# **Supporting Information**

## Ultrathin Fe-MOF modified by Fe<sub>9</sub>S<sub>10</sub> for highly efficient oxygen evolution reaction

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## 1. Calculation formulas

The calculation formulas used in this paper were showed as follows:

Overpotential:  $\eta = E_{RHE}$ -1.23 V,  $E_{RHE} = E_{Ag/AgCl}$ + 0.059\*pH+0.197 V

where  $E_{RHE}$  referred to reversible hydrogen electrode potential.

Tafel slope:  $\eta = b \log j + a$ ,

where b was the Tafel slope. It was derived from the LSV curve, log j (j was the current density) as the abscissa and  $\eta$  as the ordinate, and the resulting slope was called the Tafel slope.

 $2C_{dl}$  is estimated by plotting  $\Delta J=(Ja-Jc)$  at 0.94 V against the scan rates.

Faradic efficiency: FE=(V/V<sub>m</sub>)/n,

Where V represents the actual volume of  $O_2$  collected,  $V_m$  represents the molar volume of gas at the corresponding temperature at the time of the test. n represents the number of molars of gas theoretically released.

Turnover Number: TON =  $n_{O_2}/n_{cat}$ , where  $n_{O_2}$  is the number of molars of generated O<sub>2</sub>,  $n_{cat}$  is the number of molars of catalyst.

Turnover Frequency: TOF=TON/t, where t is time

# 2. Chemical

Urea (CH<sub>4</sub>N<sub>2</sub>O,  $\geq$ 99.0%), ammonium fluoride (NH<sub>4</sub>F,  $\geq$ 98.0%), iron nitrate nonahydrate (Fe(NO<sub>3</sub>)<sub>3</sub>·9H<sub>2</sub>O,  $\geq$ 99.9%), sodium sulfide nonahydrate (Na<sub>2</sub>S·9H<sub>2</sub>O,  $\geq$ 98.0%), 1,4benzenedicarboxylic acid (C<sub>8</sub>H<sub>6</sub>O<sub>4</sub>,  $\geq$ 99.0%), sodium hydroxide (NaOH,  $\geq$ 97.0%), and N,Ndimethylformamide (DMF,  $\geq$ 99.5%) were purchased from Aladdin (Shanghai). Hydrochloric acid (HCl, 36%~38%) and ethanol ( $C_2H_5OH$ ,  $\geq$ 99.5%) were bought from Sinopharm Chemical Reagent Co., Ltd (Shanghai). Ultrapure water and nickel foam (NF) were bought from Crystal chemical (Nanjing). The above chemical was used directly without further treatment.

#### 3. The preparation of NF

Due to the presence of dirt and oxidation layers on the surface of NF, it was necessary to clean before using. The NF purchased was cut to the size of 1\*1.5 cm<sup>2</sup>, and then it was immersed in the3 M HCl solution prepared in advance. Cleaning with an ultrasonic cleaner for 20 minutes was employed. Then deionized water and ethanol were used for ultrasonic cleaning for 10min respectively, and then the NF was dried under vacuum at 60 °C for 12 h.



Fig. S1. XRD patterns of a) precursor, b) Fe<sub>9</sub>S<sub>10</sub>/NF and c) Fe-MOF/NF



Fig. S2. SEM images of a,) Fe-MOF/NF, b)  $Fe_9S_{10}/NF$ 



Fig. S3. TEM images of a, b)  $Fe_9S_{10}/NF$ , c, d) Fe-MOF/NF



Fig. S4. XPS spectra of C1s, O1s, Fe2p, S2p in Fe<sub>9</sub>S<sub>10</sub>/Fe-MOF/NF-2 before OER



**Fig. S5.** CV curves of a) Fe-MOF, b) Fe<sub>9</sub>S<sub>10</sub>/NF, c) Fe<sub>9</sub>S<sub>10</sub>/Fe-MOF/NF-1, d) Fe<sub>9</sub>S<sub>10</sub>/Fe-MOF/NF-2, e) Fe<sub>9</sub>S<sub>10</sub>/Fe-MOF/NF-3 at different scan rates



Fig. S6 a) Faradic efficiency, b) the device of drainage gas collection method, c-h) the volume of oxygen collected over time in 1 M KOH



Fig. S7. SEM images of  $Fe_9S_{10}$ /Fe-MOF/NF-2 after OER



Fig S8 a) Faradic efficiency, b) the device of drainage gas collection method, c-h) the volume of oxygen collected over time in 1 M KOH+0.5M NaCl



Fig. S9. Photos of  $Fe_9S_{10}$ /Fe-MOF/NF-2 when folded and unfolded



Fig. S10. XPS spectra of C1s, O1s in  $Fe_9S_{10}/Fe-MOF/NF-2$  before OER



Fig. S11. a, b) TEM images, c) HRTEM images, d, e, f, g) elemental mapping of  $Fe_9S_{10}/Fe-MOF/NF$  after OER



Fig. S12. XRD patterns of a) Fe-MOF/NF, b) Fe<sub>9</sub>S<sub>10</sub>/NF before and after reaction



Fig. S13. survey spectrum of a) Fe-MOF/NF, c)  $Fe_9S_{10}/NF$ , and the changes in the content of elements of b) Fe-MOF/NF, d)  $Fe_9S_{10}/NF$  before and after OER test



Fig. S14. TEM images of a) Fe-MOF/NF, c) Fe<sub>9</sub>S<sub>10</sub>/NF and HRTTM images of b) Fe-MOF/NF, d)  $Fe_9S_{10}/NF$  after reaction

**Table S1.** Comparison of OER performance in this work with recently reported materials introduced into S by the postprocessing of precursors electrocatalysts

|           |                                |          |           | -       |           |
|-----------|--------------------------------|----------|-----------|---------|-----------|
|           | Overpotential                  | Tafel    |           |         |           |
| Catalysts | (mV)/ current                  | slope    | Stability | Support | Reference |
|           | density (mA cm <sup>-2</sup> ) | (mV/dec) | time (h)  |         |           |
|           |                                |          |           |         |           |

| Fe <sub>9</sub> S <sub>10</sub> /Fe-MOF/NF-2                            | 202/10, 225/50 | 34   | 90  | NF | This work |
|---|----------------|------|-----|----|-----------|
| Ni <sub>3</sub> S <sub>2</sub> /MIL-53(Fe)                              | 214            | 33.8 | 23  | NF | 1         |
| CoMoO <sub>4</sub> /Fe-MOF  | 238            | 49   | 30  | NF | 2         |
| NiMoO <sub>4</sub> /Ni-MOF/NF   | 218            | 68   | 20  | NF | 3         |
| Co <sub>3</sub> O <sub>4</sub> @Co-MOF                                  | 277            | 79   | -   | GC | 4         |
| NM50-Ni <sub>3</sub> S <sub>4</sub> /NF                                 | 257            | 67   | 300 | NF | 5         |
| S-NiFe(CN)5NO-500   | 274            | 54   | 30  | GC | 6         |
| Fe <sub>0.5</sub> Ni <sub>0.5</sub> Co <sub>2</sub> -S/NF               | 251/50         | 73   | 25  | NF | 7         |
| NiCo <sub>2</sub> S <sub>4</sub> /CC                                    | 290/50         | 139  | 10  | CC | 8         |
| CoS <sub>2</sub> -MoS <sub>2</sub> HNAs/Ti                              | 266            | 104  | 24  | TF | 9         |
| Cr-NiS <sub>2</sub> /C@NF   | 207            | 43   | 80  | NF | 10        |
| PBA-SA  | 261            | 38   | 15  | RD | 11        |
| FeNi <sub>2</sub> -400-S  | 214            | 80   | 12  | GC | 12        |
| Co <sub>9</sub> S <sub>8</sub> /Zn <sub>0.8</sub> Co <sub>0.2</sub> S@C | 292            | 52   | 5.5 | GC | 13        |
| NiCo <sub>2</sub> S <sub>4</sub> /FeOOH                                 | 200            | 71   | 20  | CC | 14        |

- NF: Ni Foam
- CC: Carbon Cloth
- GC: Glassy Carbon
- TF: Ti Foil

**RD:** Rotating Disk

Table S2 The TON and TOF of catalysts

| Catalysts                                    | TON   | <b>TOF (s<sup>-1</sup>)</b> |
|--|-------|-----------------------------|
| Fe <sub>9</sub> S <sub>10</sub> /Fe-MOF/NF-2 | 122.4 | 0.034                       |
| Fe-MOF/NF                                    | 3.6   | 0.0011                      |
| Fe <sub>9</sub> S <sub>10</sub> /NF          | 25.2  | 0.0072                      |

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