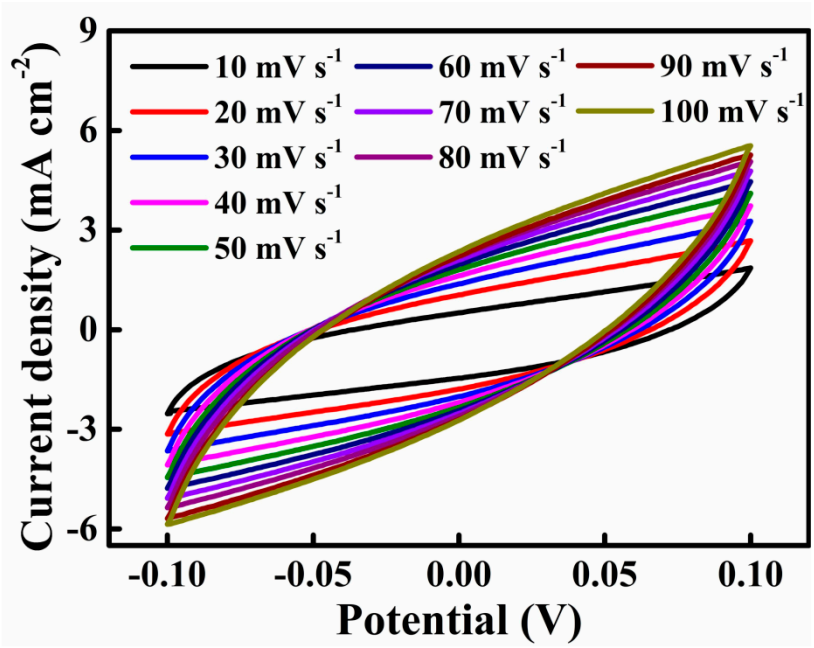


Figure S15 Rate CV curves of CoMn-3||CoMn-3 symmetric cell.