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Supporting Information

In-Situ Construction of Ohmic/Schottky-Type MoS_2/S_v-$ZnIn_2S_4/Cu(OH)_2$ Dual-Junction Photocatalysts with Boosting Water Splitting into Hydrogen Generation Activity

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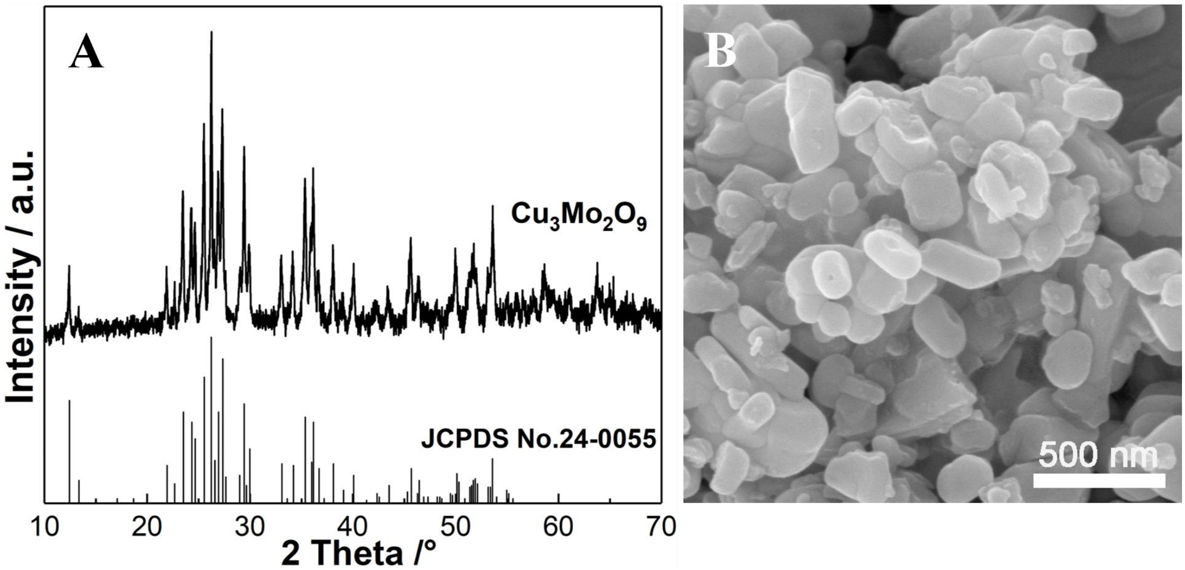

Figure S1 (A) XRD pattern and (B) FESEM image of pristine CMO.

2. FESEM Images

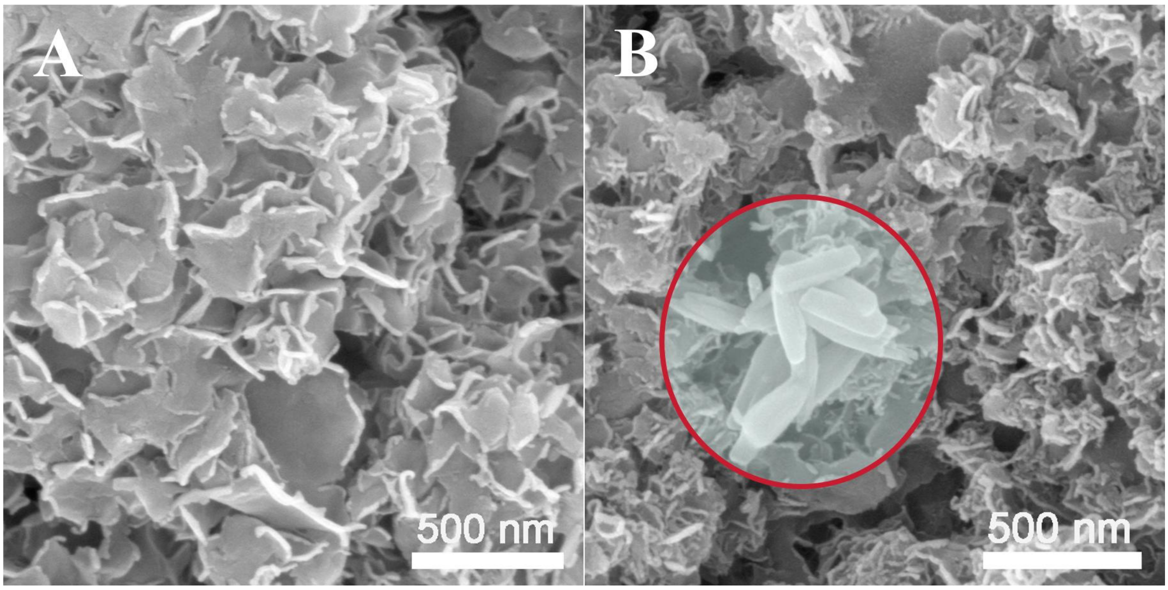

Figure S2. Typical FESEM images of (A) CMO-0.2/ZIS and (B) CMO-2.0/ZIS.

3. XRD Patterns of ZIS and CMO-x/ZIS

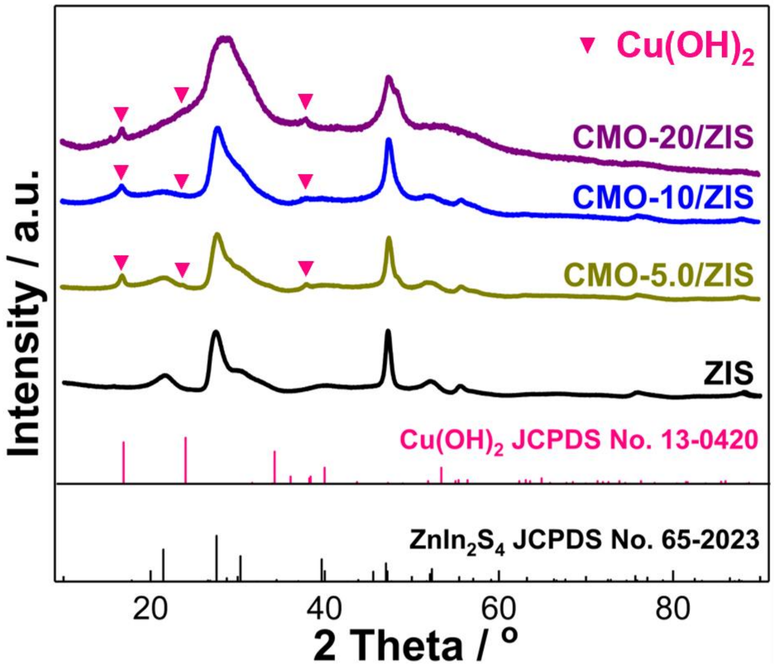

Figure S3 XRD patterns of pristine ZIS and CMO-x/ZIS samples.

4. XPS Characterization

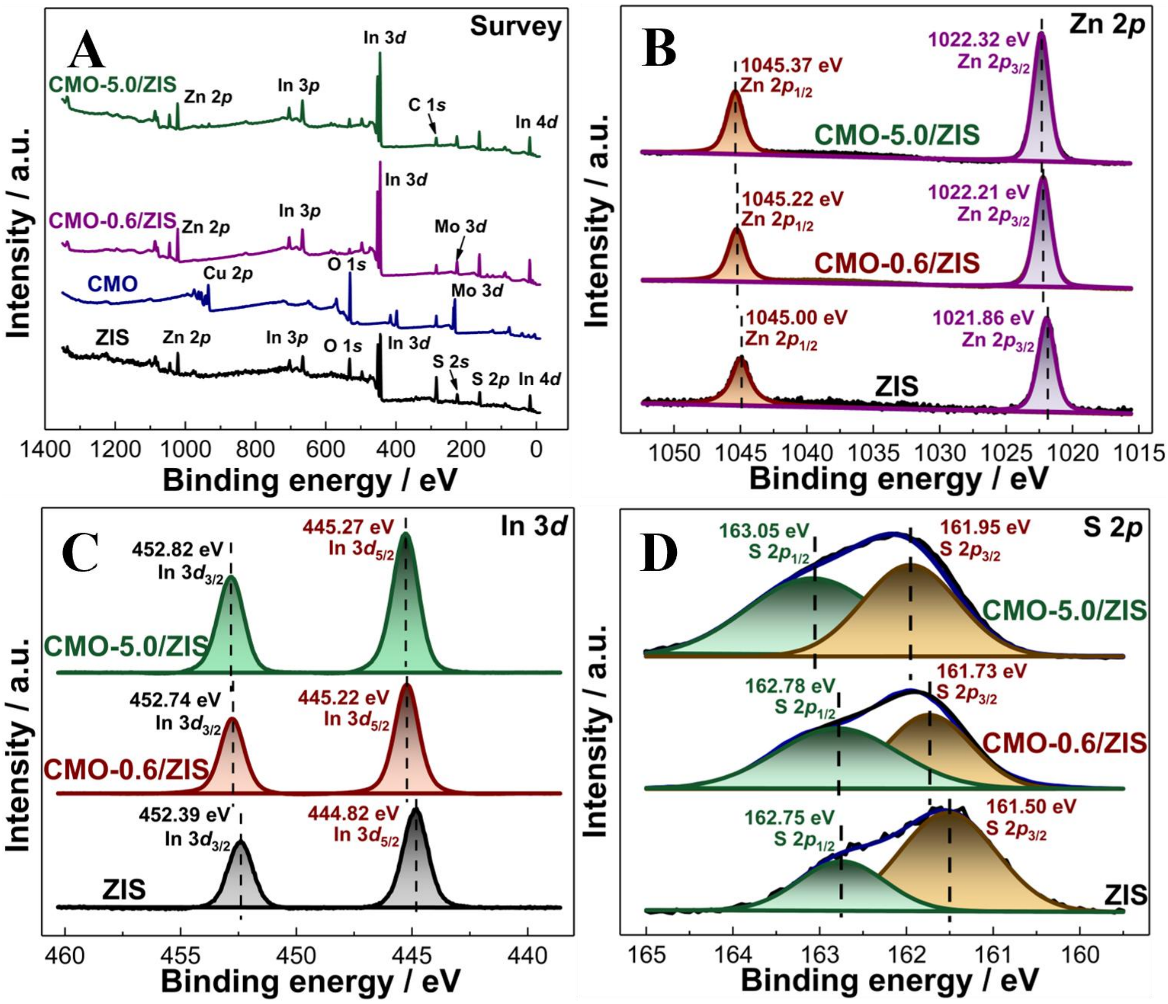

Figure S4 (A) XPS survey spectra of pristine ZIS, pristine CMO, CMO-0.6/ZIS and CMO-5.0/ZIS samples; narrow-scanned XPS spectra of (B) Zn 2*p*, (C) In 3*d* and (D) S 2*p* for different samples.

5. Hydrogen Evolution Rates of Photocatalysts

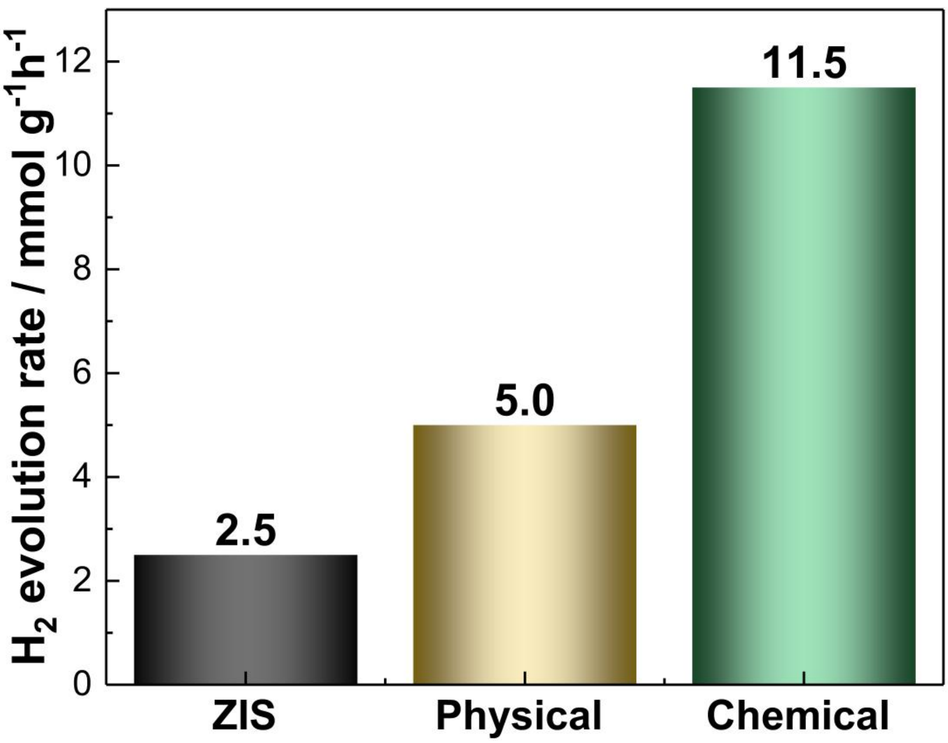

Figure S5. Hydrogen evolution rates of the catalyst prepared by different methods.

6. XRD、SEM、TEM and XPS Results of Initial and Irradiated Photocatalysts

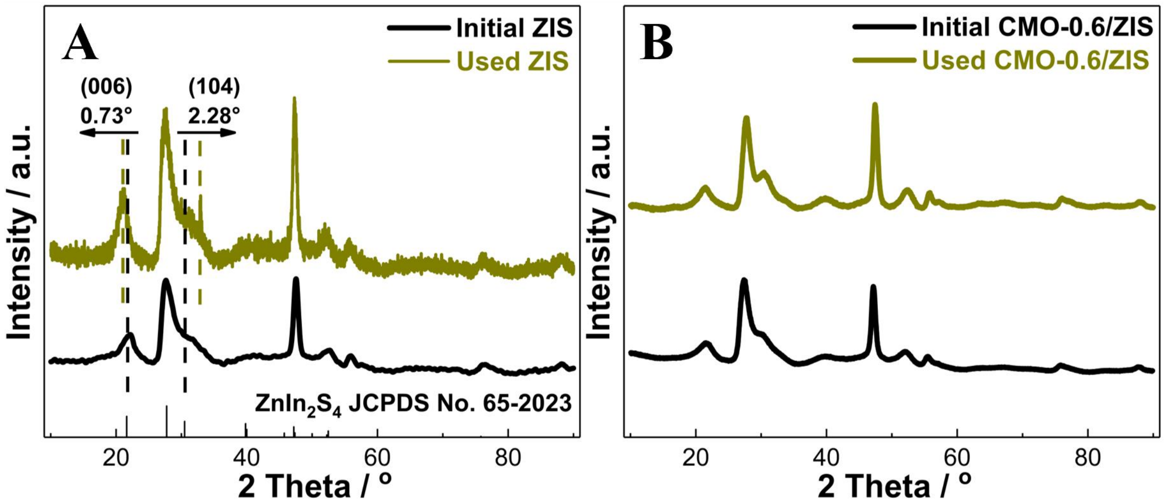

Figure S6. XRD patterns of (A) initial ZIS & irradiated ZIS for four runs and (B) initial CMO-0.6/ZIS & irradiated CMO-0.6/ZIS for four runs.

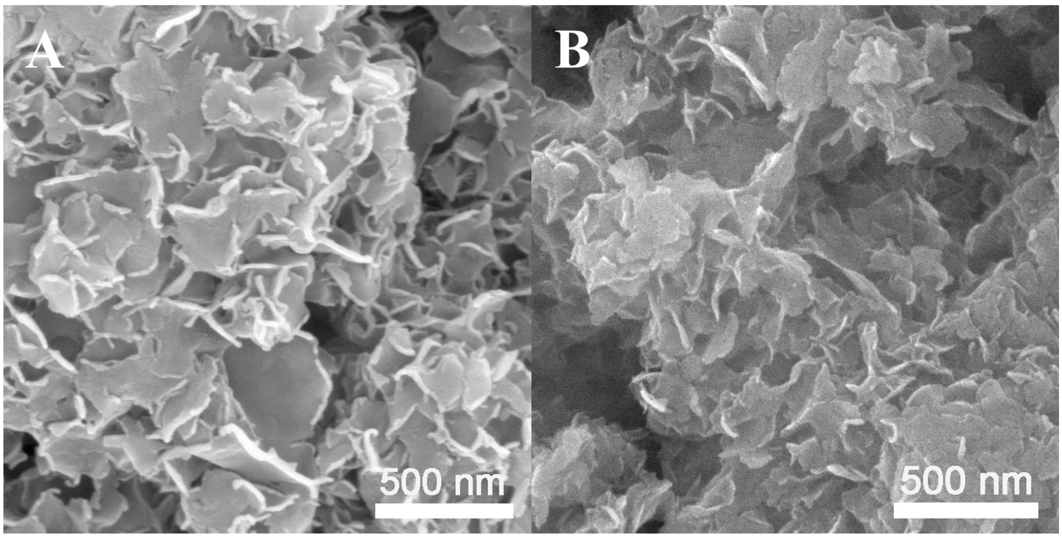

Figure S7. FESEM images of (A) initial CMO-0.6/ZIS and (B) irradiated CMO-0.6/ZIS for four runs.

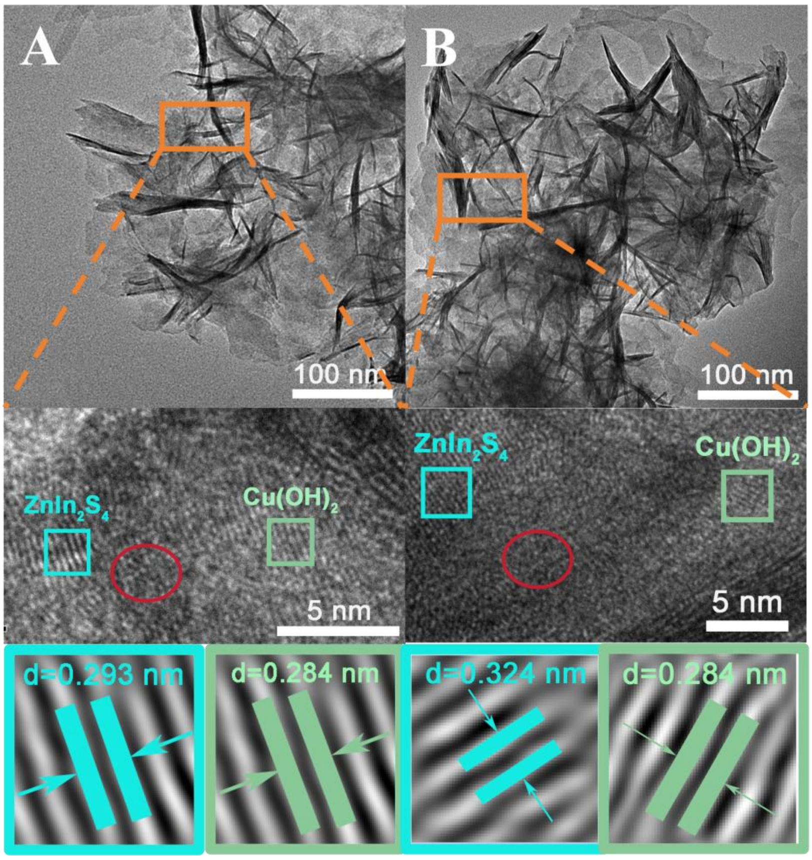

Figure S8. HRTEM images of (A) initial CMO-0.6/ZIS and (B) irradiated CMO-0.6/ZIS for four runs.

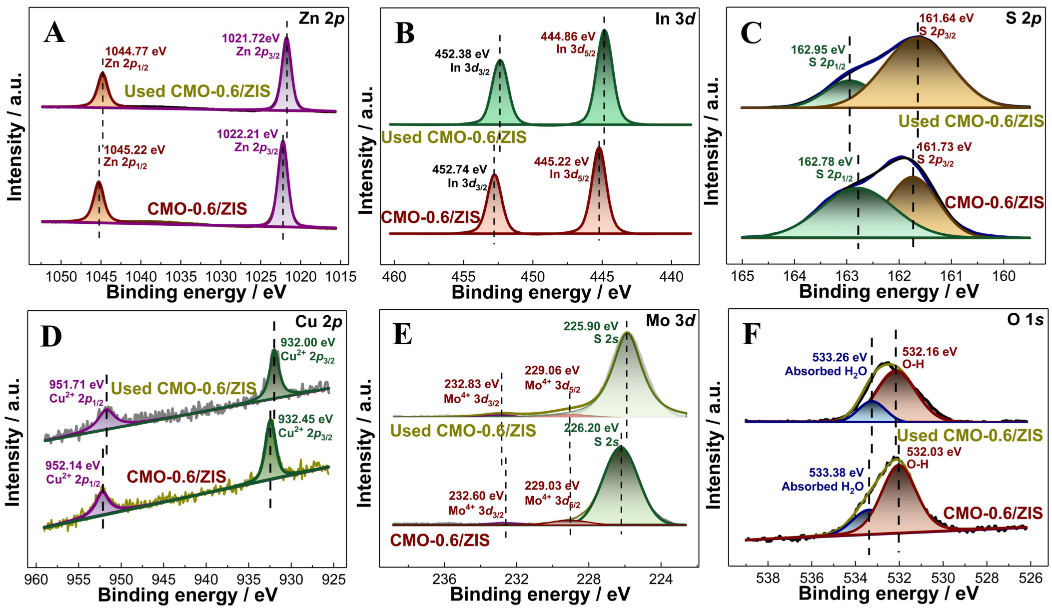

Figure S9. XPS patterns of initial CMO-0.6/ZIS and irradiated CMO-0.6/ZIS for four runs.

7. Calculated *AQE* Values

Table S1 *AQE* values of pristine ZIS and CMO-0.6/ZIS catalysts at different wavelengths.

AQE	ZIS	CMO-0.6/ZIS
420 nm	1.82%	2.15%
450 nm	1.02%	2.15%
475 nm	0.23%	1.54%
500 nm	0.04%	0.66%

AQE measurement was carried out under the same reaction conditions as photocatalytic hydrogen production experiments except that the incident light was supplied by a 300W Xe lamp equipped with specific band-pass filters to get the monochromatic incident wavelength (λ = 420, 450, 475 and 500 nm). *AQE* values for pristine ZIS and CMO-0.6/ZIS samples were roughly calculated as follows:

(1) λ = 420 nm

ZIS:

$$\mathrm{N} = \frac{E\lambda}{hc} = \frac{10.4 \times 5.28 \times 10^{-3} \times 3 \times 3600 \times 420 \times 10^{-9}}{6.626 \times 10^{-34} \times 3 \times 10^{8}} = 12.53 \times 10^{20}$$

$$\mathrm{AQE} = \frac{the\ number\ of\ reacted\ electrons}{the\ number\ of\ incident\ photons} \times 100\%$$

$$= \frac{2 \times \text{the number of evolved } \mathrm{H_2} \text{ molecules}}{\mathrm{N}} \times 100\%$$

$$= \frac{2 \times 6.02 \times 10^{23} \times 18.9 \times 10^{-6}}{12.53 \times 10^{20}} \times 100\% = 1.82\%$$

CMO-0.6/ZIS:

$$\mathrm{N} = \frac{E\lambda}{hc} = \frac{10.4 \times 5.28 \times 10^{-3} \times 3 \times 3600 \times 420 \times 10^{-9}}{6.626 \times 10^{-34} \times 3 \times 10^{8}} = 12.53 \times 10^{20}$$

$$\mathrm{AQE} = \frac{the\ number\ of\ reacted\ electrons}{the\ number\ of\ incident\ photons} \times 100\%$$

$$= \frac{2 \times \text{the number of evolved } \mathrm{H_2} \text{ molecules}}{\mathrm{N}} \times 100\%$$

$$=\frac{2\times 6.02\times 10^{23}\times 22.36\times 10^{-6}}{12.53\times 10^{20}}\times 100\%=2.15\%$$

(2) λ = 450 nm

ZIS:

$$\mathrm{N}=\frac{E\lambda}{hc}=\frac{9.7\times 5.28\times 10^{-3}\times 3\times 3600\times 450\times 10^{-9}}{6.626\times 10^{-34}\times 3\times 10^{8}}=12.52\times 10^{20}$$

$$\mathrm{AQE}=\frac{the\ number\ of\ reacted\ electrons}{the\ number\ of\ incident\ photons}\times 100\%$$

$$=\frac{2\times \text{the number of evolved } \mathrm{H_2} \text{ molecules}}{\mathrm{N}}\times 100\%$$

$$=\frac{2\times 6.02\times 10^{23}\times 10.61\times 10^{-6}}{12.52\times 10^{20}}\times 100\%=1.02\%$$

CMO-0.6/ZIS:

$$\mathrm{N}=\frac{E\lambda}{hc}=\frac{9.7\times 5.28\times 10^{-3}\times 3\times 3600\times 450\times 10^{-9}}{6.626\times 10^{-34}\times 3\times 10^{8}}=12.52\times 10^{20}$$

$$\mathrm{AQE}=\frac{the\ number\ of\ reacted\ electrons}{the\ number\ of\ incident\ photons}\times 100\%$$

$$=\frac{2\times \text{the number of evolved } \mathrm{H_2} \text{ molecules}}{\mathrm{N}}\times 100\%$$

$$=\frac{2\times 6.02\times 10^{23}\times 22.32\times 10^{-6}}{12.52\times 10^{20}}\times 100\%=2.15\%$$

(3) λ = 475 nm

ZIS:

$$\mathrm{N}=\frac{E\lambda}{hc}=\frac{18.5\times 5.28\times 10^{-3}\times 3\times 3600\times 475\times 10^{-9}}{6.626\times 10^{-34}\times 3\times 10^{8}}=25.21\times 10^{20}$$

$$\mathrm{AQE}=\frac{the\ number\ of\ reacted\ electrons}{the\ number\ of\ incident\ photons}\times 100\%$$

$$=\frac{2\times \text{the number of evolved } \mathrm{H_2} \text{ molecules}}{\mathrm{N}}\times 100\%$$

$$=\frac{2\times 6.02\times 10^{23}\times 4.88\times 10^{-6}}{25.21\times 10^{20}}\times 100\%=0.23\%$$

CMO-0.6/ZIS:

$$\mathrm{N}=\frac{E\lambda}{hc}=\frac{18.5\times 5.28\times 10^{-3}\times 3\times 3600\times 475\times 10^{-9}}{6.626\times 10^{-34}\times 3\times 10^{8}}=25.21\times 10^{20}$$

$$\mathrm{AQE}=\frac{the\ number\ of\ reacted\ electrons}{the\ number\ of\ incident\ photons}\times 100\%$$

$$=\frac{2\times \text{the number of evolved } \mathrm{H_2} \text{ molecules}}{\mathrm{N}}\times 100\%$$

$$= \frac{2 \times 6.02 \times 10^{23} \times 32.21 \times 10^{-6}}{25.21 \times 10^{20}} \times 100\% = 1.54\%$$

(4) λ = 500 nm

ZIS:

$$N = \frac{E\lambda}{hc} = \frac{15.4 \times 5.28 \times 10^{-3} \times 3 \times 3600 \times 500 \times 10^{-9}}{6.626 \times 10^{-34} \times 3 \times 10^{8}} = 22.09 \times 10^{20}$$

$$\text{AQE} = \frac{the\ number\ of\ reacted\ electrons}{the\ number\ of\ incident\ photons} \times 100\%$$

$$= \frac{2 \times \text{the number of evolved } H_2 \text{ molecules}}{N} \times 100\%$$

$$= \frac{2 \times 6.02 \times 10^{23} \times 0.649 \times 10^{-6}}{22.09 \times 10^{20}} \times 100\% = 0.04\%$$

CMO-0.6/ZIS:

$$N = \frac{E\lambda}{hc} = \frac{15.4 \times 5.28 \times 10^{-3} \times 3 \times 3600 \times 500 \times 10^{-9}}{6.626 \times 10^{-34} \times 3 \times 10^{8}} = 22.09 \times 10^{20}$$

$$\text{AQE} = \frac{the\ number\ of\ reacted\ electrons}{the\ number\ of\ incident\ photons} \times 100\%$$

$$= \frac{2 \times \text{the number of evolved } H_2 \text{ molecules}}{N} \times 100\%$$

$$= \frac{2 \times 6.02 \times 10^{23} \times 12.1 \times 10^{-6}}{22.09 \times 10^{20}} \times 100\% = 0.66\%$$

8. Photogenerated Charge Carrier Dynamics

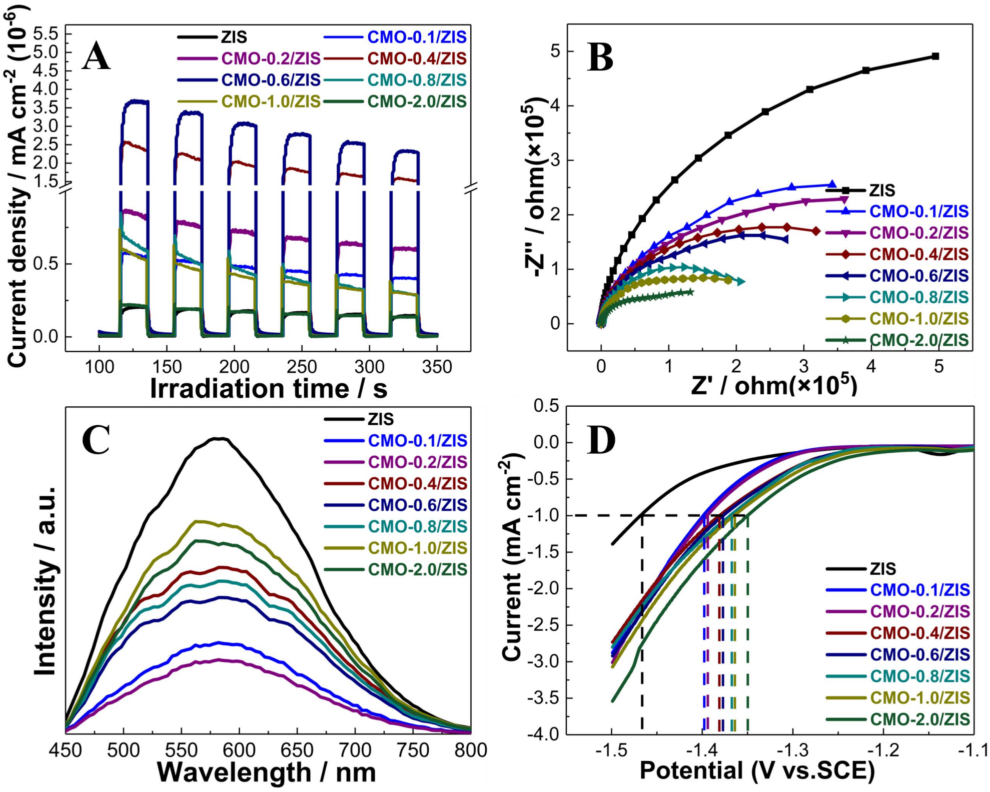

Figure S10. Photoelectronic dynamics characterization. (A) Transient photocurrent curves, (B) EIS Nyquist plots, (C) steady-state PL spectra, (D) LSV curves of different photocatalysts.

9. Calculated TRPL Data

Table S2 The summary of TRPL.

	τ_1	A_1	τ_2	A_2	τ_{ave}
ZIS	0.91	0.54	30.87	0.30	29.36
CMO-0.2/ZIS	1.04	0.67	39.63	0.26	37.19
CMO-0.6/ZIS	0.95	0.54	53.70	0.37	52.37
CMO-2.0/ZIS	1.32	0.47	37.43	0.35	35.80

Notes: The average PL emission lifetime (τ_{ave}) could be calculated according to the following formula: $\tau_{ave} = \frac{A_1\tau_1^2 + A_2\tau_2^2}{A_1\tau_1 + A_2\tau_2}$

10. UV-vis Absorption Spectra and Tauc's Plots

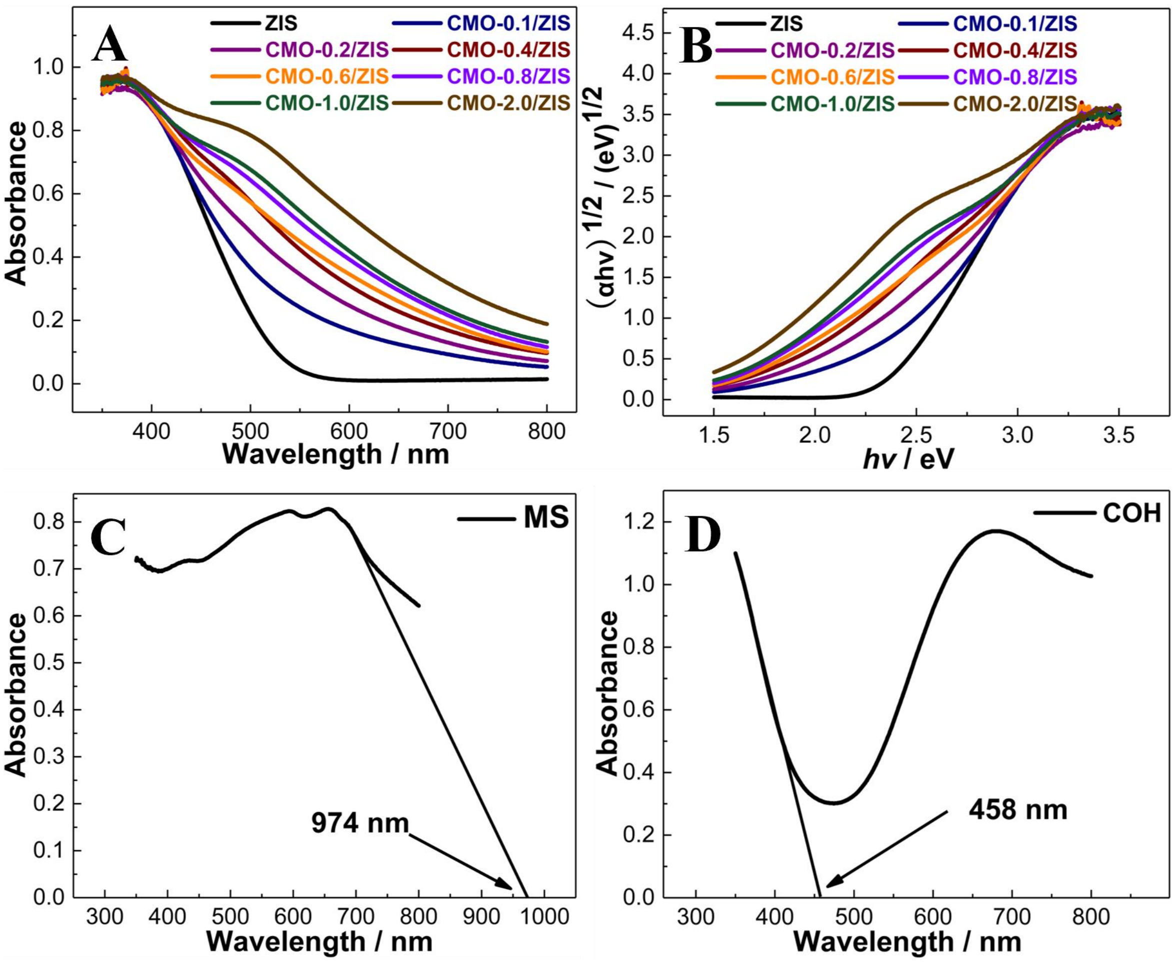

Figure S11. (A) UV-vis absorption spectra and (B) Tauc's plots of CMO-x/ZIS composite; UV-vis absorption spectra of (C) MS and (D) COH.

11. Absorption Edges and Band Gaps of Different Samples

Table S3 Absorption edges and band gaps of ZIS, MS, COH and CMO-x/ZIS samples.

Samples	Band gaps / eV	Absorption edges / nm
ZIS	2.38	522
MS	1.27	974
COH	2.70	458
CMO-0.1/ZIS	2.29	543
CMO-0.2/ZIS	2.09	596
CMO-0.4/ZIS	1.90	653
CMO-0.6/ZIS	1.86	667
CMO-0.8/ZIS	1.82	683
CMO-1.0/ZIS	1.77	699
CMO-2.0/ZIS	1.62	767

12. Optical Images of Different Photocatalysts

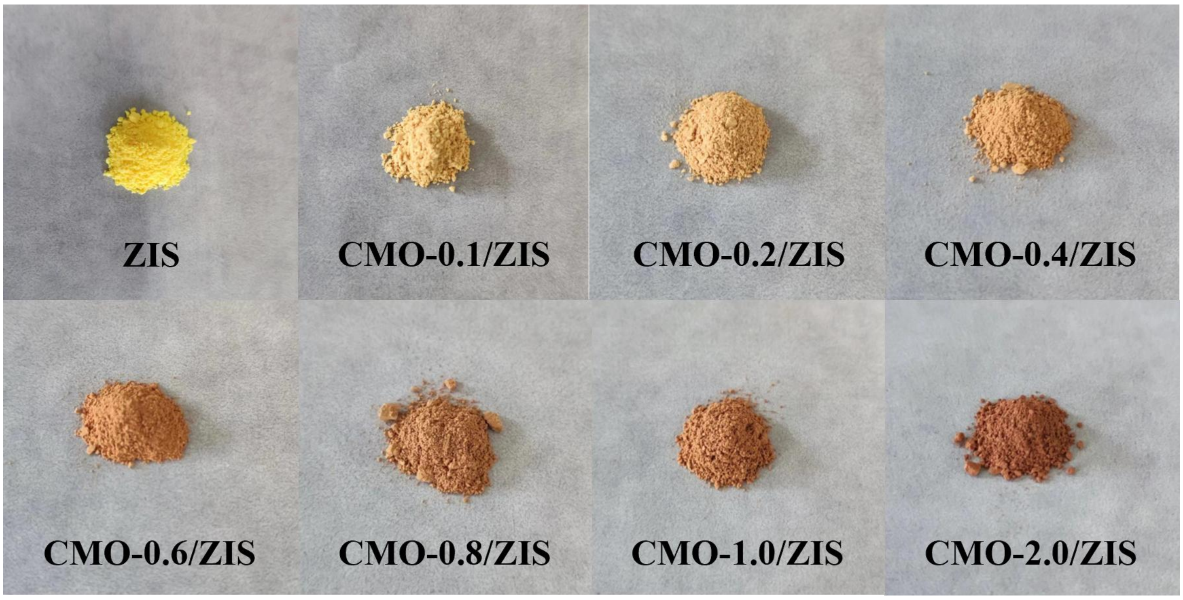

Figure S12. Optical images of different photocatalysts.

13. Carrier Concentration

Table S4 The summary of carrier concentration.

Samples	N_d-500 / $cm^{-3}(10^{23})$	N_d-800 / $cm^{-3}(10^{23})$	N_d-1000 / $cm^{-3}(10^{23})$	N_d-ave / $cm^{-3}(10^{23})$
ZIS	2.88	2.39	1.96	2.41
CMO-0.6/ZIS	4.09	3.30	2.57	3.32
CMO-0.6/ZIS	4.06	2.81	2.27	3.05
CMO-0.6/ZIS	3.88	2.54	2.00	2.81

Notes: The carrier concentration (N_d) could be determined using the following equation:

$$N_d = \left(\frac{2}{e\varepsilon\varepsilon_0}\right)\left[\frac{d(E_s)}{d(\frac{1}{C^2})}\right]$$

where e = 1.6 × 10^{-19} C, ε_0 = 8.86 × 10^{-14} F cm^{-1}, ε = 4.7.

14. UPS Results

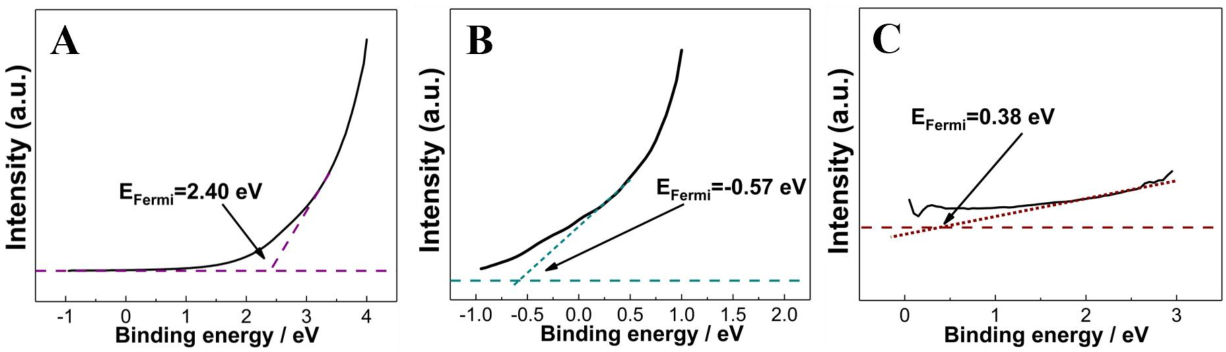

Figure S13. Magnified UPS views of (A) pristine ZIS (B) pristine MS (C) pristine COH.

15. Narrow-scanned Zn 2*p* , Cu 2*p* and Mo 3*d* XPS Spectra

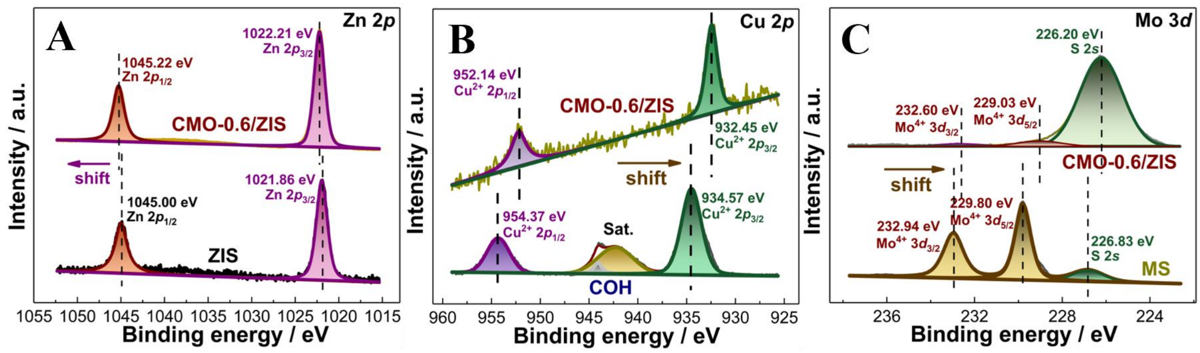

Figure S14. (A) Narrow-scanned Zn 2*p* XPS spectra of pristine ZIS and CMO-0.6/ZIS; (B) narrow-scanned Cu 2*p* XPS spectra of pristine COH and CMO-0.6/ZIS; (C) narrow-scanned Mo 3*d* XPS spectra of pristine MS and CMO-0.6/ZIS.