# **Electronic Supporting Information (ESI)**

# Fabrication of amorphous CoMoS<sub>4</sub> as a bifunctional electrocatalyst for water splitting in strong alkaline condition

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Fig. S1 X-Ray diffraction (XRD) pattern of the as-prepared amorphous CoMoS<sub>4</sub>.



Fig. S2 Large-size SEM image for the as-prepared amorphous CoMoS<sub>4</sub> deposited on a Cu foil.



**Fig. S3** EDX spectrum of the as-synthesized amorphous CoMoS<sub>4</sub>. The signal of Cu arises from the TEM grid made of elemental Cu.

	Atomic ratio of	Weight ratio of	Weight
Analysis	Co/ Mo / S	Co / Mo /S	percentage of C
			(%)
ICP-AES	1: 0.87: 3.60	1:1.42:1.95	
XPS	1: 1.08: 3.40	1:1.75:1.85	11.2
EDX	1: 1.03: 3.53	1:1.68:1.92	
EA			10.6

 Table. S1
 Elemental mole ratio in the sample determined by different analytical methods.



Fig. S4 Raman spectrum of the as-synthesized amorphous CoMoS<sub>4</sub>.



**Fig. S5** X-Ray diffraction (XRD) pattern for amorphous  $CoMoS_4$  after it was immersed into 1.0 M KOH. The observation of (001) diffraction plane of hexagonal  $\beta$ -Co(OH)<sub>2</sub> suggests that the material contains  $CoMoS_4/\beta$ -Co(OH)<sub>2</sub> composites as active catalyst.



Fig. S6 SEM images of (a) carbon cloth, (b) the surface of the CoMoS<sub>4</sub>/CC electrode.



Fig. S7 Polarization curves at different loading weight for hydrogen evolution reaction (HER).



Fig. S8 Polarization curves at different loading weight for oxygen evolution reaction (OER).



Fig. S9 First and 2000th cyclic voltammograms of the CoMoS<sub>4</sub>/CC electrode with CoMoS<sub>4</sub>/ $\beta$ -Co(OH)<sub>2</sub> composites for HER



**Fig. S10** XRD patterns, SEM images and TEM images of the as-synthesized CoS (a, c, e) and  $MoS_2$  (b, d, f), respectively.



Fig. S11 XRD pattern, SEM and TEM images of the as-synthesized  $\beta$ -Co(OH)<sub>2</sub> according to Ref. 27 in maintext.



Fig. S12 Polarization curves of the CoMoS<sub>4</sub>/CC electrode with CoMoS<sub>4</sub>/ $\beta$ -Co(OH)<sub>2</sub> composites, CoS, MoS<sub>2</sub> and  $\beta$ -Co(OH)<sub>2</sub> at the same mass loading in 1.0 M KOH for HER.



Fig. S13 First and 2000th cyclic voltammograms of the CoMoS<sub>4</sub>/CC electrode with CoMoS<sub>4</sub>/ $\beta$ -Co(OH)<sub>2</sub> composites for OER.



**Fig. S14** (a) Polarization curves of the CoMoS<sub>4</sub>/CC electrode with CoMoS<sub>4</sub>/ $\beta$ -Co(OH)<sub>2</sub> composites, CoS, MoS<sub>2</sub> and  $\beta$ -Co(OH)<sub>2</sub> at the same mass loading in 1.0 M KOH for OER, and (b) stability of CoS with an initial polarization curve and after 500 cycles in 1.0 M KOH.



**Fig. S15** (a) HER and (b) OER liner sweep voltammetry curves of amorphous  $CoMoS_4$  with hydrothermal process (blue) comparing to the ones of direct precipitation sample (red).



**Fig. S16**. Nyquist plots of various catalysts/CC electrodes at an overpotential of 200 mV. Z' is real impedance and Z'' is imaginary impedance.

### **TOF** calculation.

The number of active sites (n) is examined using CVs with 1 M KOH at a scan rate of 50 mV s<sup>-1</sup>. When the number of voltammetric charges (Q) is obtained after deduction of the blank value, n (mol) can be calculated with the following equation:

n= Q/2F

where F is Faraday constant (96485 C mol<sup>-1</sup>). For the sample of CoMoS<sub>4</sub> with hydrothermal process (black), Q is  $2.413 \times 10^{-3}$  C (obtained from Fig. S17), n (mol) =  $2.413 \times 10^{-3}$ / (2×96485) mol=  $1.25 \times 10^{-8}$  mol. And for the sample of direct precipitation (red), Q is  $1.299 \times 10^{-3}$  C, n (mol) =  $1.299 \times 10^{-3}$ / (2×96485) mol=  $6.73 \times 10^{-9}$  mol.



**Fig. S17** Cyclic voltammograms of blank CC (blue), amorphous  $CoMoS_4$  with hydrothermal process (black) and direct precipitation (red) recorded in 1.0 M KOH; scan rate: 50 mV s<sup>-1</sup>

When the number of active sites is obtained, the per-site turnover frequencies (in s<sup>-1</sup>) were calculated with the following equation:

TOF= I/(2Fn)

I — Current (in A) during the linear sweep measurement.

F — Faraday constant (96485 C mol<sup>-1</sup>).

n —Number of active sites (in mol).

The factor 1/2 in the equation represents that two electrons are required to form one hydrogen molecule from two protons ( $2H^+ + 2e^{-1} = H_2$ ).



**Fig. S18** Calculated turnover frequencies for amorphous CoMoS<sub>4</sub> with hydrothermal process (black) and direct precipitation (red).

# **Current calibration.**

The capacitances of the double layer at the solid–liquid interface of electrodes with different mass loading were measured by cyclic voltammograms (CVs) collected in the region of 0. 0-0.10 V at scan rate of 5 mV/s, where the current response should be only due to the charging of the double layer (Fig. S19). All the LSV curves were calibrated with capacitive current reduction.



**Fig. S19** CVs for CoMoS<sub>4</sub>/CC electrode with CoMoS<sub>4</sub>/ $\beta$ -Co(OH)<sub>2</sub> composites with different mass loading in 1.0 M KOH solution.



**Fig. S20** SEM images of the CoMoS<sub>4</sub>/CC electrode with CoMoS<sub>4</sub>/ $\beta$ -Co(OH)<sub>2</sub> composites after HER cycles (a) and after OER cycles (b), respectively.



**Fig. S21** (a) XRD patterns of the CoMoS<sub>4</sub>/ $\beta$ -Co(OH)<sub>2</sub> composites before cycles (blue), after HER cycles (green) and after OER cycles (red) with standard JCPDS No. 74-1057, (b) HRTEM images of CoMoS<sub>4</sub>/ $\beta$ -Co(OH)<sub>2</sub> composites after 2000 OER cycles and (c-f) HAADF-STEM image and STEM-EDS elemental mapping images of Co, Mo and S after 2000 OER cycles.

The samples after cycles were obtained by loading amorphous  $CoMoS_4$  onto Ni foam (Ni foam was applied as substrate here because the diffraction peaks of carbon cloth is so strong that the rest peaks would not be easily showed), followed by running 2000 CV cycles and washing with absolute ethanol, then the active material was transferred into ethanol under ultrasound for further characterization.

# Loss of active material during long time electrolysis.

An electrolyzer with carbon cloth (1×1 cm<sup>2</sup>) loading 1.0 mg cm<sup>-2</sup> amorphous CoMoS<sub>4</sub> as both anode and cathode, 1.0 M KOH as electrolyte, worked at current density of 10 mA cm<sup>-2</sup> for 12 hours electrolysis, and Co, Mo, S can be detected in the electrolyte after digesting by HNO<sub>3</sub>. The results showed ~7% total mass loss for the electrolyzer.

		,	,
elements	Со	Мо	S
Total amount/µg	5.7	61.5	75.1

Table. S2 Elemental content detected in the electrolyte after 12 hours electrolysis.



**Fig. S22** Comparison of XPS spectra between the  $CoMoS_4/CC$  electrode with  $CoMoS_4/\beta$ - $Co(OH)_2$  composites before cycles (dark), after HER (red) and after OER (blue): (a) survey spectra, (b) Mo 3d, (c) Co 2p, and (d) S 2p spectra.