Electronic Supporting Information

Ultrathin 2D/2D $Ti_3C_2T_x$/semiconductor dual-functional photocatalysts for simultaneous imine production and H_2 evolution

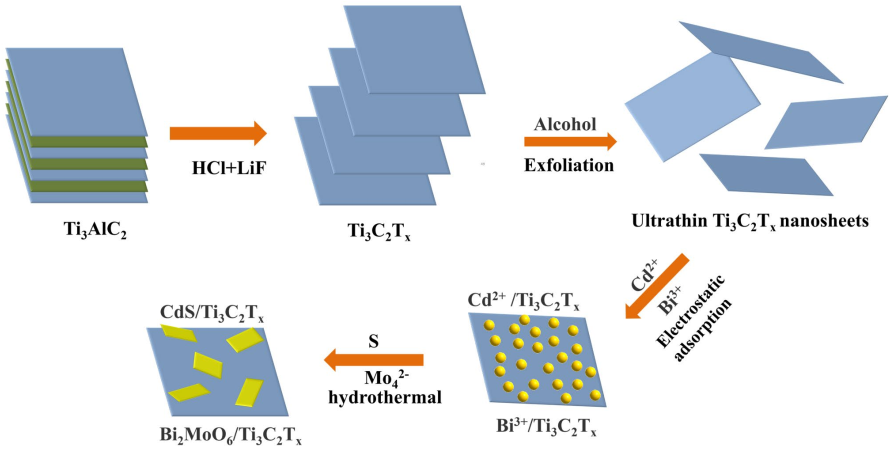
*Hao Wang, [a] Peng Hu, [a] Jie Zhou, [a] Maarten B. J. Roeffaers, [c] Bo Weng, [*c] Yongqing Wang [*a] and Hongbing Ji [*a, b]*

[a] *Fine Chemical Industry Research Institute, School of Chemistry, Sun Yat-sen University, Guangzhou 510275, P. R. China.*

[b] *School of Chemical Engineering, Guangdong University of Petrochemical Technology, Maoming 525000, P. R. China.*

[c] *cMACS, Department of Microbial and Molecular Systems, KU Leuven, Celestijnenlaan 200F, 3001 Leuven, Belgium*

E-mail: bo.weng@kuleuven.be; wangyq223@mail.sysu.edu.cn; jihb@mail.sysu.edu.cn



Scheme S1. Schematic illustration of the synthesis of 2D $Ti_3C_2T_x$/2D semiconductor hybrids.

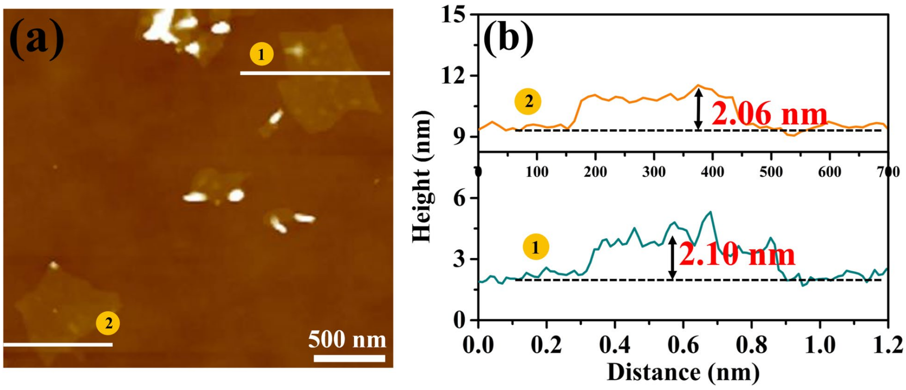


Fig. S1. (a) AFM image and (b) height cutaway view of $Ti_3C_2T_x$.

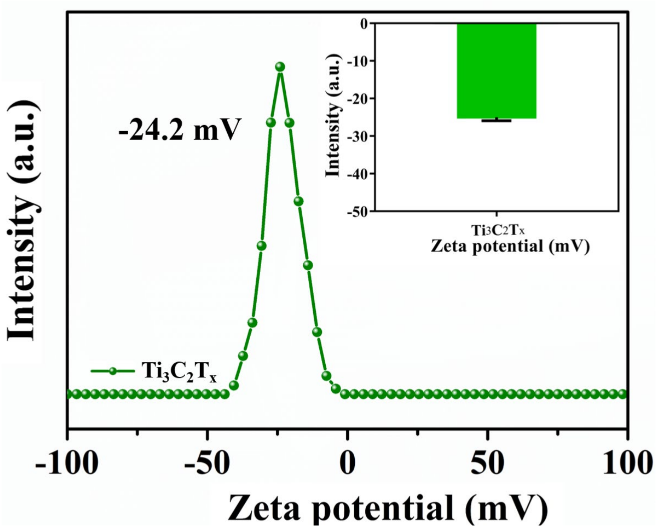


Fig. S2. Zeta potential of $Ti_3C_2T_x$.

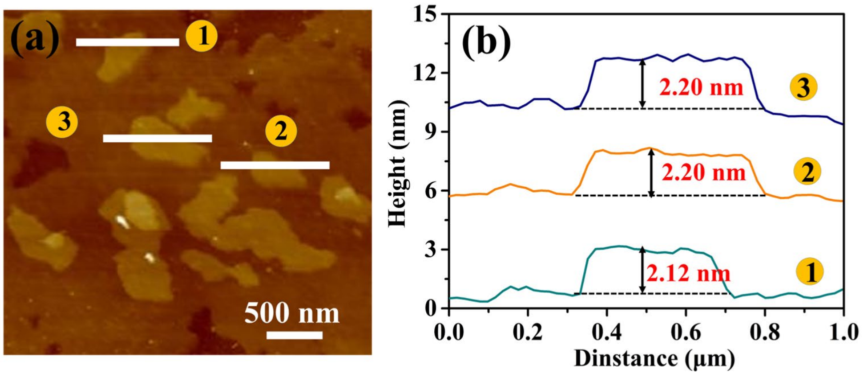


Fig. S3. (a) AFM image and (b) height cutaway view of CdS.

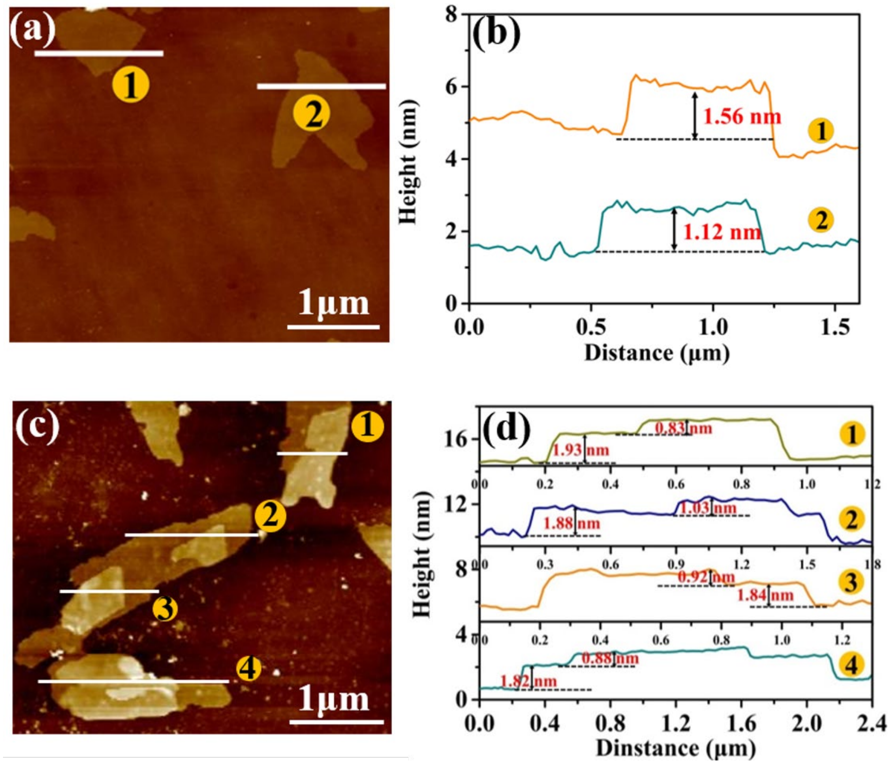


Fig. S4. AFM images and height cutaway views of (a, b) Bi_2MoO_6 and (c, d) TM1.5.

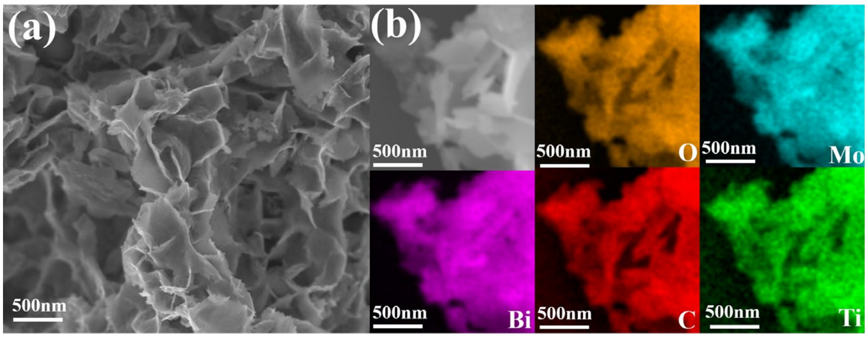


Fig. S5. SEM images of (a) Bi_2MoO_6 and (b) TM1.5 samples with the corresponding element mappings.

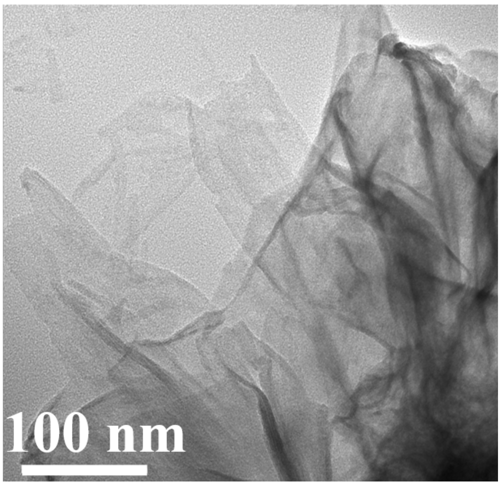


Fig. S6. Aberration-corrected STEM image of CdS.

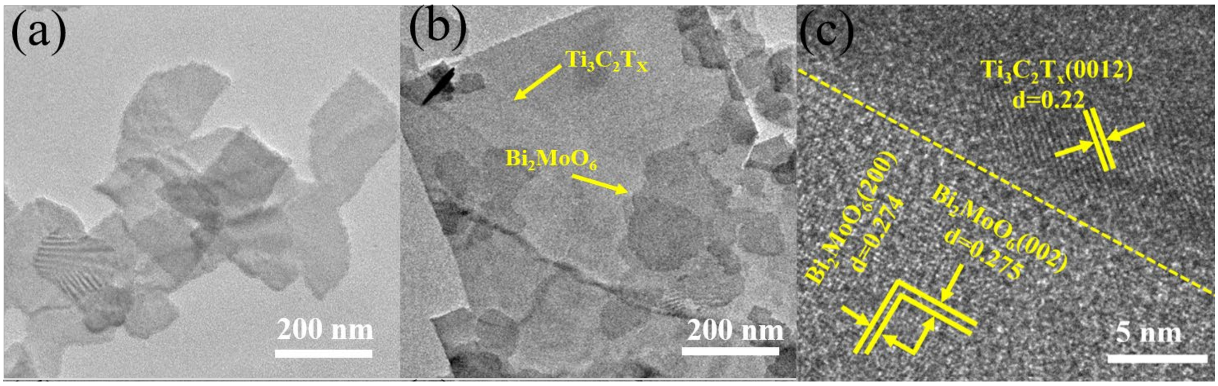


Fig. S7. Aberration-corrected STEM images of (a) Bi_2MoO_6 and (b-c) TM1.5 samples.

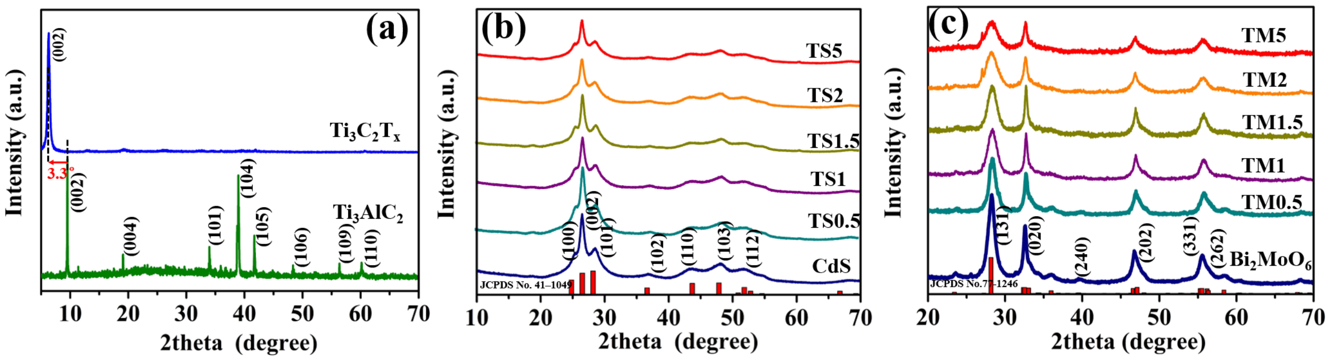


Fig. S8. (a) XRD patterns of $Ti_3C_2T_x$ and Ti_3AlC_2 samples; (b) XRD patterns of CdS and TSx composites (where x represents the addition amount of $Ti_3C_2T_x$); (c) XRD patterns of Bi_2MoO_6 and TMx composites (where x represents the addition amount of $Ti_3C_2T_x$).

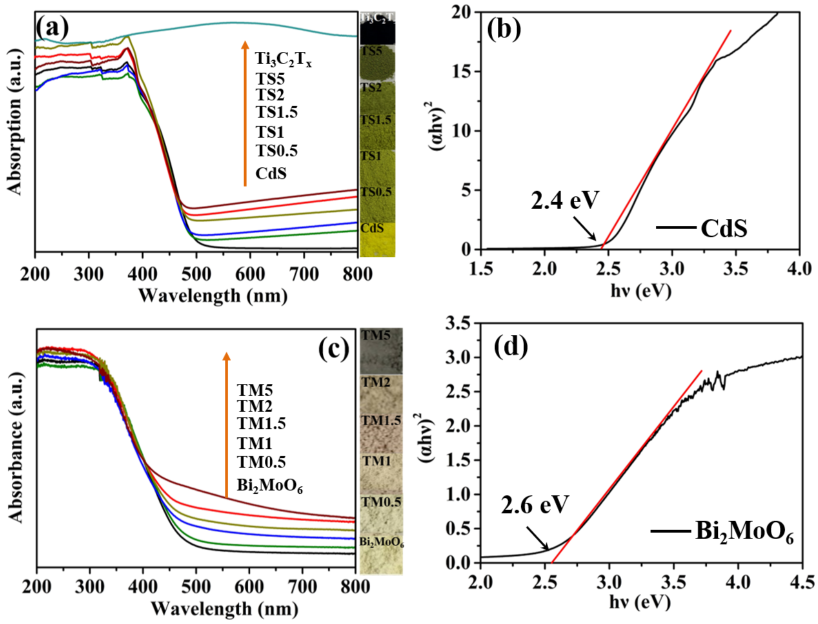


Fig. S9. (a) UV-Vis DRS spectra of $Ti_3C_2T_x$, CdS and TSx composites and photographs of different samples; (b) Tauc plot of CdS sample; (c) UV/Vis DRS spectra of Bi_2MoO_6 and TMx composites and photographs of different samples; (d) Tauc plot of Bi_2MoO_6 sample.

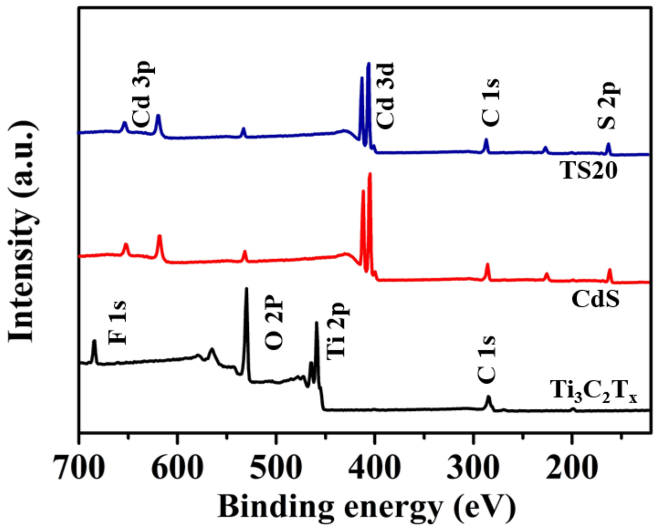


Fig. S10. XPS survey spectra of TS20, CdS and $Ti_3C_2T_x$

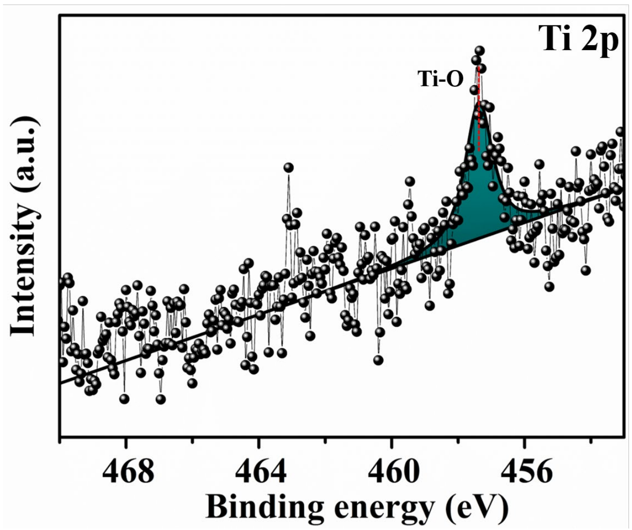


Fig. S11. High-resolution XPS spectrum of Ti 2p over TS20 sample.

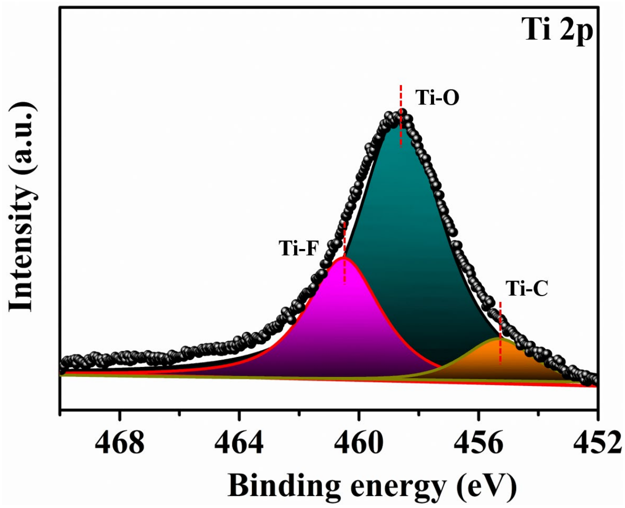


Fig. S12. High-resolution XPS spectrum of Ti 2p over TM1.5 sample.

Table S1: The activity of photocatalytic benzylamine oxidation integrated with H_2 production under different reaction conditions.

Entry	Catalyst	hv	Yield $_{(Imines)}$/μmolg^{-1}h^{-1}	Yield$_{(H_2)}$/μmolg^{-1}h^{-1}
1	TS1.5	-	-	-
2	TM2	-	-	-
3	-	+	-	-

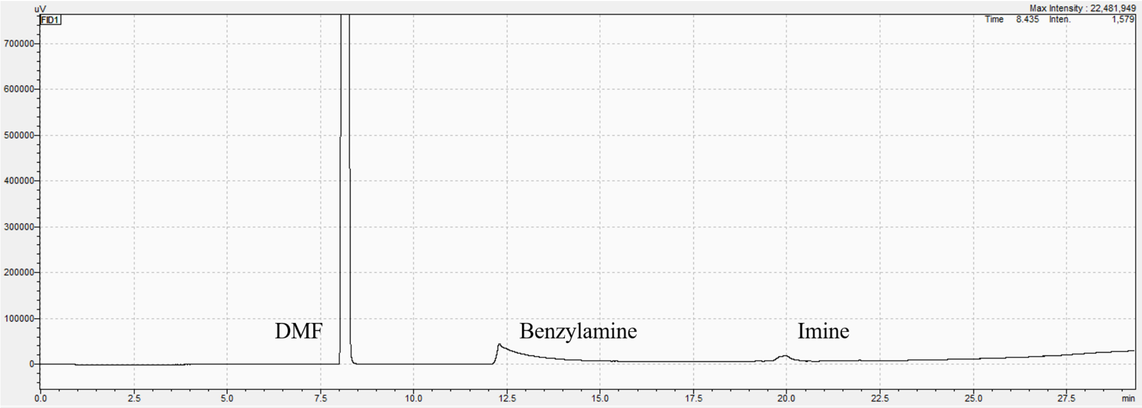


Fig. S13. GC spectrum of products after photocatalytic reaction over TS1.5 sample.

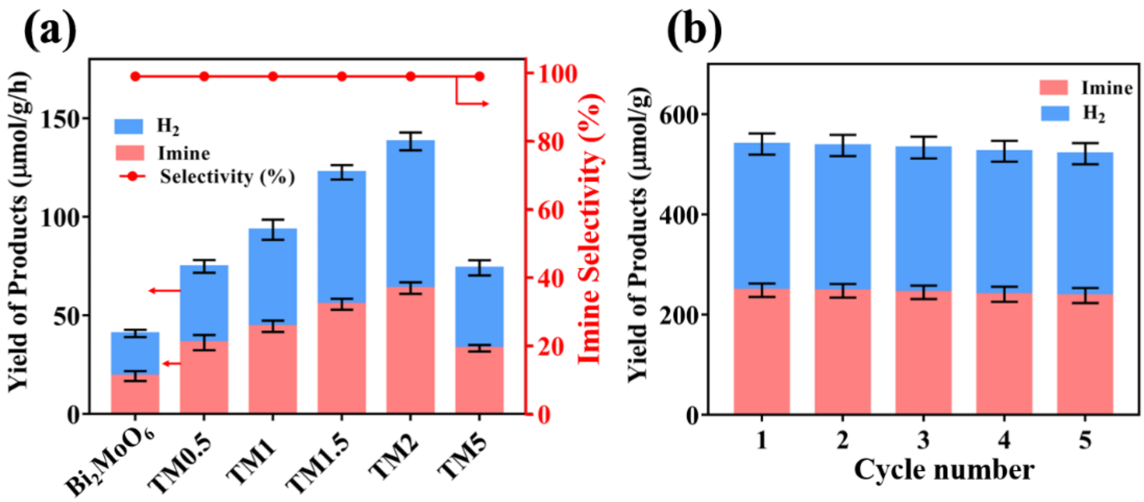


Fig. S14. (a) Photocatalytic activity for benzylamine oxidation and H_2 generation over Bi_2MoO_6 and TMx composites under visible light irradiation ($\lambda \geq 420$ nm); (b) Recycling tests over TM2 composite for photocatalytic benzylamine oxidation integrated with H_2 production (each cycle is 4 h).

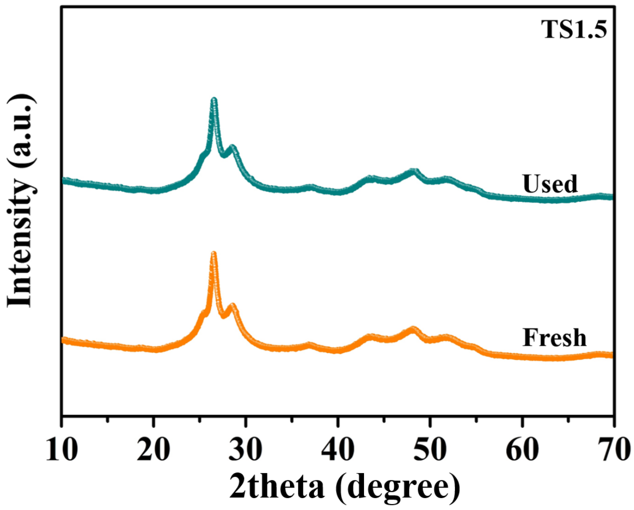


Fig. S15. XRD patterns of the TS1.5 sample before and after photocatalytic reaction.

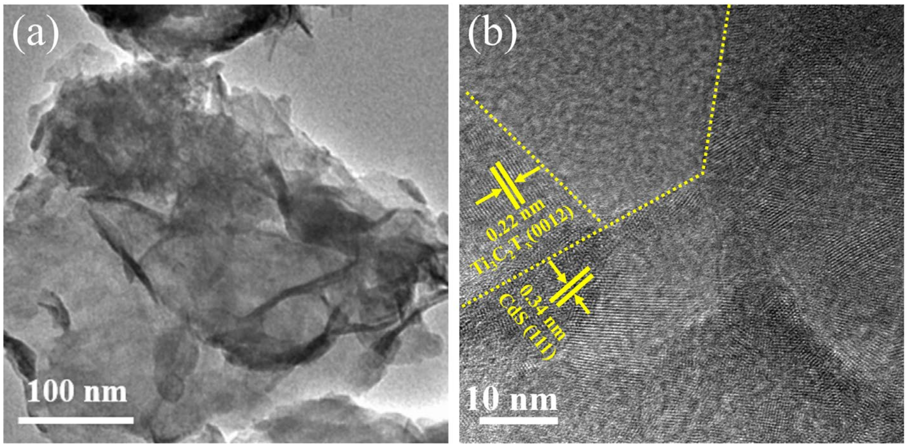


Fig. S16. Aberration-corrected STEM patterns of the TS1.5 sample after photocatalytic reaction.

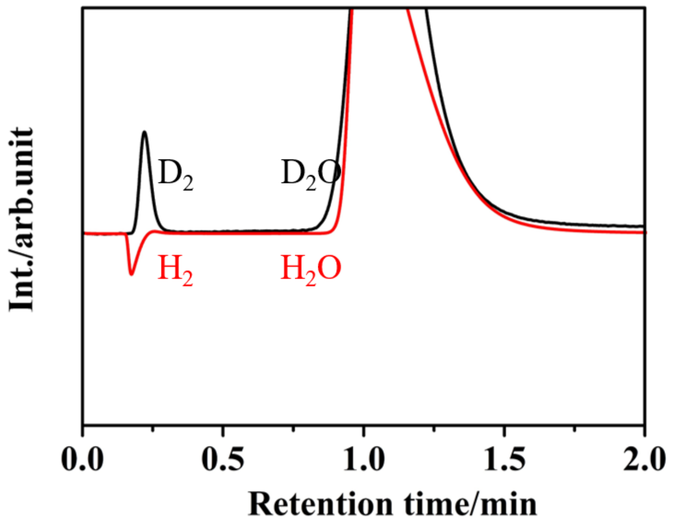


Fig. S17. GC analysis of gas phase products for photocatalytic benzylamine coupling and H_2O (or D_2O) splitting.

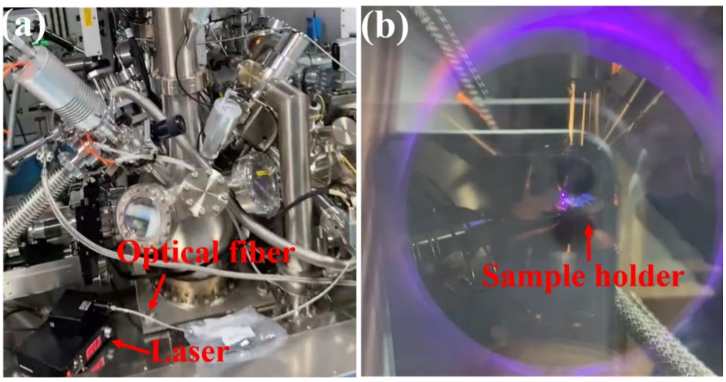


Fig. S18. In-situ irradiation XPS setup of TS20 sample.

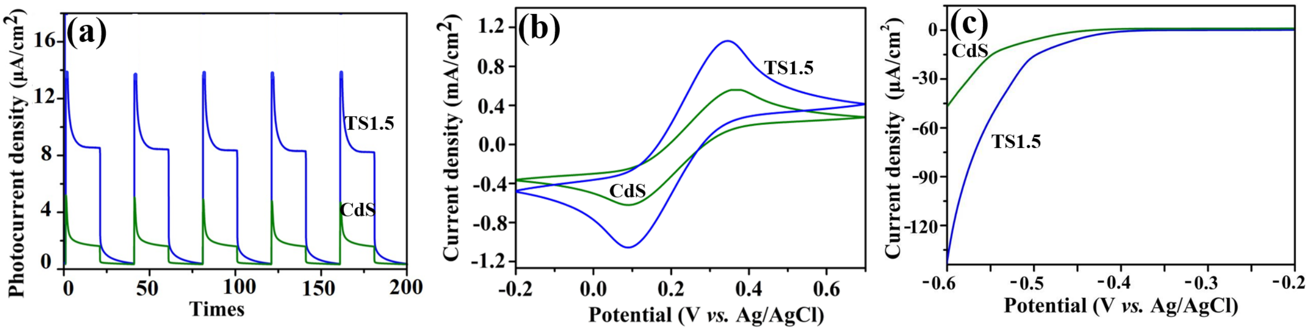


Fig. S19. (a) Transient photocurrent spectra, (b) CV and (c) LSV of TS1.5 and CdS samples.

Table S2: Photocatalytic hydrogen production uses different catalysts

photocatalysts	Light sources	Reaction conditions	Efficiencies (μmol g^{-1} h^{-1})	Ref.
CdS/$Ti_3C_2T_x$	300 W Xe lamp λ ≥ 420 nm	10mg; DMF-based solution with H_2O;5 mL	219.7	This work
Pt/F-TiO_2	300 W Xe lamp 500 nm ≥ λ ≥300 nm	15 mg; Perchloric acid aqueous solution	160	1
Ag/Co_3O_4/Cu_2O	100W Xe lamp	100 mg; aqueous solution;100mL	97.2	2
MoS_2QDs@$ZnIn_2S_4$@RGO	300 W Xe lamp 780 nm≥λ≥320 nm	100 mg; aqueous solution	37.8	3
$ZnO_{0.6}S_{0.4}$	300 W Xe lamp λ ≥ 400 nm	50 mg; aqueous solution; 50 mL	49.2	4
Pt/TiO_2	450 W Xe lamp (UV−vis)	10 mg; aqueous solution; 215 mL	101	5
TiO_2	300 W Xe lamp (UV−vis)	100 mg; aqueous solution; 120 mL	19.4	6
CdS/CdS/GCN	300 W Xe lamp λ > 420 nm;	100 mg; aqueous solution; 80 mL	5	7
Pt/PCN-777	300 W Xe lamp (UV−vis)	10 mg; DMF-based solution with H_2O; 5.1 mL	335; 1.7	8

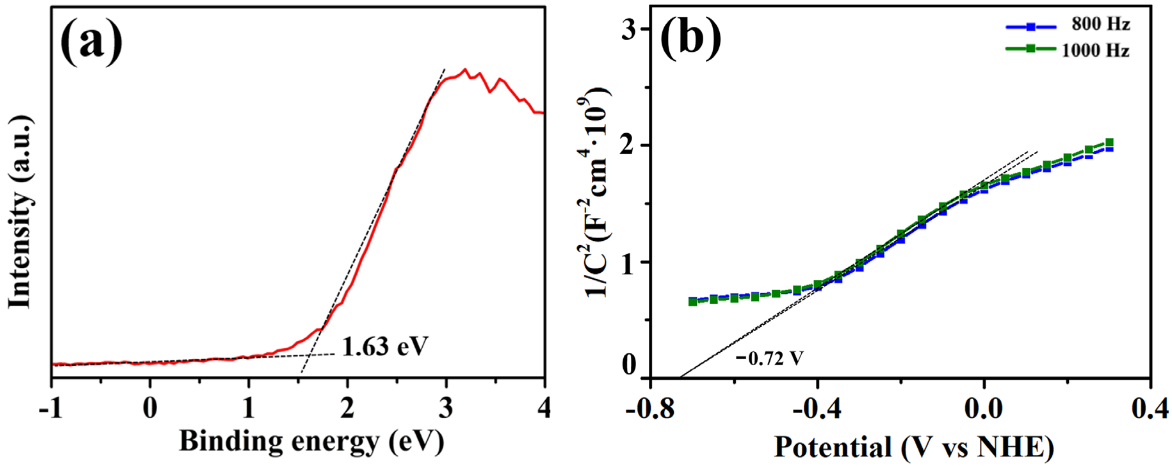


Fig. S20. (a) Ultraviolet photoelectron spectroscopy (UPS) spectrum and (b) Mott-Schottky plots of CdS sample.

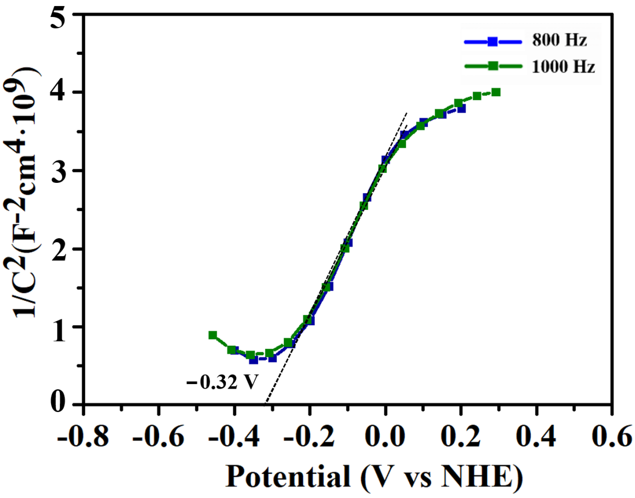


Fig. S21. (a) Mott-Schottky plots of Bi_2MoO_6 sample.

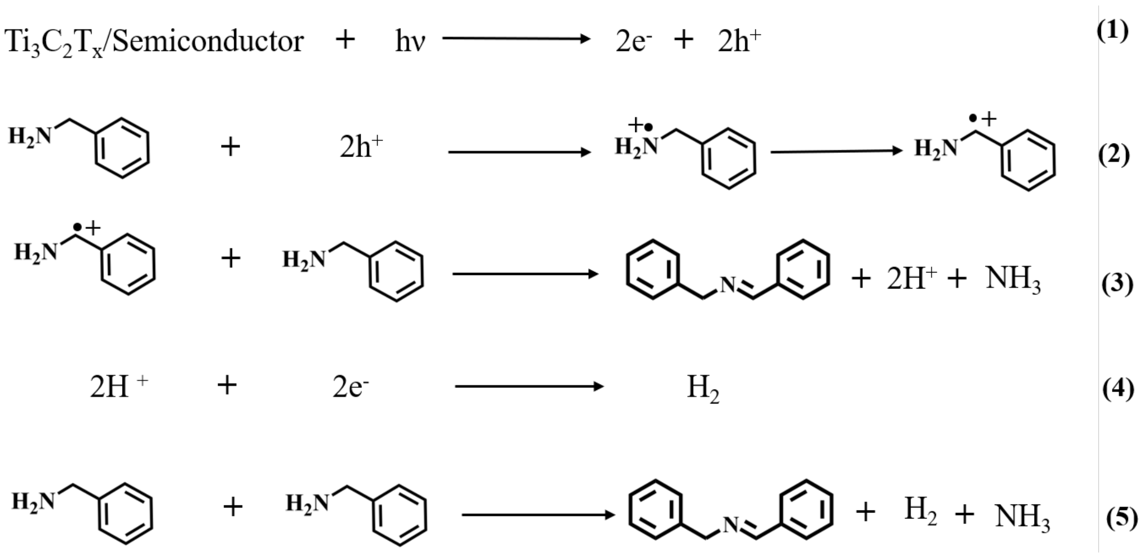


Fig. S22. Proposed reaction pathway for photocatalytic imine production and H_2 evolution over $Ti_3C_2T_x$/semiconductors.

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