Supporting Information

Amorphous MoS_x Nanosheets with Abundant Interlayer Dislocations for Enhanced Photolytic Hydrogen Evolution Reaction

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Received: 31 July 2025
Revised: 6 September 2025
Accepted: 8 September 2025
Published: 13 September 2025

Citation: Xu, X.;Wu, Z.; Hu,W.; Sun, N.; Li, Z.; Feng, Z.; Zhao, Y.; Wang, L. Amorphous MoS_x Nanosheets with Abundant Interlayer Dislocations for Enhanced Photolytic Hydrogen Evolution Reaction. *Catalysts* **2025**, *15*, 879. https://doi.org/10.3390/catal15090879.

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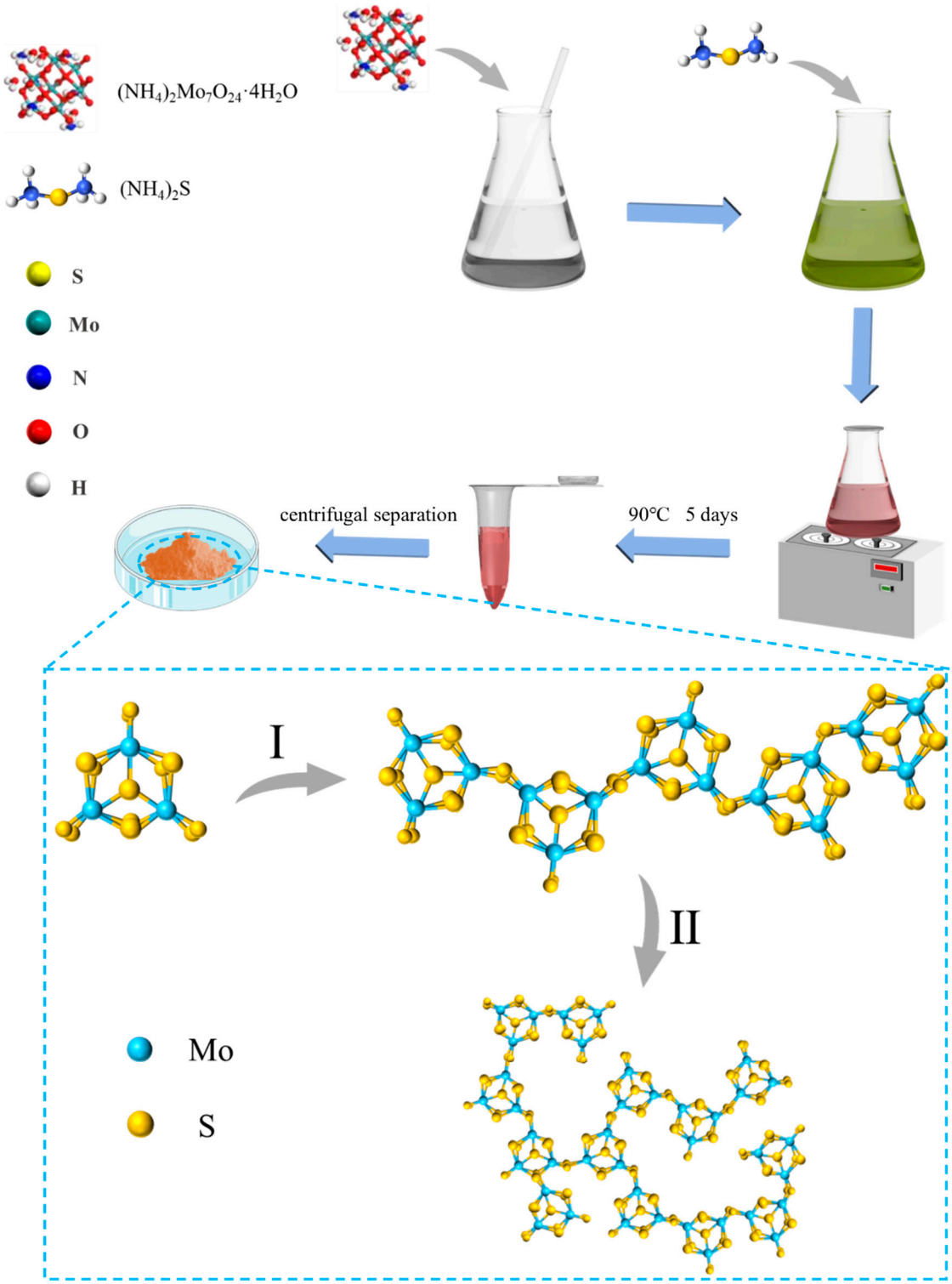

Figure S1. Schematic illustration of the synthesis process for amorphous MoS_x materials. The blue dashed lines indicate the potential linkage modes of $[Mo_3S_{13}]^{2-}$ clusters during the synthesis.

As indicated by the blue dashed boxes in Figure S1, $[Mo_3S_{13}]^{2-}$ clusters can assemble into diverse architectures via varying linkage modes during the thermal synthesis process. The interconnection of $[Mo_3S_{13}]^{2-}$ clusters generates chain-like structures (I), where molybdenum and

sulfur atoms are chemically bonded to form linearly aligned configurations. This process demonstrates how individual clusters can expand into larger aggregates through chemical bonding, ultimately developing extended one-dimensional chain structures. Furthermore, these chain structures can undergo additional interconnections to evolve into two-dimensional sheet-like architectures (II). In such configurations, partial clusters become bridged via sulfur atoms along planar directions, resulting in materials with substantially increased specific surface areas and homogeneous planar distributions. The formation of sheet-like structures is typically associated with relatively lower synthesis temperatures and higher solvent concentrations. The particular arrangement of clusters within these layered structures may endow the material with exceptional electrocatalytic performance for certain reactions, owing to optimized electronic configurations and exposed active sites.

Table S1 Comparison of HER performance of MoS_x catalytic system with various reported Mo-S-based catalysts

photocatalyst	Hydrogen evolution efficiency（mmol g^{-1} h^{-1}）	light source	bibliography
MoS_x	4.53	300 W Xenon lamp	This work
MoS_2/TiO_2	0.03	300 W Xenon lamp	[19]
Colloidal MoS_2	—	300 W Xenon lamp	[20]
CdSe-MoS_2	0.8	300 W Xenon lamp	[21]
MoS_2/g-C_3N_4	1.155	300 W Xenon lamp	[22]
MoS_2-CdS	1.472	300 W Xenon lamp	[23]
3 wt% $MoS_2/CuInS_2$	0.316	300 W Xenon lamp	[24]
5 wt% MSQD-CN	0.2229	300 W Xenon lamp	[25]
p-MoS_2/n-Rgo	0.16	300 W Xenon lamp	[26]
MoS_2	0.05	100 W Halogen lamp	[27]
NEG-MoS_2	0.83	100 W Halogen lamp	[27]
EG-MoS_2	0.54	100 W Halogen lamp	[27]
RGO-MoS_2	3	100 W Halogen lamp	[27]
EY-G/MoS_2	0.091	250 W Xenon lamp	[28]
TiO_2/MoS_2/graphene	0.1653	350 W Xenon lamp	[29]
Multiphasic MoS_2	750 nmol min^{-1}	sunlight	[30]

Reference

19. Kanda, S.; Akita, T.; Fujishima, M.; Tada, H. Facile Synthesis and Catalytic Activity of MoS_2/TiO_2 by a Photodeposition-Based Technique and Its Oxidized Derivative MoO_3/TiO_2 with a Unique Photochromism. *J. Colloid Interface Sci.* **2010**, *354*, 607–610. https://doi.org/10.1016/j.jcis.2010.11.007.
20. Zong, X.; Na, Y.; Wen, F.; Ma, G.; Yang, J.; Wang, D.; Ma, Y.; Wang, M.; Sun, L.; Li, C. Visible Light Driven H_2 Production in Molecular Systems Employing Colloidal MoS_2 Nanoparticles as Catalyst. *Chem. Commun.* **2009**, 4536–4538. https://doi.org/10.1039/b907307h.
21. Frame, F.A.; Osterloh, F.E. CdSe-MoS_2: A Quantum Size-Confined Photocatalyst for Hydrogen Evolution from Water under Visible Light. *J. Phys. Chem. C* **2010**, *114*, 10628–10633. https://doi.org/10.1021/jp101308e.
22. Yuan, Y.; Shen, Z.; Wu, S.; Su, Y.; Pei, L.; Ji, Z.; Ding, M.; Bai, W.; Chen, Y.; Yu, Z.; et al. Liquid Exfoliation of g-C_3N_4 Nanosheets to Construct 2D-2D MoS_2/g-C_3N_4 Photocatalyst for Enhanced Photocatalytic H_2 Production Activity. *Appl. Catalysis B Environ. Energy* **2019**, *246*, 120–128. https://doi.org/10.1016/j.apcatb.2019.01.043.
23. Chen, J.; Wu, X.; Yin, L.; Li, B.; Hong, X.; Fan, Z.; Chen, B.; Xue, C.; Zhang, H. One-pot Synthesis of CdS Nanocrystals Hybridized with Single-Layer Transition-Metal Dichalcogenide Nanosheets for Efficient Photocatalytic Hydrogen Evolution. *Angew. Chemie* **2014**, *127*, 1226–1230. https://doi.org/10.1002/ange.201410172.
24. Yuan, Y.; Chen, D.; Huang, Y.; Yu, Z.; Zhong, J.; Chen, T.; Tu, W.; Guan, Z.; Cao, D.; Zou, Z. MoS_2 Nanosheet-modified $CuInS_2$ Photocatalyst for Visible-light-driven Hydrogen Production from Water. *ChemSusChem* **2016**, *9*, 1003–1009. https://doi.org/10.1002/cssc.201600006.
25. Liu, Y.; Zhang, H.; Ke, J.; Zhang, J.; Tian, W.; Xu, X.; Duan, X.; Sun, H.; Tade, M.O.; Wang, S. 0D (MoS_2)/2D (g-C_3N_4) Het-erojunctions in Z-Scheme for Enhanced Photocatalytic and Electrochemical Hydrogen Evolution. *Appl. Catalysis B En Vironmental* **2018**, *228*, 64–74. https://doi.org/10.1016/j.apcatb.2018.01.067.
26. Meng, F.; Li, J.; Cushing, S.K.; Zhi, M.; Wu, N. Solar Hydrogen Generation by Nanoscale p–n Junction of p-Type Molybdenum Disulfide/n-Type Nitrogen-Doped Reduced Graphene Oxide. *J. Am. Chem. Soc.* **2013**, *135*, 10286–10289. https://doi.org/10.1021/ja404851s.
27. Maitra, U.; Gupta, U.; De, M.; Datta, R.; Govindaraj, A.; Rao, C. Highly Effective Visible-Light-Induced H_2 Generation by Single-Layer 1T-MoS_2 and a Nanocomposite of Few-Layer 2H-MoS_2 with Heavily Nitrogenated Graphene. *Angew. Chemie Int. Ed.* **2013**, *52*. https://doi.org/10.1002/anie.201306918.
28. Liu, X.; Zhao, L.; Lai, H.; Wei, Y.; Yang, G.; Yin, S.; Yi, Z. Graphene Decorated MoS_2 for Eosin Y-Sensitized Hydrogen Evolution from Water under Visible Light. *RSC Advances* **2017**, *7*, 46738–46744. https://doi.org/10.1039/C7RA09009A.
29. Xiang, Q.; Yu, J.; Jaroniec, M. Synergetic Effect of MoS_2 and Graphene as Cocatalysts for Enhanced Photocatalytic H_2 Production Activity of TiO_2 Nanoparticles. *J. Am. Chem. Soc.* **2012**, *134*, 6575–6578. https://doi.org/10.1021/ja302846n.
30. Chou, S.S.; Sai, N.; Lu, P.; Coker, E.N.; Liu, S.; Artyushkova, K.; Luk, T.S.; Kaehr, B.; Brinker, C.J. Understanding Catalysis in a Multiphasic Two-Dimensional Transition Metal Dichalcogenide. *Nat. Commun.* **2015**, *6*, 8311. https://doi.org/10.1038/ncomms9311.