## **Supporting Information**

## Three-dimension Cross-linked Co-MoS<sub>2</sub> Catalyst on Carbon

## **Cloth for Efficient Hydrogen Evolution Reaction**

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\*Corresponding author. *E-mail address: gaohong65cn@126.com*  **Chemicals.** Thiourea (CH<sub>4</sub>N<sub>2</sub>S, Aladdin Chemistry Co., Ltd, AR), cobalt nitrate hexahydrate (Co(NO<sub>3</sub>)<sub>2</sub>·6H<sub>2</sub>O, Shanghai Macklin Biochemical Technology Co., Ltd, AR), urea (CH<sub>4</sub>N<sub>2</sub>O, Aladdin Chemistry Co., Ltd, AR), ammonium molybdate tetrahydrate ((NH<sub>4</sub>)<sub>6</sub>Mo<sub>7</sub>O<sub>24</sub>·4H<sub>2</sub>O, Aladdin Chemistry Co., Ltd, ACS) and potassium hydroxide (KOH, Shanghai Macklin Biochemical Technology Co., Ltd, AR) were used directly without any further purification. The first-grade deionized (DI) water used in all experiments.

Synthesis of  $MoS_2/CC$ . For comparative studies, pure  $MoS_2$  was synthesized to attain insights into the relationship between architecture and catalytic activity. Except without adding CoCH precursor, pure  $MoS_2$  was prepared using a process similar to Co-MoS<sub>2</sub>-4/CC.



Figure S1. (a-c) High magnificent SEM images of CoCH-1/CC, CoCH-

4/CC, and CoCH-8/CC, respectively.



Figure S2. (a-b) Low and high magnificent SEM images of bare MoS<sub>2</sub>.



**Figure S3**. (a-c) EDS data of Co-MoS<sub>2</sub>-1, Co-MoS<sub>2</sub>-4, and Co-MoS<sub>2</sub>-8, respectively; insets show the corresponding stoichiometric ratios of S, Mo, and Co.



**Figure S4.** XRD patterns of the 3DSC Co-MoS<sub>2</sub>-4/CC nanostructure

synthesized at different reduction temperatures.



**Figure S5.** X-ray photoelectron spectroscopy (XPS) results of MoS<sub>2</sub>/CC: (a) survey, (b) Mo 3d, and (c) S 2p regions.



**Figure S6.** Comparison of HER activities of samples synthesized at changing annealing temperatures in 1 M KOH electrolyte. (a) The HER polarization curves and (b) Tafel plots for a series of Co-MoS<sub>2</sub>-200/CC, Co-MoS<sub>2</sub>-350/CC, and Co-MoS<sub>2</sub>-500/CC electrocatalysts in 1 M KOH electrolyte. (c) Plots of  $\Delta j/2$  versus scan rates. (d) Nyquist plots at an overpotential of 200 mV for electrodes.



**Figure S7.** (a) Double layer capacitance estimated from the linear slope between  $\Delta j$  (=  $j_a - j_c$ ) and scan rates. (b-f) CV curves at different scan rates for all synthesized catalysts.



Figure S8. The final catalytic activity of a series of  $Co-MoS_2/CC$  electrodes after excluding the electrochemical surface area effects. Electrolyte: 1M KOH.



**Figure S9.** Potential-dependent TOF values of a series of  $Co-MoS_2/CC$  electrodes.

Catalyst	$\eta_{10}(mV)$	Reference
Co-MoS <sub>2</sub> -4/CC	40	This Work
$MoS_2/Ni_2O_3H$	84	Small, 2020, 16, 2002212
Co <sub>9</sub> S <sub>8</sub> -MoS <sub>2</sub> /NF	110	Adv. Funct. Mater., 30, 2020, 2002536
Ni/M-MoS <sub>2</sub>	145	ChemElectroChem, 2020, 7, 3606-3615
$1T-MoS_2/CoS_2$	71	Small, 2020, 16, 2002850
MoS <sub>2</sub> /CoS <sub>2</sub> NTs	85	J. Mater. Chem., A, 2019, 7, 13339-13346
1T-MoS <sub>2</sub> QS/Ni(OH) <sub>2</sub>	57	Adv. Funct. Mater., 30, 2020, 2000551
MoS <sub>2</sub> /FNS/FeNi	120	Adv. Mater., 30, 2018, 1803151
Co-Ni <sub>3</sub> S <sub>2</sub> -MoS <sub>2</sub> /CA	89	Small, 2021, 17, 2006730
$Co_3O_4/MoS_2$	205	Appl. Catal. B-Environ., 2019, 248, 202-210
Co <sub>4</sub> S <sub>3</sub> /MoC-NSC-2	82.5	Appl. Catal. B-Environ., 2020, 260, 118197
N-NiS/NiS <sub>2</sub>	185	Chem. Eng. J., 2020, 397, 125507
$Ni_2P/Ni_3S_2$	80	Nano Energy, 2018, 51, 26-36

Table S1. Comparison of HER activity of Co-MoS<sub>2</sub>-4/CC catalyst with other reported electrocatalysts in 1 M KOH electrolyte.