Electronic Supplementary Information

Experimental section

Materials: $Co(NO_3)_2 \cdot 6H_2O$, $Fe(NO_3)_3$, urea and $NaS_2 \cdot 9H_2O$ were purchased from Aladdin. Pt/C (20 wt% Pt on Vulcan XC-72R) Carbon cloth was purchased from Sigma-Aldrich. The ultrapure water used throughout all experiments through a Millipore system. All chemicals were used as received without further purification.

Preparation of hydroxide precursor: In a typical synthesis process, 1 mmol $Co(NO_3)_2 \cdot 6H_2O$, 0.1 mmol $Fe(NO_3)_3$ and 5 mmol urea were dissolved in 35 mL deionized water under vigorous stirring for 30 min. Then the solution was transferred to a 60 mL Teflonlined stainless steel autoclave with a piece of carbon cloth (3 cm × 4 cm). Then autoclave was sealed and maintained at 120 °C for 12 h in an oven. After the autoclave cooled down 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. Then the hydroxide precursor was obtained. The hydroxide precursor of CoS_2/CC were made under the same conditions without using $Fe(NO_3)_3$.

Preparation of Fe-CoS_2/CC and CoS_2/CC: 8 mmol $NaS_2 \cdot 9H_2O$ were dissolved in 40 mL deionized water under vigorous stirring for 30 min. Then the solution was transferred to a 60 mL Teflonlined stainless steel autoclave with the hydroxide precursor. Then autoclave was sealed and maintained at 120 °C for 4 h in an oven. After the autoclave cooled down to room temperature, the sulfide was taken out and thoroughly washed with deionized water and ethanol several times, then dried at 60 °C for 6 h in air. **CoS_2/CC** was made under identical conditions.

Preparation of RuO_2 electrode: RuO_2 was prepared according to previous publication. Briefly, 2.61 g of $RuCl_3 \cdot 3H_2O$ and 30.0 mL KOH (1.0 M) were added into 100 mL distilled water and stirred for 45 min at 100 ^oC. 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 ^oC overnight and then annealed at 300 ^oC in air atmosphere for 3 h. For a typical synthesis of RuO_2/CC electrode, 50 mg RuO_2 was dispersed in 1 mL ethane/water (v:v = 1:1)

solution with sonication for 30 min. Then 10 μL catalytic inks were dropped on CC (0.5*0.5 cm), and dried at 80 ^{o}C for 4 h.

Characterization: The X-ray diffraction (XRD) patterns were obtained from a LabX XRD-6100 X-ray diffractometer with Cu Kα radiation (40kV, 30mA) of wavelength 0.154 nm (SHIMADZU, 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 ESCALABMK II x-ray photoelectron spectrometer using Mg as the exciting source.

Electrochemical measurement: The electrochemical measurements were performed on a CHI 660E electrochemical workstation (Chenhua, Shanghai). A three-electrode system was used in the experiment: a mercuric oxide electrode (Hg/HgO) was used as the reference electrode, a graphite rod was used as the counter electrode and the as-prepared Fe-CoS_2/CC was used as the working electrode. All tests were carried out at room temperature. The potentials reported in this work were calibrated to RHE other than especially explained, using the following equation: E (RHE) = E (Hg/HgO) + (0.098 + 0.059 × pH) V.

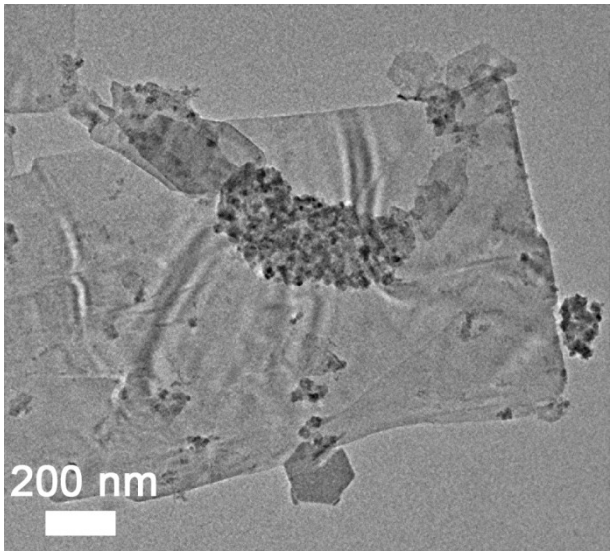

Fig. S1. TEM image of Fe-CoS_2 nanosheet.

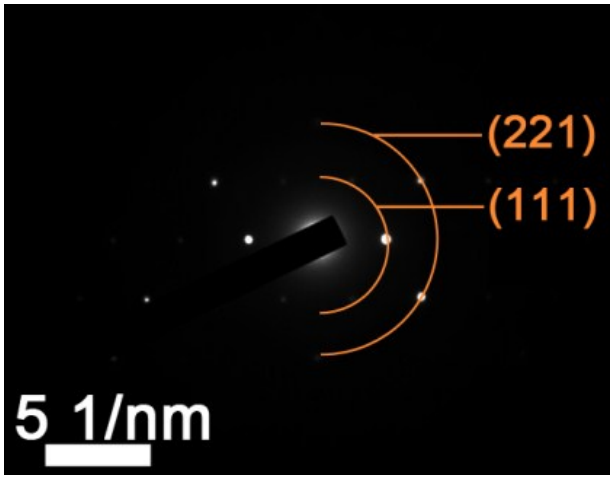

Fig. S2. SAED pattern taken from Fe-CoS_2 nanosheet.

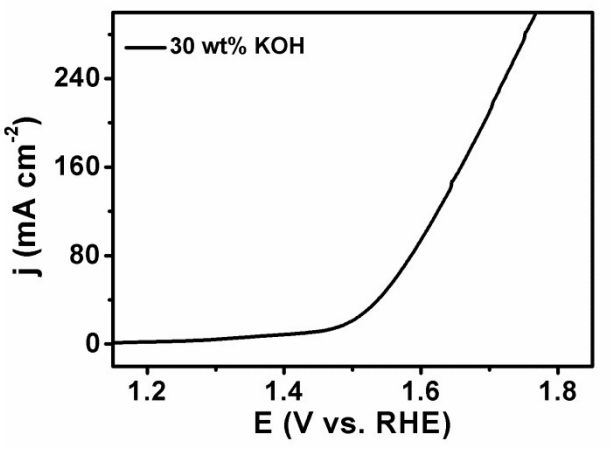

Fig. S3. LSV curves for Fe-CoS_2/CC in 30 wt% KOH.

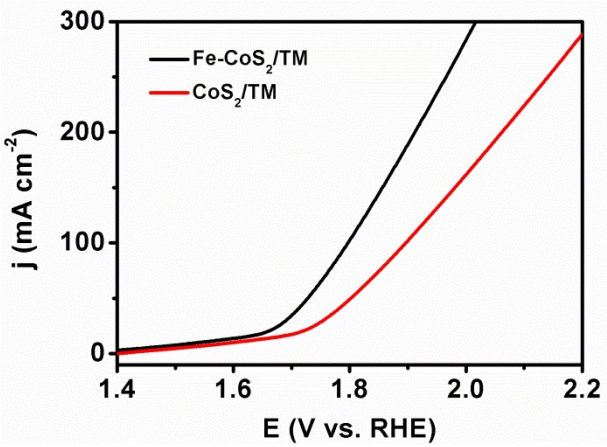

Fig. S4. LSV curves of CoS_2/TM and Fe-CoS_2/TM with a scan rate of 5 mV s^{-1} in 1.0 M KOH.

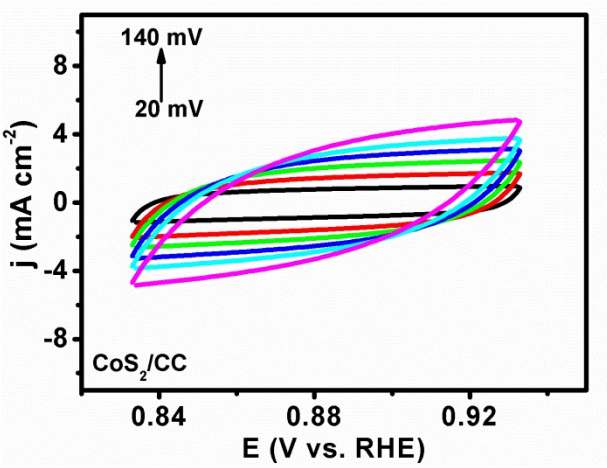

Fig. S5. CVs of CoS_2/CC with various scan rates.

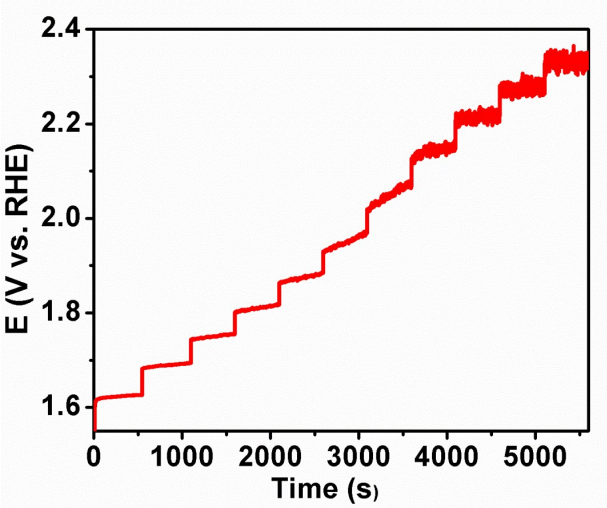

Fig. S6. Multi-current process of Fe-CoS_2/CC without iR correction. The current density started at 20 mA cm^{-2} and ended at 200 mA cm^{-2}, with an increment of 20 mA cm^{-2} per 500 s.

Table S1. The comparison of OER performance for Fe-CoS_2/CC with other non-noble-metal electrocatalysts at 1.0 M KOH.

Catalyst	j (mA cm^{-2})	η (mV)	Reference
Fe-CoS_2/CC	10	302	This work
CoS_2//CC	10	387	
Co_3S_4-L	10	360	1
Co–FeS_2/CoS_2	10	278	2
S–CoO	10	360	3
N, S–codoped graphene supported CoS_2	10	393	4
$Fe_{0.5}Co_{0.5}Se_2$ spheres	10	290	5
Co_3O_4/N-doped porous carbon	10	330	6
Co_3O_4@GF_O_3	10	450	7
Co_3O_4 nanocubes/graphene	10	400	8
$NiCo_2O_4$	10	350	9
CoS_2 hollow nanospheres	10	290	10

References

1 M. Zhu, Z. Zhang, H. Zhang, H. Zhang, X. Zhang, L. Zhang and S. Wang, *J. colloid interf. sci.*, 2018, **509**, 522-528.

2 K. Wang, W. Guo, S. Yan, H. Song and Y. Shi, *RSC Adv.,* 2018, **8**, 28684-28691.

3 X. Kuang, B. Kang, Z. Wang, L. Gao, C. Guo, J. Lee, X. Sun and Q. Wei, *Chem.-Eur. J.,* 2018, **24**, 17288-17292.

4 B. Chen, Z. Jiang, L. Zhou, B. Deng, Z.-J. Jiang, J. Huang, M. Liu, *J. Power Source,* 2018, **389**, 178-187.

5 J. Q. Chi, X. Shang, W. K. Gao, B. Dong, K. L. Yan, X. Li, Y. R. Liu, Y. M. Chai and C. G. Liu, *Int. J. Hydrogen Energy,* 2017, **42**, 15189-15195.

6 G. Zhuang, Y. Gao, X. Zhou, X. Tao, J. Luo, Y. Gao, Y. Yan, P. Gao, X. Zhong and J. Wang, *Chem. Eng. J.* 2017, **330**, 1255-1264.

7 M. P. Araújo, M. Nunes, I. M. Rocha, M. F. R. Pereira, C. Freire, *Chemistry Select,* 2018, **3**, 10064-10076.

8 P. Zhang, X. Han, H. Hu, J. Gui, M. Li and J. Qiu, *Catal. Commun.* 2017, **88**, 81-84.

9 C. Broicher, F. Zeng, J. Artz, H. Hartmann, A. Besmehn, S. Palkovits and R. Palkovits, *Chemcatchem,* 2019, **11**, 412-416.

10 X. Ma, W. Zhang, Y. Deng, C. Zhong, W. Hu and X. Han, *Nanoscale,* 2018, **10**, 4816-4824.