

Electronic Supplementary Information

Experimental section

Materials: NH_4F , $\text{Ni}(\text{NO}_3)_2 \cdot 6\text{H}_2\text{O}$ were purchased from Sinopharm Chemical Reagent Co., Ltd. Urea was purchased from Nanjing Chemical Reagent Co., Ltd. $\text{Fe}(\text{NO}_3)_3 \cdot 9\text{H}_2\text{O}$ was purchased from Aladdin Ltd. (Shanghai, China). $\text{RuCl}_3 \cdot 3\text{H}_2\text{O}$ ($\geq 43\%$) was purchased from Sigma–Aldrich Chemical Reagent Co., Ltd. The plain weave 100 mesh Ti mesh with the diameter of 0.10mm was provided by Hongshan District, Wuhan Instrument Surgical Instruments Business. All reagents were analytical reagent grade and used as received without further purification.

Preparation of hydroxide precursor: In a typical synthesis process, 0.58 g $\text{Ni}(\text{NO}_3)_2 \cdot 6\text{H}_2\text{O}$, 0.09 g $\text{Fe}(\text{NO}_3)_3 \cdot 9\text{H}_2\text{O}$, 0.60 g urea, and 0.22 g NH_4F were dissolved in 40 mL deionized water under vigorous stirring for 10 min. Ti mesh was selected as the substrate due to its excellent chemical stability, acceptable electronic conductivity, three dimensional structure with high surface area, and open structure allowing solvent good access at the reaction interface. Ti mesh was cleaned by sonication in water and ethanol for 10 min, was immersed into the solution. Then the solution was transferred to a 50 mL Teflonlined stainless steel autoclave with a piece of Ti mesh (2 cm \times 4 cm). Third, the autoclave was sealed and maintained at 120 °C for 6 h in an electric oven. After the autoclave cooled down naturally to room temperature, the NiFe–LDH/TM was taken out and thoroughly washed with deionized water, finally dried at 60 °C for 6 h in air. The hydroxide precursor of FeS/TM and NiS/TM were synthesised under same condition without $\text{Ni}(\text{NO}_3)_2 \cdot 6\text{H}_2\text{O}$ and $\text{Fe}(\text{NO}_3)_3 \cdot 9\text{H}_2\text{O}$, respectively.

Preparation of FeS–NiS/TM, FeS/TM and NiS/TM: FeS–NiS/TM, FeS/TM and NiS/TM were obtained from corresponding hydroxide precursor via hydrothermal sulfurization reaction. In a typical synthesis process, 1.92 g Na_2S were dissolved in 40 mL deionized water. Then the solution was transferred to a 50 mL Teflonlined stainless steel autoclave with the hydroxide precursor. Third, the autoclave was sealed and maintained at 120 °C for 4 h in an electric oven. After the autoclave cooled down

naturally to room temperature, the hydroxide precursor was taken out and thoroughly washed with deionized water and ethanol several times, then dried at 60 °C for 6 h in air.

Preparation of RuO₂/TM electrode: Preparation RuO₂ by 2.61 g of RuCl₃·3H₂O and 30 mL KOH (1.0 M) were dissolved in 100 mL solution under vigorous stirring for 10 min at 100 °C. Then the above solution was centrifuged for 10 minutes and filtered. The precipitates were collected and washed with water several times. Finally, the product was dried at 80 °C overnight and then annealed at 300 °C in air atmosphere for 3 h. For a typical synthesis of RuO₂/TM electrode, 50 mg RuO₂ was dispersed in 1 mL ethane/water (v:v = 1:1) solution with sonication for 30 min. Then dropped on Ti mesh (0.5 × 0.5 cm), and dried at 80°C for 4 h.¹

Characterization: The X-ray diffraction (XRD) patterns were obtained from were obtained from MiniFlex600 diffractometer (Rjgaku, Japan). Scanning electron microscope (SEM) measurements were recorded on a XL30 ESEM FEG scanning electron microscope at an accelerating voltage of 20 kV. The structures of the samples were determined by transmission electron microscopy (TEM) images on a HITACHI H-8100 electron microscopy (Hitachi, Tokyo, Japan) operated at 200 kV. X- ray photoelectron spectroscopy (XPS) data of the samples was collected on an Thermo ESCALAB 250XI x- ray photoelectron spectrometer using Mg as the exciting source.

Electrochemical measurements: Electrochemical measurements were performed with a CHI 660E electrochemical analyzer (CH Instruments, Inc., Shanghai) in a standard three-electrode system using FeS–NiS/TM as working electrode, graphite rod as the counter electrode, and a saturated Hg/HgO used as the reference electrode. All experiments were carried out at room temperature. All potentials measured were calibrated to RHE using the following equation: $E(\text{RHE}) = E(\text{Hg/HgO}) + (0.098 + 0.059 \times \text{pH}) \text{ V}$.

TOF calculations: The slope is derived from the linear relationship, the number of activity species is calculated by the formula: $\text{slope} = n^2 F^2 m / 4RT$, n represents the number of electrons transferred, we assuming that the electron transferred process is

one electron process between FeS–NiS/TM and NiFe–LDH/TM, n is 1, F represents the faradaic constant (96485 C mol^{-1}), m is the amount of active species, R and T represent ideal gas constant and the absolute temperature, respectively. The TOF is employed by the following equation: $\text{TOF} = jA/4Fm$ (j is the current density; A is the geometrical area of the electrode, and 4 indicates the moles of electron consumption for one mole oxygen evolution).

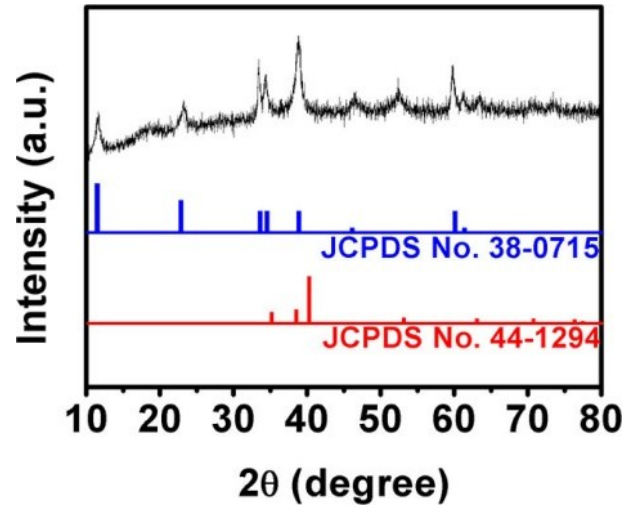


Fig. S1. XRD pattern of NiFe-LDH/TM.

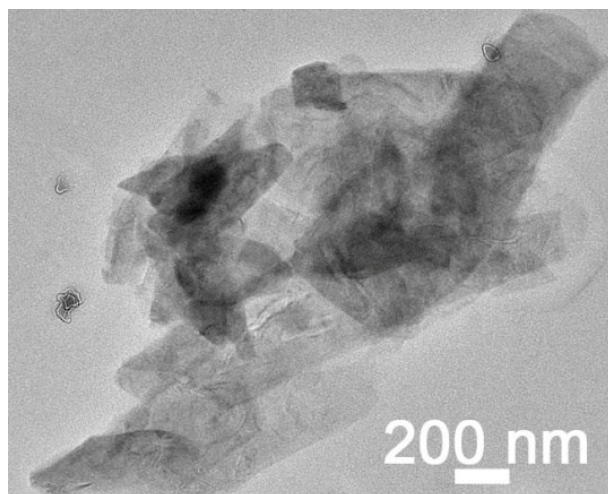


Fig. S2. TEM image for NiFe-LDH nanosheet.

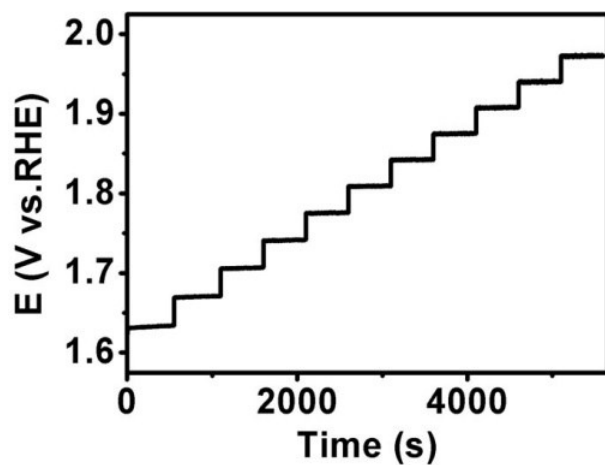


Fig. S3. Multi-current process of FeS–NiS/TM without iR correction.

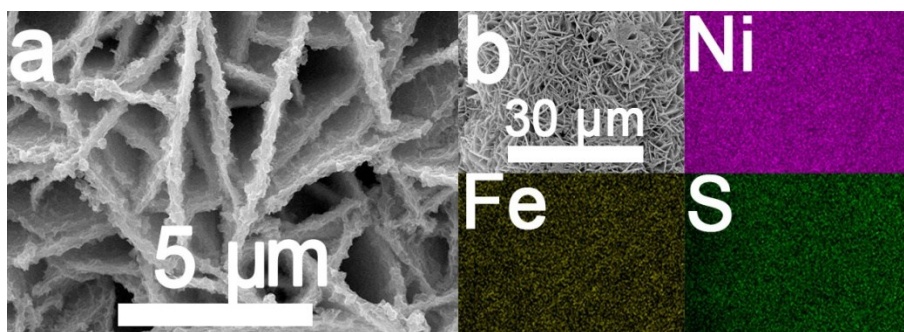


Fig. S4. (a) SEM image and (b) EDX elemental mapping images of Ni, Fe and S for FeS-NiS/TM after the durability test.

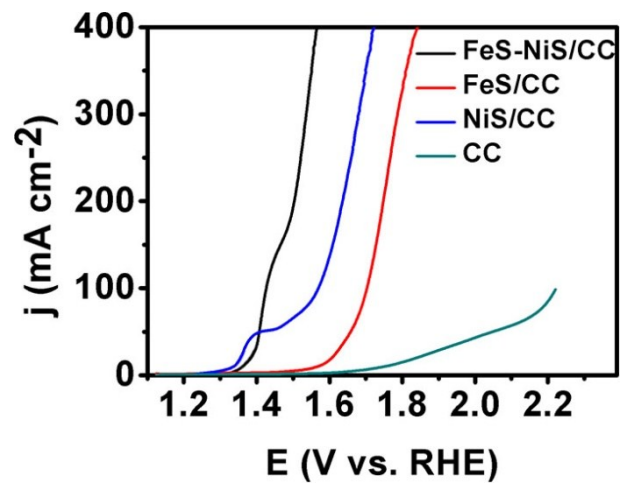


Fig. S5. LSV curves of CC and FeS-NiS/CC with a scan rate of 5 mV s⁻¹ in 1.0 M KOH.

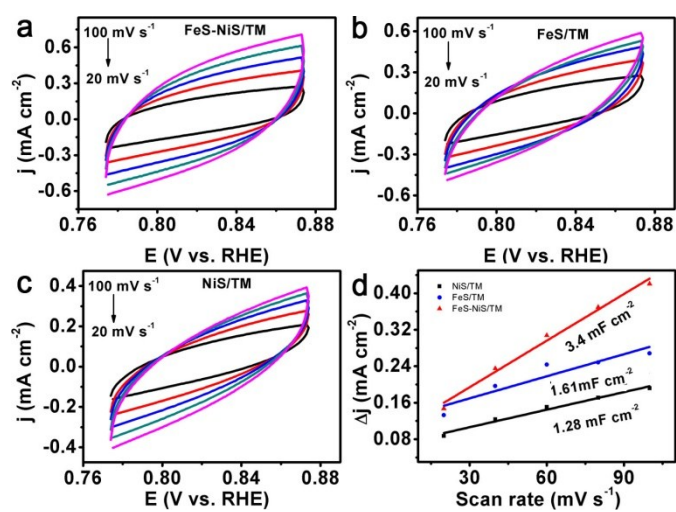


Fig. S6. CVs of (a) FeS–NiS/TM, (b) FeS/TM and (c) NiS/TM with various scan rates. (d) Capacitive current with various rates.

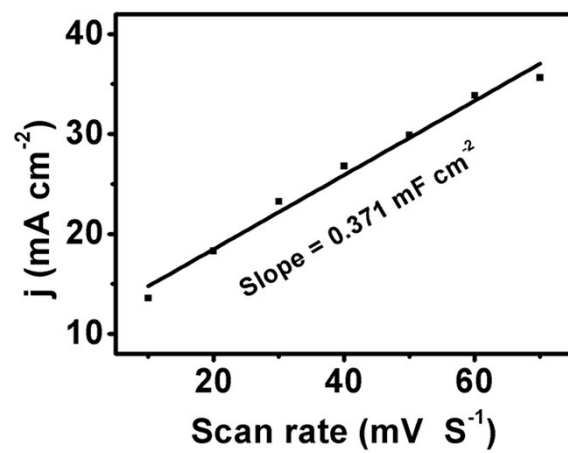


Fig. S7. Oxidation peak current versus scan rate plot for FeS–NiS/TM.

Table S1. Comparison of OER performance for FeS–NiS/TM with other non-noble-metal OER electrocatalysts in alkaline media.

Catalyst	j (mA cm ⁻²)	η (mV)	Electrolyte	Ref.
FeS–NiS/TM	10	260	1.0 M KOH	This work
NiPS ₃	10	294	1.0 M KOH	2
Ni@graphene	10	~370	1.0 M KOH	3
Ni ₂ P	10	290	1.0 M KOH	4
NiS@N/S–C	10	417	1.0 M KOH	5
Co ₃ FeP _x O	10	291	1.0 M KOH	6
Co–P@NC-800	10	370	1.0 M KOH	7
NiFe/NF	10	262	1.0 M KOH	8
CoS ₂	10	308	1.0 M KOH	9
CoO _x –4h	10	306	1.0 M KOH	10
Ni–Fe–P	10	~309	30 wt% KOH	11
Fe–CoOOH/G	10	330	1.0 M KOH	12
Ni _{0.69} Fe _{0.31} O _x /C	10	280	1.0 M KOH	13
CoMnON/N–rGO	10	350	1.0 M KOH	14
Ni _{0.5} Co _{0.5} O _x	10	360	1.0 M KOH	15
PtNi	10	350	0.1 M KOH	16
Au ₁ N _x singlesite/C ₃ N ₄	10	~450 mV	1.0 M KOH	17
Co–doped Ni–Mn LDH	10	~310	1.0 M KOH	18
NiS	10	354	1.0 M KOH	19
FeNiP@N–CFs	10	300	1.0 M KOH	20
Ni ₅₉ Cu ₁₉ P ₉	10	307	1.0 M KOH	21
Co ₉ S ₈	10	285	1.0 M KOH	22
SL–Co(OH) ₂	10	350	1.0 M KOH	23
a–CoVOx	10	347	1.0 M KOH	24
Cu ₂ O@C	10	330	1.0 M KOH	25

Table S2. Comparison of TOF for FeS–NiS/TM with other non-noble-metal OER electrocatalysts in alkaline media.

Catalyst	j (mA cm ⁻²)	TOF (mol O ₂ s ⁻¹)	Electrolyte	Ref.
FeS–NiS/TM	500	0.521	1.0 M KOH	This work
NiCo ₂ O ₄ @Ni–Co–B/CC	500	0.019	1.0 M KOH	26
Co–MoF/NF	400	0.180	1.0 M KOH	27
Ni ₃ S ₂ @G	220	0.035	1.0 M KOH	28
Co ₉ S ₈	285	0.0268	1.0 M KOH	29
NiTe	400	0.324	1.0 M KOH	30
NiFe	400	0.075	1.0 M KOH	31

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