

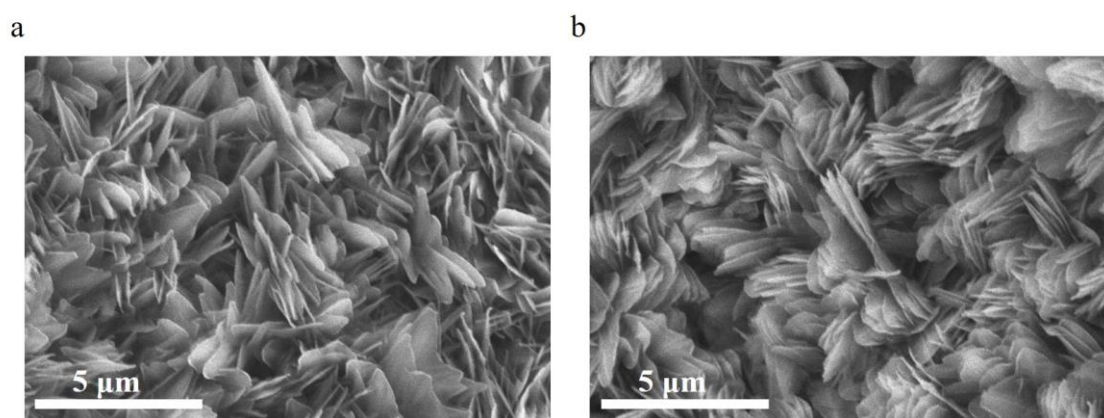
Supplementary materials

# Shape-Preserved CoFeNi–MOF/NF Exhibiting Superior Performance for Overall Water Splitting Across Alkaline and Neutral Conditions

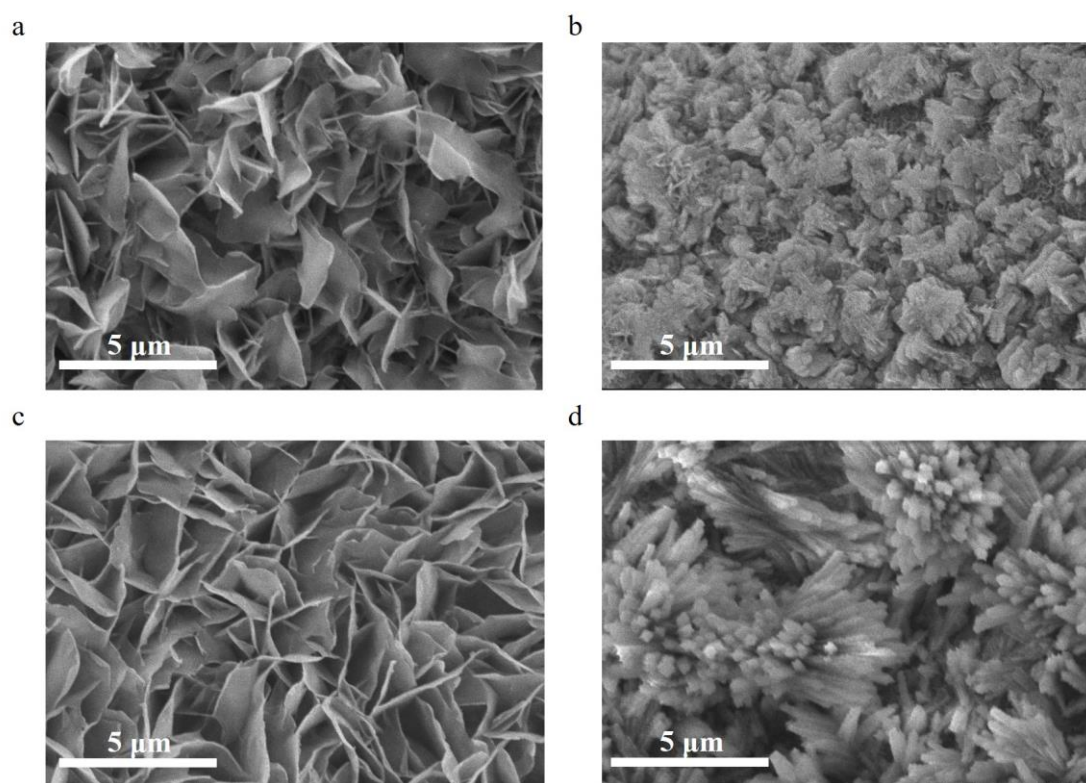
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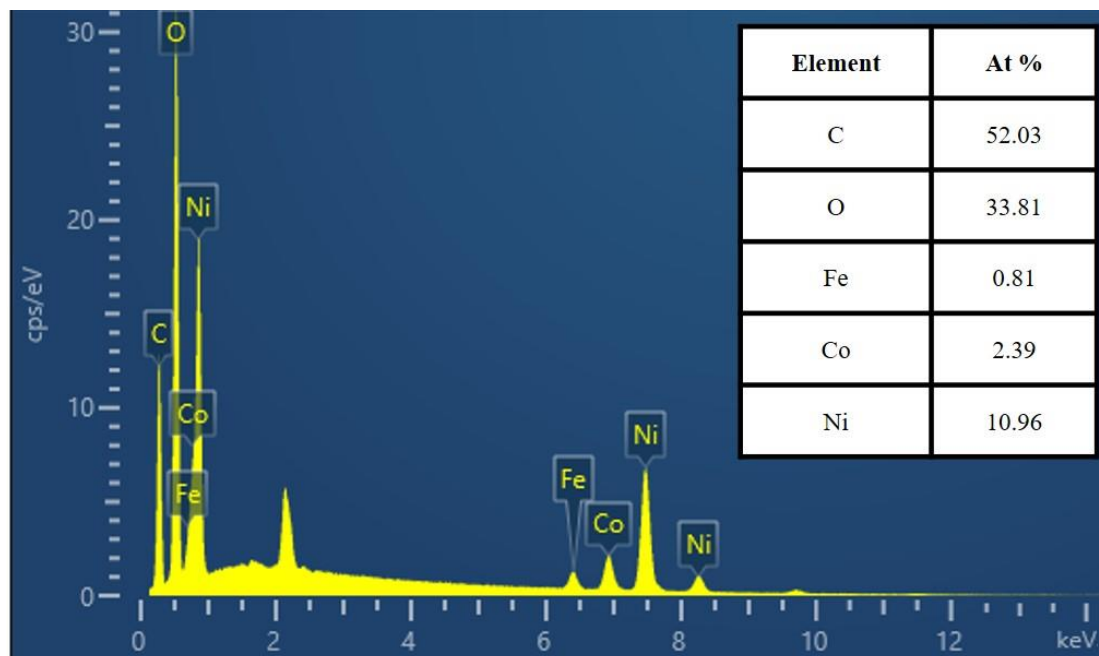
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**Figure S1.** SEM images of (a) Co<sub>0.45</sub>Fe<sub>0.45</sub>Ni<sub>0.9</sub>–LDH/NF and (b) Co<sub>0.45</sub>Fe<sub>0.45</sub>Ni<sub>0.9</sub>–MOF/NF.



**Figure S2.** SEM images of (a)  $\text{Fe}_{0.45}\text{Ni}_{0.9}\text{-MOF/NF}$ ; (b)  $\text{Co}_{0.9}\text{Fe}_{0.45}\text{Ni}_{0.9}\text{-MOF/NF}$ ; (c)  $\text{Co}_{0.45}\text{Ni}_{0.9}\text{-MOF/NF}$ ; and (d)  $\text{Co}_{0.45}\text{Fe}_{0.9}\text{Ni}_{0.9}\text{-MOF/NF}$ .



**Figure S3.** EDS spectrum of  $\text{Co}_{0.45}\text{Fe}_{0.45}\text{Ni}_{0.9}\text{-MOF/NF}$ .

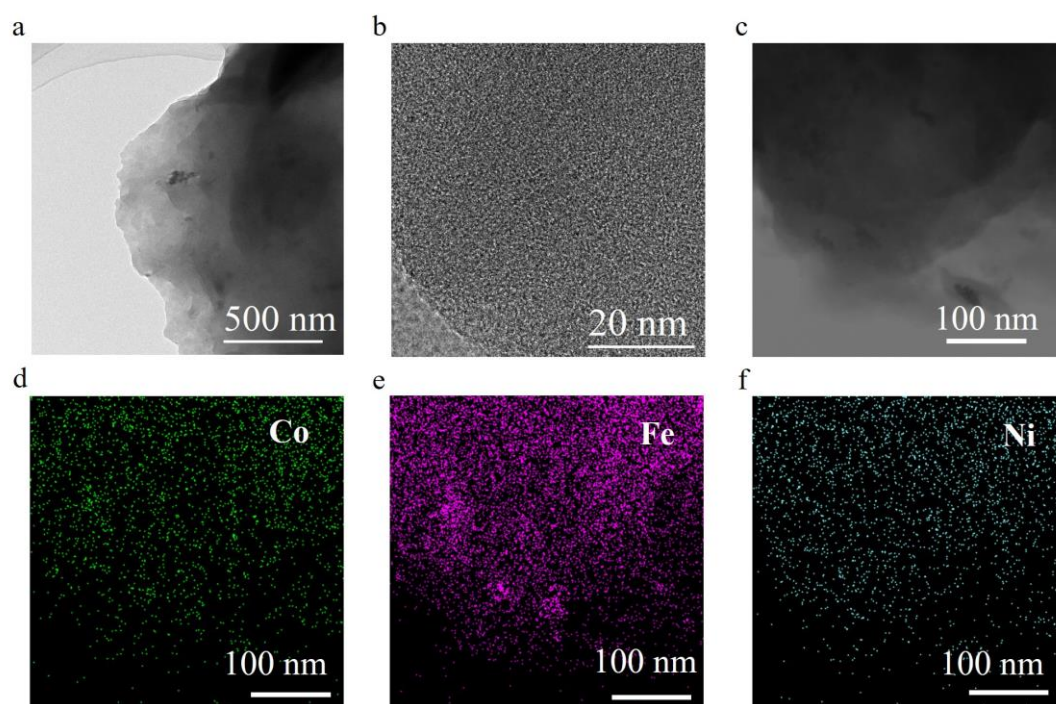


Figure S4. (a,b) HRTEM images and (c–f) EDS elemental mapping of  $\text{Co}_{0.45}\text{Fe}_{0.45}\text{Ni}_{0.9}\text{-MOF/NF}$ .

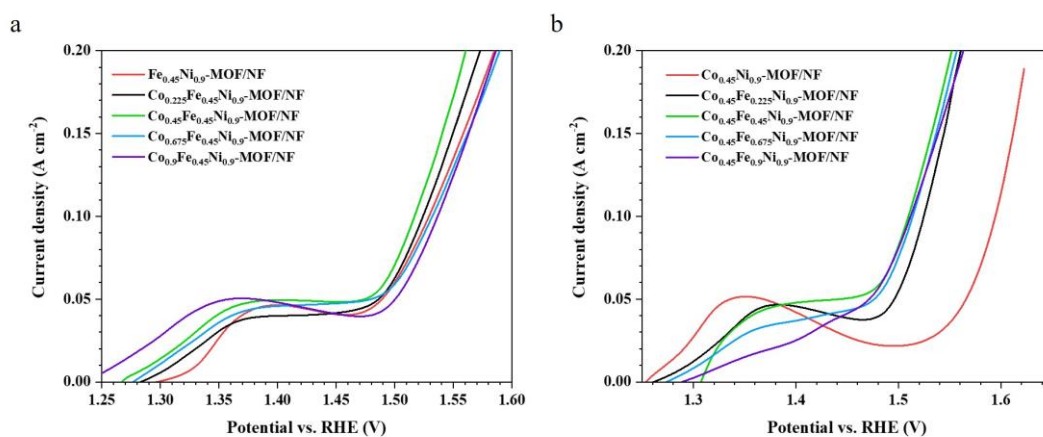
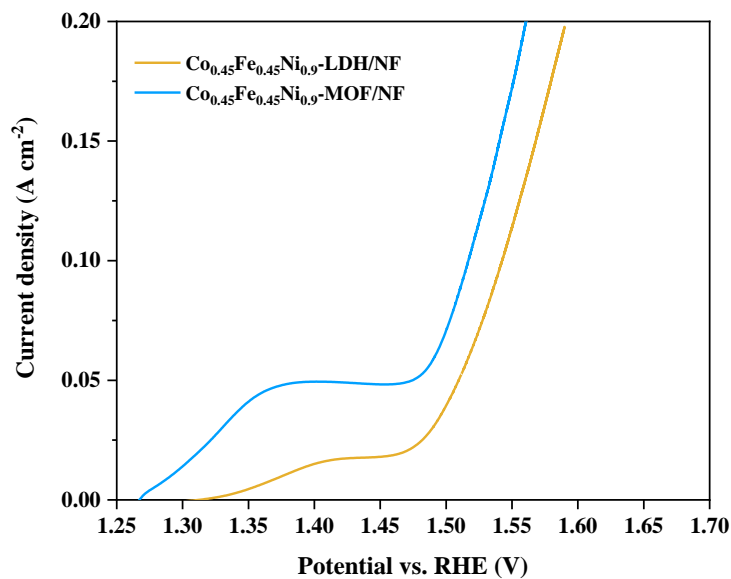
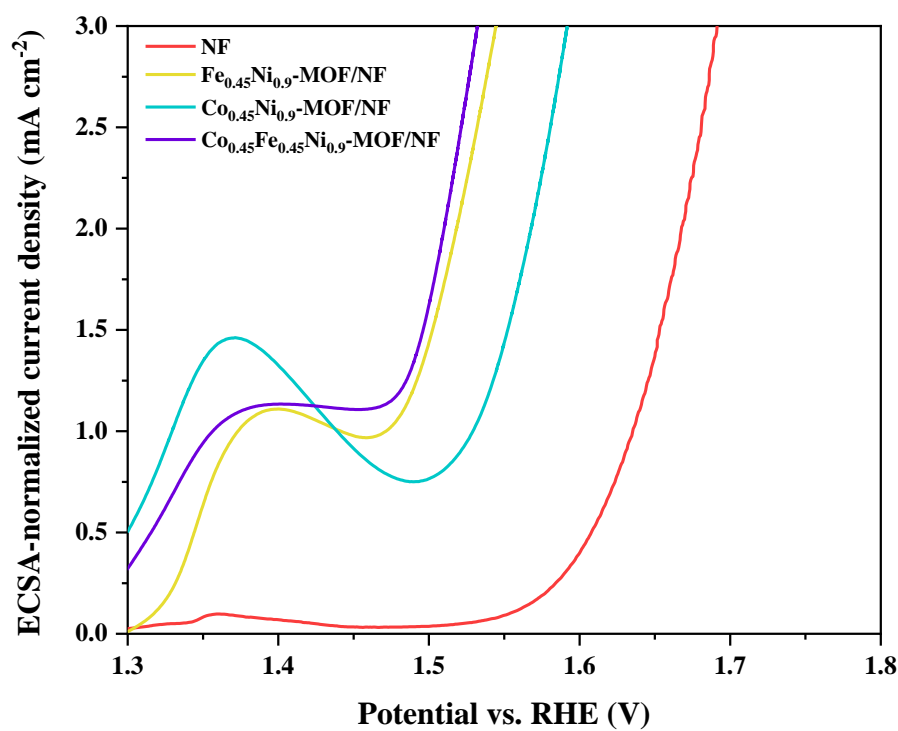


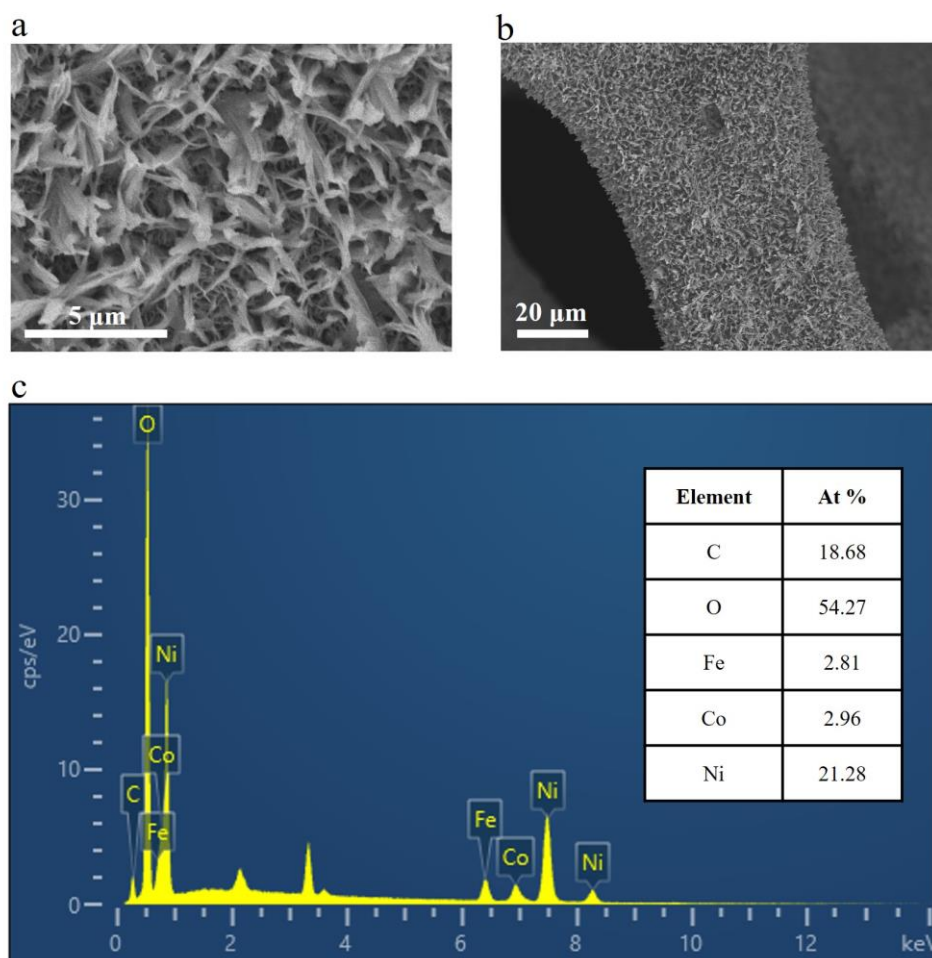
Figure S5. LSV curves of OER under 1 M KOH for the catalysts with different dosages of (a) Co and (b) Fe.



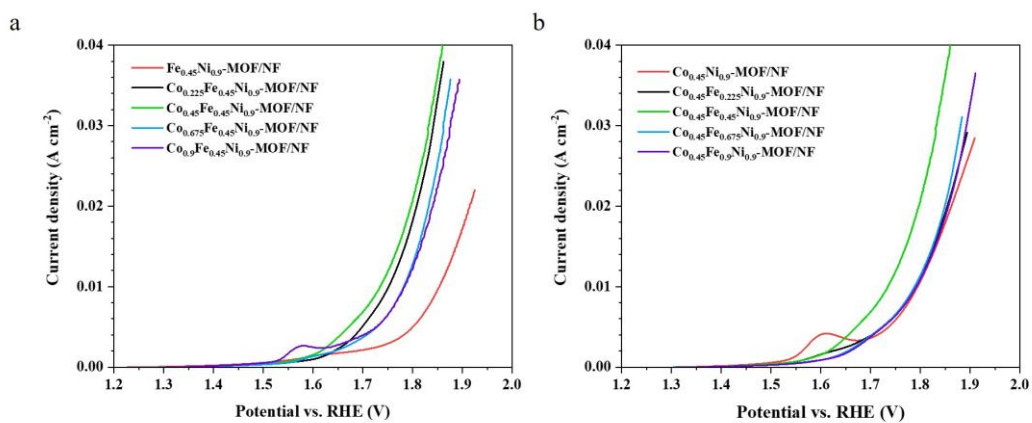
**Figure S6.** LSV curves of OER under 1 M KOH for Co<sub>0.45</sub>Fe<sub>0.45</sub>Ni<sub>0.9</sub>-LDH/NF and Co<sub>0.45</sub>Fe<sub>0.45</sub>Ni<sub>0.9</sub>-MOF/NF.



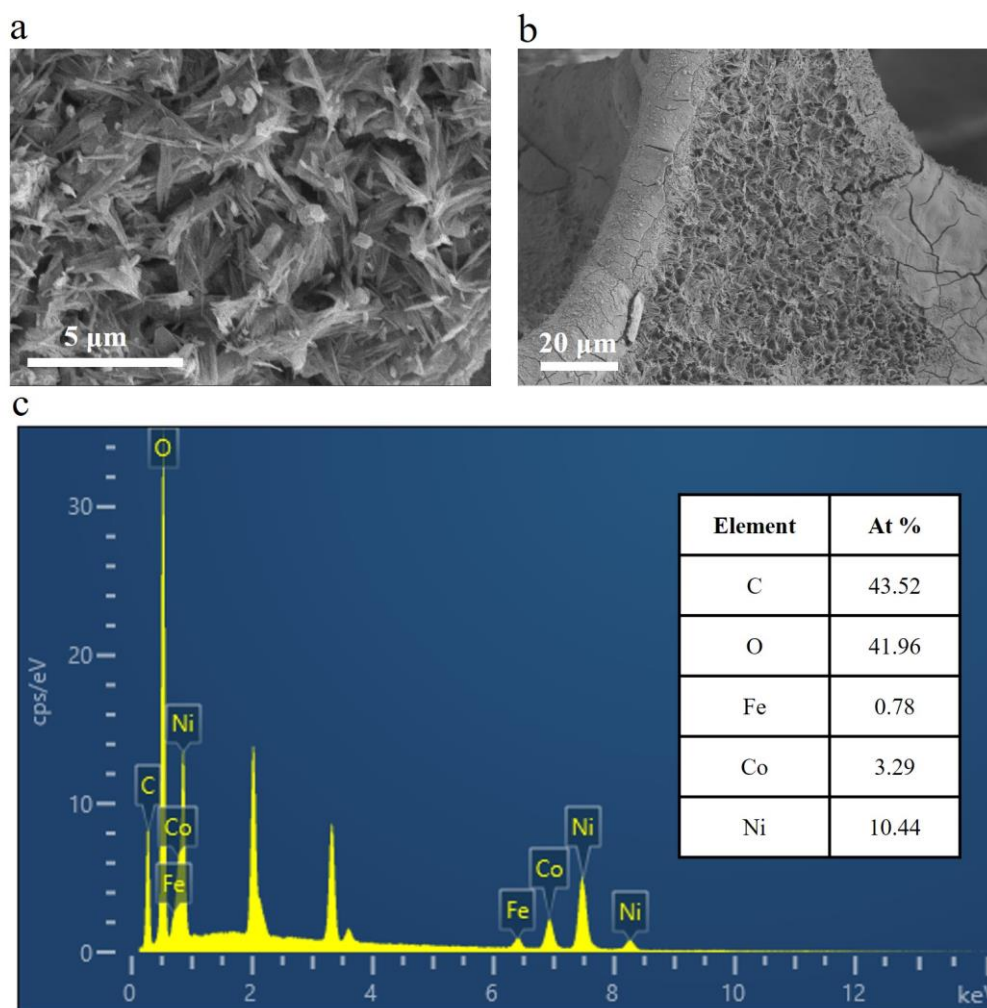
**Figure S7.** ECSA-normalized LSV curves of NF, Fe<sub>0.45</sub>Ni<sub>0.9</sub>-MOF/NF, Co<sub>0.45</sub>Ni<sub>0.9</sub>-MOF/NF, and Co<sub>0.45</sub>Fe<sub>0.45</sub>Ni<sub>0.9</sub>-MOF/NF.



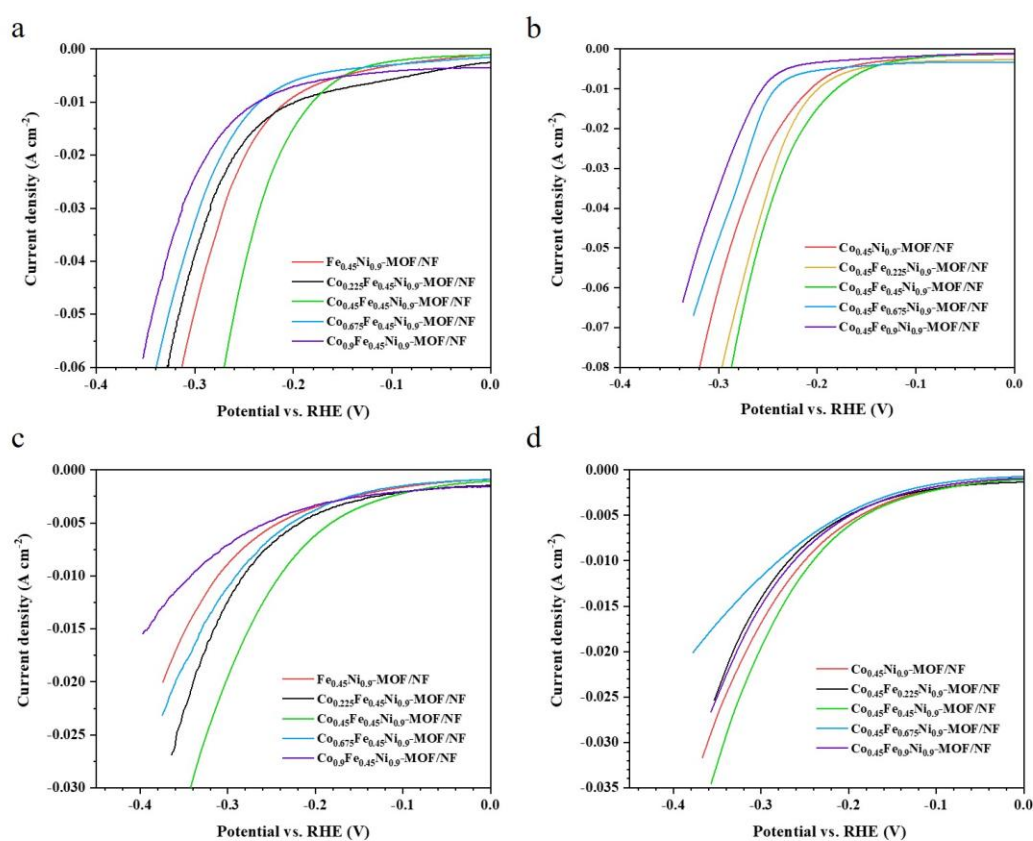
**Figure S8.** (a,b) SEM images of Co<sub>0.45</sub>Fe<sub>0.45</sub>Ni<sub>0.9</sub>-MOF/NF after OER i-t test under 1 M KOH with different magnifications and (c) EDS spectrum of (b).



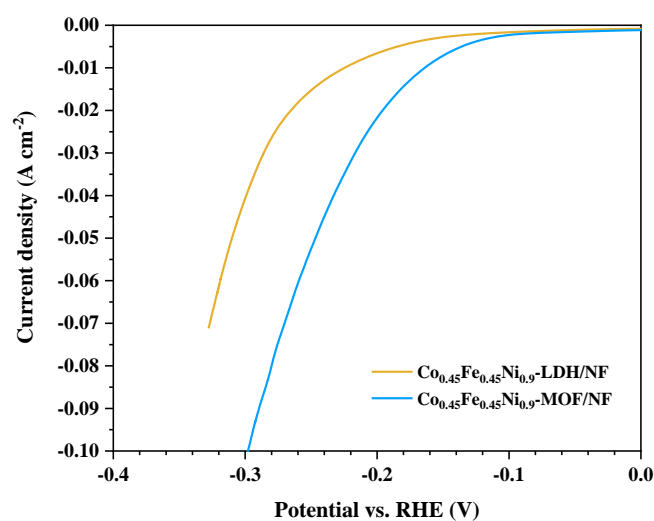
**Figure S9.** OER LSV curves under 1 M PBS for different dosages of (a) Co and (b) Fe.



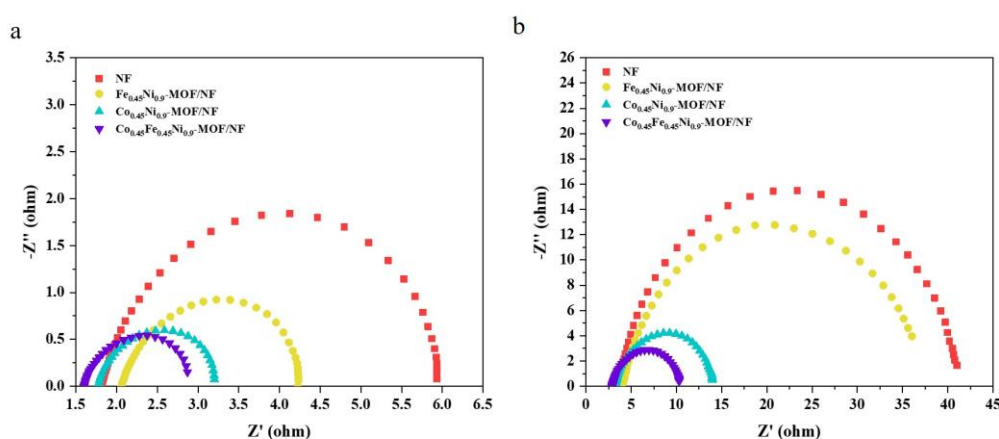
**Figure S10.** (a,b) SEM images of  $\text{Co}_{0.45}\text{Fe}_{0.45}\text{Ni}_{0.9}\text{-MOF/NF}$  after OER i-t test under 1 M PBS with different magnifications and (c) EDS spectrum of (b).



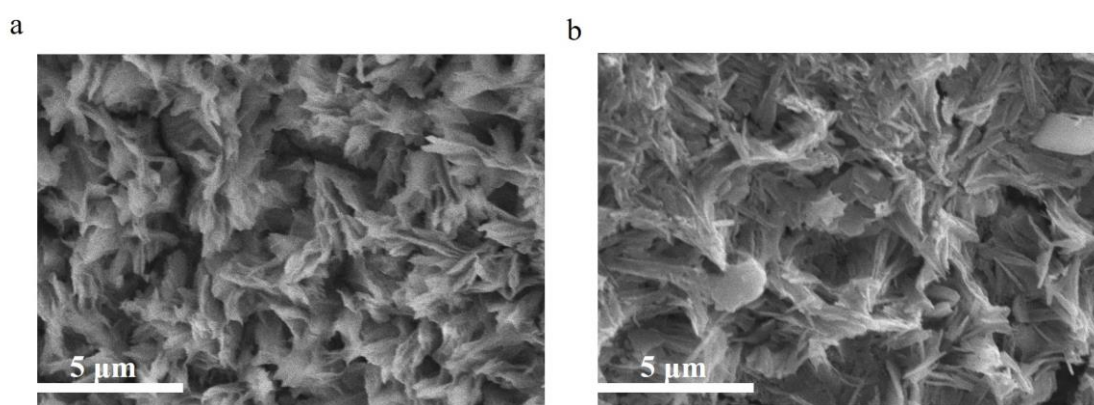
**Figure S11.** HER LSV curves under 1 M KOH with different dosages of (a) Co and (b) Fe. HER LSV curves at 1 M PBS with different dosages of (c) Co and (d) Fe.



**Figure S12.** LSV curves of HER under 1 M KOH for  $\text{Co}_{0.45}\text{Fe}_{0.45}\text{Ni}_{0.9}\text{-LDH/NF}$  and  $\text{Co}_{0.45}\text{Fe}_{0.45}\text{Ni}_{0.9}\text{-MOF/NF}$ .



**Figure S13.** Nyquist plots measured at  $-0.3$  V vs. RHE of NF,  $\text{Fe}_{0.45}\text{Ni}_{0.9}\text{-MOF/NF}$ ,  $\text{Co}_{0.45}\text{Ni}_{0.9}\text{-MOF/NF}$ , and  $\text{Co}_{0.45}\text{Fe}_{0.45}\text{Ni}_{0.9}\text{-MOF/NF}$  under (a) 1 M KOH and (b) 1 M PBS, respectively.



**Figure S14.** SEM images of  $\text{Co}_{0.45}\text{Fe}_{0.45}\text{Ni}_{0.9}\text{-MOF/NF}$  after the  $i$ - $t$  test of HER under (a) 1 M KOH and (b) 1 M PBS, respectively.

**Table S1.** ICP-OES test of metallic elements in  $\text{Co}_{0.45}\text{Fe}_{0.45}\text{Ni}_{0.9}\text{-MOF/NF}$ .

Element	Mass concentration (mg/L)	Average mass concentration (mg/L)	Average atomic percentage (mol%)
Co	5.4	5.45	14.26
	5.5		
Fe	3.9	3.9	10.76
	3.9		
Ni	28.2	28.55	74.98
	28.9		

**Table S2.** Comparison of OER performance for  $\text{Co}_{0.45}\text{Fe}_{0.45}\text{Ni}_{0.9}\text{-MOF/NF}$  with other non-noble-metal-based catalysts under alkaline conditions.

Catalyst	$j$ ( $\text{mA cm}^{-2}$ )	$\eta$ (mV)	Electrolyte	Ref.
$\text{Co}_{0.45}\text{Fe}_{0.45}\text{Ni}_{0.9}\text{-MOF/NF}$	50	244	1 M KOH	This work
	100	287		
$\text{Ni}_{10}\text{Co-BTC/KB}$	10	347	1 M KOH	[62]
Co-MOFs	50	348	1 M KOH	[57]

FeNi <sub>2</sub> S <sub>4</sub>	10	316	1 M KOH	[63]
NiCo-MOF/NF	50	270	1 M KOH	[64]
CVN/CC	10	263	1 M KOH	[65]
Ni(Fe)MOF/mKB14	50	291	1 M KOH	[66]
CoNi-CNT/NC-800	10	300	1 M KOH	[67]
Te-Ni(OH) <sub>2</sub> /NF	10	270	1 M KOH	[68]
CoCu-MOF NBs	10	271	1 M KOH	[28]
Ni <sub>2</sub> Mo <sub>3</sub> N/NF	50	336	1 M KOH	[69]

**Table S3.** Comparison of OER performance for Co<sub>0.45</sub>Fe<sub>0.45</sub>Ni<sub>0.9</sub>-MOF/NF with other non-noble-metal-based catalysts under near-neutral conditions.

Catalyst	<i>j</i> (mA cm <sup>-2</sup> )	<i>η</i> (mV)	Electrolyte	Ref.
Co <sub>0.45</sub> Fe <sub>0.45</sub> Ni <sub>0.9</sub> -MOF/NF	10	505	1 M PBS	This work
Co-Pi/ITO	1	410	0.1 M KP <sub>i</sub>	[70]
2D-Co-MOF	2	548	0.1 M SPB	[71]
Mn <sub>5</sub> O <sub>8</sub> NP/FTO	5	580	0.3 M NaP <sub>i</sub>	[72]
a-Co <sub>2</sub> P	10	592	0.1 M PBS	[73]
Sub-10 MnO NC/FTO	5	530	0.3 M NaP <sub>i</sub>	[74]
MnCat/ITO	1	590	0.1 M KP <sub>i</sub>	[75]
Co <sub>0.7</sub> Fe <sub>0.3</sub> P/CNT	10	500	1 M PBS	[76]
Co-Ci-2D	1	480	0.1 M PBS	[77]
Mn <sub>3</sub> (PO <sub>4</sub> ) <sub>2</sub> ·3H <sub>2</sub> O/FTO	0.316	680	0.5 M NaP <sub>i</sub>	[78]
Ni-Fe-Mg	10	514	0.5 M KHCO <sub>3</sub>	[79]

**Table S4.** Comparison of HER performance for Co<sub>0.45</sub>Fe<sub>0.45</sub>Ni<sub>0.9</sub>-MOF/NF with other non-noble-metal-based catalysts under alkaline conditions.

Catalyst	<i>j</i> (mA cm <sup>-2</sup> )	<i>η</i> (mV)	Electrolyte	Ref.
Co <sub>0.45</sub> Fe <sub>0.45</sub> Ni <sub>0.9</sub> -MOF/NF	10	164	1 M KOH	This work
FeNi <sub>2</sub> S <sub>4</sub>	10	299	1 M KOH	[63]
CoNi-MOF-PCG	10	265	1 M KOH	[80]
MoS <sub>2</sub> /MoO <sub>3</sub>	10	210	1 M KOH	[81]
S-C-MoO <sub>2</sub> /MoO <sub>3-x</sub>	10	160	1 M KOH	[82]
N, S co-doped graphitic	10	230	0.1 M KOH	[83]
CuO@CQDs@CHNS	10	159	1 M KOH	[84]
CoS <sub>2</sub> -600	10	270	1 M KOH	[85]
2D CoNi(1:1)-MOF	10	376	1 M KOH	[86]
Ni-doped CoP/Co <sub>2</sub> P	10	161	1 M KOH	[87]
Co-BDC/MoS <sub>2</sub>	10	248	1 M KOH	[88]

**Table S5.** Comparison of HER performance for Co<sub>0.45</sub>Fe<sub>0.45</sub>Ni<sub>0.9</sub>-MOF/NF with other non-noble-metal-based catalysts under near-neutral conditions.

Catalyst	<i>j</i> (mA cm <sup>-2</sup> )	<i>η</i> (mV)	Electrolyte	Ref.
Co <sub>0.45</sub> Fe <sub>0.45</sub> Ni <sub>0.9</sub> -MOF/NF	10	241	1 M PBS	This work
Ni-Cr-Mo-Fe	10	297	1 M PBS	[89]
CoP/CC	2	65	1 M PBS	[90]
NiP <sub>x</sub> /CC	10	230	0.1 M ABS	[91]
CoO-CoSe <sub>2</sub>	10	337	0.5 M PBS	[92]
Ni <sub>0.1</sub> Co <sub>0.9</sub> P	10	125	1 M PBS	[93]
SiO <sub>2</sub> /PPy NTs-CFs	10	183	1 M PBS	[94]

FePN/CNT-200	10	158	1 M PBS	[95]
Ni-Co-Fe-P/NF	10	201	1 M PBS	[96]
CoFe@NC	10	340	1 M PBS	[97]
NiCo-MOF/NF	10	252	0.5 M Na <sub>2</sub> SO <sub>4</sub>	[18]

**Table S6.** Comparison of overall water splitting performance for Co<sub>0.45</sub>Fe<sub>0.45</sub>Ni<sub>0.9</sub>-MOF/NF with other non-noble-metal-based catalysts under alkaline conditions.

Catalyst	<i>j</i> (mA cm <sup>-2</sup> )	Cell voltage (V)	Electrolyte	Ref.
Co <sub>0.45</sub> Fe <sub>0.45</sub> Ni <sub>0.9</sub> -MOF/NF	10	1.59	1 M KOH	This work
FeNi <sub>2</sub> S <sub>4</sub>	10	1.63	1 M KOH	[63]
CVN/CC	10	1.64	1 M KOH	[65]
CuO@CQDs@CHNS	10	1.83	1 M KOH	[84]
Ni <sub>3</sub> Fe <sub>0.9</sub> Cr <sub>0.1</sub> /CACC	10	1.59	1 M KOH	[98]
Co-CeO <sub>2</sub> @CNF/ Ni <sub>2</sub> Fe@CNF	10	1.59	1 M KOH	[99]
NiCo-LDH-OH	10	1.89	1 M KOH	[100]
FeVO(OH)/Ni(OH) <sub>2</sub>	20	1.65	1 M KOH	[101]
BP@MOF	10	1.63	1 M KOH	[102]
CoFe10%-P/NF	10	1.61	1 M KOH	[103]
FeMn <sub>6</sub> Ce <sub>0.5</sub> -MOF-74/NF	10	1.65	1 M KOH	[104]