

Supporting Information(SI)

Amorphous MoS_x modified $\text{g-C}_3\text{N}_4$ composite for efficient photocatalytic hydrogen evolution

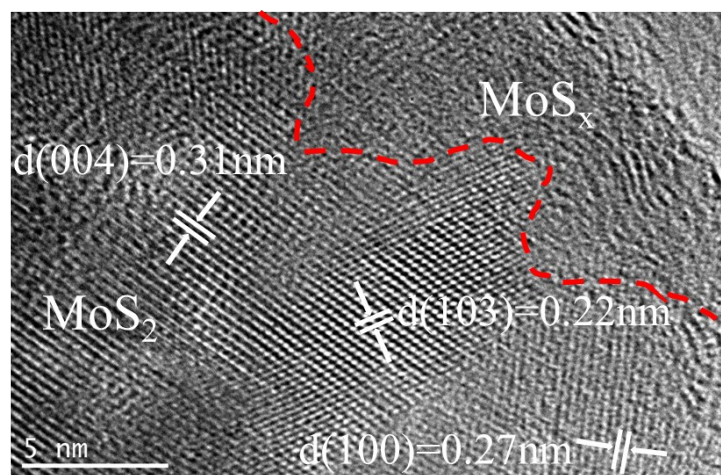
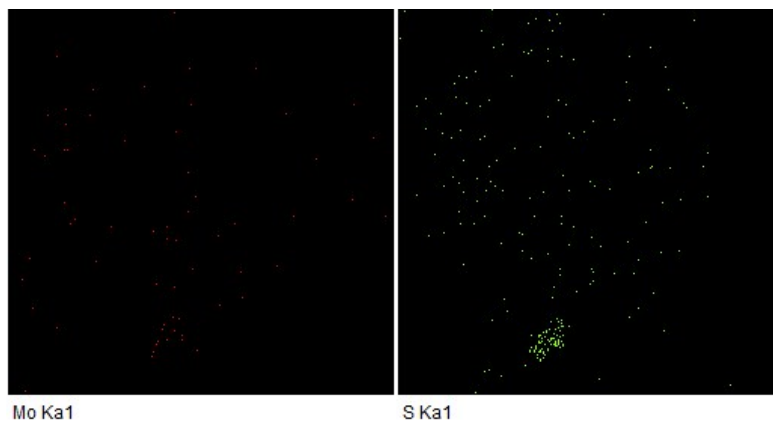
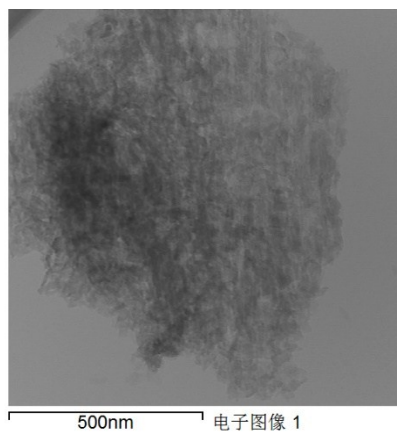


Fig. S1 TEM image of $\text{MoS}_x/\text{MoS}_2$



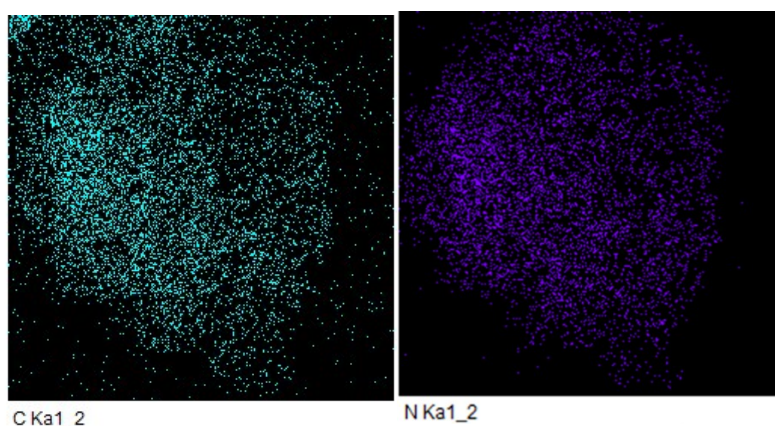


Fig. S2. Element mapping of UCN/MoS_x

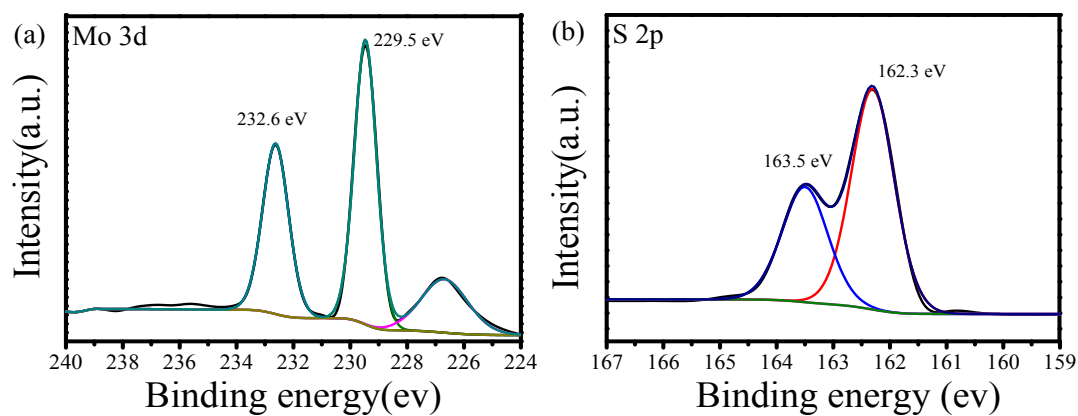


Fig. S3. XPS spectra of (a) Mo 3d and (b) S 2p

Table S1 Summary of photocatalytic activity of g-C₃N₄/MoS₂ composite

Catalyst	Light source	Sacrificial reagent	H ₂ yield ($\mu\text{mol h}^{-1}$ g^{-1})	Reaction time	Reference
20 mg MoS _x /UCN	300 W Xe lamp, $\lambda > 400$ nm	10% triethanolamine in water	1586	5h	This work
20 mg MoS ₂ /mpg-CN	300 W Xe lamp, $\lambda > 420$ nm	10% triethanolamine in water	1030	4h	[1]
100 mg MoS ₂ /C ₃ N ₄ (1 wt% Pt)	300 W Xe lamp, $\lambda > 400$ nm	25% methano in water	231	6h	[2]

10 mg g-C ₃ N ₄ /MoS ₂	300 W Xe lamp, $\lambda > 400$ nm (0.6w cm ⁻²)	10% triethanolamine in water	252	6h	[3]
50 mg a-MoS _x /g-C ₃ N ₄	3w LEDs, $\lambda > 420$ nm	10% lactic acid in water	273.1	3h	[4]
20 mg MoS ₂ /HCNS	300 W Xe lamp, $\lambda > 420$ nm	10% lactic acid in water	1340	4h	[5]
50 mg MoS ₂ /CN-Py	300 W Xe lamp, $\lambda > 420$ nm	10% triethanolamine in water	500	3h	[6]
10 mg SSCN@MoS ₂	300 W Xe lamp, $\lambda > 420$ nm	15% triethanolamine in water	91	5h	[7]
50 mg 1T-MoS ₂ @g-C ₃ N ₄	300 W Xe lamp, without filter	10% lactic acid in water	948	5h	[8]
10 mg 1T-MoS ₂ /O-g-C ₃ N ₄	300 W Xe lamp, $\lambda > 400$ nm	10% triethanolamine in water	1841	5h	[9]
10 mg MoS ₂ /CNNS	450 W Xe lamp, AM 1.5G filter (100 mW cm ⁻²)	10% lactic acid in water	1420	5h	[10]
50 mg MSQD-CN (2 wt% Pt)	300W Xe lamp, $\lambda > 420$ nm (320 mW/cm ²)	25% methanol in water	557	4h	[11]
10 mg MoO ₃ /MoS ₂ /g-C ₃ N ₄ (3 wt% Pt)	300W Xe lamp, $\lambda > 420$ nm	10% triethanolamine in water	513	4h	[12]
3 mg g-C ₃ N ₄ /MoS ₂ -nanodot	Xe lamp, $\lambda > 400$ nm (350 mW/cm ²)	20% lactic acid in water	660	3h	[13]

Table S2 BET surface area and pore volume of UCN and MoS_x/UCN

Samples	UCN	MoS _x /UCN
SA/m ² ·g ⁻¹	89.29	72.56
Pore volume (m ³ /g)	0.31	0.18

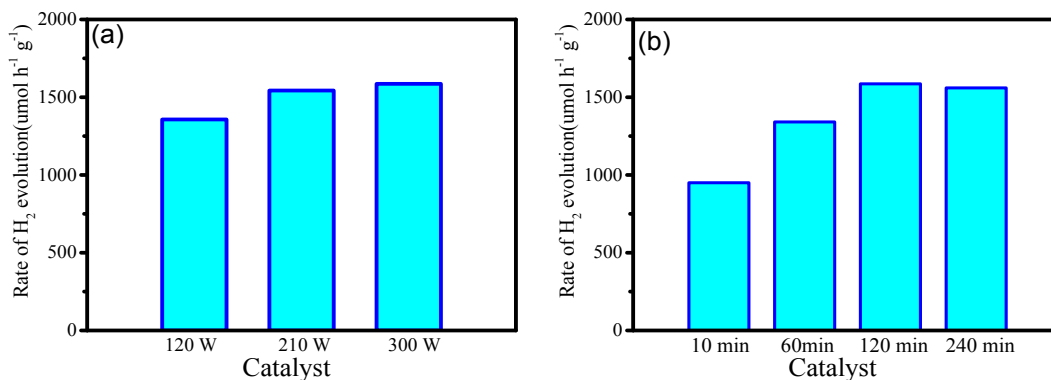


Fig. S4. The photocatalytic H₂ evolution rate of samples: (a) MoS_x/UCN (7 wt%) prepared with different ultrasonic power (b) MoS_x/UCN (7 wt%) prepared with different ultrasonic time

Both the ultrasonic power and time can affect the degree of exfoliation. We synthesized samples with different time and power to research their effects on the performance of hydrogen evolution. In the first group, MoS_x/UCN (7 wt%) was prepared with different ultrasonic power (120 W, 210 W, 300 W), and ultrasonic time was fixed at 2h, the results are shown in Fig. S4(a). In the second group, MoS_x/UCN (7 wt%) was prepared with different ultrasonic time (10 min, 60 min, 120 min, 240min), and ultrasonic power was fixed at 300 W, the results are shown in Fig. S4 (b). It can be seen that proper increase of time and power is beneficial to the improvement of performance, but excess ultrasonic time and ultrasonic power will not be beneficial to the performance.

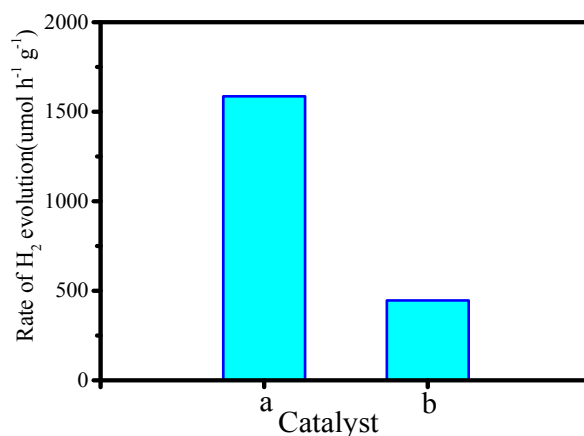


Fig. S5. The photocatalytic H₂ evolution rate of MoS_x/UCN(7 wt%) under aqueous solution containing different sacrificial reagent: (a) triethanolamine (b) lactic acid

In the photocatalytic hydrogen evolution system, the sacrificial agent is a good electron donor, which can effectively improve the quantum efficiency of photocatalysis. The choice of sacrificial agent will greatly affect the photocatalytic results. Currently, researchers prefer to choose triethanolamine and lactic acid as sacrificial agents in the photocatalytic hydrogen evolution system of g-C₃N₄. We found that MoS_x/UCN(7 wt%) show better photocatalytic performance under aqueous solution containing triethanolamine than under aqueous solution containing lactic acid (see Fig. S5). Therefore,

we use triethanolamine as sacrificial reagent in this study.

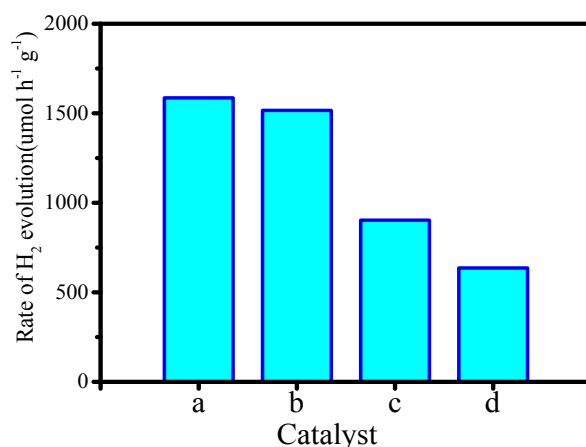


Fig. S6. The photocatalytic H₂ evolution rate of samples:(a) MoS_x/UCN(7 wt%) (b) 1-MoS_x/UCN(7 wt%) (c) 2-MoS_x/UCN(7 wt%) (d) 3-MoS_x/UCN(7 wt%)

In order to research the hydrothermal temperature and reaction precursor of influence on the formation of molybdenum sulfide, we prepared MoS_x samples under different conditions. On the basis of preparing MoS_x, we prepared 1-MoS_x by setting the hydrothermal temperature at 200 °C, prepared 2-MoS_x by reducing the mole ratio of CH₃CSNH₂ : Na₂MoO₄·H₂O to 3 : 1, prepared 3-MoS_x by removing H₄SiO₄(W₃O₉)₄ from the precursor. And the hydrogen evolution performance of samples was shown in figure S6. The results suggest that change of the hydrothermal temperature shows little influence on the co-catalytic performance of MoS_x, ratio of CH₃CSNH₂ : Na₂MoO₄·H₂O and the presence of H₄SiO₄(W₃O₉)₄ shows great influence on the co-catalytic performance of MoS_x. High ratio of CH₃CSNH₂ to Na₂MoO₄·H₂O and presence of H₄SiO₄(W₃O₉)₄ are beneficial to the co-catalytic performance of MoS_x. By comparing XPS of various MoS_x (figure S7), we think that the change of hydrothermal temperature did not cause obvious change in structure of MoS_x. High ratio of CH₃CSNH₂ to Na₂MoO₄·H₂O and presence of H₄SiO₄(W₃O₉)₄ can increase the percent of S atoms with high binding energy in MoS_x, which is a reason why MoS_x and 1-MoS_x show better co-catalytic performance than 2-MoS_x and 3-MoS_x. The TEM images (figure S7) of various MoS_x are show in figure S8. It can find that the size of 4-MoS_x is bigger than MoS_x, 2-MoS_x and 3-MoS_x, which suggests that H₄SiO₄(W₃O₉)₄ can reduce the size of the MoS_x.

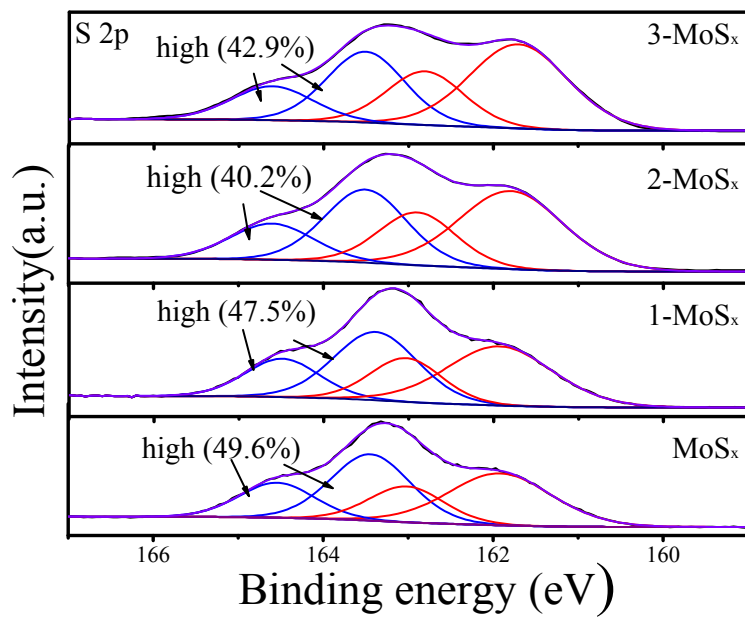


Fig.S7. High-resolution XPS spectra of S 2p

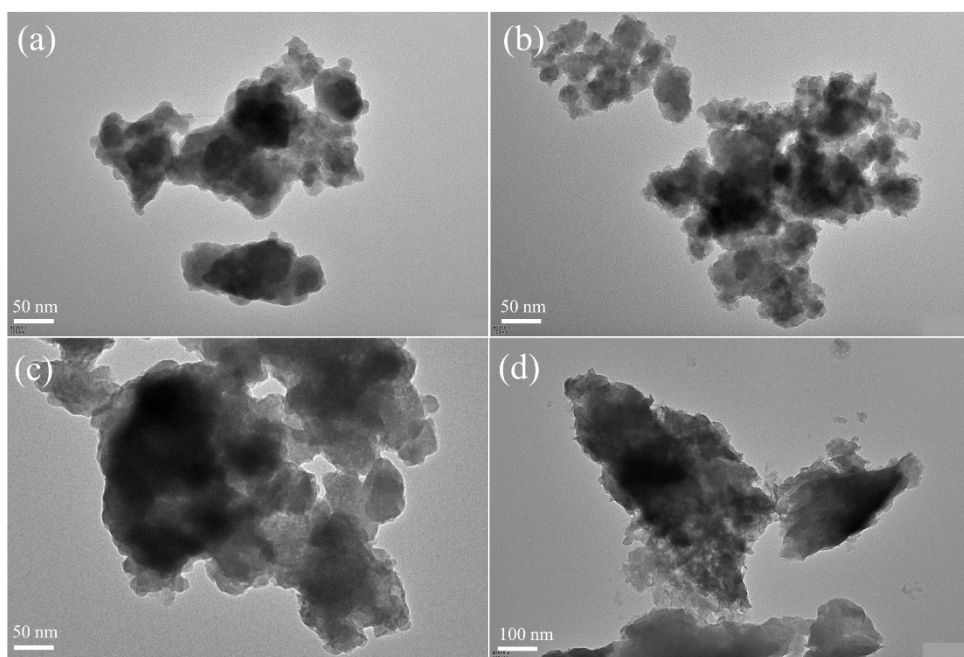


Fig.S8. TEM images: (a) MoS_x (b) 1-MoS_x (c) 2-MoS_x (d) 3-MoS_x.

References

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