Electronic Supplementary Information (ESI)

Dual-defective Strategy Directing In-situ Assembly for Effective Interfacial Contacts in MoS_2 Cocatalyst/In_2S_3 Light Harvester Layered Photocatalyst

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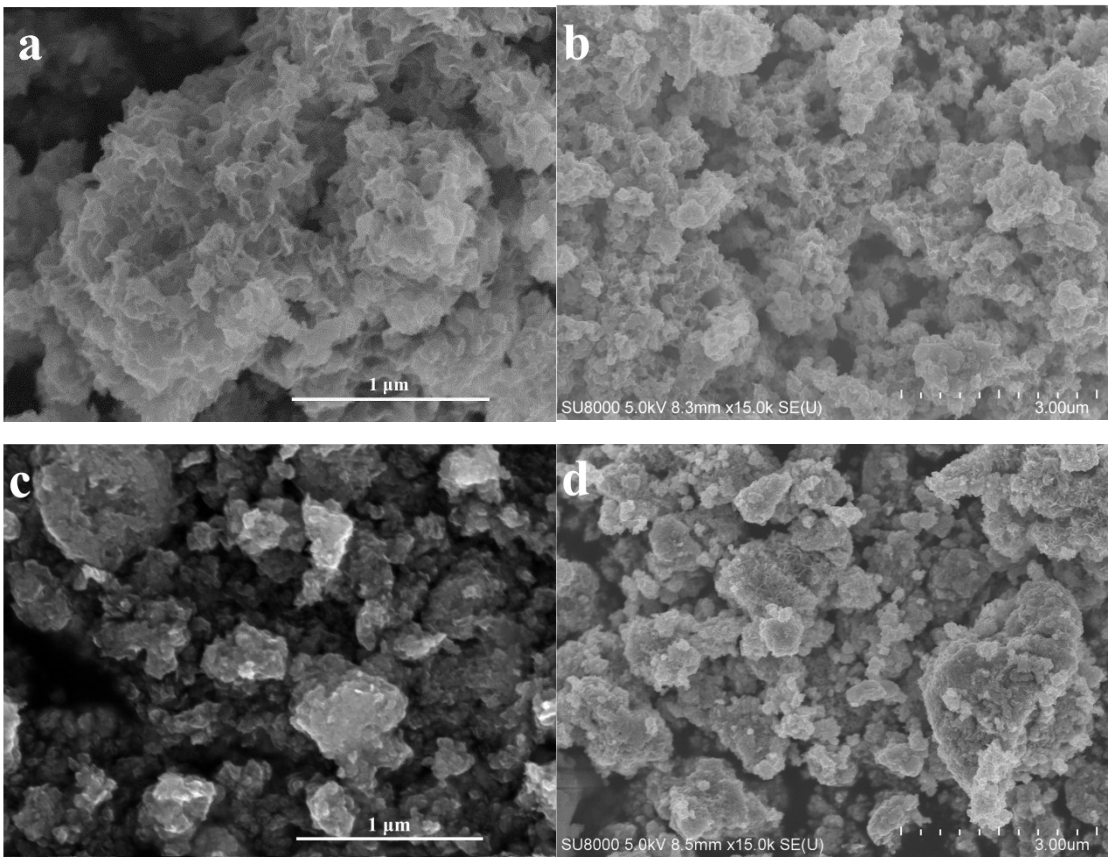

Figure S1. SEM images of the as-prepared samples with or without CTAB assistance: (a) and (b) IS (with CTAB); (c) and (d) b-IS (without CTAB).

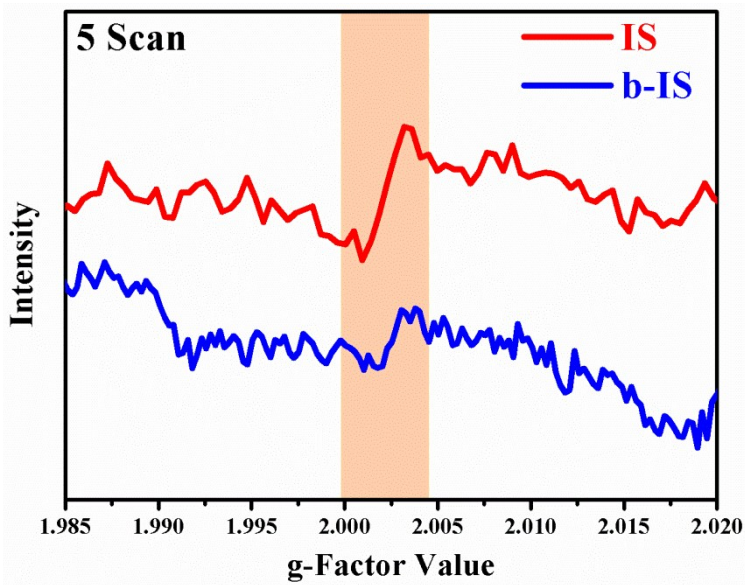

Figure S2. Intensified room-temperature ESR spectra conducted with 5 scans.

Since b-IS is not perfectly crystallized, trace amounts of defects like S vacancies should still exist in it. By increasing scan times, we can observe a faint ESR signal of S vacancies in b-IS sample, which is much weaker than that in IS sample (Figure S2). This result correlates with their different crystallinities revealed by XRD results.

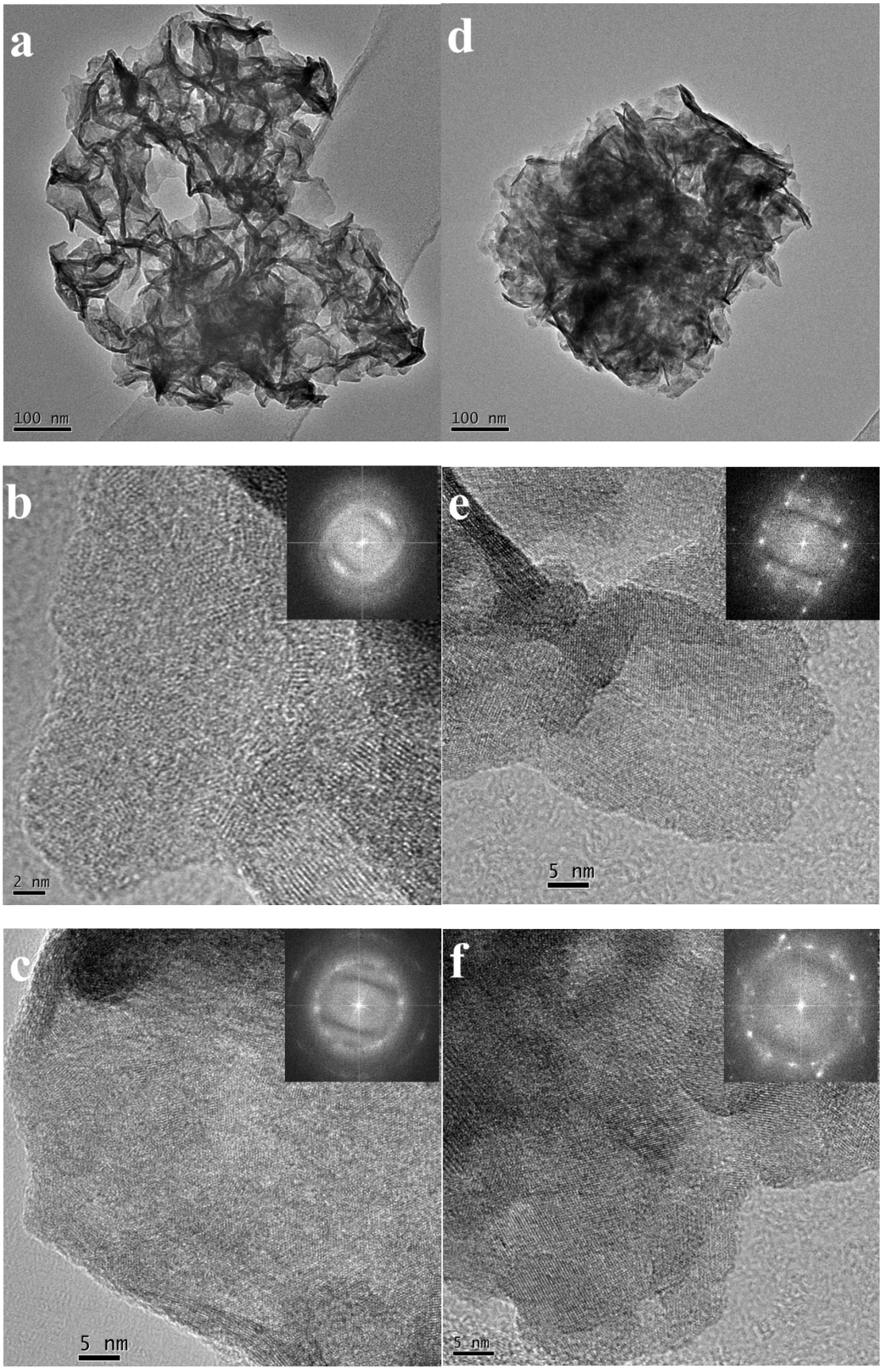

Figure S3. TEM and HRTEM images of In_2S_3 samples with different disordered degrees: (a-c) IS sample; (d-f) b-IS samples; inserts: corresponding FFT patterns.

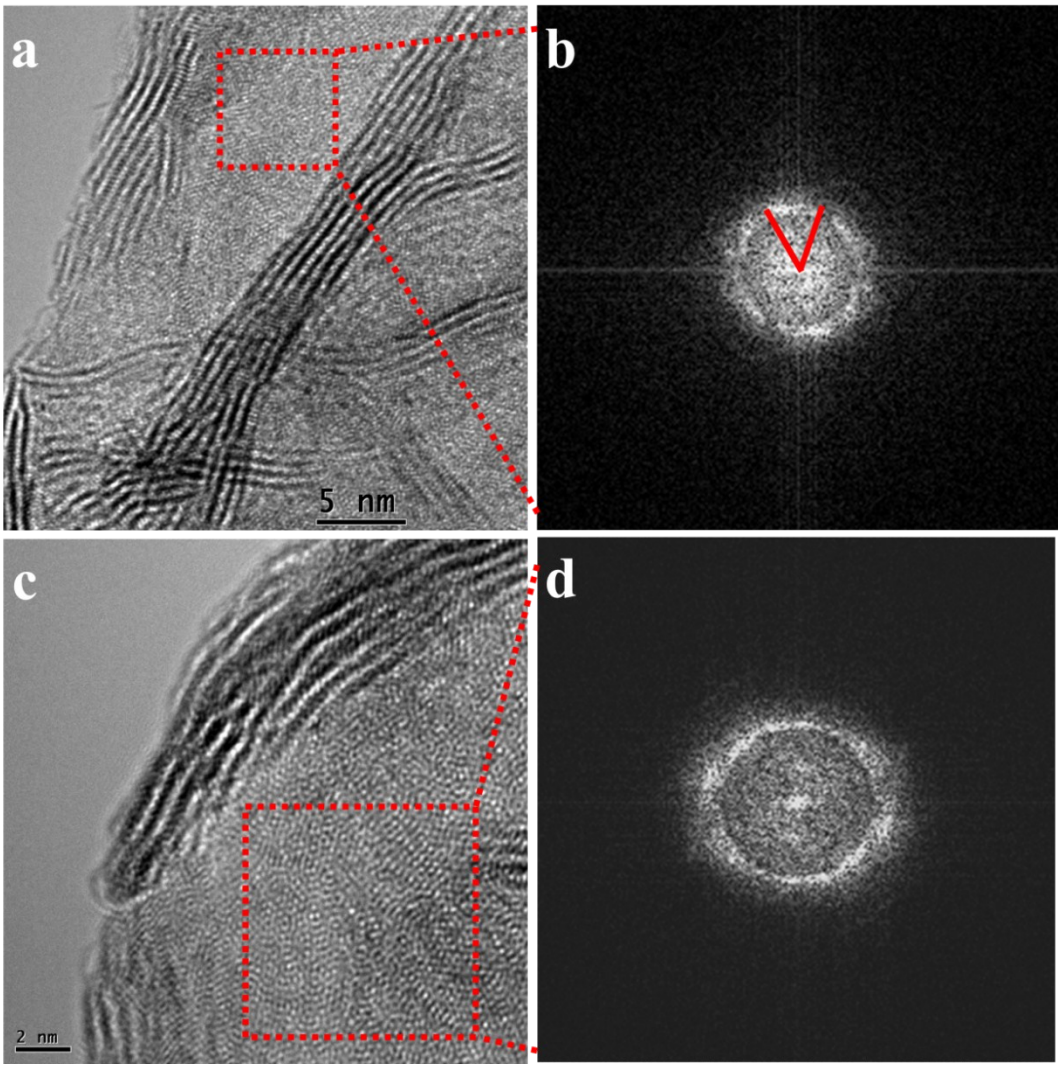

Figure S4. HRTEM images (a and b) and corresponding FFT patterns (c and d) for the as-prepared defective MoS_2 (dMS).

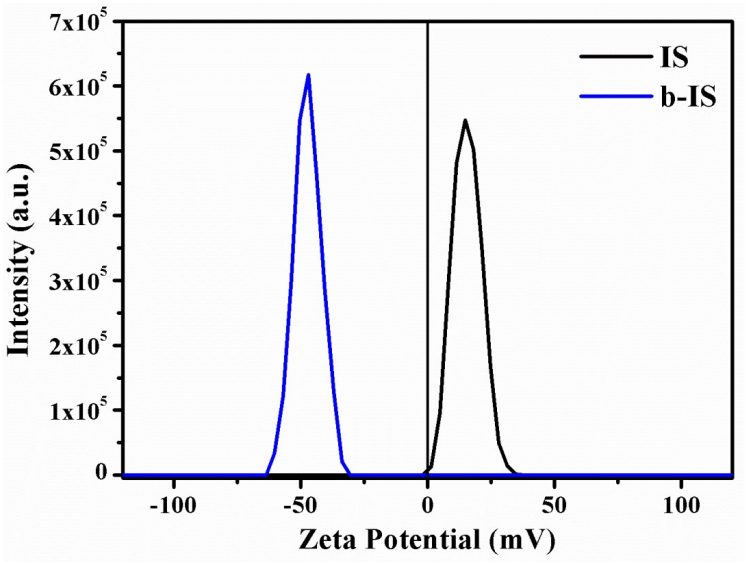

Figure S5. Zeta potentials of the as-prepared In_2S_3 samples with or without CTAB assistance.

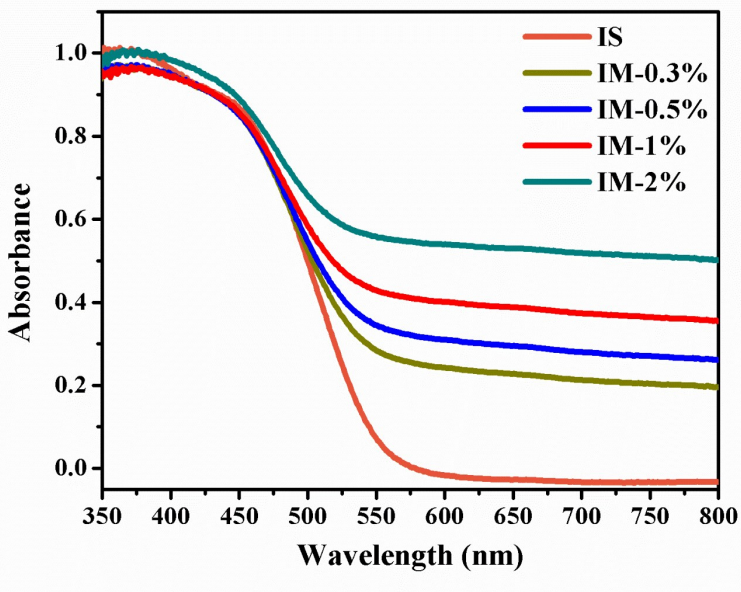

Figure S6. UV-vis diffuse reflectance spectra (DRS) of bare In_2S_3 sample and composite samples loaded with varied amount of MoS_2.

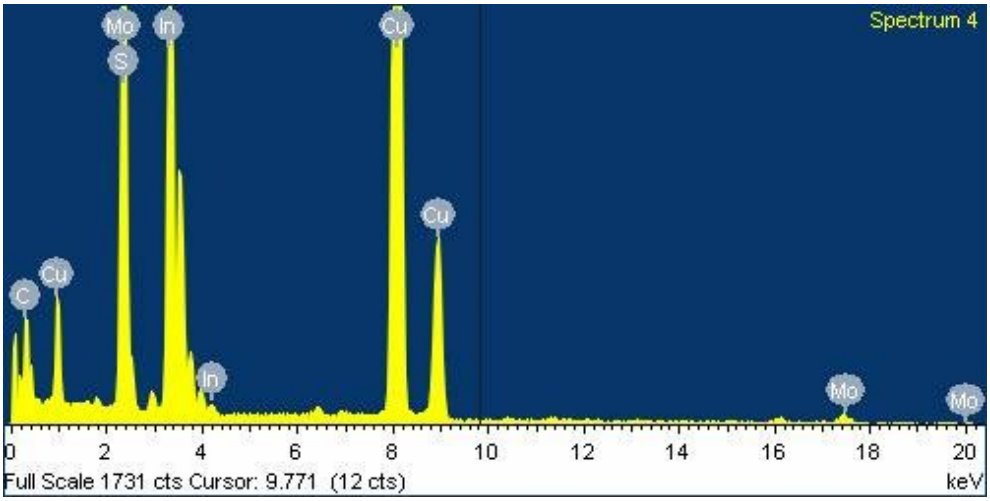

Figure S7. Energy dispersive spectroscopy (EDS) of IM-2% sample corresponding to Figure 5b.

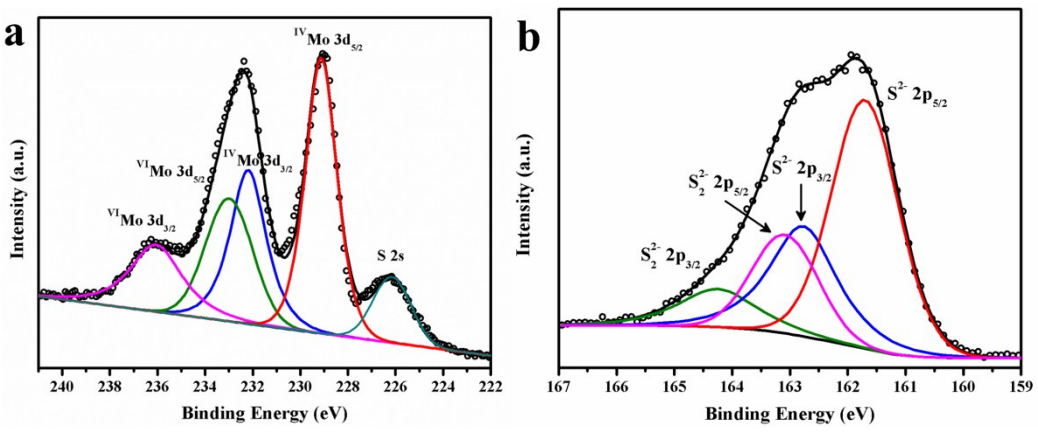

Figure S8. XPS spectra of Mo 3d orbitals (**a**) and S 2p orbitals (**b**) for defective MoS_2 (dMS).

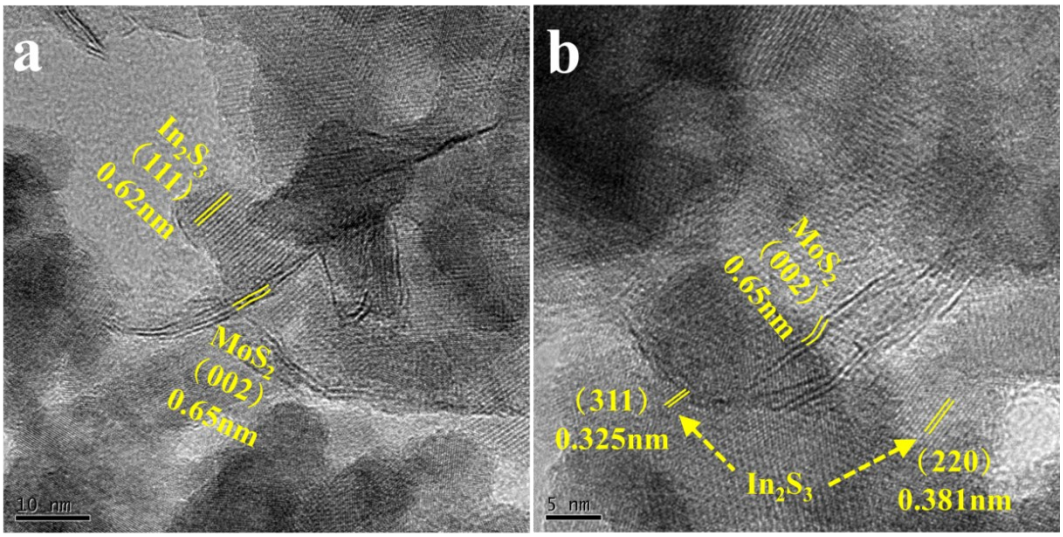

Figure S9. HRTEM images of b-IM1% sample.

Table S1. BET surface area of the samples

Sample	BET Surface Area ($m^2 \cdot g^{-1}$)
IS	64.2005
IM-0.3%	66.9216
IM-0.5%	69.4746
IM-1%	67.2880
IM-2%	68.5181
b-IS	129.2601
b-IM1%	110.2031

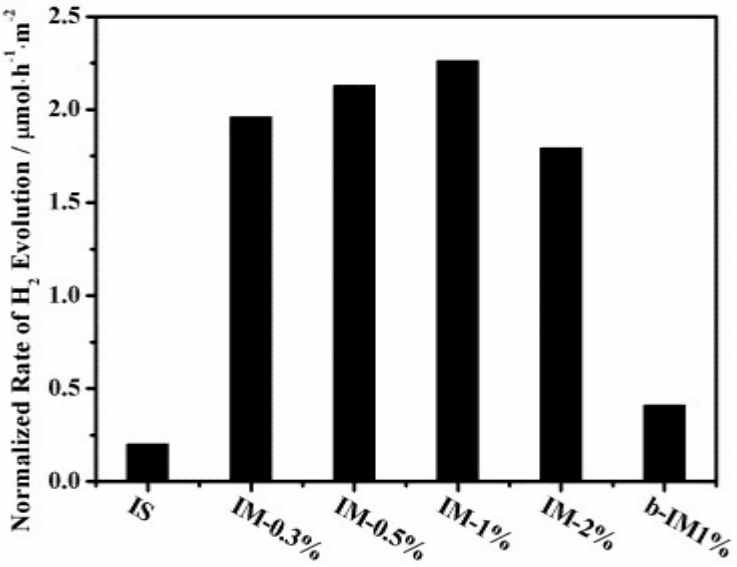

Figure S10. Surface-area-normalized rate of H_2 generation for the photocatalysts

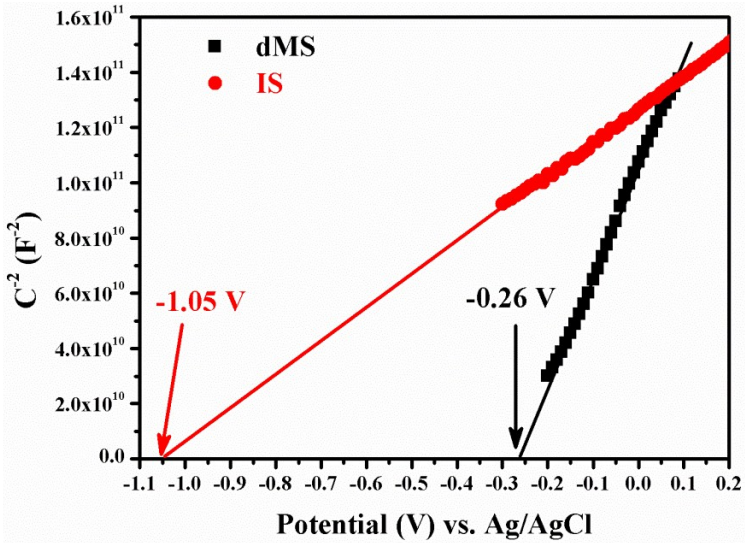

Figure S11. Mott-Schottky curves of IS electrode and dMS electrode.

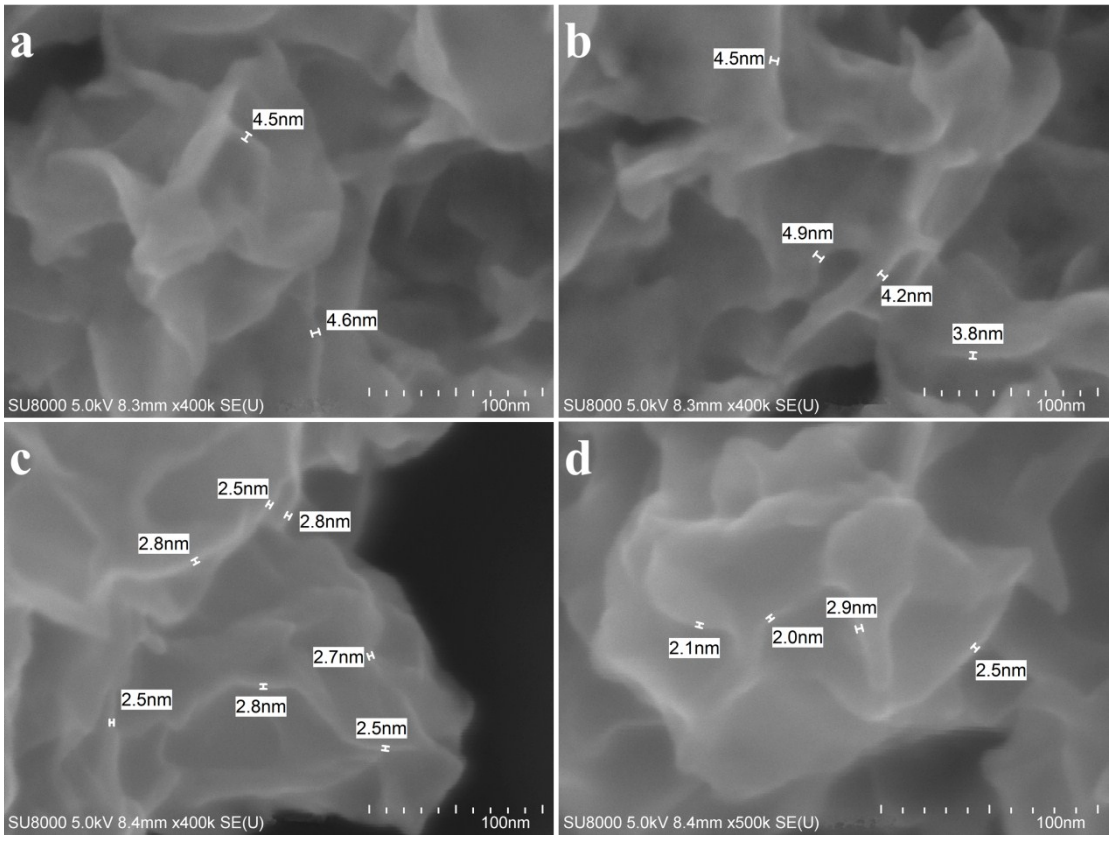

Figure S12. High-magnification SEM images of as-prepared IS (a and b) and dMS (c and d) sample for thickness measurements.

Measured from the following SEM images of IS sample (Figure S12ab in the revised ESI), the average thickness of In_2S_3 nanosheets is 4.42 nm. Since the as-prepared In_2S_3 nanosheets are not totally monodispersed but partially integrated with each other as shown in Figure 1a and Figure S1ab, it should be hard to define and measure the size of them. Likewise, the average thickness of MoS_2 nanosheets is measured to be 2.55

nm, indicating a layer number of 4 for the as-prepared MoS_2 nanosheets. This is in accord with the HRTEM results in Figure 4d and Figure S4ac, confirming the few-layered nature of the defective MoS_2.

Table S2. Data for the calculation of apparent quantum yields (AQYs) of IM-1% photocatalyst

Wavelength (nm)	Irradiance ($\mu W \cdot cm^{-2}$)	H_2 yield ($\mu L/h$)	AQY (%)
400	20599.97	59.9369	0.191
450	27962.15	62.1667	0.130
500	29000.49	22.8184	0.0413
550	31670.75	2.51	0.00379
600	34096.80	0	0
650	33753.42	0	0

The apparent quantum yield (AQY) was measured under the same photocatalytic reaction condition with incident light at different wavelengths by using band-pass filters (λ ± 15 nm) and a 300 W Xe lamp. The AQY was calculated according to the following equation:

$$AQY\ [\%] = \frac{2 \times number\ of\ H_2\ molecules}{number\ of\ incident\ photons} \times 100$$

$$= \frac{2 \times V_{H_2}/22.4 \times N_A}{\frac{I \times A \times t}{hc/\lambda}} \times 100$$

in which V_{H_2} is volume of the produced H_2, N_A is Avogadro's constant, I is the measured irradiance of incident light, A is the irradiation area (11.3 cm^{-2}), t is the irradiation time, h is Planck constant, c is the speed of light and λ is the wavelength of incident light.

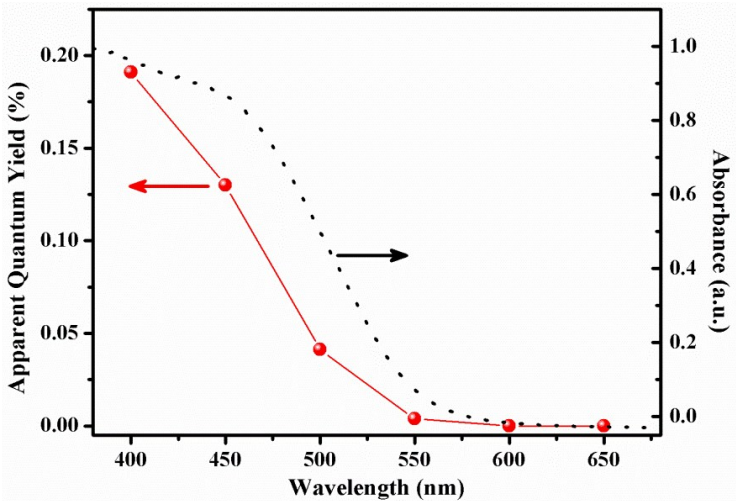

Figure S13. The light-absorbance-depended AQY distribution of IM-1% photocatalyst.

Although the AQYs of IM-1% at different monochromatic incident lights are low, a considerate dependence on the light absorbance is manifested obviously.

Table S3. Product Yields of as-prepared samples

Sample	IS	b-IS	IM-1%	b-IM1%	dMS	dMS-IM1%	cMS-IM1%
Yield	106.3%	94.0%	80.6%	89.8%	102.4%	114.2%	111.0%

Notably, yields of sample IS, dMS are greater than 100%, suggesting that the stoichiometric ratios of their composing elements should not exactly follow 2 : 3 for In : S and 1 : 2 for Mo : S due to their amorphous natures. And similar reasons can explain the cases of dMS-IM1% and cMS-IM1%, both of which undergo the fabrication process of amorphous In_2S_3 (see in Experimental Details of the revised main text).