Supporting Information

## A Bulky and Flexible Electrocatalyst for Highly Efficient Hydrogen Evolution based on the Growth of MoS<sub>2</sub> Nanoparticles on Carbon Nanofibers Foam

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Figure S1. SEM image of the MoS<sub>2</sub>/CNFs hybrid by replacing the DMF with water as solvent.



Figure S2. Polarization curve of  $MoS_2$  nanoparticles physically mixed with carbon black (7:3 w/w) and  $MoS_2/CNFs$  hybrid by replacing the DMF with water as the solvent.



Figure S3. (a) XRD patterns of  $MoS_2$  nanoparticles and  $MoS_2/CNFs$  composite. (b) Raman spectroscopy of  $MoS_2/CNFs$  composite.



Figure S4. TG curves of the  $MoS_2/CNFs$ , the bare  $MoS_2$  nanoparticles and the bare CNFs.



Figure S5. The compression-recovery processes of  $MoS_2/CNFs$ .



Figure S6. XRD pattern of (a) the as-prepared  $MoS_2/CNFs$  and (b) the  $MoS_2/CNFs$  composite after annealed at 800 °C for 2 h.



Figure S7. The polarization curves and the Tafel slope (inset) of as-prepared  $MoS_2/CNFs$  and  $MoS_2/CNFs$  annealed at 800 °C.



Figure S8. The turnover frequencies and the cyclic voltammograms (inset) of asprepared MoS<sub>2</sub>/CNFs and MoS<sub>2</sub>/CNFs annealed at 800 °C.

## Assessment of turnover frequency (TOF)

The turnover frequencies (in s<sup>-1</sup>) were calculated with the following equation:

$$TOF = \frac{i \ 1}{Fn2}$$

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

F- Faraday constant (in 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  $(2H^+ + 2e^{-1} = H_2)$ .



Figure S9. The durability test of the 3D MoS<sub>2</sub>/CNFs electrode.



Figure S10. The GC curve to detect gas production at the potential of -0.35 V (vs. RHE).

Table S1. Comparion of HER performance in acidic media for  $MoS_2/CNFs$  with other  $MoS_2$ -based HER electrocatalysts.

Catalyst	Tafel slope (mV/dec)	Current density (i)(m A cm <sup>-2</sup> )	Overpotential at the corresponding	Referenc e
MoS control	185	(I)(IIIA CIII ) 71	200	(6)
sponge	105	/1	200	(0)
double-gyriod	50	2	190	(7)
MoS <sub>2</sub> /FTO				
MoO <sub>3</sub> -MoS <sub>2</sub> /FTO	50-60	10	300	(8)
MoS <sub>2</sub> /mesoporous	42	100	200	(18)
graphene				
MoS <sub>x</sub> /graphene -				
protected 3D Ni	42.8	45	200	(19)
foams				
MoS <sub>2</sub> nanoflower	95	10	250	(20)
/rGO				
MoS <sub>2</sub> /CNT	44.6	14	200	(21)
MoS <sub>2</sub> /RGO	41	10	150	(23)
MoS <sub>2</sub> /CNF fiber	45	25	120	(25)
mats				
MoS <sub>2</sub> /CNFs	44	16	230	This work

## Supplementary Movies

**Movie S1**. This movie shows  $MoS_2/CNFs$  foam operated from +0.12 V to -0.23 V *vs*. RHE to drive HER.

**Movie S2**. This movie shows pristine CNF operated from +0.12 V to -0.23 V vs. RHE to drive HER.