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Supporting information

Interfacial Electron Redistribution of 2D Nanopetal $ZnIn_2S_4$ with (110) Edge-decorated Ni Clusters for Accelerated Photocatalytic H_2 Evolution

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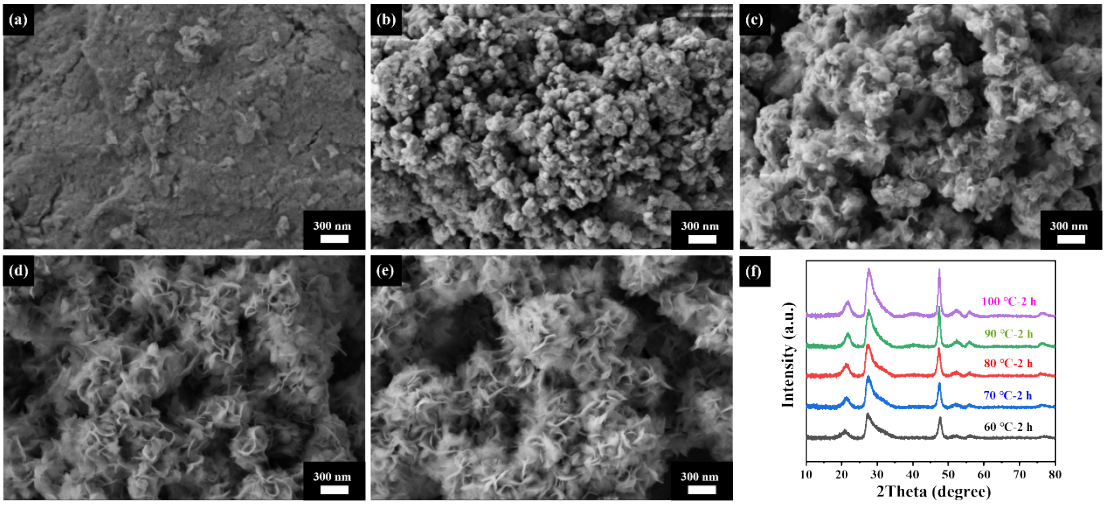


Fig. S1. SEM images of samples prepared in various temperature gradients under the same conditions for the remaining variables. (a) 60 °C-2 h, (b) 70 °C-2 h, (c) 80 °C-2 h, (d) 90 °C-2 h, (e) 100 °C-2 h, (f) XRD pattern of samples by gradient at various temperature.

The nano-flower-like spheres ZIS-H was prepared by a hydrothermal method[1]. $ZnCl_2$, $InCl_3 \cdot 4H_2O$ and thioacetamide were dispersed in 60 mL deionized water under

vigorous stirring for 10 min. After that, the mixture solution was transferred into a 100 mL Teflon lined stainless steel autoclave, which was then heated at 180 °C for 12 h. After cooling down to room temperature, the nano-flower-like spheres ZIS-H was collected by filtering and washing with deionized water and ethanol for 4 times and then dried at 60 °C overnight.

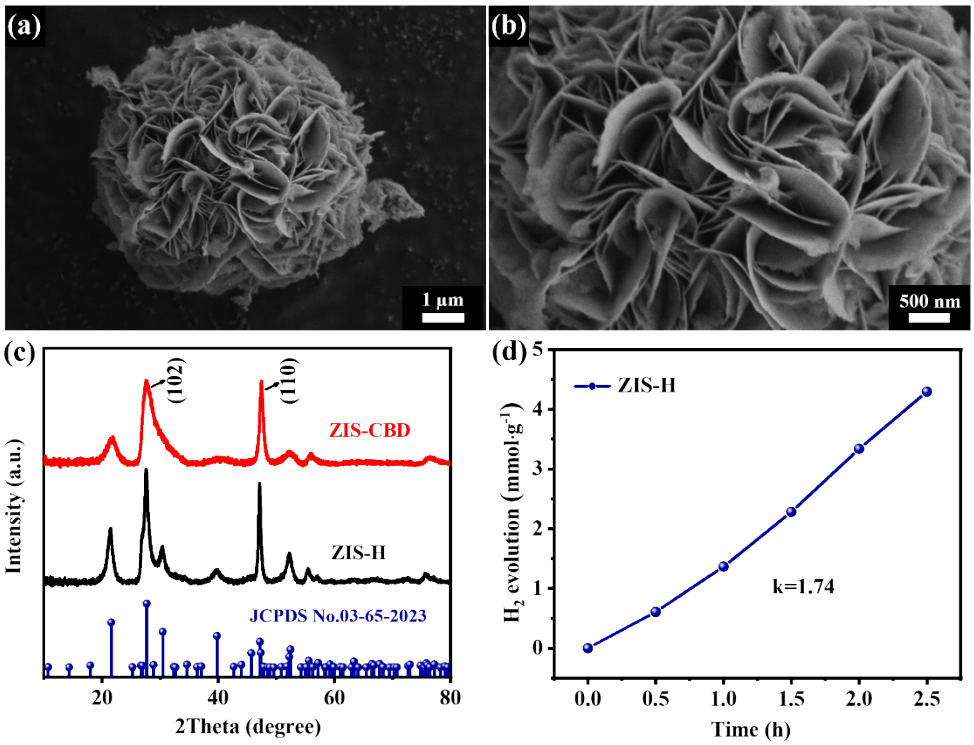


Fig. S2. (a) and (b) are SEM images of nano-flower-like spheres prepared by conventional hydrothermal method at different magnifications of ZIS-H, (c) XRD images of ZIS-H and ZIS-CBD, (d) is hydrogen production property of ZIS-H.

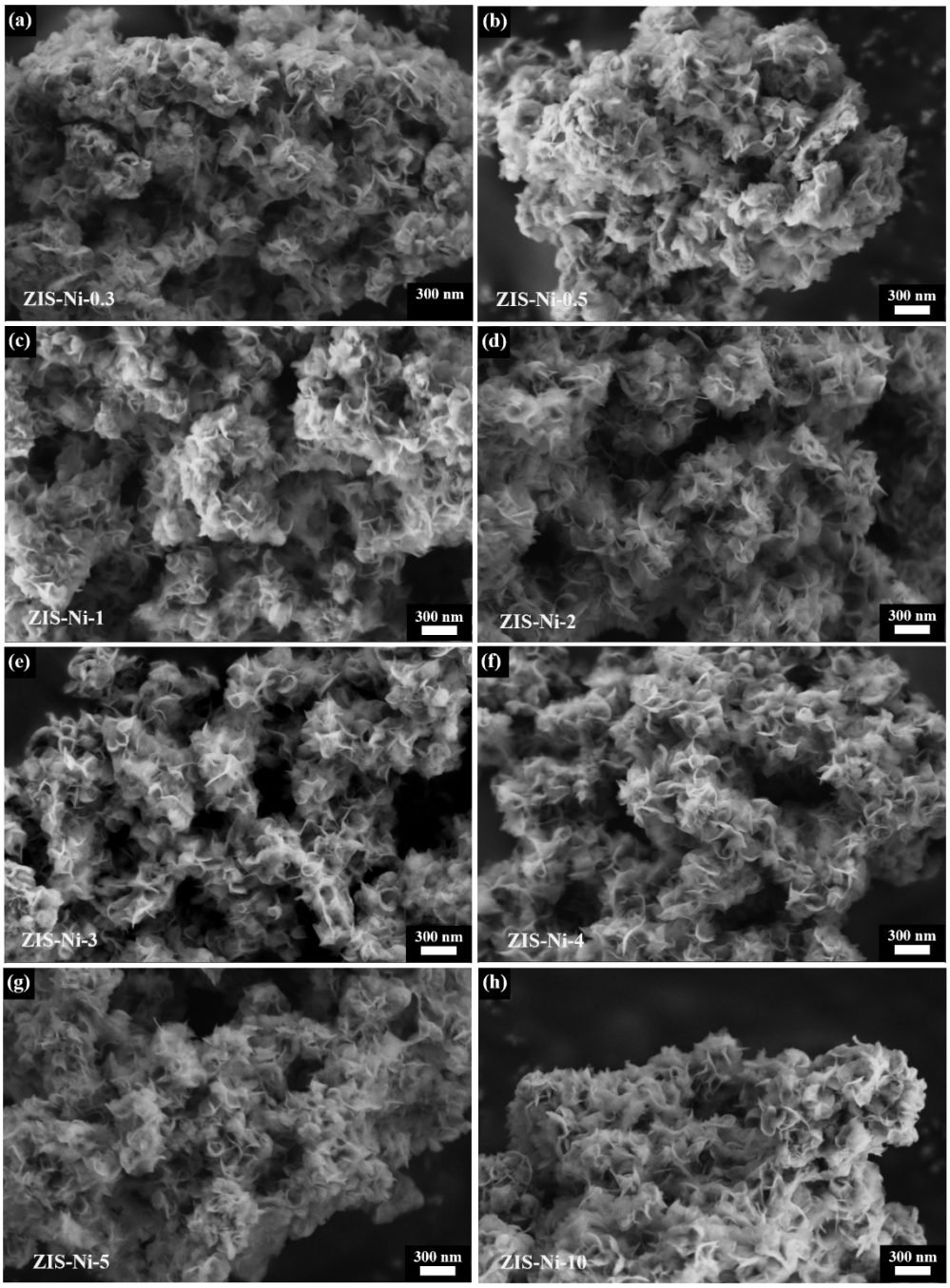


Fig. S3. SEM images of (a) ZIS-Ni-0.3; (b) ZIS-Ni-0.5; (c) ZIS-Ni-1; (d) ZIS-Ni-2; (e) ZIS-Ni-3; (f) ZIS-Ni-4; (g) ZIS-Ni-5; (h) ZIS-Ni-10.

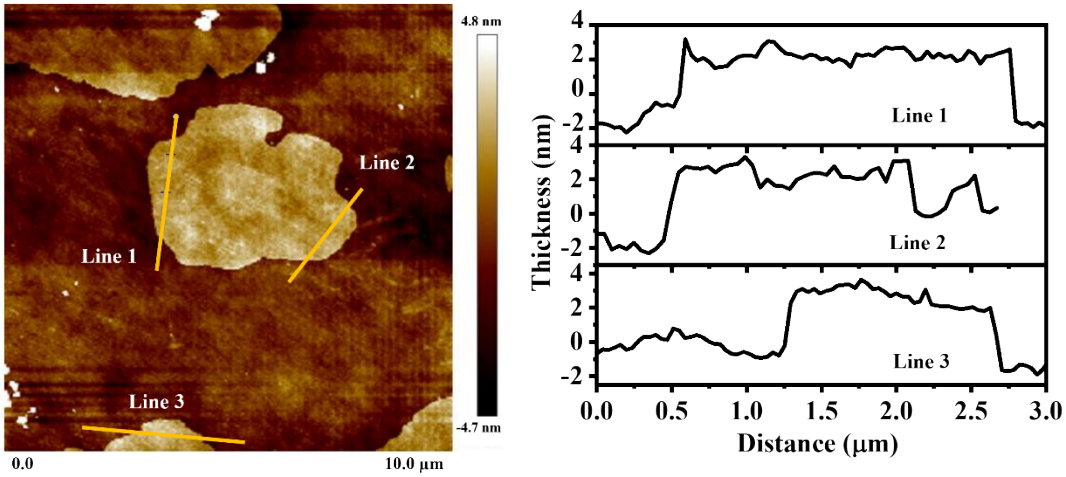


Fig. S4. AFM topographic image of prepared ZIS nanosheet with corresponding height profiles for Line1-Line3.

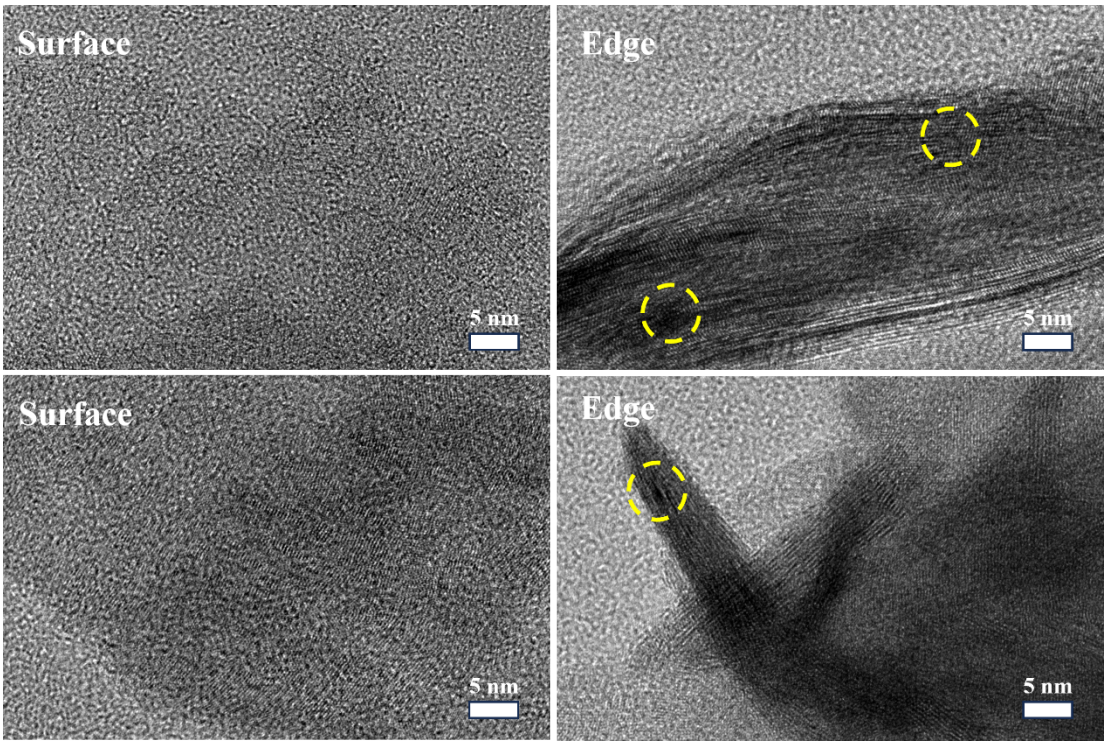
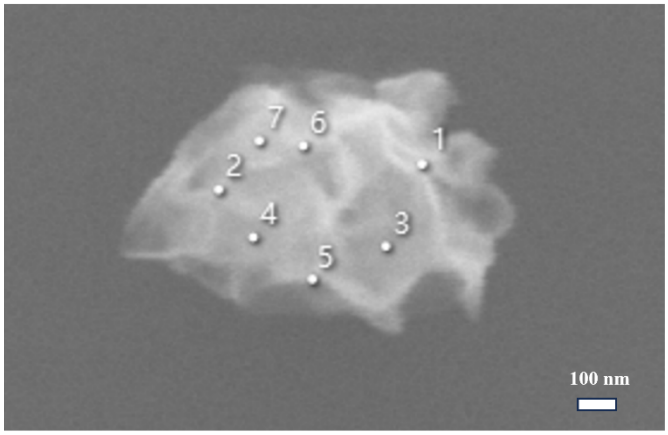


Fig. S5. Comparison of ZIS-Ni-3 surface and edge via HRTEM images.



EDS-point	Ni (wt%)
1	7.23
2	6.67
3	0.00
4	4.50
5	7.40
6	5.10
7	1.40

Fig. S6. EDS spot analysis and Ni content at each point.

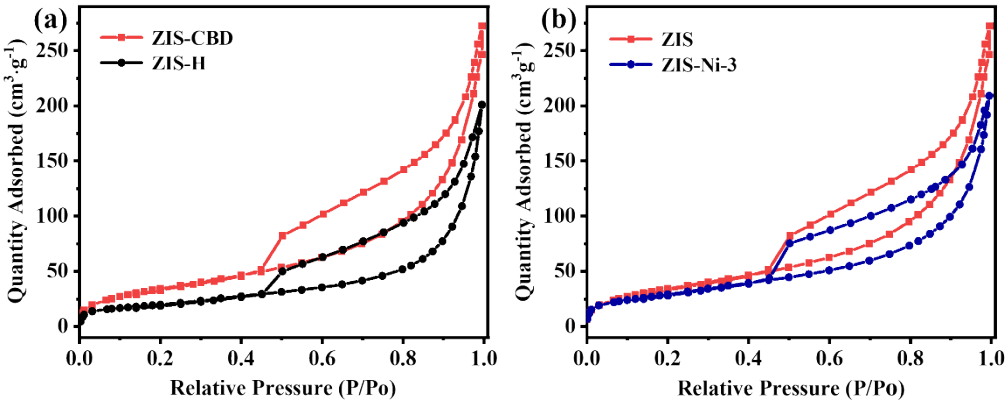


Fig. S7. The N_2 adsorption–desorption isotherms of (a) ZIS-CBD and ZIS-H, (b) ZIS and ZIS-Ni-3.

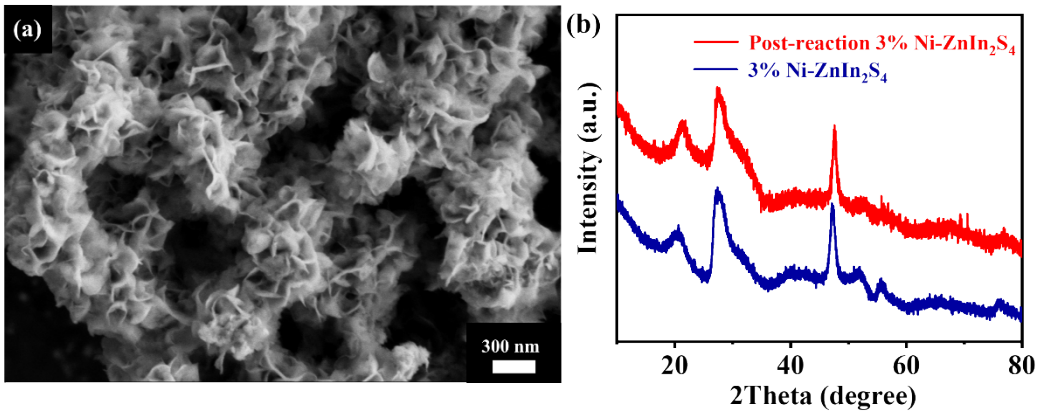


Fig. S8. ZIS-Ni-3 SEM (a) and XRD (b) plot after cyclic hydrogen production testing.

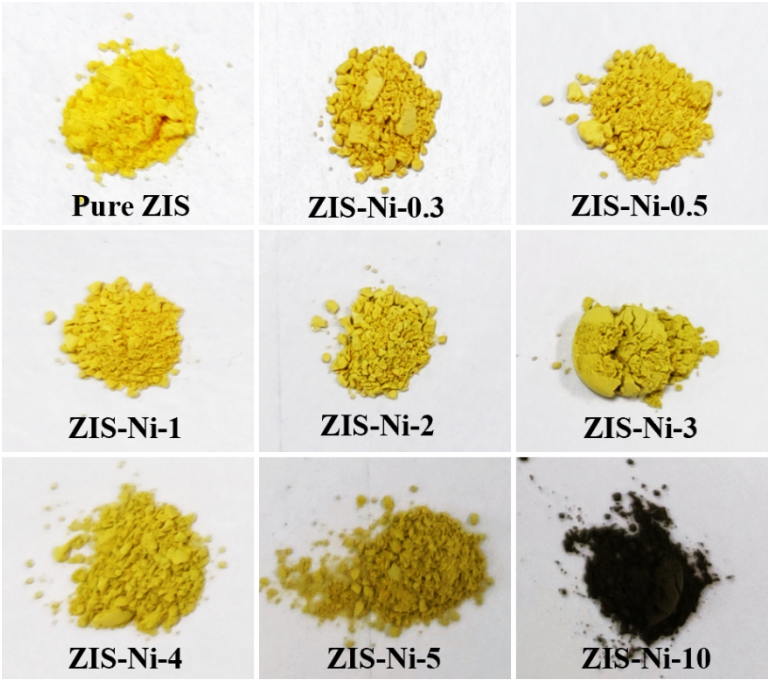


Fig. S9. Digital images of ZIS-Ni photocatalysts with different Ni contents.

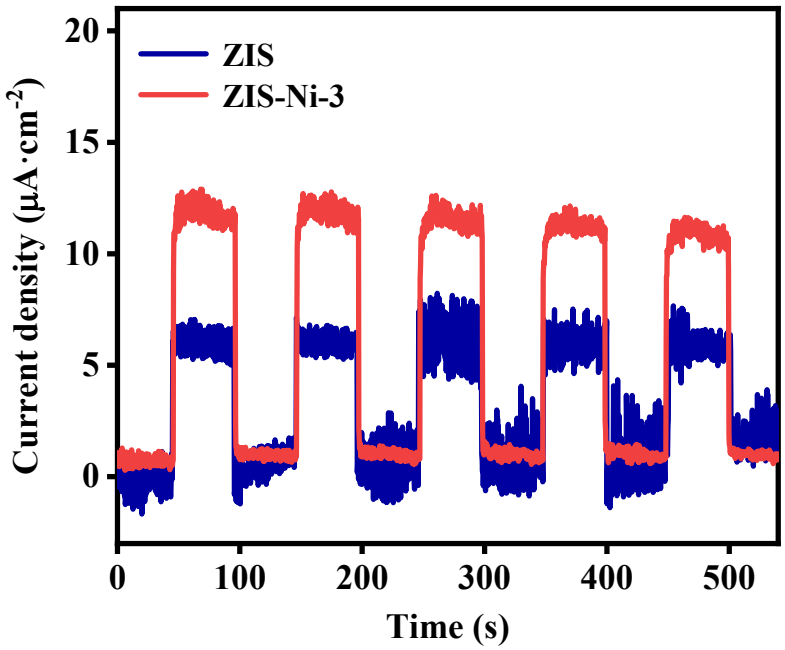


Fig. S10. The transient photocurrent responses of ZIS (a) and ZIS-Ni-3 (b).

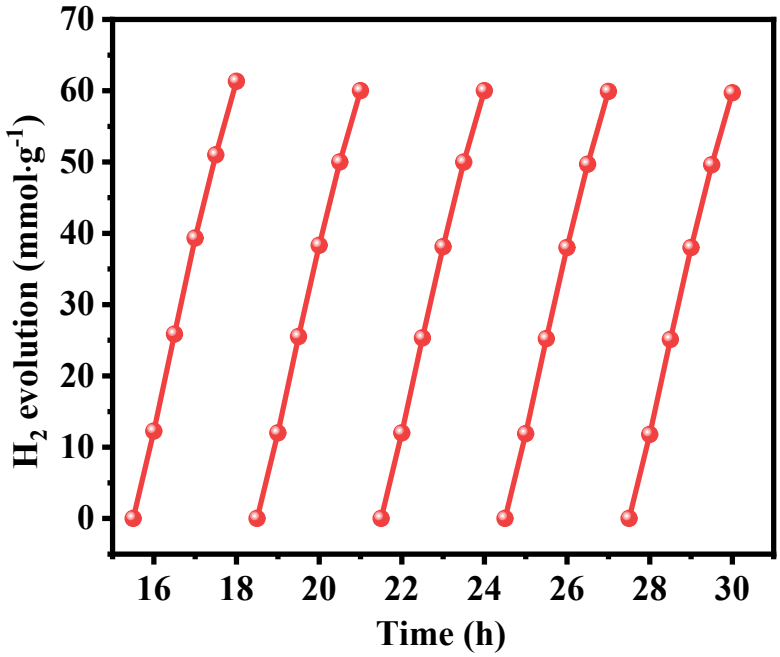


Fig. S11. Cyclic stability testing of optimized ZIS-Ni-3.

Table S1. The content of Ni determined by ICP-OES.

Sample	Ni (wt%) ICP-OES	Ni (wt%) theoretical
ZIS-Ni-0.3	0.27	0.3
ZIS-Ni-0.5	0.46	0.5
ZIS-Ni-1	0.61	1
ZIS-Ni-2	1.11	2
ZIS-Ni-3	1.50	3
ZIS-Ni-4	1.87	4
ZIS-Ni-5	2.32	5
ZIS-Ni-10	4.63	10

Table S2. Comparison of the hydrogen production performance of $ZnIn_2S_4$-based photocatalytic materials.

Catalysts	**Light source**	**Sacrificial agent**	**H_2 ($mmol \cdot g^{-1} \cdot h^{-1}$)**	**AQY (%) @ 420 nm**	**Ref.**
Ni atom-$ZnIn_2S_4$	300 W Xenon lamp (λ>420 nm)	10 vol% TEOA	4.22	19.70	[2]
$ZnIn_2S_4/TiO_2$	300 W Xenon lamp	TEOA	6.03	10.49	[3]
Ni/In-doped $ZnIn_2S_4$	300 W Xenon lamp (λ>420 nm)	10 vol% TEOA	10.97	-	[4]
NiS/CdS/g-C_3N_4	300 W Xenon lamp (λ>400 nm)	20 vol% TEOA	3.02	5.90	[5]
$ZnIn_2S_4$@SiO_2@TiO_2	300 W Xenon lamp (AM 1.5 filter)	10 vol% TEOA	0.62	17.56	[6]
NiS/ZnIn2S_4/g-C_3N_4	full-spectrum light irradiation	20 vol% TEOA	5.02	30.50	[7]
Ultrathin $ZnIn_2S_4$	300 W Xenon lamp (λ>420 nm)	20 vol% TEOA	1.94	10.10	[8]
NiSAs/$ZnIn_2S_4$	300 W Xenon lamp (λ>420 nm)	10 vol% TEOA	1.79	9.60	[9]
$NiSe_2/ZnIn_2S_4$	300 W Xenon lamp (λ>420 nm)	10 vol% TEOA	1.49	6.10	[10]
ZIS-Ni	300 W Xenon lamp (AM 1.5 filter)	10 vol% TEOA	26.80	61.68	This work

Table S3. PL decay spectroscopy specific values of the parameters obtained after the fit.

	A_1	τ_1(ns)	A_2	τ_2(ns)
ZIS	0.1188	11.8955	113.8656	1.0331
ZIS-Ni3.0	0.2919	5.3188	338.9376	0.8524

Reference

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