Low-cost and Multi-level Structured NiFeMn Alloy@NiFeMn Oxyhydroxide Electrocatalysts for Highly-efficient Overall Water Splitting

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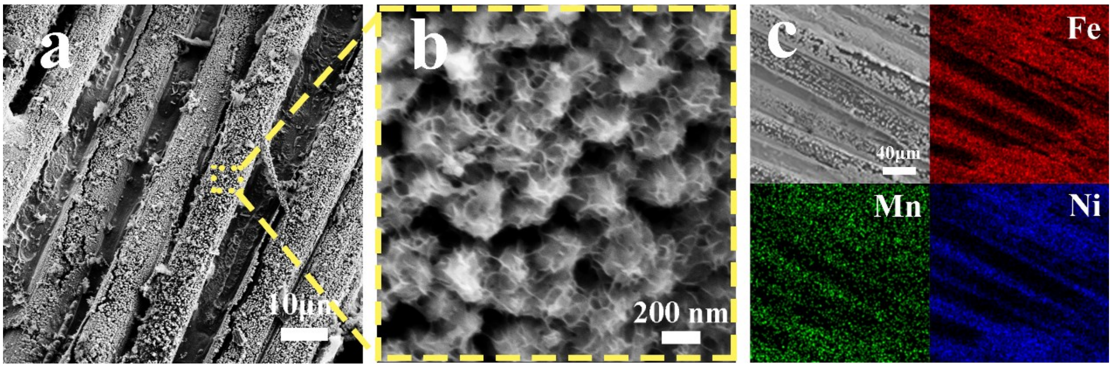


Fig. S1 (a, b) SEM images of the NiFeMn-AOs on carbon cloth. (c) EDX elemental mapping images from SEM.

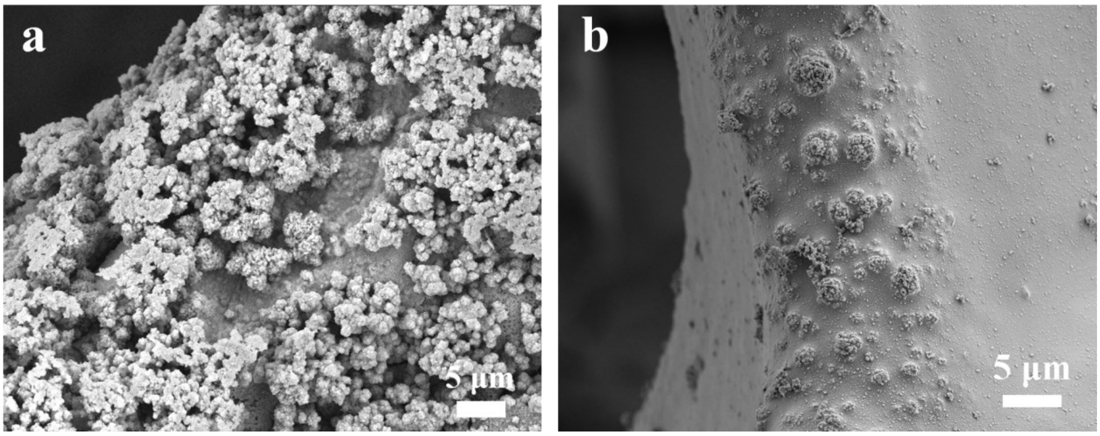


Fig. S2 (a) surface geometry of the NiFeMn-AOs from SEM (b) surface geometry of the NiFe-AOs from SEM.

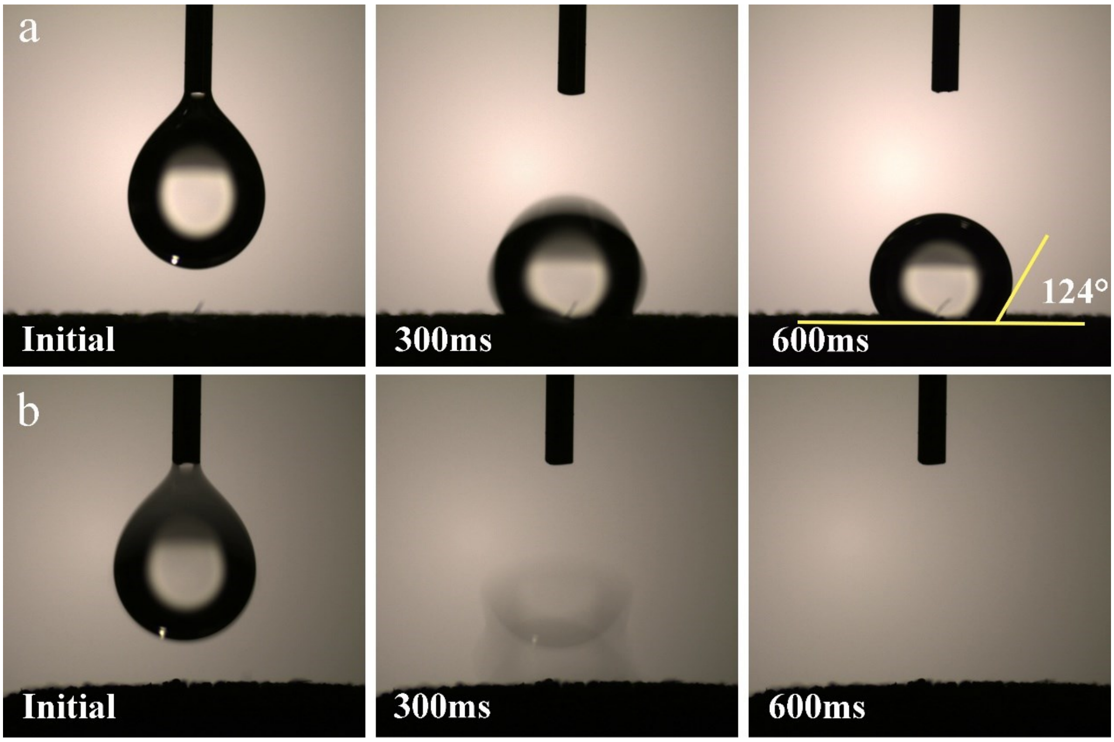
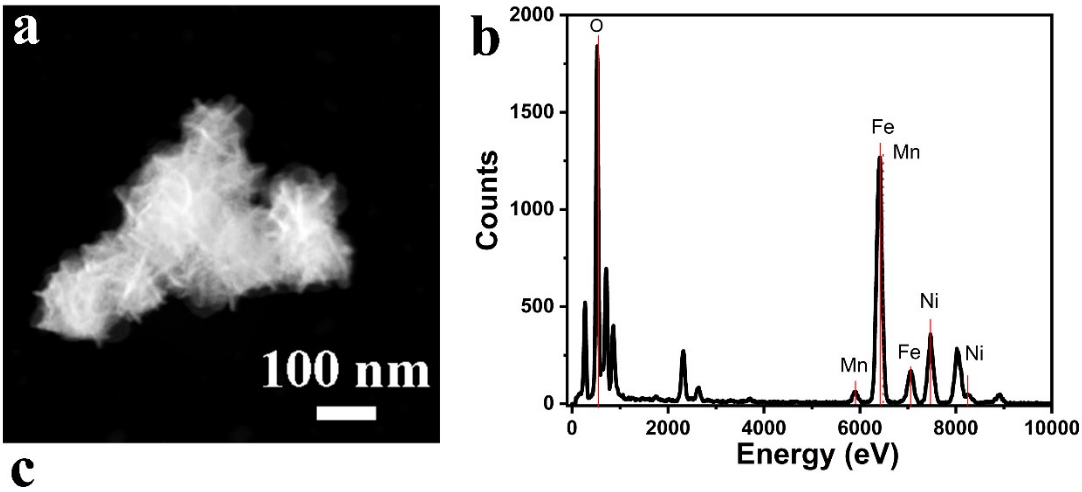


Fig. S3 Contact angle measurements of a droplet (1 M KOH electrolyte) placed on the surface of (a) commercial nickel foam and (b) NiFeMn-AOs to investigate the wettability between the electrolyte and the catalyst surface.



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Element	Series	unn. C[wt.%]	norm. C[wt.%]	Atom. C[at.%]
Nickel	K-series	27.25	31.60	18.55
Iron	K-series	28.12	32.62	20.12
Manganese	K-series	8.88	10.30	6.46
Oxygen	K-series	21.97	25.48	54.87

Fig. S4 (a, b) HAADF-STEM image and the corresponding elemental selected area of Mn, Ni, Fe, and O in NiFeMn-AOs. (c) EDX element selected area from SEM.

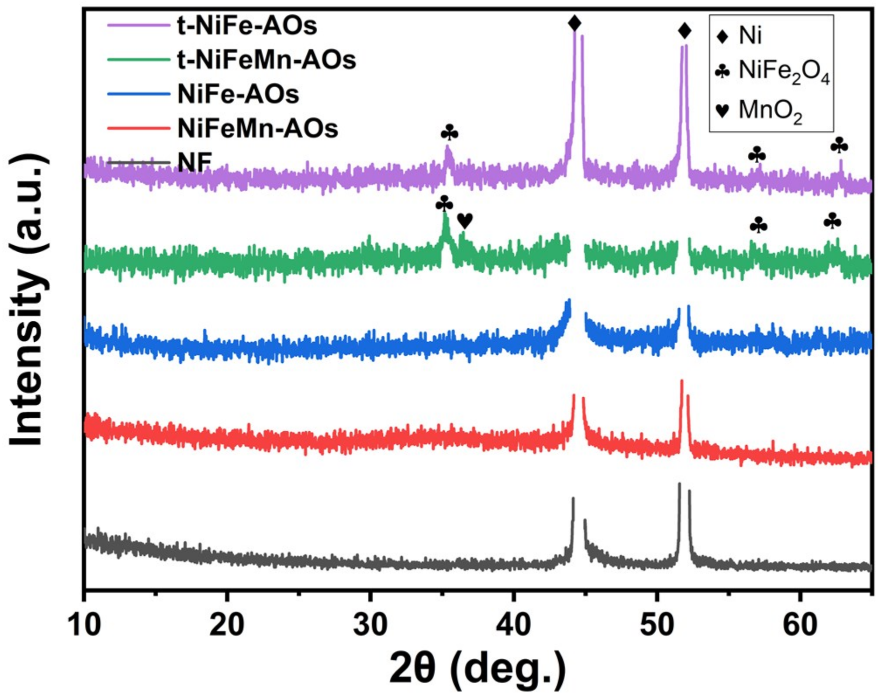

Fig. S5 XRD Pattern of nickel foam (NF), NiFe-AOs, NiFeMn-AOs, t-NiFe-AOs (after annealing) and t-NiFeMn-AOs (after annealing).

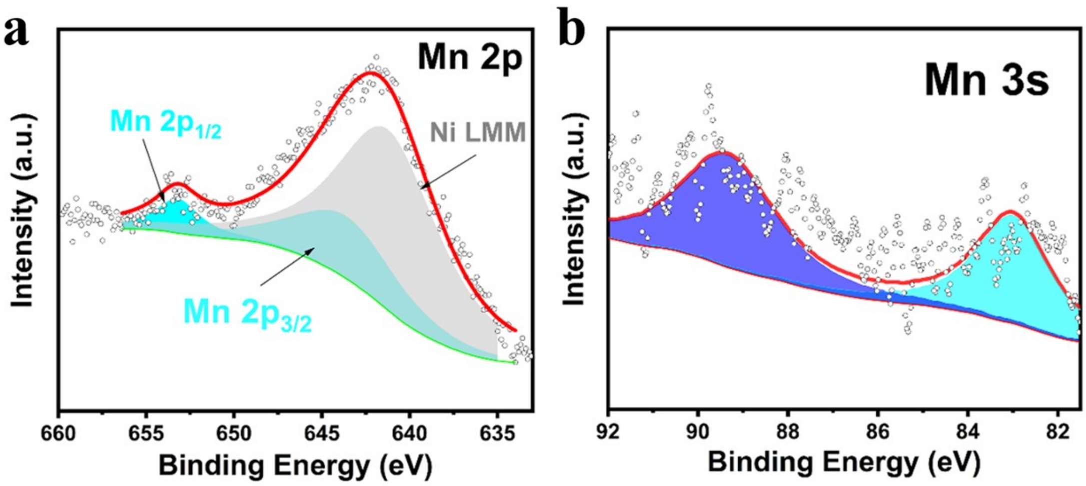

Fig. S6 (a) XPS high-resolution scans of Mn 2p, (b) XPS high-resolution scans of Mn 3s.

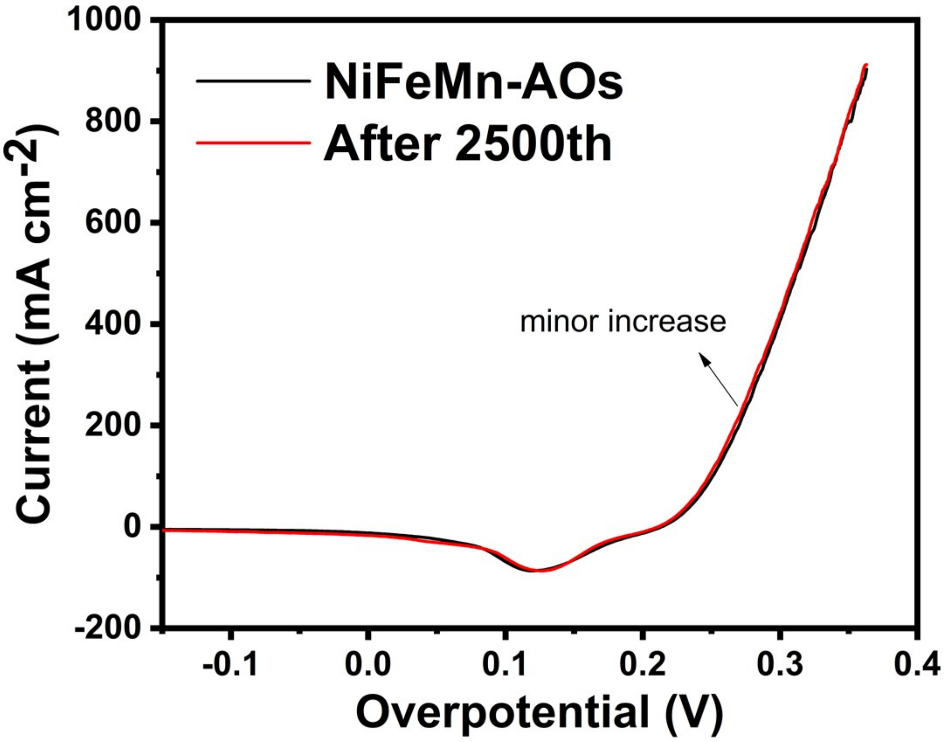

Fig. S7 Polarization data for NiFeMn-AOs before and after 2500th CV cycles.

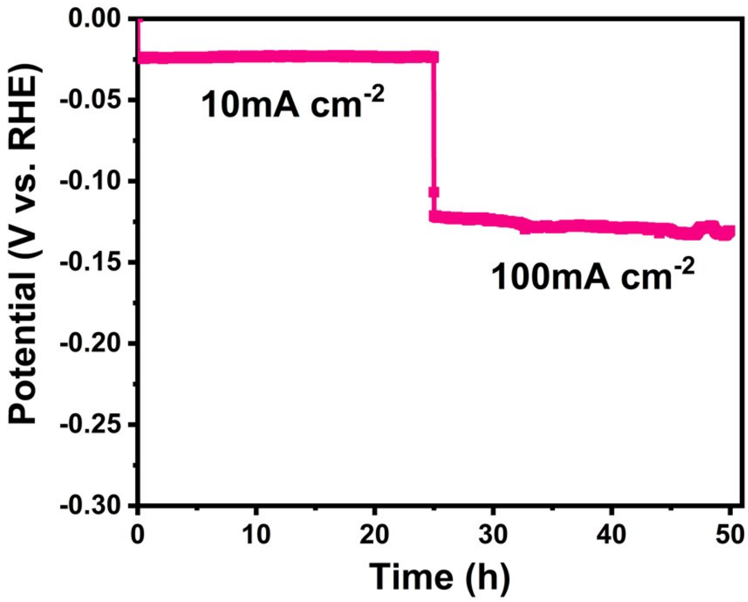

Fig. S8 Long-term HER performance stability of NiFeMn-AOs over 50 h by chronopotentiometry test.

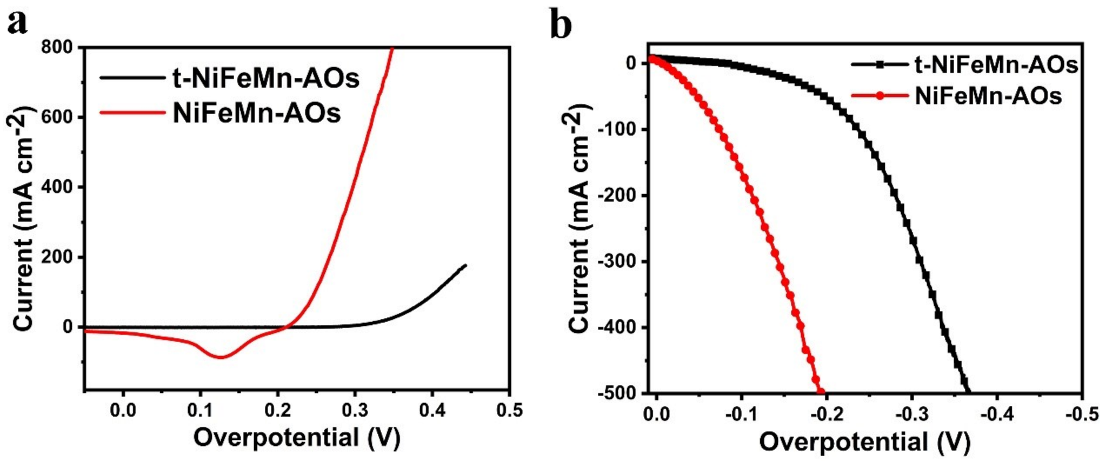

Fig. S9 (a) Comparison of OER performance of NiFeMn-AOs performance before and after annealing at 500°C. (b) Comparison of HER performance of NiFeMn-AOs performance before and after annealing at 500°C.

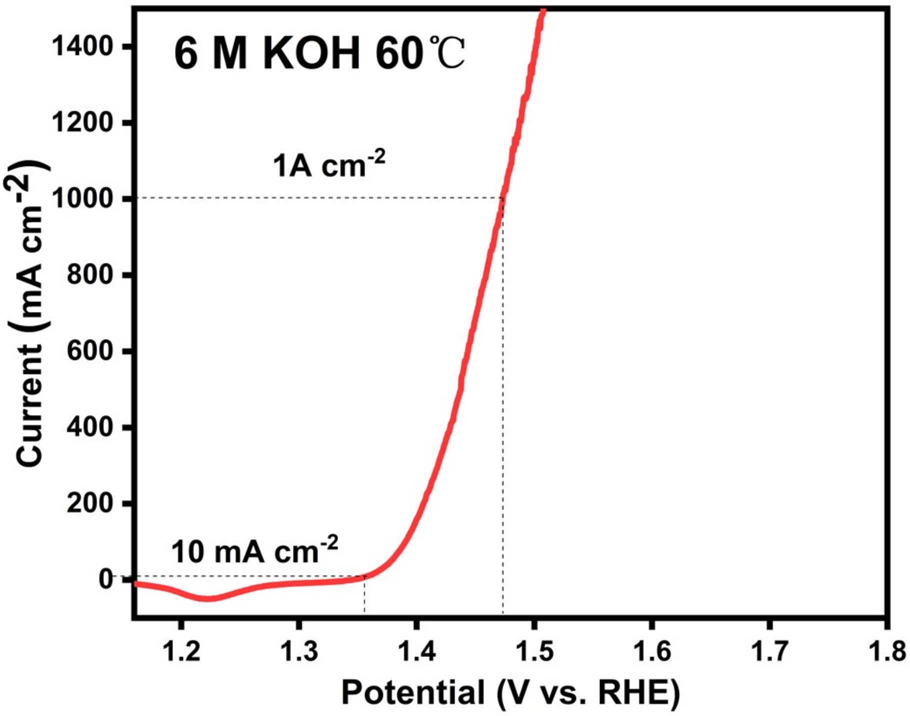

Fig. S10 OER polarization curve of NiFeMn-AOs in 6 M KOH at 60℃.

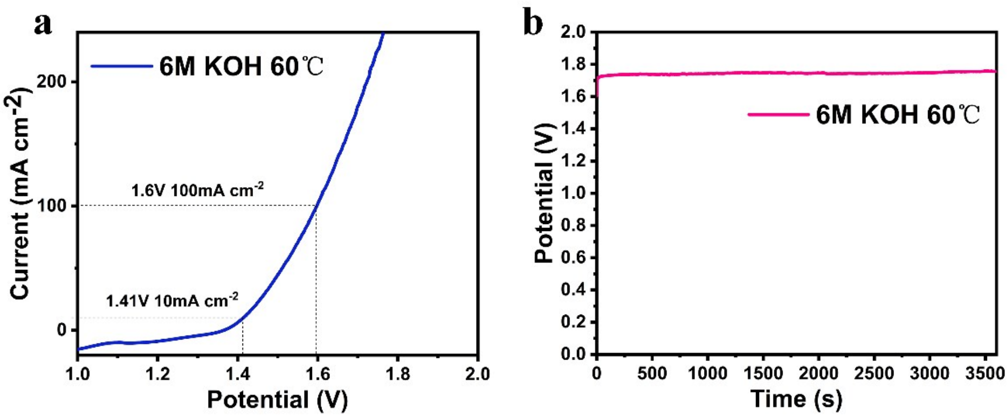

Fig. S11 (a) The polarization curves of NiFeMn-AOs// NiFeMn-AOs electrolyzer in 6 M KOH at 60℃ (without iR-compensation). (b) Chronopotentiometry curves of NiFeMn-AOs in a 6 M KOH electrolyte at 60℃.

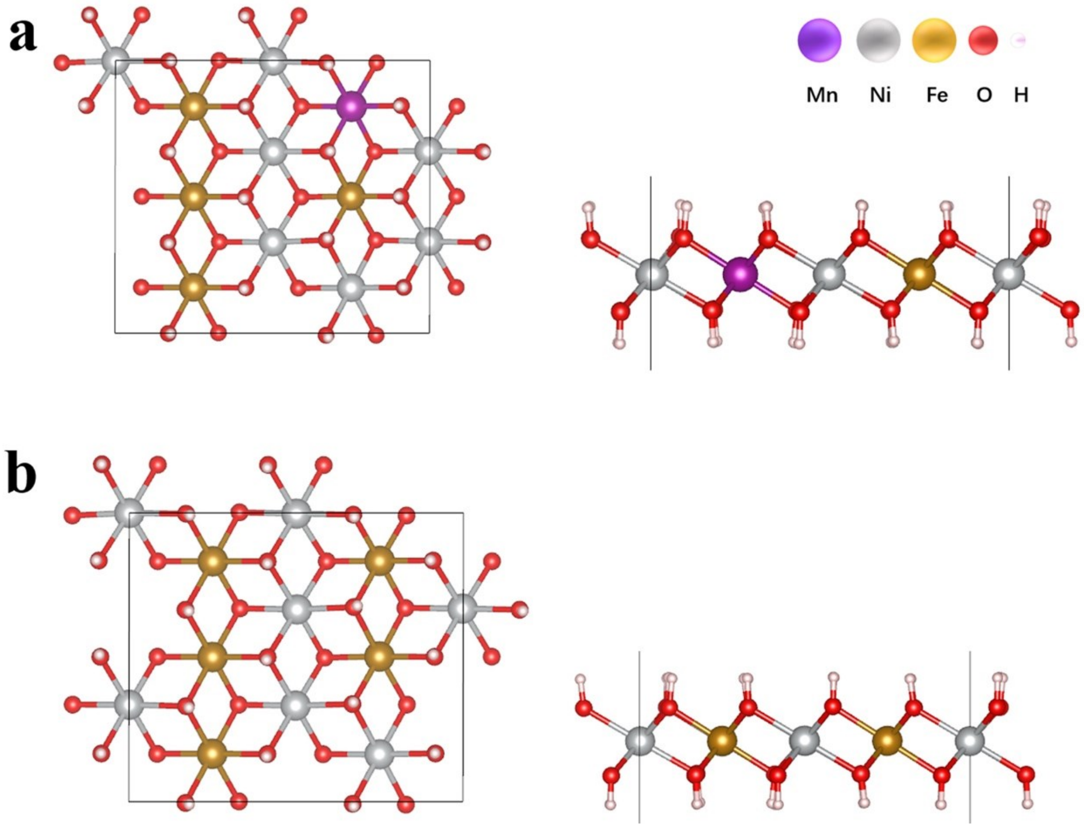

Fig. S12 Surface model of (a) NiFeMn-AOs and (b)NiFe-AOs.

Table S1. The alkaline OER activities of the NiFeMn-AOs and the reported non-noble based electrocatalysts

Electrocatalysts	**Overpotential at 10 mA cm^{-2}**	**Tafel slope (mV dec^{-1})**	**Reference**
NiFe/NiFe-OH	222 mV	41	*Appl. Catal. B: Environ.* **2019,** 257, 117899
$Ni_{60}Fe_{30}Mn_{10}$-alloy	200 mV	62	*Energy Environ. Sci.* **2016**, *9*, 540
Ni@NiFe LDH	218 mV	66	*J. Mater. Chem. A.* **2019**, *7*, 21722
FeOOH(Se)	221 mV	54	*J. Am. Chem. Soc.* **2019**, *141*, 7005
Ni_3Se_4@NiFe LDH	223 mV	55	*Nanoscale Horiz.* **2019***, 4, 1132*
CoFe@NiFe/NF	190 mV	46	*Appl. Catal. B.* **2019**, 253, 131
NiFe LDH/$NiFe_2O_4$	202 mV	47	*ACS Appl. Mater. Interfaces* **2018**, 10, 26283
CoFeZr oxides	248 mV	54	*Adv. Mater.* **2019**, 31, 1901439
$NiFeMoO_x$	255 mV	35	*Adv. Sci.* **2020**, 7, 1902034
CuO_x@$NiMnO_x$	225 mV	80	*J. Mater. Chem. A.* **2020**, 8, 16463
NiFeMn-AOs	220 mV	26	(This work)

Table S2. The alkaline HER activities of the NiFeMn-AOs and the reported non-noble based Electrocatalysts.

Electrocatalysts	Overpotential at 10 mA cm^{-2}	Tafel slope (mV dec^{-1})	Reference
Ni@NCNT/NiMoN/NF	15 mV	68	*J. Mater. Chem. A,* **2019**, 7, 13671
Ni@NiFe LDH	92 mV	72	*J. Mater. Chem. A.* **2019**, *7*, 21722
NiFe LDH@Ni_3N	30 mV	79	*J. Mater. Chem. A* **2020**, *33*, 17202
Ni_3Se_4@NiFe LDH	85 mV	98.6	*Nanoscale Horiz.* **2019***, 4, 1132*
CoFe@NiFe/NF	240 mV	88.9	*Appl. Catal. B: Environ.* **2019**, 253, 131
NiFe LDH/$NiFe_2O_4$	101 mV	67.1	*ACS Appl. Mater. Interfaces* **2018**, 10, 26283
CoFeZr oxides	104 mV	119	*Adv. Mater.* **2019**, 31, 1901439
$NiFeMoO_x$	22 mV	76	*Adv. Sci.* **2020**, 7, 1902034
CuO_x@$NiMnO_x$	71.6 mV	63	*J. Mater. Chem. A* **2020**, 8, 16463
NiFeMn-AOs	19 mV	45	(This work)

Table S3. The alkaline water splitting activities of NiFeMn-AOs//NiFeMn-AOs coupled electrolyzer and the reported electrolyzer

Electrocatalysts	**Overpotential at 10 mA cm^{-2}**	**Overpotential at 100 mA cm^{-2}**	**Reference**
$NiS_{0.5}Se_{0.5}$	1.55 V	1.73 V	*Adv. Mater.* **2020**, 2000231
Ni/Mo_2C	1.57 V	>1.8 V	*Adv. Energy Mater.* **2019**, 9, 1803185
Ni@NiFe LDH	1.56 V	1.61 V (with iR compensation)	*J. Mater. Chem. A.* **2019**, *7*, 21722
$CuCo_2S_4$	1.61 V (20 mA cm^{-2})	>1.8 V	*J. Mater. Chem. A.* **2020**. 8, 1799
Ni_3Se_4@NiFe LDH	1.54 V	1.75 V	*Nanoscale Horiz.* **2019**, *4, 1132*
CoFe@NiFe/NF	1.59 V	>1.8 V	*Appl. Catal. B.* **2019**, 253, 131
NiFe LDH/$NiFe_2O_4$	1.53 V	1.74 V	*ACS Appl. Mater. Interfaces* **2018**, 10, 26283
CoFeZr oxides	1.63 V	1.78 V	*Adv. Mater.* **2019**, 31, 1901439
$NiFeMoO_x$	1.50 V	1.63 V	*Adv. Sci.* **2020**, 7, 1902034
CuO_x@$NiMnO_x$	1.62 V	1.82 V	*J. Mater. Chem. A.* **2020**, 8, 16463
NiFeMn-AOs	1.46 V (15 mA cm^{-2})	1.63 V	(This work)