## **1. Experimental section**

#### 1.1. Raw materials

All the raw materials, including nickel sulfide (Ni<sub>3</sub>S<sub>2</sub>), nickel(II) oxide (NiO), nickel hydroxide (Ni(OH)<sub>2</sub>), nickelic oxide (Ni<sub>2</sub>O<sub>3</sub>), potassium hydroxide (KOH), Ir/C (20 wt.% in Ir), Nafion solution (5.0 wt.% in ethanol), sodium sulfide (Na<sub>2</sub>S), nickel foam, and hydrochloric acid were purchased from commercial sources.

## 1.2. Electrochemical evaluation on Ni<sub>3</sub>S<sub>2</sub>, NiO, Ni(OH)<sub>2</sub>, and Ni<sub>2</sub>O<sub>3</sub>

Electrochemical evaluation was performed by using a three-electrode system controlled by a CHI 760E electrochemistry station at room temperature. The counter electrode and the reference electrode were a platinum sheet and a saturated calomel electrode (SCE), respectively. The working electrode was a rotating disk electrode (RDE) (Pine Research Instrument, USA). The RDE was a glassy carbon electrode with a diameter of 5.0 mm and area of 0.196 cm<sup>2</sup>. All the recorded potentials were calibrated to the reversible hydrogen electrode (RHE) according to the following equation:

$$E_{\rm RHE} = E_{\rm SCE} + 0.241 + 0.0592 \,\text{pH} \tag{1}$$

where  $E_{\text{RHE}}$  and  $E_{\text{SCE}}$  are the potentials relevant to RHE and SCE, respectively.

To prepare the working electrode, the electrocatalysts were coated onto the disk electrode via a drop-casting method. Specifically, 5.0 mg electrocatalyst was mixed with 0.95 mL ethanol and 50  $\mu$ L Nafion solution (5.0 wt.% in ethanol), followed by ultrasonication for 30 min to form a homogenous suspension. Then, 10.0  $\mu$ L of the suspension was dropped onto the disk electrode, where the electrode was mechanically

polished and ultrasonically washed by deionized water and ethanol in advance. After full evaporation of the solvent, the working electrode was successfully prepared for further electrochemical evaluations.

The electrolyte was O<sub>2</sub>-saturated 0.10 mol L<sup>-1</sup> KOH aqueous solution. The rotating speed of the working electrode was 1600 rpm throughout the measurements. The electrocatalytic oxygen evolution activity was evaluated via 95% *iR*-recompensed linear sweep voltammetry (LSV) measurements performed at a scan rate of 10.0 mV s<sup>-1</sup> from -0.01 to 0.80 V vs SCE.

Based on the data collected from the LSV measurements, Tafel slopes were calculated according to Tafel equation:

$$\eta = b \log(j/j_0) \tag{2}$$

where  $\eta$  is the overpotential ( $\eta$ =*E*<sub>RHE</sub>-1.23 V), *b* is the Tafel slope, *j* and *j*<sub>0</sub> are the recorded current density and exchange current density, respectively.

The electrochemical impedance spectroscopy (EIS) titration was carried out at a series of potentials. The potential sequence was from lower potential to higher potential (1.54, 1.55, 1.56, 1.57, 1.58, 1.60, 1.62, 1.64, 1.66, 1.68, and 1.70 V vs RHE). The frequency range was from  $10^{-1}$  to  $10^{5}$  Hz. The voltage amplitude was 5.0 mV.

The electrochemical active surface area (ECSA) was tested via comparing the double-layer capacitance ( $C_{dl}$ ) of the electrocatalysts. Typically, the electrocatalysts with higher  $C_{dl}$  correspond to larger ECSA. To determine the  $C_{dl}$ , cyclic voltammetry (CV) with a fixed scan window form -0.025 to 0.025 V vs SCE was performed at a series of scan rates of 10, 20, 40, 60, 80, and 100 mV s<sup>-1</sup>.  $C_{dl}$  was half of the slope

calculated by fitting the current density difference at 0.000 V vs. SCE against the scan rate. ECSA can be calculated via the equation 'ECSA =  $C_{dl}/C_s$ ', where  $C_s = 40 \ \mu F \ cm^{-2}$ .

#### 1.3. Synthesis of Ni<sub>3</sub>S<sub>2</sub>-1h, Ni<sub>3</sub>S<sub>2</sub>-10h, and Ni<sub>3</sub>S<sub>2</sub>-100h

For the synthesis of Ni<sub>3</sub>S<sub>2</sub>-1h, 100 mg Ni<sub>3</sub>S<sub>2</sub> was mixed with 3.0 mL ethanol and ultrasonicated for 5.0 min to form a homogenous suspension. After that, the suspension was uniformly dropped onto a conductive carbon paper  $(5.0 \times 5.0 \text{ cm}^2)$ . After full evaporation of the solvent, a carbon cloth (5.0×5.0 cm<sup>2</sup>, WOS1002, CeTech) was mechanically pressed onto the Ni<sub>3</sub>S<sub>2</sub>-coated carbon paper neatly to construct the electrode. A three-electrode system was further set up with the Ni<sub>3</sub>S<sub>2</sub> electrode as the working electrode, the saturated calomel electrode (SCE) as the reference electrode, and the platinum sheet as the counter electrode, respectively. The electrolyte was 0.10 M KOH aqueous solution. Constant potential of 0.54 V vs SCE was applied on the working electrode for an hour. After that, the electrocatalysts in the working electrode was scraped, collected, washed thoroughly with deionized water and ethanol for three times respectively, and dried at 45°C overnight. After that, Ni<sub>3</sub>S<sub>2</sub>-1h was finally obtained. Ni<sub>3</sub>S<sub>2</sub>-10h and Ni<sub>3</sub>S<sub>2</sub>-100h were synthesized following otherwise identical procedures as Ni<sub>3</sub>S<sub>2</sub>-1h except replacing the OER duration time from 1 h to 10 or 100 h, respectively.

## 1.4. Material characterization

Scanning electron microscope (SEM) images were recorded on a JSM 7401F

(JEOL Ltd., Tokyo, Japan) SEM operated at 3.0 kV. Transmission electron microscope (TEM) images were recorded on a JEM 2010 (JEOL Ltd., Tokyo, Japan) TEM operated at 120.0 kV or a FEI Titan Cubic Themis G2 300 TEM operated at 300.0 kV. Aberrationcorrected high-angle annular dark field scanning transmission electron microscope (HADDF–STEM) images were recorded on the FEI Titan Cubic Themis G2 300 TEM equipped with double spherical aberration correctors and a HADDF detector with the convergence angle being 24 mrad and the collection angel between 90 and 240 mrad. The working voltage was 300.0 kV. Energy-dispersive X-ray spectrometer (EDS) analysis and corresponding elemental mapping were carried out using the JSM 7401F SEM or the FEI TEM equipped with an Oxford Instrument energy dispersive X-ray spectrometer.

Nitrogen adsorption-desorption isotherms were tested on an Autosorb-IQ2-MP-C system at 77K. The specific surface area was evaluated following the multipoint Brunauer–Emmett–Teller (BET) method. The pore-size distribution was calculated using the Quenched Solid Density Function Theory (DFT) model according to the data of the adsorption branch. X-ray diffraction (XRD) patterns were recorded by using a Bruker D8 Advanced diffractometer with Cu-K<sub> $\alpha$ </sub> radiation at 40.0 kV and 120 mA as the X-ray source. Electron probe micro analysis (EPMA) was accomplished by using a JEOL JXA8230 micro analyzer.

Time-of-flight secondary ion mass spectrometry (TOF-SIMS) analysis was carried out via a TESCAN S9000 scanning electronic microscopy equipped with focused ion beam (FIB) and TOF-SIMS accessories. The (FIB) was used for surface etching and further elemental analysis in depth. The current for FIB ranged from 100 pA to 1 nA. The FIB gun was in 55° angle with the TOF-SIMS detector. Auger electron spectroscopy (AES) analysis was carried out by using PHI-700. The Ar<sup>+</sup> sputtering rate for the AES depth-profiling calibrated on SiO<sub>2</sub>/Si was *ca.* 3 nm min<sup>-1</sup>. X-ray photoelectron spectroscopy (XPS) measurements were performed via Escalab 250xi. All the samples were cleaned with argon plasma in advance. All the XPS results were calibrated using C 1s line at 284.6 eV. Ni L-edge X-ray absorption near edge structures (XANES) characterizations were performed from BL12B- $\alpha$  of Hefei National Synchrotron Radiation Laboratory (NSRL).

## 1.5. Fabrication and electrochemical evaluation of Ni<sub>3</sub>S<sub>2</sub>-NF and Ir/C-NF

For the fabrication of Ni<sub>3</sub>S<sub>2</sub>-NF, in-situ vulcanization was performed on nickel foam (NF). Specifically, a NF was cut into the size of 3.0×3.0 cm<sup>2</sup> and then washed in 3.0 M hydrochloric acid for 20 min to remove the surface oxide layer. After that, the NF was washed with deionized water for three times. Then, the cleaned NF was immersed into 40 mL 0.10 M Na<sub>2</sub>S aqueous solution in a hydrothermal kettle. The hydrothermal kettle was placed at 140°C for 6.0 h. After cooling to room temperature, the NF was washed with deionized water and ethanol for four times respectively and dried at 50°C overnight. After that, Ni<sub>3</sub>S<sub>2</sub>-NF was finally obtained.

For the fabrication of Ir/C-NF, a suspension of the Ir/C electrocatalyst was firstly prepared. 9.0 mg Ir/C was dispersed into 0.95 mL ethanol and 0.05 mL Nafion solution (5.0 wt% in ethanol) followed by 10 min sonication to form a homogeneous suspension.

Then, the suspension was dropped onto a NF with a size of  $3.0 \times 3.0$  cm<sup>2</sup>. After the solvent was fully evaporated, Ir/C-NF was obtained.

Electrochemical evaluation on the above monolithic electrodes was identical to that for RDE evaluation except using the monolithic electrodes directly as the working electrode and utilizing O<sub>2</sub>-saturated 1.0 mol  $L^{-1}$  KOH aqueous solution as the electrolyte.

## 2. Supplementary note

Numerical simulation on the Ni<sub>3</sub>S<sub>2</sub> electrocatalyst was carried out based on a onedimensional semi-infinite plate model. Specifically, considering the thickness of the oxysulfide layer is significantly lower than that of the particle size of the Ni<sub>3</sub>S<sub>2</sub> electrocatalyst as well as the flattened surface of the Ni<sub>3</sub>S<sub>2</sub> electrocatalyst, the Ni<sub>3</sub>S<sub>2</sub> electrocatalyst is regarded as a solid semi-infinite plate with initially uniform ion concentration. The Ni<sub>3</sub>S<sub>2</sub> electrocatalyst is further exposed into the liquid electrolyte, where OER occurs on the solid–liquid interface. According to the experimental results, OER induces surface reconstruction to afford relative stable surface ion concentrations as the boundary conditions. During continuous OER process, sulfur and oxygen ions diffuse driven by the concentration gradient to afford consequent ion redistribution and depth structural reconstruction inside the bulk of the Ni<sub>3</sub>S<sub>2</sub> electrocatalyst (Figure S38). The mass transfer equations are as follows:

The conservation equations of sulfur and oxygen ions in Ni<sub>3</sub>S<sub>2</sub> electrocatalyst:

$$\frac{\partial c(S)}{\partial t} = -\nabla(N_S) + R_{VS}$$
$$\frac{\partial c(0)}{\partial t} = -\nabla(N_O) + R_{VO}$$

Where *t* is the OER duration, c(S) and c(O) are the concentrations of sulfur and oxygen ions in the Ni<sub>3</sub>S<sub>2</sub> electrocatalyst, *N*s and *N*o are the molar fluxes of sulfur and oxygen ions in the bulk of Ni<sub>3</sub>S<sub>2</sub> electrocatalyst, and *R*<sub>VS</sub> and *R*<sub>VO</sub> are the reaction rates to consume/generate sulfur and oxygen ions, respectively. Since sulfur and oxygen ions in the bulk phase of Ni<sub>3</sub>S<sub>2</sub> electrocatalyst cannot be consumed or generated:

$$R_{\rm VS} = 0$$
$$R_{\rm VO} = 0$$

No bulk flow is in the Ni<sub>3</sub>S<sub>2</sub> electrocatalyst (v=0, v is the flow velocity of the solid

phase). The molar fluxes of sulfur and oxygen ions can be expressed as:

$$N_{\rm S} = J_{\rm S} + c({\rm S})v = J_{\rm S}$$
$$N_{\rm O} = J_{\rm O} + c({\rm O})v = J_{\rm O}$$

where  $J_{\rm S}$  and  $J_{\rm O}$  are the molar diffusion fluxes of sulfur and oxygen ions, respectively.

Assuming that the density of the electrocatalyst ( $\rho$ ) and ion diffusion coefficient (*D*) in the electrocatalyst are constant. The molar diffusion fluxes of sulfur and oxygen ions can be expressed following Fick's law:

$$J_{\rm S} = -\frac{\rho}{M_{\rm S}} D_{\rm S} \frac{\partial \omega_{\rm S}}{\partial x} = -D_{\rm S} \frac{\partial c({\rm S})}{\partial x}$$
$$J_{\rm O} = -\frac{\rho}{M_{\rm O}} D_{\rm S} \frac{\partial \omega_{\rm O}}{\partial x} = -D_{\rm O} \frac{\partial c({\rm O})}{\partial x}$$

where  $M_S$  and  $M_O$  are the molar masses of sulfur and oxygen ions,  $\omega_S$  and  $\omega_O$  are the mass fractions of sulfur and oxygen ions,  $D_S$  and  $D_O$  are the diffusion coefficients of sulfur and oxygen ions in the electrocatalyst, and x is the depth of electrocatalyst.

The conservation equations of sulfur and oxygen ions can be written as:

$$\frac{\partial c(S)}{\partial t} = D_S \frac{\partial^2 c(S)}{\partial x^2}$$
$$\frac{\partial c(0)}{\partial t} = D_0 \frac{\partial^2 c(0)}{\partial x^2}$$

The boundary conditions of the above differential equations are listed as follows:

$$c(S)(x, 0) = c(S)_{i} (t = 0)$$
  

$$c(S)(0, t) = c(S)_{s} (x = 0)$$
  

$$c(S)(\infty, t) = c(S)_{i} (x = \infty)$$

$$c(0)(x,0) = 0 (t = 0)$$
  

$$c(0)(0,t) = c(0)_{s} (x = 0)$$
  

$$c(0)(\infty,t) = 0 (x = \infty)$$

where  $c(S)_i$  refers to the concentration of sulfur ion in pure Ni<sub>3</sub>S<sub>2</sub>,  $c(S)_s$  refers to the concentration of sulfur ion at the surface of the Ni<sub>3</sub>S<sub>2</sub> electrocatalyst under working OER conditions, and  $c(O)_s$  refers to the concentration of oxygen ion at the surface of the Ni<sub>3</sub>S<sub>2</sub> electrocatalyst under working OER conditions. Specifically, according to the material characterization results,  $c(S)_s$  is *ca*. 0.2 times of  $c(S)_i$ .

To solve the above partial differential equations, dimensionless concentrations are defined as follows:

$$f(S) = \frac{c(S)_{i} - c(S)}{c(S)_{i} - c(S)_{s}}$$
$$f(0) = \frac{c(0)}{c(0)_{s}}$$

The solutions of above equations are:

$$f(S) = 1 - \operatorname{erf}(q_S)$$
$$f(0) = 1 - \operatorname{erf}(q_O)$$

where erf(q) is Gauss error function:

$$\operatorname{erf}(q) = \frac{2}{\sqrt{\pi}} \int_{0}^{q} e^{-\eta^{2}} d\eta$$
$$q_{\mathrm{S}} = \frac{x}{2\sqrt{D_{\mathrm{S}}t}}$$
$$q_{0} = \frac{x}{2\sqrt{D_{\mathrm{S}}t}}$$

Hence, the functional relationships of c(S) and c(O) to depth (x) and time (t) are:

$$c(S)(x,t) = c(S)_{i} - [c(S)_{i} - c(S)_{s}][1 - erf(\frac{x}{2\sqrt{D_{S}t}})]$$
$$c(0)(x,t) = c(0)_{s}[1 - erf(\frac{x}{2\sqrt{D_{0}t}})]$$

For further simulation, the values of the following parameters are set as follows:

$$D_{\rm S} = 1.0 \times 10^{-17} {\rm cm}^2 {\rm s}^{-1}$$
  
 $D_{\rm O} = 1.0 \times 10^{-17} {\rm cm}^2 {\rm s}^{-1}$ 

$$c(S)_{S} = 0.2 c(S)_{i}$$
  
 $t = 1, 10, \text{ or } 100 \text{ h}$ 

The numerical simulation results are presented in Figure S39.

# 3. Supplementary figures



Figure S1. SEM images of Ni<sub>3</sub>S<sub>2</sub>.



Figure S2. TEM images of Ni<sub>3</sub>S<sub>2</sub>.



Figure S3. XRD patterns of Ni<sub>3</sub>S<sub>2</sub> and standard nickel sulfide.



**Figure S4.** 95% *iR*-compensated LSV profiles of the Ni<sub>3</sub>S<sub>2</sub> electrocatalyst at different scans in oxygen-saturated 0.1 M KOH electrolyte.



Figure S5. EIS titration results of the  $Ni_3S_2$  electrocatalyst with a potential range of 1.54–1.70 V and voltage step of 10/20 mV before and after OER.



Figure S6. SEM images of the Ni<sub>3</sub>S<sub>2</sub> electrocatalyst after OER.



Figure S7. Pore size distribution of the  $Ni_3S_2$  electrocatalyst before and after OER.



Figure S8. XRD patterns of the Ni<sub>3</sub>S<sub>2</sub> electrocatalyst before and after OER.



Figure S9. SEM and EDS mapping images of the Ni<sub>3</sub>S<sub>2</sub> electrocatalyst after OER.



Figure S10. EPMA mapping images of the Ni<sub>3</sub>S<sub>2</sub> electrocatalyst after OER.



Figure S11. SEM images of (a) Ni(OH)<sub>2</sub>, (b) Ni<sub>2</sub>O<sub>3</sub>, and (c) NiO electrocatalysts.



**Figure S12.** Tafel plots of Ni<sub>3</sub>S<sub>2</sub>, Ni(OH)<sub>2</sub>, Ni<sub>2</sub>O<sub>3</sub>, and NiO electrocatalysts according to the LSV data in Figure 1f.



**Figure S13.** (a) ECSA and (b) ECSA-normalized OER LSV profiles of Ni<sub>3</sub>S<sub>2</sub>, Ni(OH)<sub>2</sub>, Ni<sub>2</sub>O<sub>3</sub>, and NiO electrocatalysts, where  $j_{ECSA}$  refers to the ECSA-normalized current densities.



Figure S14. TEM image of the  $Ni_3S_2$  electrocatalyst after OER.



**Figure S15.** TEM image of the Ni<sub>3</sub>S<sub>2</sub> electrocatalyst after OER. The amorphous– crystalline core–shell structure can be identified.



Figure S16. HAADF-STEM and corresponding EDS elemental mapping images of the

Ni<sub>3</sub>S<sub>2</sub> electrocatalyst after OER.



Figure S17. Linear scan analysis along the dash line marked in Figure S16.

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**Figure S18.** SEM images of the Ni<sub>3</sub>S<sub>2</sub> electrocatalyst after OER (a) before and (b) after depth etching for TOF-SIMS characterization.



**Figure S19.** SEM images of the pristine Ni<sub>3</sub>S<sub>2</sub> electrocatalyst before OER (a) before and (b) after depth etching for TOF-SIMS characterization.



Figure S20. TOF-SIMS mapping on sulfur and oxygen on the surface (left) and on the inside (right) of the pristine  $Ni_3S_2$  electrocatalyst before OER.



Figure S21. Characterization area of the  $Ni_3S_2$  electrocatalyst after OER for depth-

profiling AES analysis.



Figure S22. SEM images of Ni<sub>3</sub>S<sub>2</sub>-10h.



Figure S23. SEM images of Ni<sub>3</sub>S<sub>2</sub>-100h.



Figure S24. SEM image of Ni<sub>3</sub>S<sub>2</sub>-10h after depth etching for TOF-SIMS characterization.



**Figure S25.** TOF-SIMS mapping on sulfur and oxygen on the surface (left) and on the inside (right) of Ni<sub>3</sub>S<sub>2</sub>-10h.



Figure S26. SEM image of  $Ni_3S_2$ -100h after depth etching for TOF-SIMS characterization.



Figure S27. TOF-SIMS mapping on sulfur and oxygen on the surface (left) and on the

inside (right) of Ni<sub>3</sub>S<sub>2</sub>-100h.



Figure S28. Characterization area of Ni<sub>3</sub>S<sub>2</sub>-10h for depth-profiling AES analysis.



Figure S29. Depth-profiling sulfur, oxygen, and nickel AES spectra of Ni<sub>3</sub>S<sub>2</sub>-10h.



Figure S30. Characterization area of Ni<sub>3</sub>S<sub>2</sub>-100h for depth-profiling AES analysis.



Figure S31. Depth-profiling sulfur, oxygen, and nickel AES spectra of Ni<sub>3</sub>S<sub>2</sub>-100h.



**Figure S32.** Sulfur content verses sputter time (depth of electrocatalyst) of Ni<sub>3</sub>S<sub>2</sub>-1h, Ni<sub>3</sub>S<sub>2</sub>-10h, and Ni<sub>3</sub>S<sub>2</sub>-100h. Similar sulfur contents can be detected on the surface despite different OER duration.



Figure S33. Surface AES nickel spectra of Ni<sub>3</sub>S<sub>2</sub>, Ni<sub>3</sub>S<sub>2</sub>-1h, Ni<sub>3</sub>S<sub>2</sub>-10h, and Ni<sub>3</sub>S<sub>2</sub>-100h.



Figure S34. Elemental content vs sputter time (depth of electrocatalyst) of Ni<sub>3</sub>S<sub>2</sub>-10h.



Figure S35. Elemental content vs sputter time (depth of electrocatalyst) of Ni<sub>3</sub>S<sub>2</sub>-100h.



Figure S36. TEM image of Ni<sub>3</sub>S<sub>2</sub>-10h.







**Figure S38.** The numerical simulation on Ni<sub>3</sub>S<sub>2</sub> electrocatalyst structural reconstruction based on a one-dimensional semi-infinite plate model.



**Figure S39.** Numerical simulation results of depth distribution of sulfur and oxygen elemental under different OER duration. c(O) and  $c(O)_s$  refer to the concentration of oxygen ion and the concentration of oxygen ion on the surface, respectively. c(S) and  $c(S)_i$  refer to the concentration of sulfur ion and the concentration of sulfur ion in bulk Ni<sub>3</sub>S<sub>2</sub>, respectively.



**Figure S40.** (a) and (b) TEM images of the electrocatalyst loaded on Ni<sub>3</sub>S<sub>2</sub>-NF. (c) Fourier-transformed crystalline lattice and (d) corresponding lattice spacing profile of the marked region in (b).



Figure S41. EDS patterns of the electrocatalyst loaded on Ni<sub>3</sub>S<sub>2</sub>-NF.



Figure S42. XRD patterns of Ni<sub>3</sub>S<sub>2</sub>-NF, NF, and standard Ni<sub>3</sub>S<sub>2</sub>.



Figure S43. SEM images of Ni<sub>3</sub>S<sub>2</sub>-NF.



Figure S44. Tafel plots of  $Ni_3S_2$ -NF and Ir/C-NF based on the data in Figure 5d.

## 4. Supplementary tables

monolithic electrodes. The electrolyte is 1.0 M KOH.							
Electrode	$E_{10}$	<i>E</i> 50	$E_{100}$	Tafel slope (mV dec <sup>-1</sup> )	Loading	Reference	
	(V vs	(V vs	(V vs				
	RHE)	RHE)	RHE)		$(mg cm^{-2})$		
Ni <sub>3</sub> S <sub>2</sub> -NF	1.415	1.460	1.476	50.7	1.0	41 :	
Ir/C-NF	1.556	1.655	1.748	148.2	1.0	this work	
SCFP-NF	1.54	1.59	1.61	55	0.034	1	
Co-MoS <sub>2</sub> /BCCF-21	1.49	1.56	1.58	85	2	2	
MoS <sub>2</sub> /FNS/FeNi foam	1.433	1.46	1.47	28.1	0.163	3	
Ni <sub>3</sub> N-VP <sub>2</sub> /NF	/	1.535	1.627	49	/	4	
NiTe/NiS	1.438	1.47	1.486	49	1.5	5	
CoFeZr oxides/NF	1.47	1.511	1.53		/	6	
NiFe-NM@G	/	/	1.437	/	/	7	
h-Co0.34Fe0.33Ni0.33-LDH	1.424	1.45	1.47	53	2.1	8	
γ-FeOOH/NF-6M	1.515	1.54	1.545	51	0.5	9	
NiSe/NF	1.49	1.53	1.55	64	2.8	10	
FeOOH/Co/FeOOH	1 47	1.505	1.54	32	/	11	
HNTAs-NF *	1.4/						
FeSe <sub>2</sub> /NF	1.474	1.515	1.537	/	3	12	

Table S1. OER performance comparison of Ni<sub>3</sub>S<sub>2</sub>-NF, Ir/C-NF, and other reported

W <sub>0.5</sub> Co <sub>0.4</sub> Fe <sub>0.1</sub> /NF	1.479	1.52	1.545	32	/	13
Mn <sub>3</sub> N <sub>2</sub> /NF	1.50	1.58	1.62	101	3	14
Fe/Ni/Co <sub>0.4</sub> -MIL-53/NF	/	/	1.467	71.3	/	15
CoMoS <sub>x</sub> /NF	/	/	1.574	/	/	16
MnGa4/NF	1.52	1.605	1.631	98	2	17
CoP <sub>x</sub> @CNS	/	1.515	1.545	70	/	18
NiGe/NF	1.457	1.55	1.58	56	1	19
NiFc-MOF/NF	1.425	1.458	1.471	44.1	0.8	20
P-CoNi2S4	1.512	1.543	1.557	40	1.6	21
Ni <sub>3</sub> S <sub>2</sub> /NF	1.49	/	/	/	1.6	22
Co1Mn1CH	/	1.587	1.616	/	5.6	23
R-NCO	1.47	1.515	1.54	50	2.5	24
CoMoNiS-NF-31	1.395	1.47	1.56	58	1.86	25
Co4Mo2@NC	1.56	1.6	1.63	46.1	/	26
Co5Mo1.0O NSs@NF	1.50	1.54	1.56	54.4	/	27
NC/NiMo/NiMoOx	1.513	1.56	1.615	111	20	28
NiCo2S4 NW/NF	1.49	1.57	1.61	40.1	/	29
S-NiFe <sub>2</sub> O <sub>4</sub> /NF	1.496	1.52	1.53	36.7	/	30
CoS <sub>2</sub> HNSs	1.519	1.565	/	57	1.5	31
Fe0.14Co0.86-P/CC	1.50	1.54	1.56	45.4	1.1	32
S-Ni <sub>3</sub> Se <sub>4</sub> -2	1.504	1.56	1.59	64	1	33

NiFeN <sub>x</sub> /NF	1.515	1.545	1.56	44.6	0.5	34
Co-S/NGF	1.463	1.58	1.675	138	5	35
S6M8	1.52	1.555	1.576	69.3	/	36
a-LNFBPO@NF	1.444	1.57	1.6	/	3	37
CoVFeN@NF	1.441	1.465	1.493	34.8	/	38
Mo-Ni <sub>3</sub> S <sub>2</sub> /Ni <sub>x</sub> P <sub>y</sub> /NF	/	1.467	1.499	60.6	3.15	39
Ni-ZIF/Ni-B@NF-4	1.463	1.515	1.56	57	/	40
NFN-MOF/NF	1.469	1.515	1.54	58.8	0.6	41
G-Ni4Fe/GF	1.539	1.59	1.595	50	0.05	42
F0.25C1CH/NF	1.457	1.471	1.487	42	4.17	43
Co@NC-600/Ni foam	1.56	1.62	/	43.9	0.5	44
NS-MnO <sub>2</sub>	1.55	1.625	1.67	40	1.59	45
Ni <sub>x</sub> B-300/NF	1.46	/	/	/	0.21	46
NiFe-PVP	1.526	1.57	1.58	/	48	47

\* The electrolyte is 1.0 M NaOH.

## **5.** Supplementary references

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