

### Supporting Information

Electronic coupling regulation in MOF derivatives Mn/Fe co-doped Ni<sub>2</sub>P through Mn/Fe integration  
for enhanced overall water splitting

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**Table S1.** Results of ICP-OES test of Mn-Fe-Ni<sub>2</sub>P/NF-X (X=0.09, 0.18, 0.27, 0.33, 0.43, 0.57, 0.72).

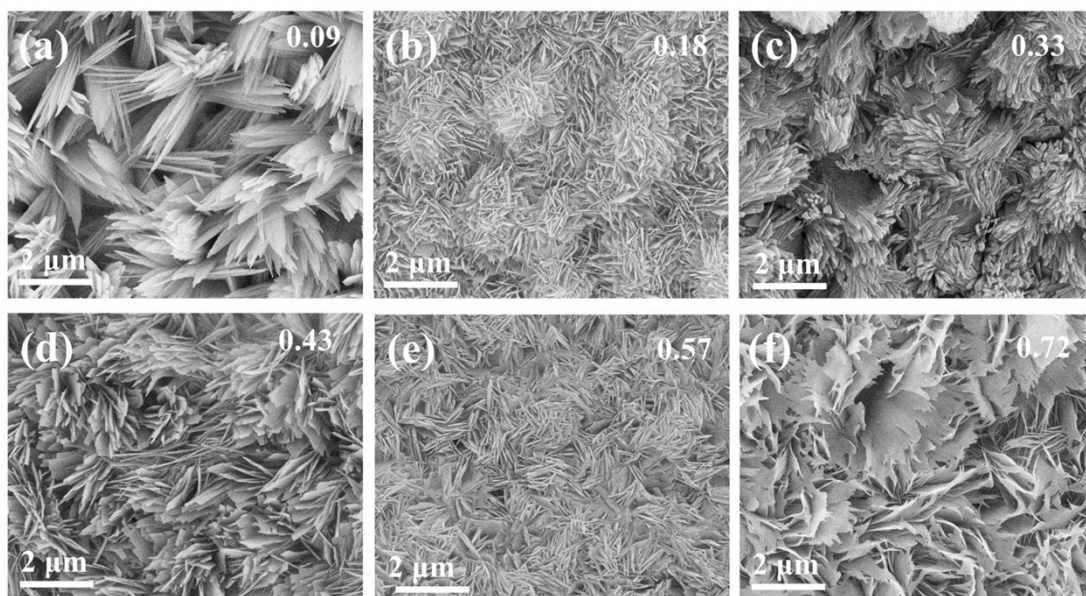
**Table S2.** C<sub>d1</sub> and ECSA of Mn-Fe-Ni<sub>2</sub>P/NF and Fe-Ni<sub>2</sub>P/NF nanosheets.

**Table S3.** EIS fitting results of Mn-Fe-Ni<sub>2</sub>P/NF-X (X=0.09, 0.18, 0.27, 0.33, 0.43, 0.57, 0.72) and Fe-Ni<sub>2</sub>P/NF in HER and OER.

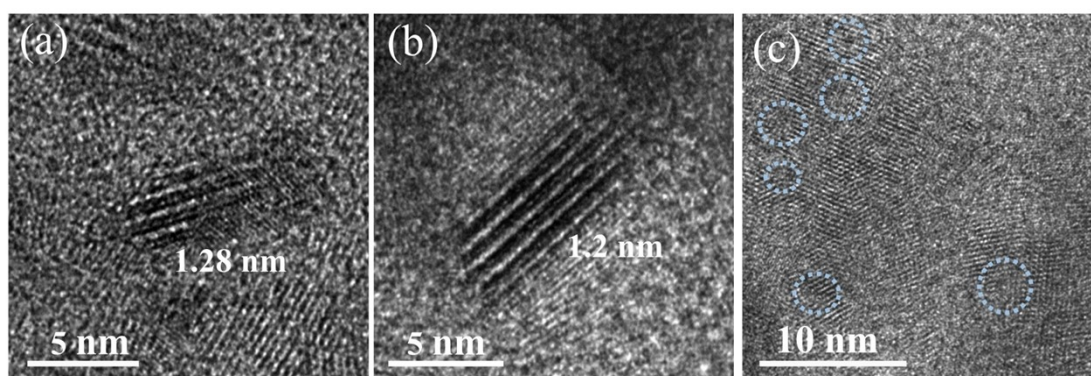
**Table S4.** Comparison of HER performances for Mn-Fe-Ni<sub>2</sub>P nanosheets with similar materials in the alkaline media.

**Table S5.** Comparison of OER performances for Mn-Fe-Ni<sub>2</sub>P nanosheets with similar materials in the alkaline media.

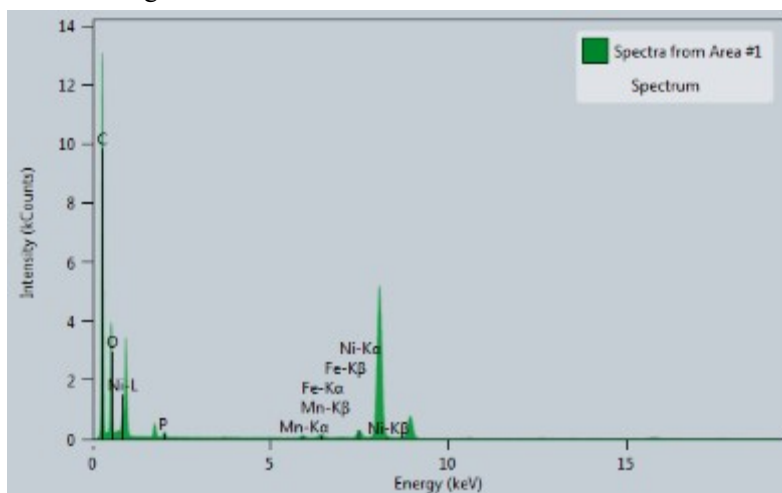
**Table S6.** Comparison of overall water splitting performances for Mn-Fe-Ni<sub>2</sub>P nanosheets with similar materials in the alkaline media.



**Fig. S1** SEM images of (a) MnFeNi-MOF/NF-0.09; (b) MnFeNi-MOF/NF-0.18; (c) MnFeNi-MOF/NF-0.33; (d) MnFeNi-MOF/NF-0.43; (e) MnFeNi-MOF/NF-0.57; (f) MnFeNi-MOF/NF-0.72.



**Fig. S2** (a-c) HRTEM images of Mn-Fe-Ni<sub>2</sub>P/NF-0.27.



**Fig. S3** EDS of Mn-Fe-Ni<sub>2</sub>P/NF-0.27.

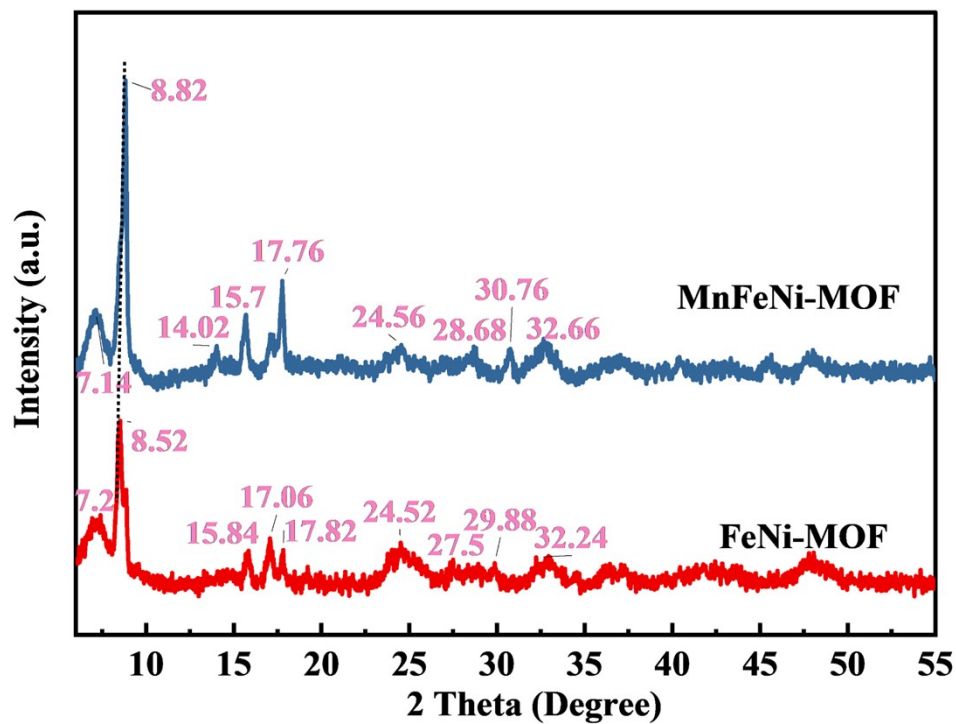


Fig. S4 XRD patterns of MnFeNi-MOF and FeNi-MOF.

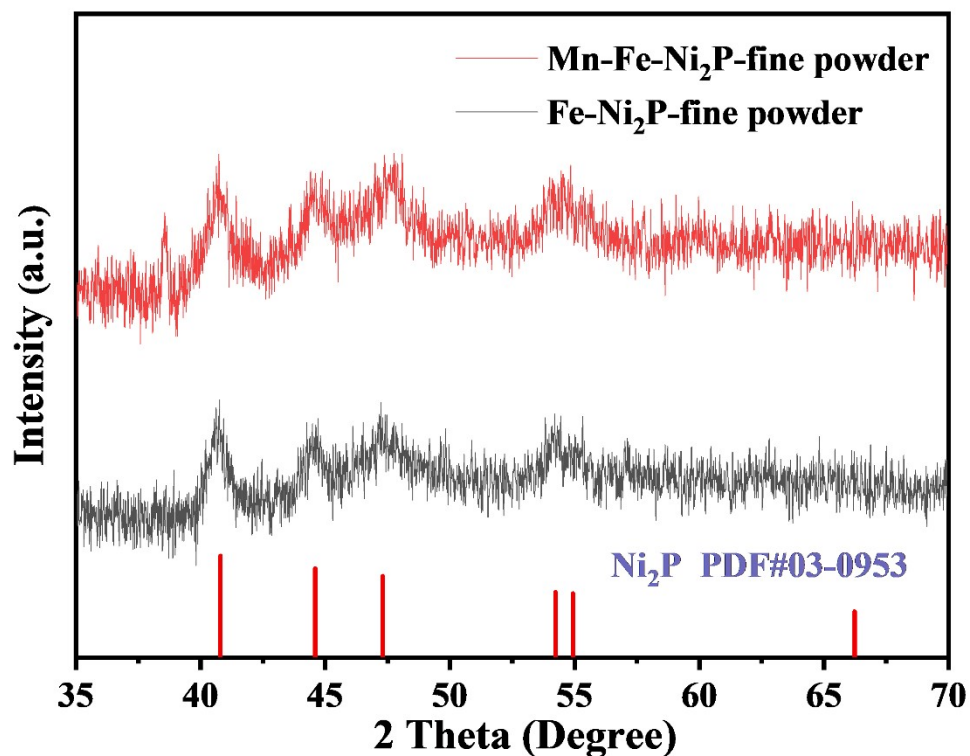


Fig. S5 XRD patterns of Mn-Fe-Ni<sub>2</sub>P-fine powder and Fe-Ni<sub>2</sub>P-fine powder.

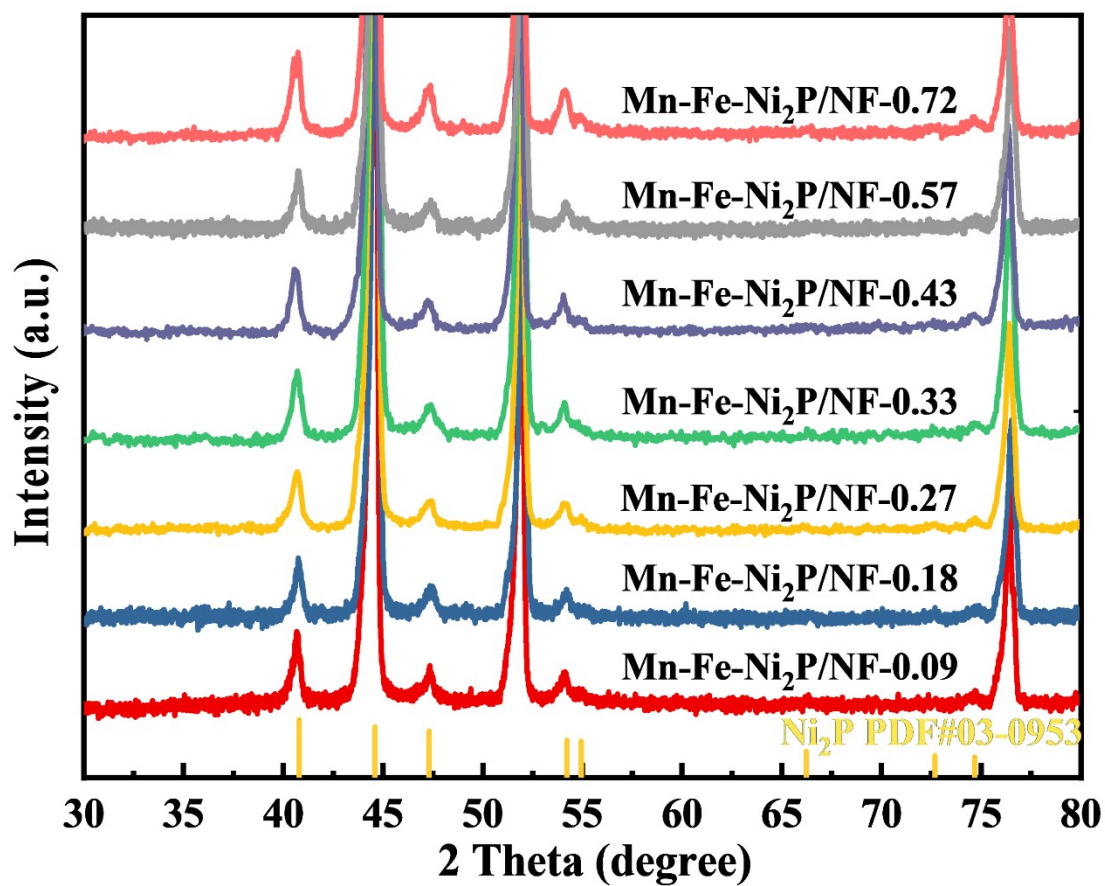
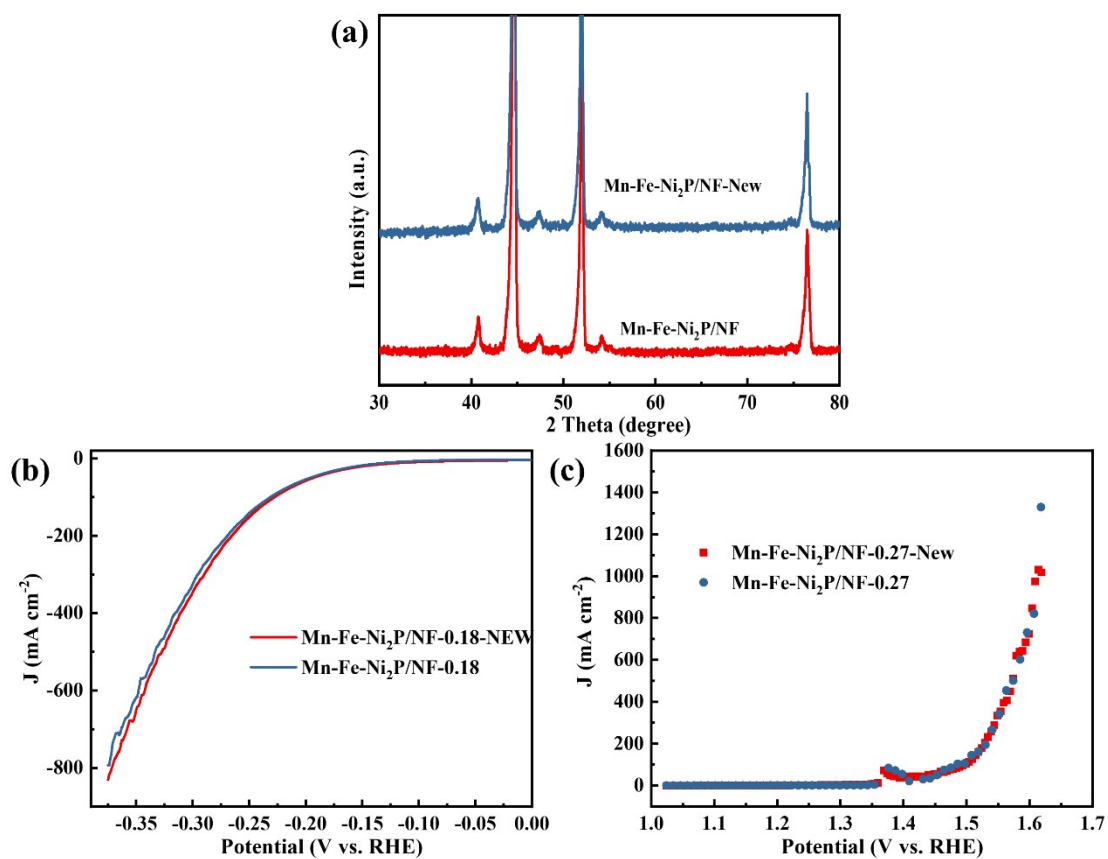
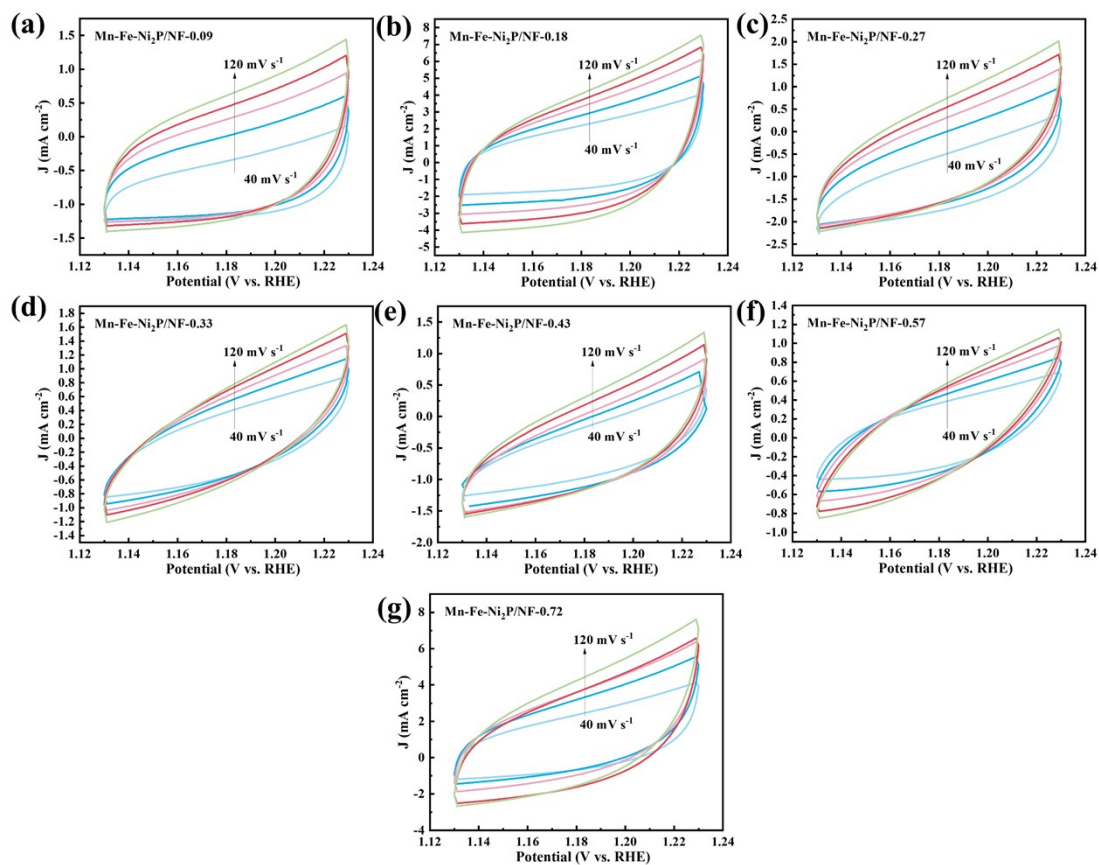


Fig. S6 XRD patterns of Mn-Fe-Ni<sub>2</sub>P/NF-X (X=0.09, 0.18, 0.27, 0.33, 0.43, 0.57, and 0.72).



**Fig. S7** Comparison of (a) XRD patterns, (b) LSV curve of HER, and (c) LSV curve of OER before and after repeated synthesis of Mn-Fe-Ni<sub>2</sub>P/NF-X.





**Fig. S8** (a-g) CV curves of Mn-Fe-Ni<sub>2</sub>P/NF-X (X=0.09, 0.18, 0.27, 0.33, 0.43, 0.57, 0.72) for estimating the  $C_{dl}$  in OER tests (1 M KOH).



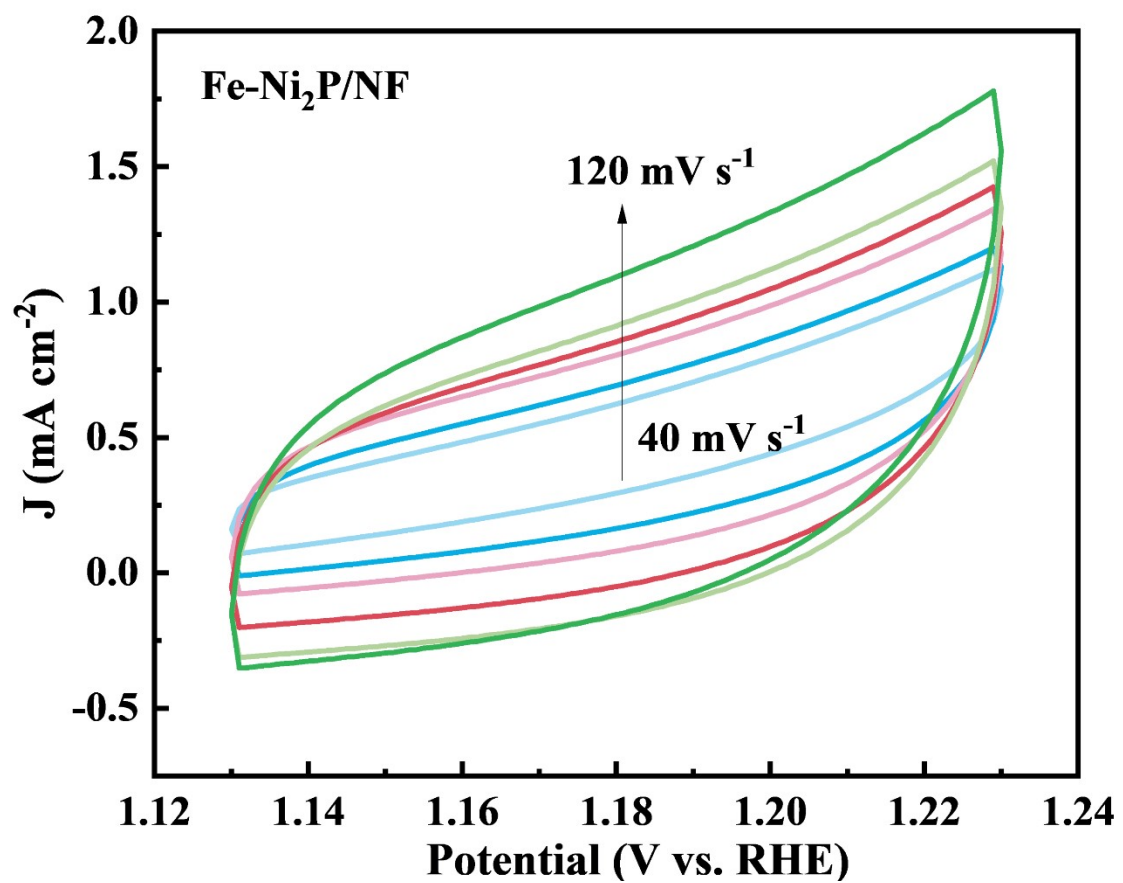
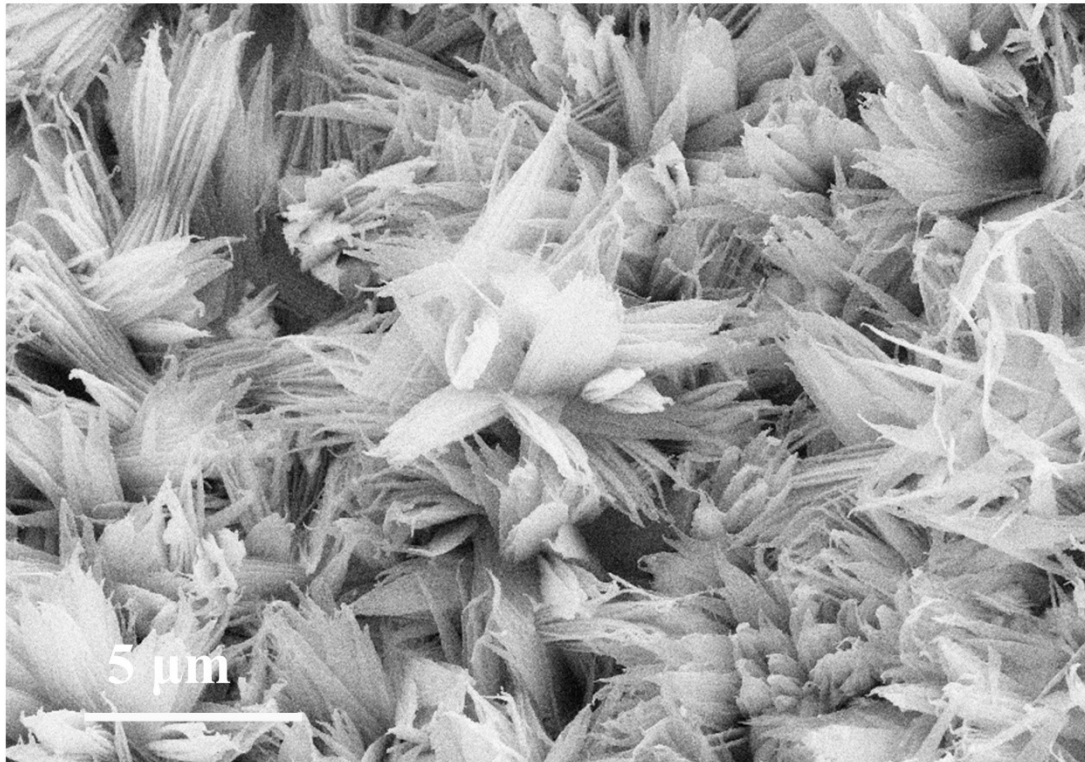
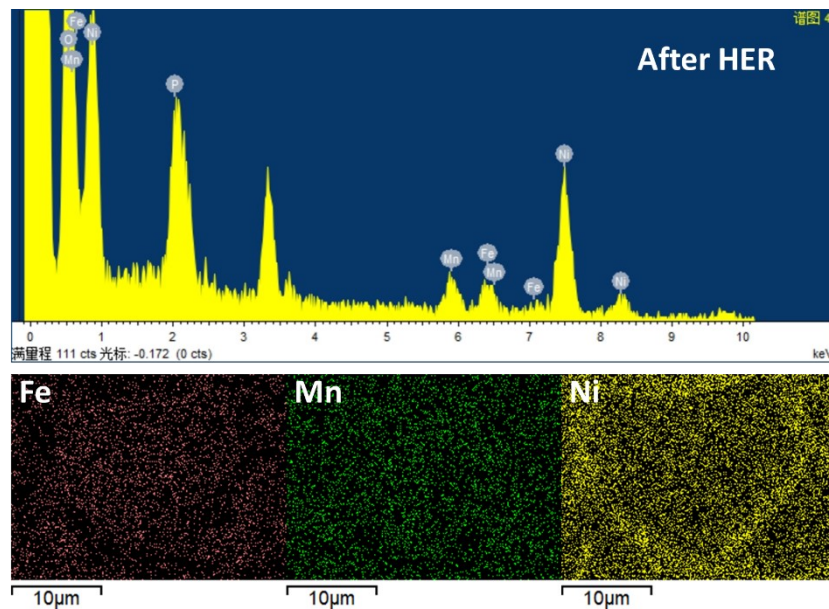


Fig. S9 CV curve of Fe-Ni<sub>2</sub>P/NF for estimating the  $C_{dl}$  in OER tests (1 M KOH).



**Fig. S10** SEM image of Mn-Fe-Ni<sub>2</sub>P/NF after HER durability testing.



**Fig. S11** EDS and Mapping of Mn-Fe-Ni<sub>2</sub>P/NF after HER durability testing.

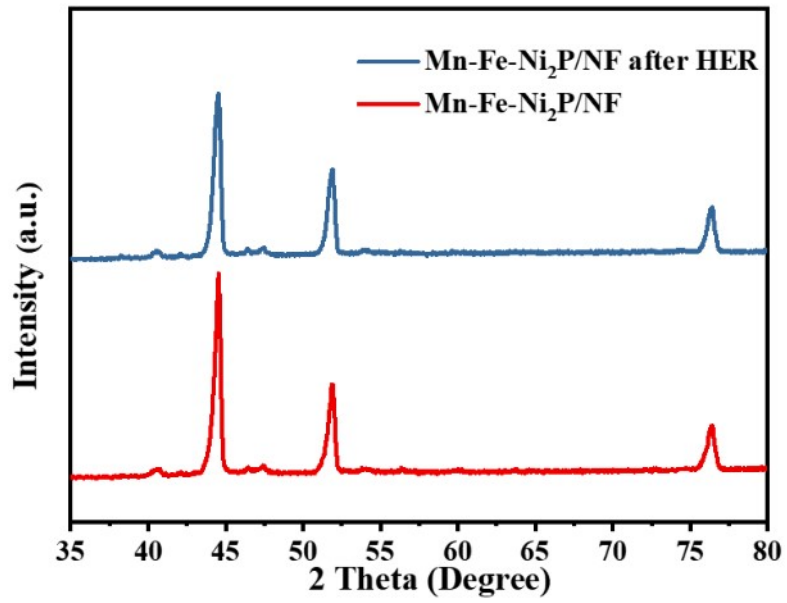


Fig. S12 XRD patterns of Mn-Fe-Ni<sub>2</sub>P/NF before and after HER durability testing.

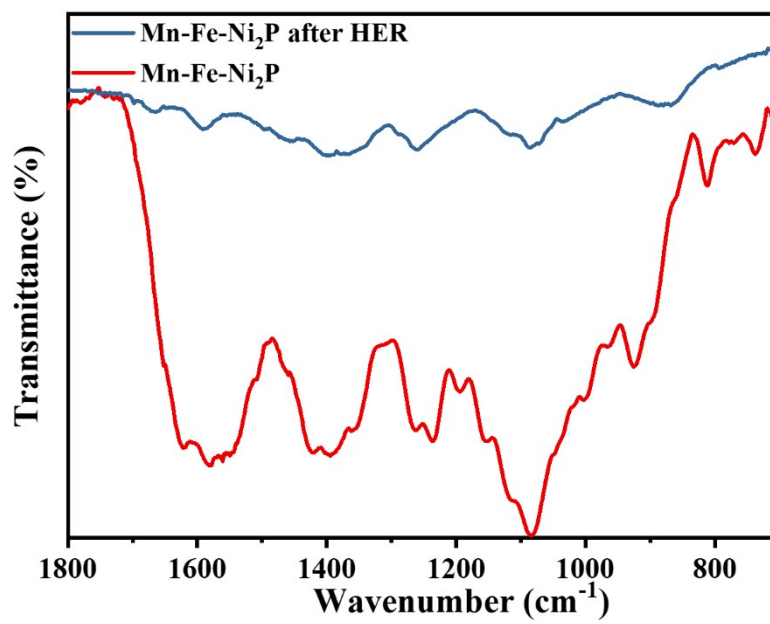
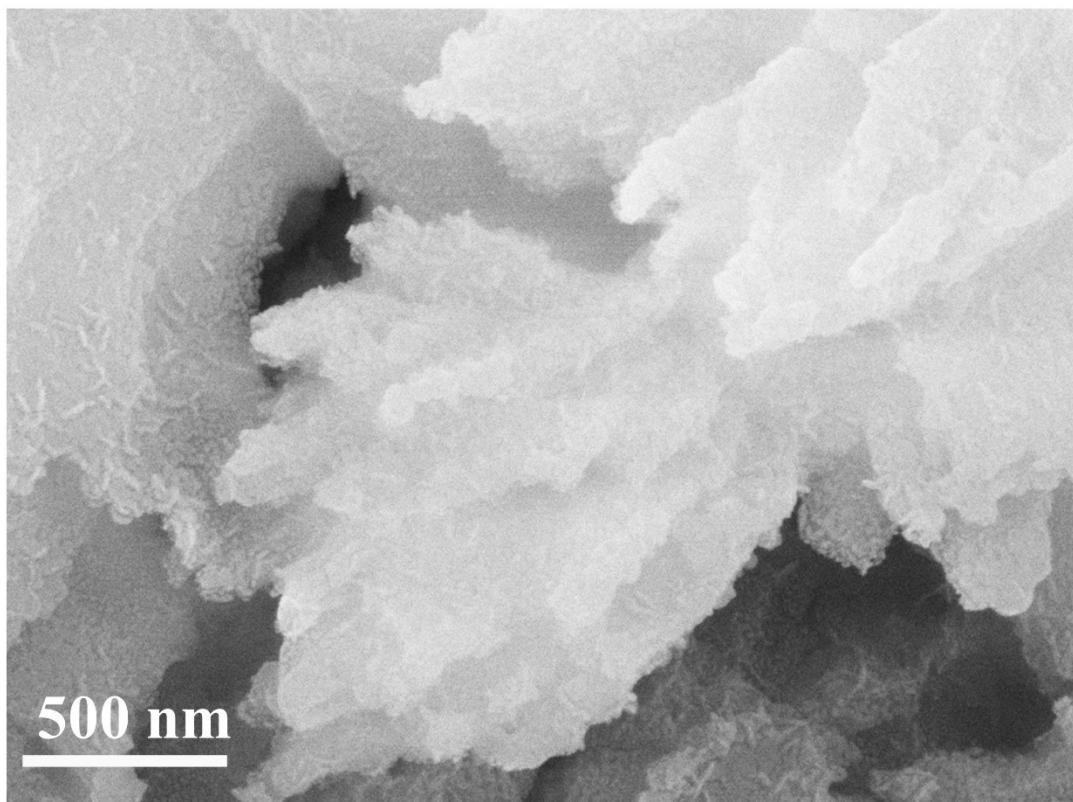
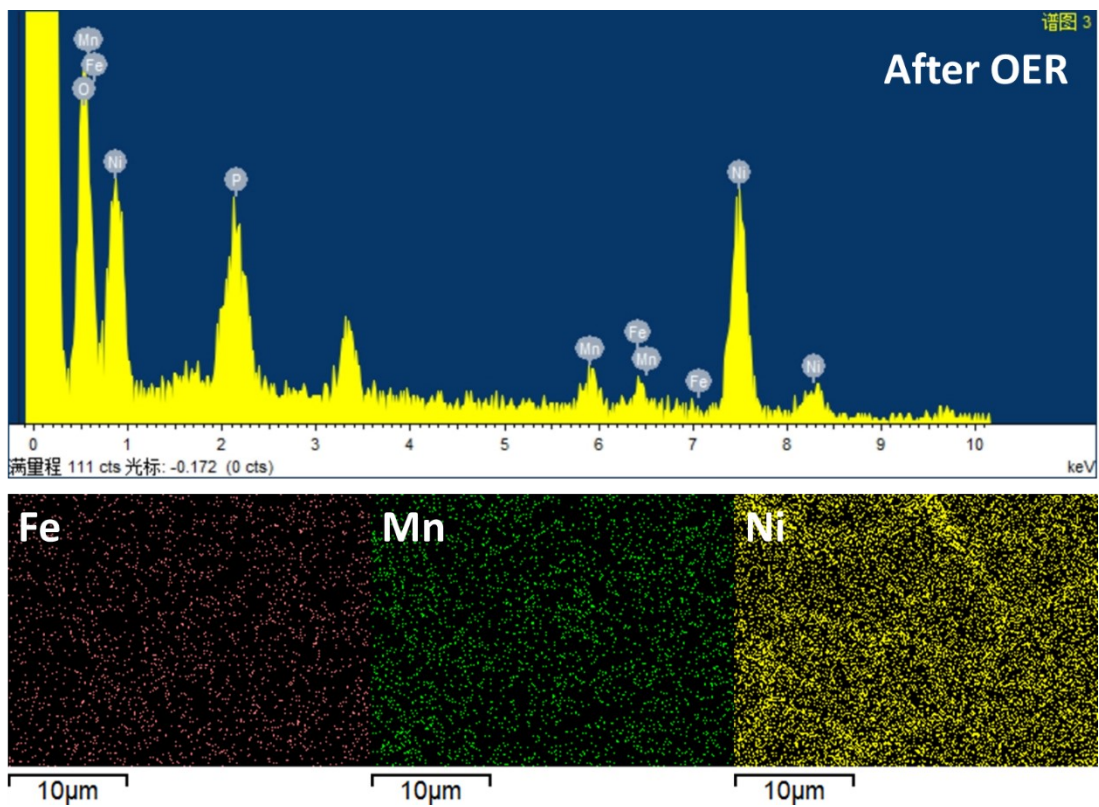


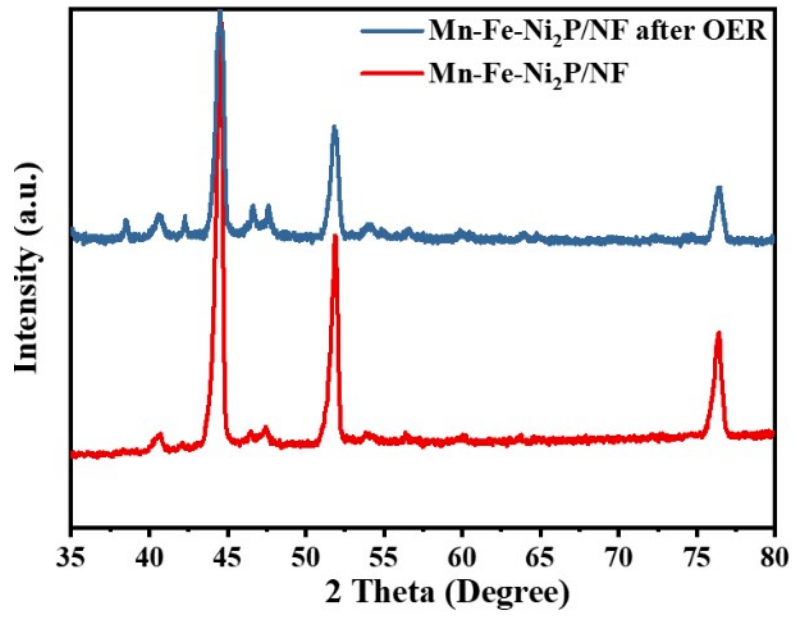
Fig. S13 FTIR spectra of Mn-Fe-Ni<sub>2</sub>P/NF before and after HER durability testing.



**Fig. S14** SEM image of Mn-Fe-Ni<sub>2</sub>P/NF after OER durability testing.

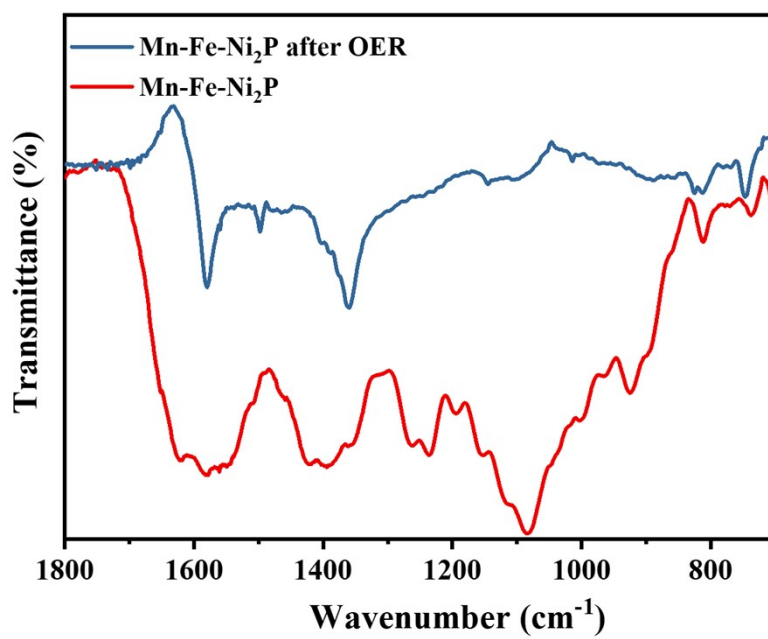


**Fig. S15** EDS and Mapping of Mn-Fe-Ni<sub>2</sub>P/NF after OER durability testing.



**Fig. S16** XRD patterns of Mn-Fe-Ni<sub>2</sub>P/NF before and after OER durability testing.





**Fig. S17** FTIR spectra of Mn-Fe-Ni<sub>2</sub>P/NF before and after OER durability testing.



**Table S1.** Results of ICP-OES test of Mn-Fe-Ni<sub>2</sub>P/NF-X (X=0.09, 0.18, 0.27, 0.33, 0.43, 0.57, 0.72).

Sample	Wt %	Mn	Fe	Ni	P	Mn/Fe
Mn-Fe-Ni <sub>2</sub> P/NF-0.09		5.22	2.73	27.56	22.33	1.91
Mn-Fe-Ni <sub>2</sub> P/NF-0.18		3.74	0.71	35.36	14.25	5.26
Mn-Fe-Ni <sub>2</sub> P/NF-0.27		2.63	1.87	27.79	22.81	1.41
Mn-Fe-Ni <sub>2</sub> P/NF-0.33		0.45	0.43	60.22	17.68	1.05
Mn-Fe-Ni <sub>2</sub> P/NF-0.43		0.92	0.87	54.25	17.34	1.06
Mn-Fe-Ni <sub>2</sub> P/NF-0.57		0.52	1.08	77.94	10.03	0.48
Mn-Fe-Ni <sub>2</sub> P/NF-0.72		3.8	1.04	27.43	21.61	3.65

**Table S2.**  $C_{dl}$  and ECSA of Mn-Fe-Ni<sub>2</sub>P/NF-X (X=0.09, 0.18, 0.27, 0.33, 0.43, 0.57, 0.72) and Fe-Ni<sub>2</sub>P/NF nanosheets.

Sample	$C_{dl}$ (mF cm <sup>-2</sup> )	ECSA
Fe-Ni <sub>2</sub> P/NF	4.5	112.5
Mn-Fe-Ni <sub>2</sub> P/NF-0.09	6.6	165
Mn-Fe-Ni <sub>2</sub> P/NF-0.18	24.3	607.5
Mn-Fe-Ni <sub>2</sub> P/NF-0.27	7.2	180
Mn-Fe-Ni <sub>2</sub> P/NF-0.33	2.9	72.5
Mn-Fe-Ni <sub>2</sub> P/NF-0.43	4.4	110
Mn-Fe-Ni <sub>2</sub> P/NF-0.57	2.0	50
Mn-Fe-Ni <sub>2</sub> P/NF-0.72	17.8	445

**Table S3.** EIS fitting results of Mn-Fe-Ni<sub>2</sub>P/NF-X (X=0.09, 0.18, 0.27, 0.33, 0.43, 0.57, 0.72) and Fe-Ni<sub>2</sub>P/NF in HER and OER.

Sample	Resistance (ohm) (HER)	Resistance (ohm) (OER)	Chi-squared (HER)	Chi-squared (OER)
Fe-Ni <sub>2</sub> P/NF	47.68	6.66	1.31×10 <sup>-3</sup>	9.33×10 <sup>-4</sup>
Mn-Fe-Ni <sub>2</sub> P/NF- 0.09	11.9	2.2	2.63×10 <sup>-4</sup>	2.58×10 <sup>-3</sup>
Mn-Fe-Ni <sub>2</sub> P/NF- 0.18	7.9	25.4	5.5×10 <sup>-3</sup>	1.48×10 <sup>-3</sup>
Mn-Fe-Ni <sub>2</sub> P/NF- 0.27	14.13	1.25	2.43×10 <sup>-4</sup>	2.52×10 <sup>-4</sup>
Mn-Fe-Ni <sub>2</sub> P/NF- 0.33	35.3	2.33	1.28×10 <sup>-3</sup>	1.18×10 <sup>-3</sup>
Mn-Fe-Ni <sub>2</sub> P/NF- 0.43	15.79	3.2	2.25×10 <sup>-3</sup>	2.23×10 <sup>-4</sup>
Mn-Fe-Ni <sub>2</sub> P/NF- 0.57	21	26.05	6.37×10 <sup>-3</sup>	2.52×10 <sup>-4</sup>
Mn-Fe-Ni <sub>2</sub> P/NF- 0.72	13.2	4.6	3.71×10 <sup>-3</sup>	6.83×10 <sup>-4</sup>

**Table S4.** Comparison of HER performances for Mn-Fe-Ni<sub>2</sub>P nanosheets with similar materials in the alkaline media.

Electrocatalysts	Current density (mA cm <sup>-2</sup> )	Overpotential (mV)	electrocatalytic substrate	Refs
<b>Mn-Fe-Ni<sub>2</sub>P</b>	<b>100</b>	<b>231</b>	<b>Nickel foam (NF)</b>	<b>This work</b>
NiOOH-Zn	100	280	Copper foam (CF)	Chem. Eng. J., 2024, 480: 148126
INF-FeCuS	100	292	Iron-nickel foam (INF)	Chem. Eng. J, 2023, 457: 141357
Fe-Ni <sub>3</sub> S <sub>2</sub>	100	254	INF	Adv. Energy Mater., 2022, 12: 2201913
NiFeSn oxyhydroxide	100	272	Carbon cloth (CC)	Nano Energy, 2024, 124: 109428.
FeCoNiCuMn HEA	100	281	\	Energy Environ. Sci., 2023,16, 619-628
CoBOx/NiCoP	100	283	NF	Chem. Eng. J. 2024: 152973
MNF-2	100	273	NF	Appl. Catal. B: Environ., 2023, 322: 122103
Ni <sub>2</sub> P@Co <sub>9</sub> S <sub>8</sub>	100	188	NF	Adv. Sci., 2023, 10(33): 2303682
Ni <sub>3</sub> S <sub>2</sub> -FeS/NF-2	100	262	NF	J. Colloid Interface Sci., 2022, 616: 422-432
FMO/NF	100	263.72	NF	J. Colloid Interface Sci., 2023, 632: 108–116
CoOx/Fe-CoP	100	267	NF	Chem. Eng. J., 2023, 478: 147374

**Table S5.** Comparison of OER performances for Mn-Fe-Ni<sub>2</sub>P nanosheets with similar materials in the alkaline media.

Electrocatalysts	Current density (mA cm <sup>-2</sup> )	Overpotential (mV)	Electrocatalytic substrate	Refs
<b>Mn-Fe-Ni<sub>2</sub>P</b>	<b>200</b>	<b>288</b>	<b>NF</b>	<b>This work</b>
Ni(OH) <sub>2</sub> /NiOOH	200	369	NF	Adv. Funct. Mater., 2024: 2407407
B-MOF-Zn-Co	100	362	Cu foam	Small, 2024, 20(22): 2308517
NiCo LDH/NiCoS	200	378	CC	Nano Res., 2022, (15): 4986-4995
Ni <sub>0.6</sub> Co <sub>1.8</sub> -MOF	200	350	Iron foam (IF)	Chem. Eng. J., 2024: 154093
NiFeSn oxyhydroxide	200	301	CC	Nano Energy, 2024, 124: 109428.
NiCoP	100	290	CC	Green Chem., 2023, 25: 4104-4112
B-MOF-Zn-Co	100	362	Cu foam	Small, 2024, 20(22): 2308517
MX-@MOF-Co <sub>2</sub> P	200	407	NF	J. Mater. Sci. Technol., 2023, (145):74-82
NiPx@HA	200	392	NF	Small, 2023, 19(11): 2205689

**Table S6.** Comparison of overall water splitting performances for Mn-Fe-Ni<sub>2</sub>P nanosheets with similar materials in the alkaline media.

Electrocatalysts	Current density (mA cm <sup>-2</sup> )	Potential (V)	electrocatalytic substrate	Refs
<b>Mn-Fe-Ni<sub>2</sub>P</b>	<b>10</b>	<b>1.54</b>	<b>NF</b>	<b>This work</b>
Fe <sub>2</sub> O <sub>3</sub> /P-CoMoO <sub>4</sub>	10	1.48	NF	Appl. Catal. B: Environ., 2024, 346: 123741
NiIrSAA-NiFe-LDH and IrSAC-NiFe-LDH	10	1.49 V	\	ACS Catalysis, 2023, 13(16): 11195-11203
NiYCe-MOF/NF	10	1.54	NF	Nano Lett. 2022, 22, 7238–7245
FeMo@CoNi-OH/Ni <sub>3</sub> S <sub>2</sub>	10	1.48 V	NF	Chem. Eng. J., 2023, 468: 143605
Mn <sub>0.25</sub> Ni <sub>0.75</sub> O  Mn <sub>0.25</sub> Ni <sub>0.75</sub> O	10	1.57	NF	J. Energy Chem., 2023, 86: 167-179
NiFeSn oxyhydroxide	10	1.55	CC	Nano Energy, 2024, 124: 109428
CoP <sub>2</sub> /Co <sub>2</sub> P@CNT-CC	10	1.55	CC	ACS Appl. Mater. Interfaces, 2022, 14: 56847-56855
CeO <sub>x</sub> @Co <sub>3</sub> O <sub>4</sub>	10	1.57	NF	Appl. Surf. Sci., 2023, 615: 156361
RuP/CoNiP <sub>4</sub> O <sub>12</sub>	10	1.56	CC	Appl. Catal. B: Environ., 2023, 368: 122447
NiCoFe-P/C	10	1.55	NF	J. Energy Chem., 2022, 74: 149-158
Ir/Ni <sub>3</sub> Fe/rGO  Ir/Ni <sub>3</sub> Fe/rGO	10	1.57	Graphite oxide	Chem. Eng. J., 2023, 451: 138548