

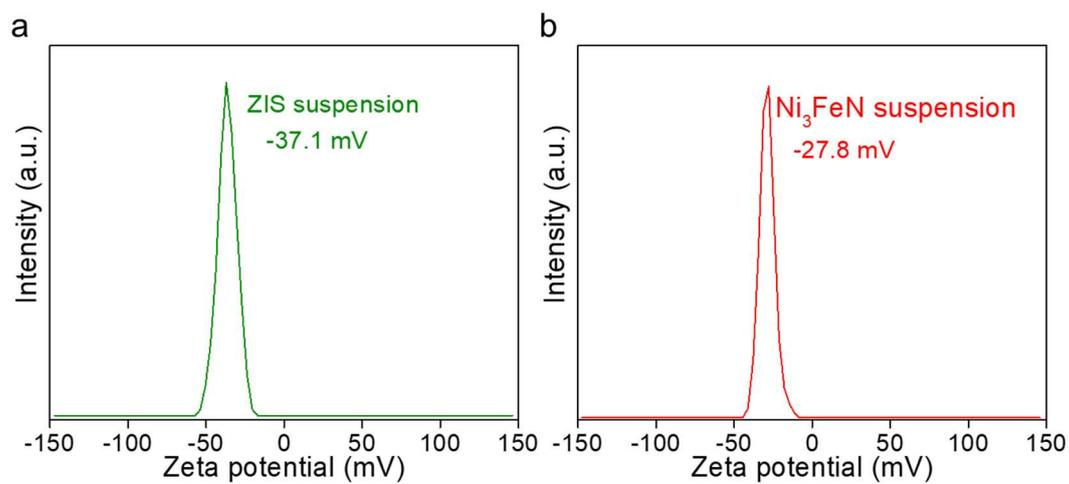
## Ultrathin $\text{ZnIn}_2\text{S}_4$ Nanosheets Supported Metallic $\text{Ni}_3\text{FeN}$ for Photocatalytic Coupled Selective Alcohol Oxidation and $\text{H}_2$ Evolution

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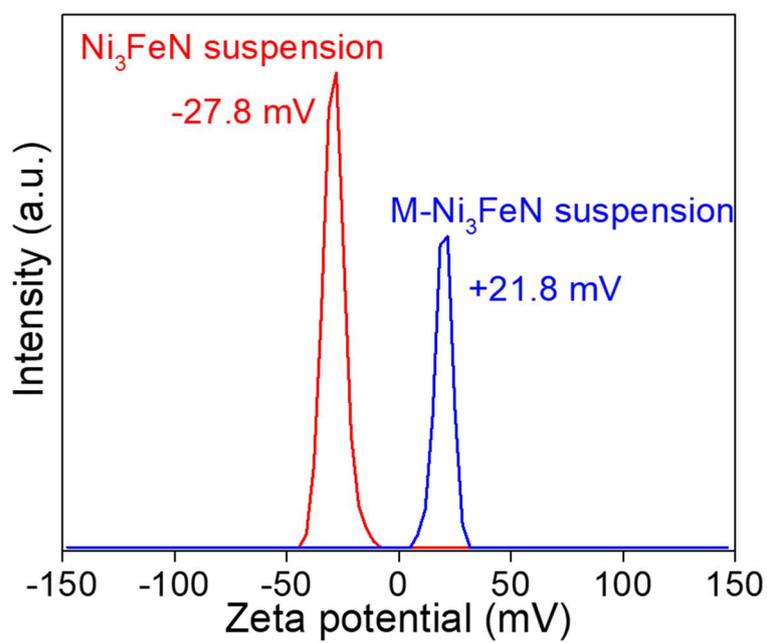
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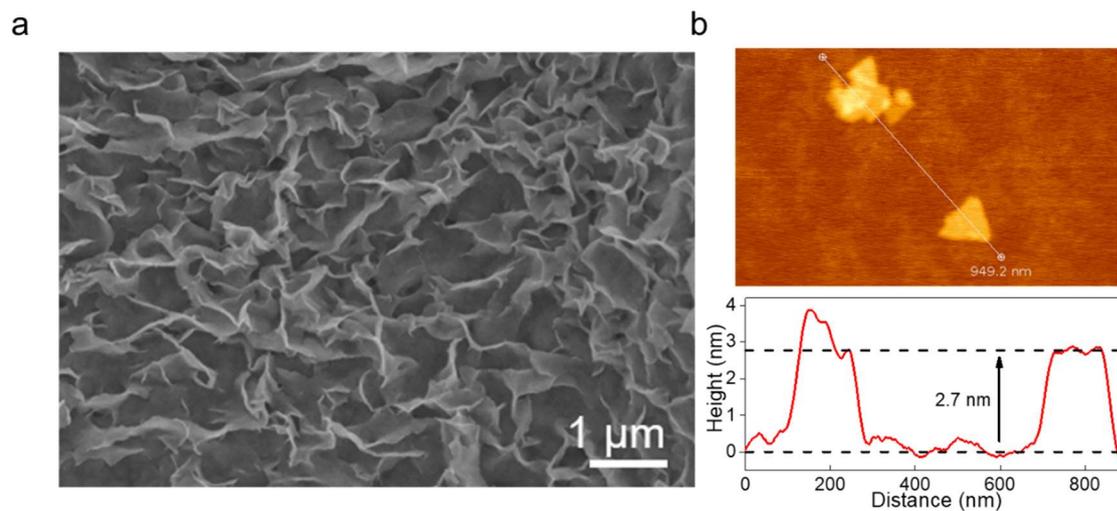
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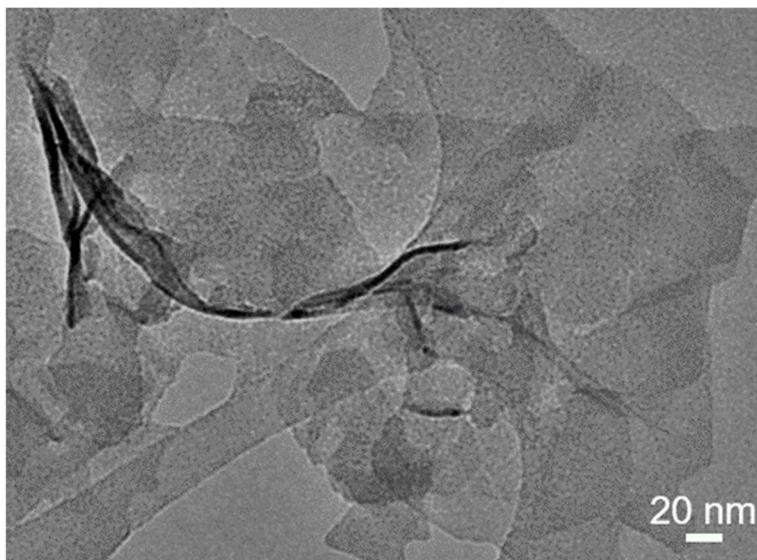
**Figure S1.** Zeta potential of ZIS and Ni<sub>3</sub>FeN suspension dispersed in DI water.



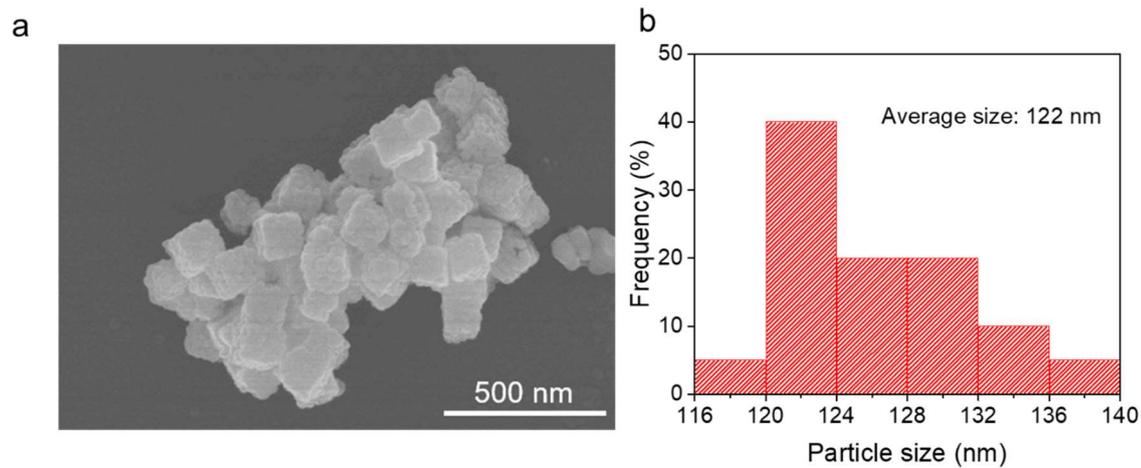
**Figure S2.** Zeta potential of Ni<sub>3</sub>FeN suspension dispersed in DI water before and after surfactant APTES modification.



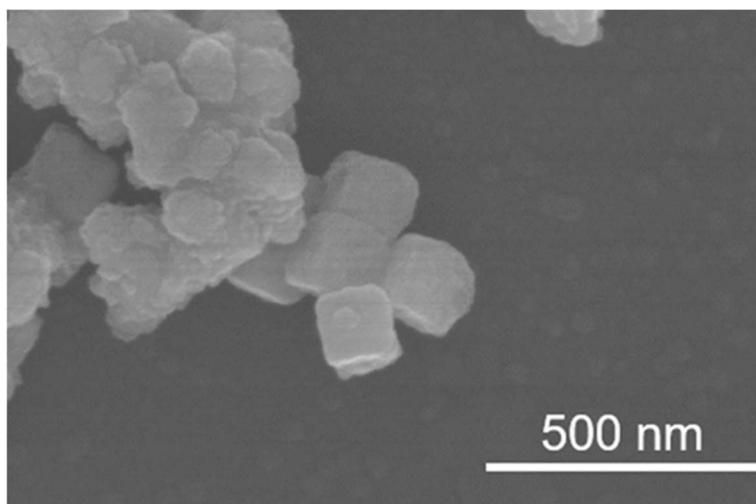
**Figure S3.** (a) SEM image and (b) AFM image of ZIS.



**Figure S4.** TEM image of bare ZIS.



**Figure S5.** (a) SEM image and (b) the statistic histogram of the size distribution of Ni<sub>3</sub>FeN NPs.



**Figure S6.** SEM of Ni<sub>3</sub>FeN after APTES modification.

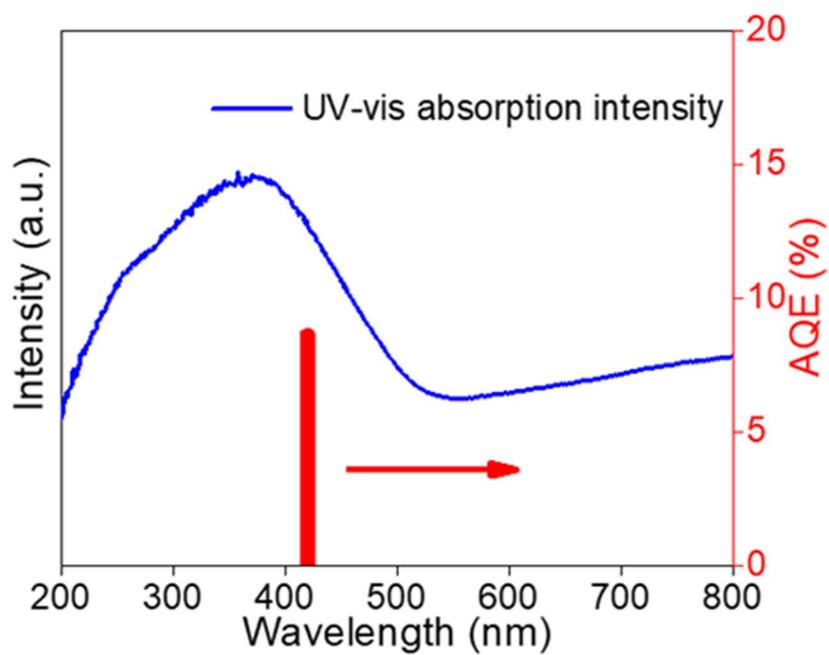
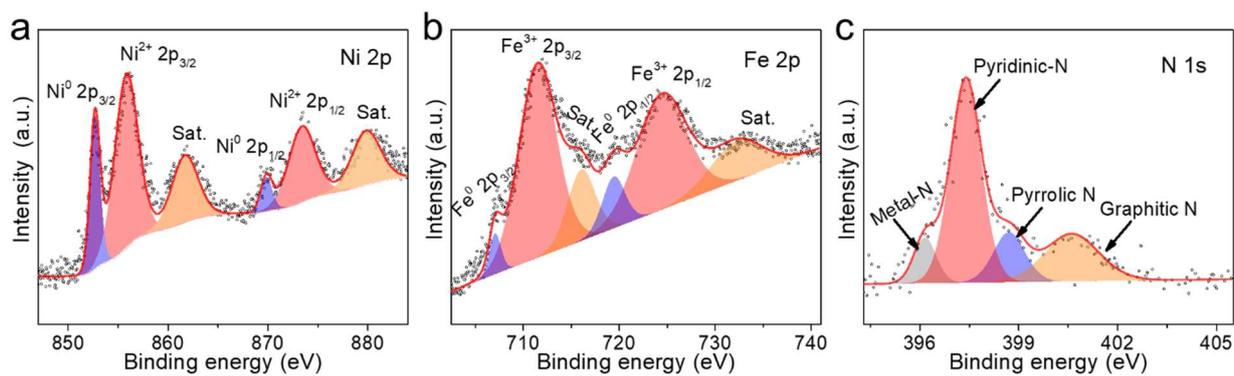
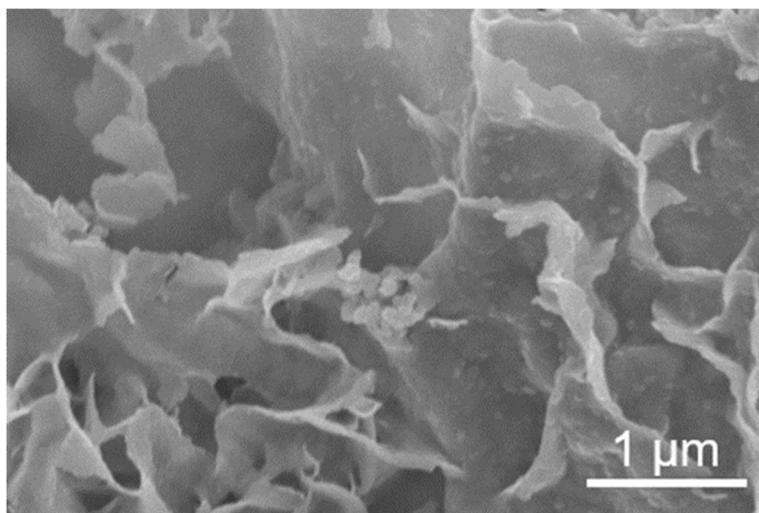


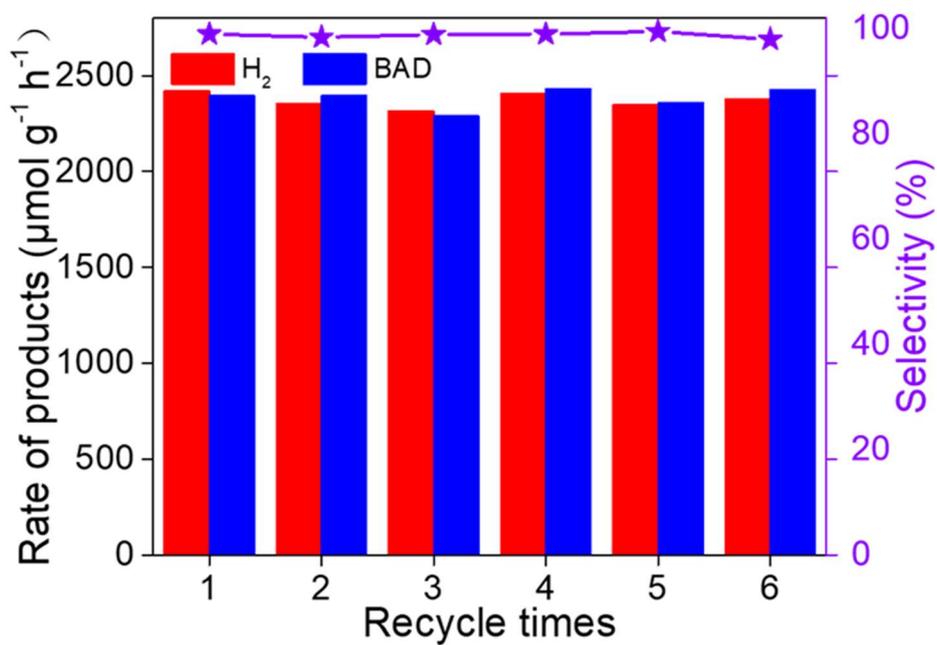
Figure S7. The apparent quantum efficiency at 420 nm of ZIS/1.5% M-Ni<sub>3</sub>FeN composite.



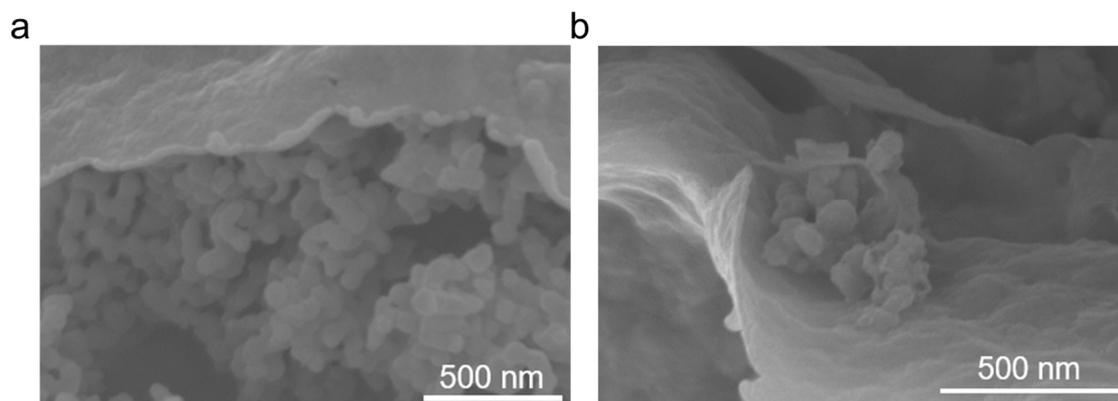
**Figure S8.** XPS spectra of (a) Ni 2p, (b) Fe 2p and (c) N 1s of ZIS/M-Ni<sub>3</sub>FeN composite after the photoactivity test.



**Figure S9.** SEM image of ZIS/M-Ni<sub>3</sub>FeN composite after the photoactivity test.



**Figure S10.** Photocatalytic cyclic test of anaerobic oxidation of benzyl alcohol to produce benzaldehyde and H<sub>2</sub> over ZIS/1.5% M-Ni<sub>3</sub>FeN composite.



**Figure S11.** SEM image of ZIS/P-Ni<sub>3</sub>FeN composite.

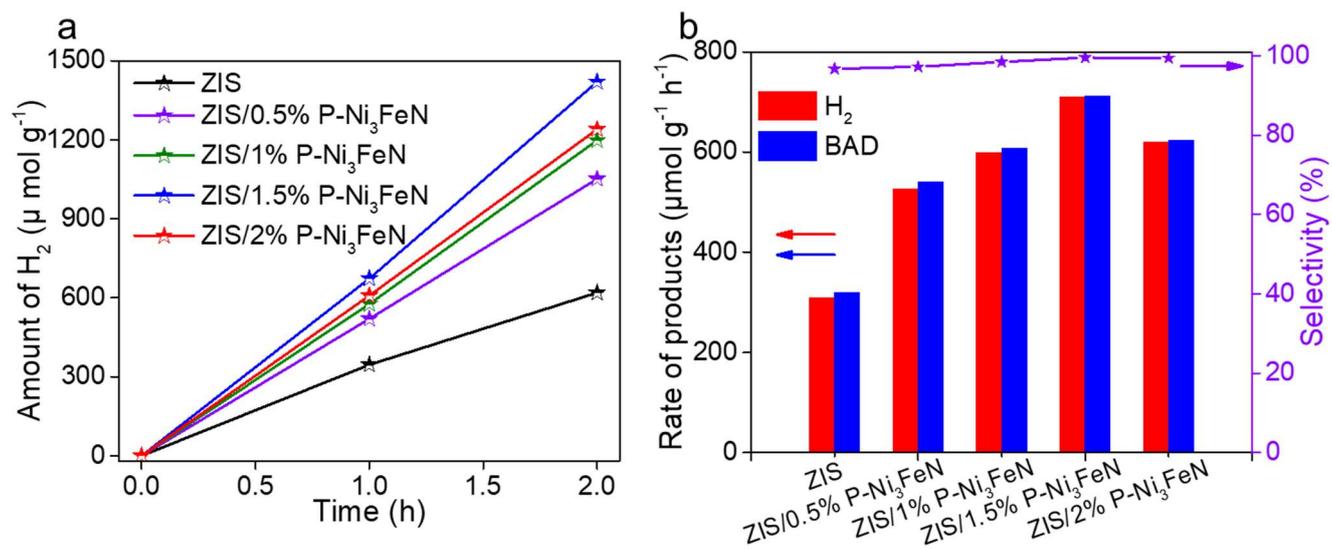


Figure S12. Photocatalytic activity for selective BA oxidation integrated with H<sub>2</sub> evolution over ZIS and ZIS/x% P-Ni<sub>3</sub>FeN hybrids.

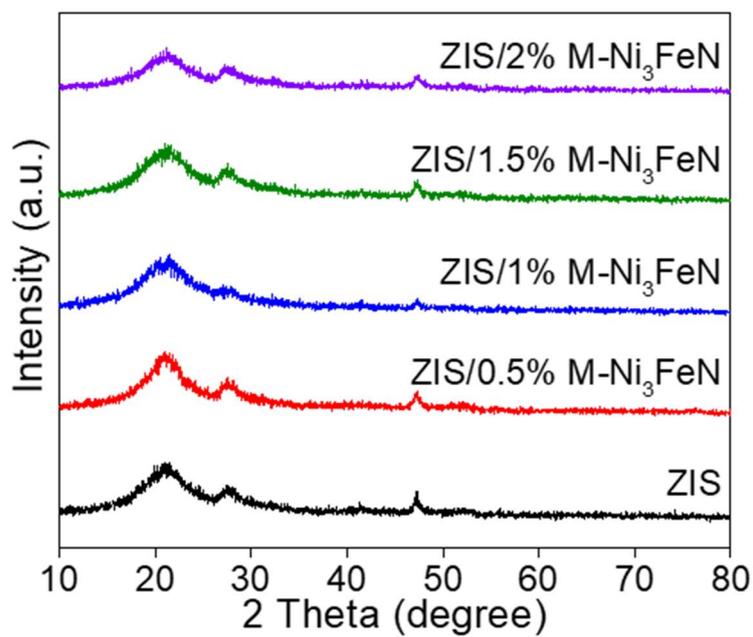


Figure S13. XRD patterns of ZIS and ZIS/x% M-Ni<sub>3</sub>FeN hybrids.

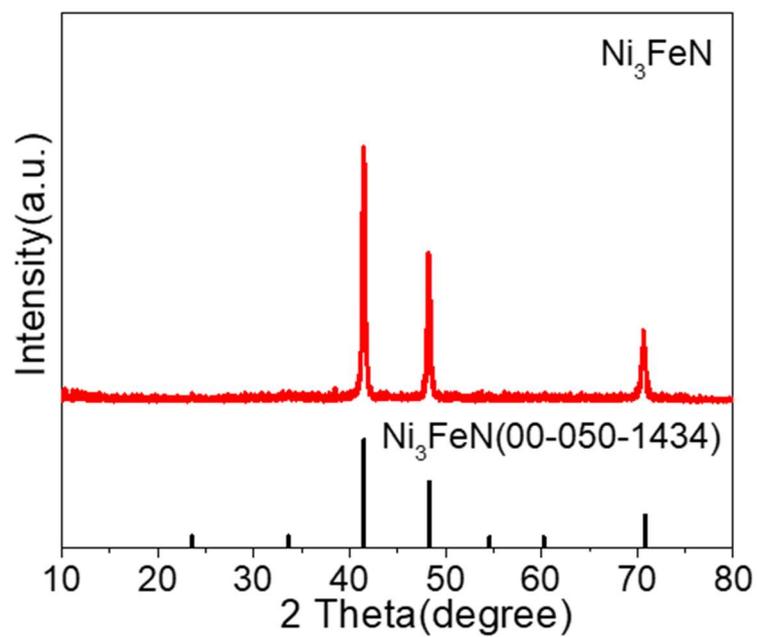


Figure S14. XRD pattern of Ni<sub>3</sub>FeN NPs.

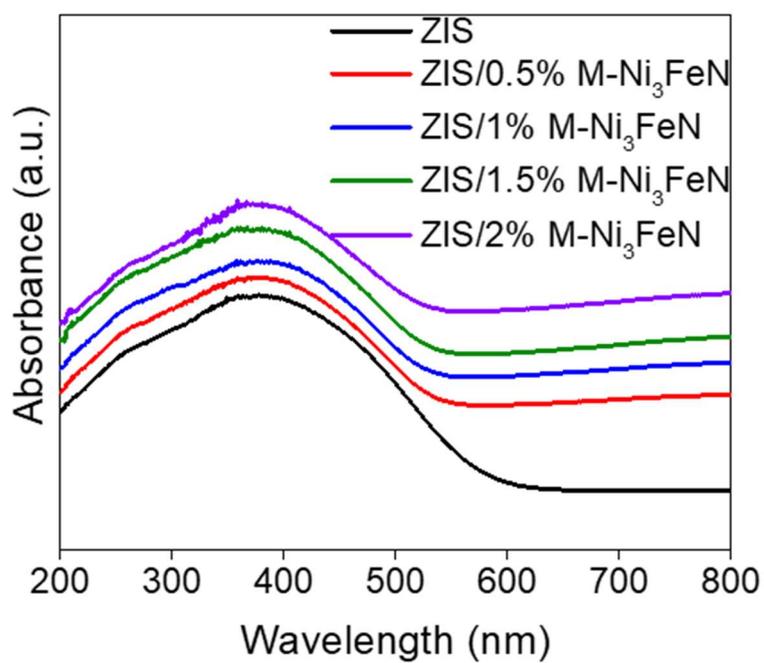


Figure S15. DRS spectra of ZIS and ZIS/x% M-Ni<sub>3</sub>FeN hybrids.

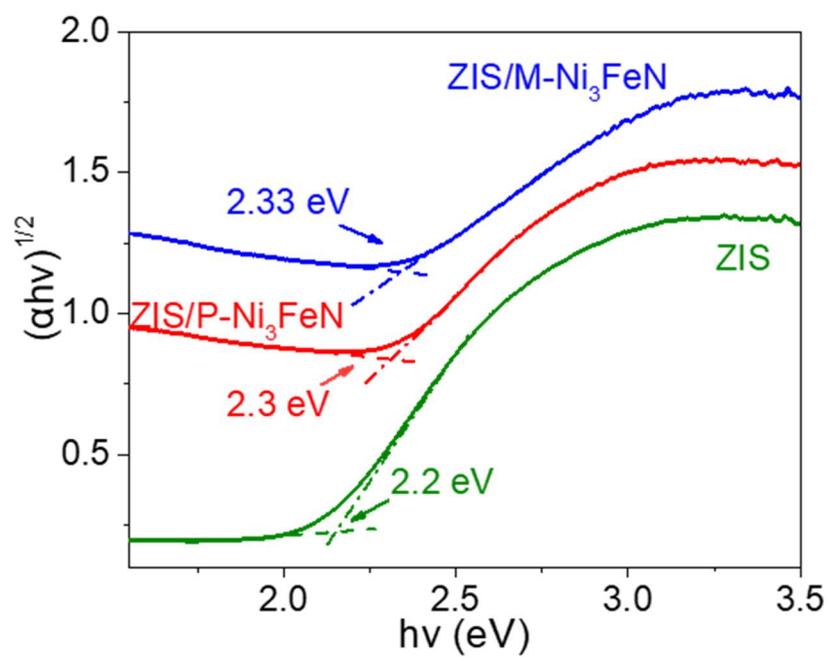


Figure S16. Tauc Plots of the  $(\alpha hv)^{1/2}$  vs. photon energy ( $h\nu$ ) of bare ZIS, ZIS/P-Ni<sub>3</sub>FeN and ZIS/M-Ni<sub>3</sub>FeN.

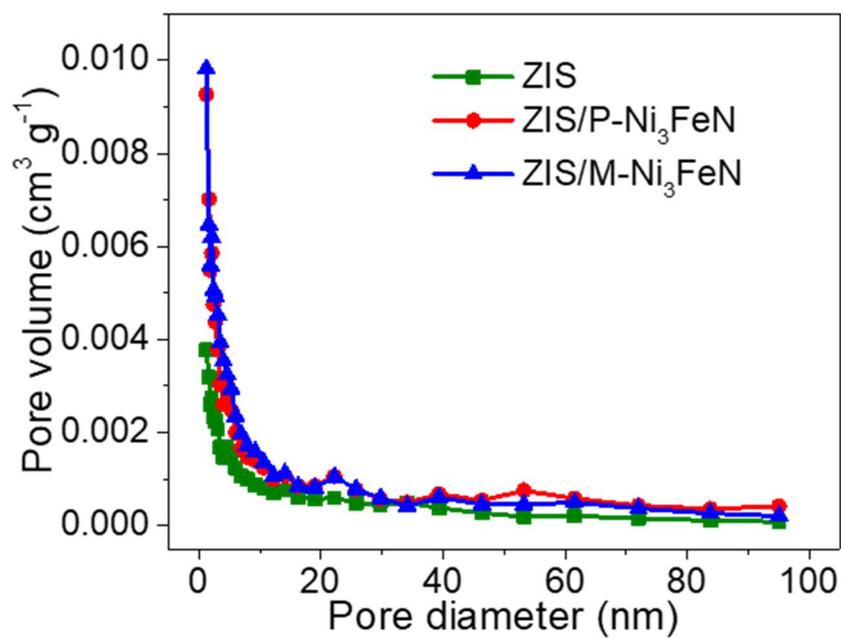


Figure S17. BJH pore size distributions of bare ZIS, ZIS/P-Ni<sub>3</sub>FeN and ZIS/M-Ni<sub>3</sub>FeN.

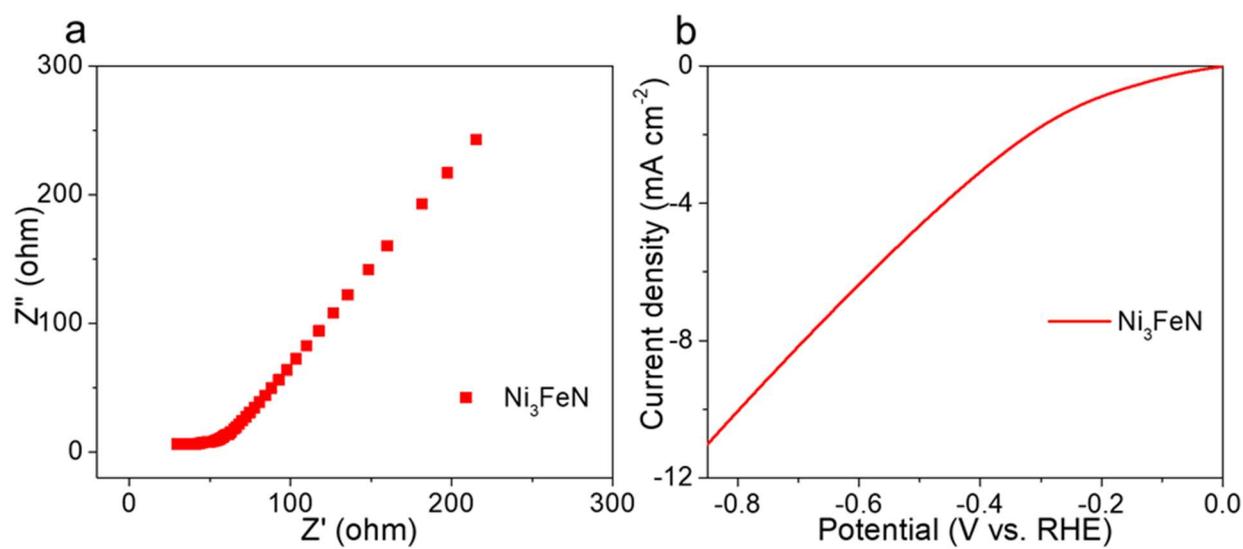
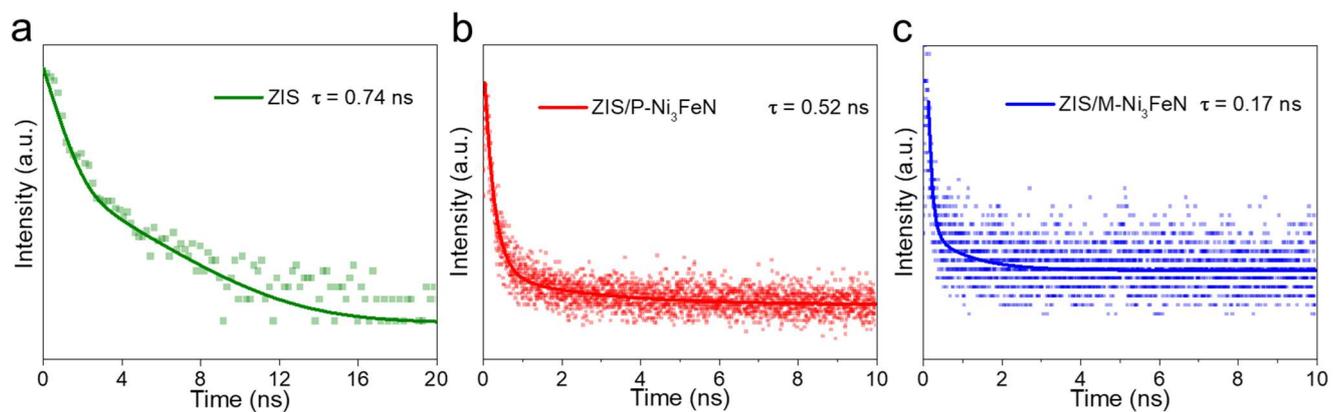
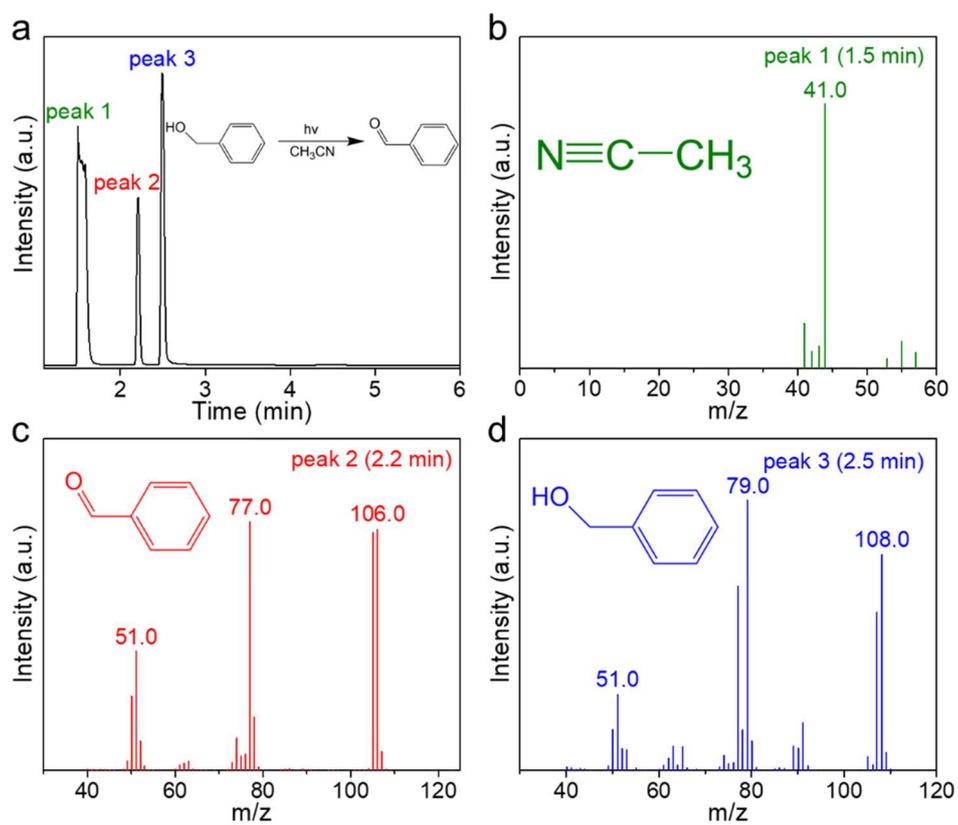


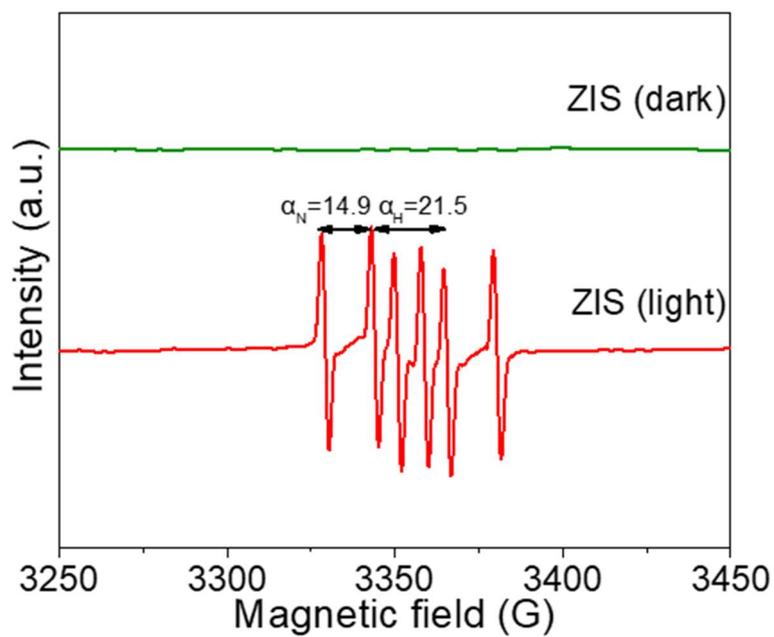
Figure S18. EIS Nyquist plot and LSV curve of Ni<sub>3</sub>FeN.



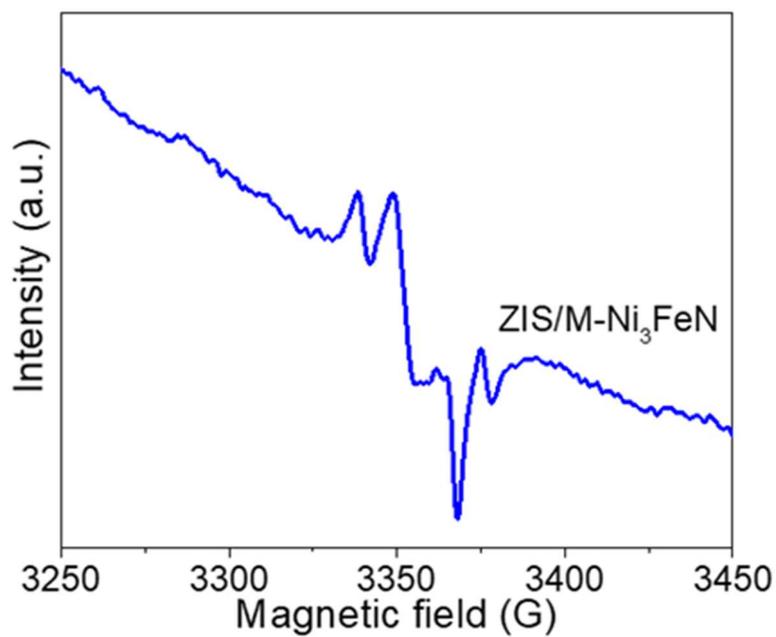
**Figure S19.** Transient PL spectra of ZIS, ZIS/P-Ni<sub>3</sub>FeN and ZIS/M-Ni<sub>3</sub>FeN composite.



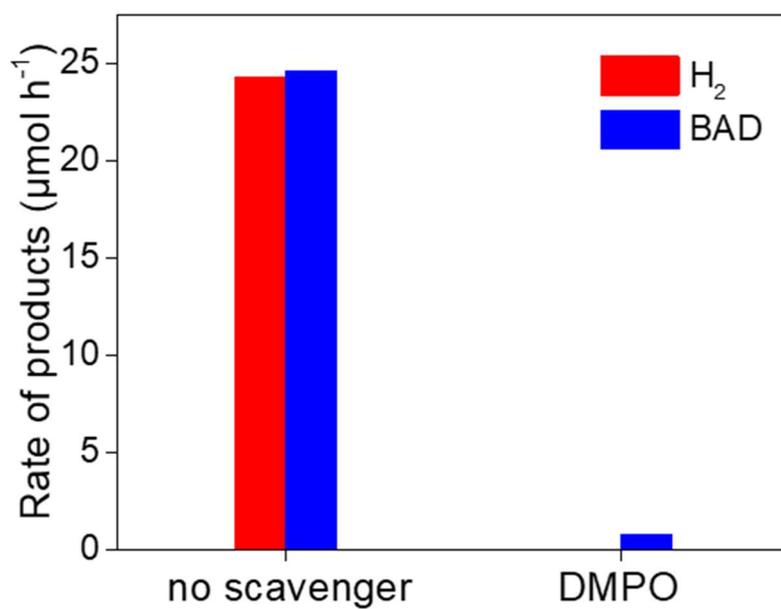
**Figure S20.** GC-MS spectra of the reaction solution after photocatalytic experiment. Conditions: 100  $\mu\text{mol}$  benzyl alcohol, 10 mg ZIS/1.5% M-Ni<sub>3</sub>FeN, 3 mL  $\text{CH}_3\text{CN}$ ,  $\lambda > 420$  nm.



**Figure S21.** EPR spectrum of ZIS in the presence of DMPO in BA-CH<sub>3</sub>CN solution under an Ar atmosphere.



**Figure S22.** EPR spectrum of ZIS/M-Ni<sub>3</sub>FeN composite in the presence of DMPO in BA-CH<sub>3</sub>CN solution under an Ar atmosphere.



**Figure S23.** The effect of DMPO on the photocatalytic performance of H<sub>2</sub> evolution and BAD generation over ZIS/1.5% M-Ni<sub>3</sub>FeN composite.

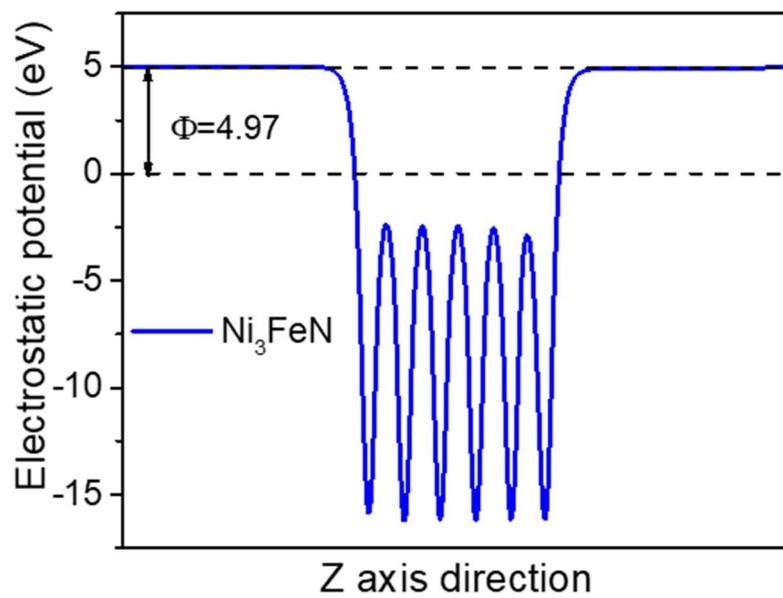


Figure S24. Work function of Ni<sub>3</sub>FeN.

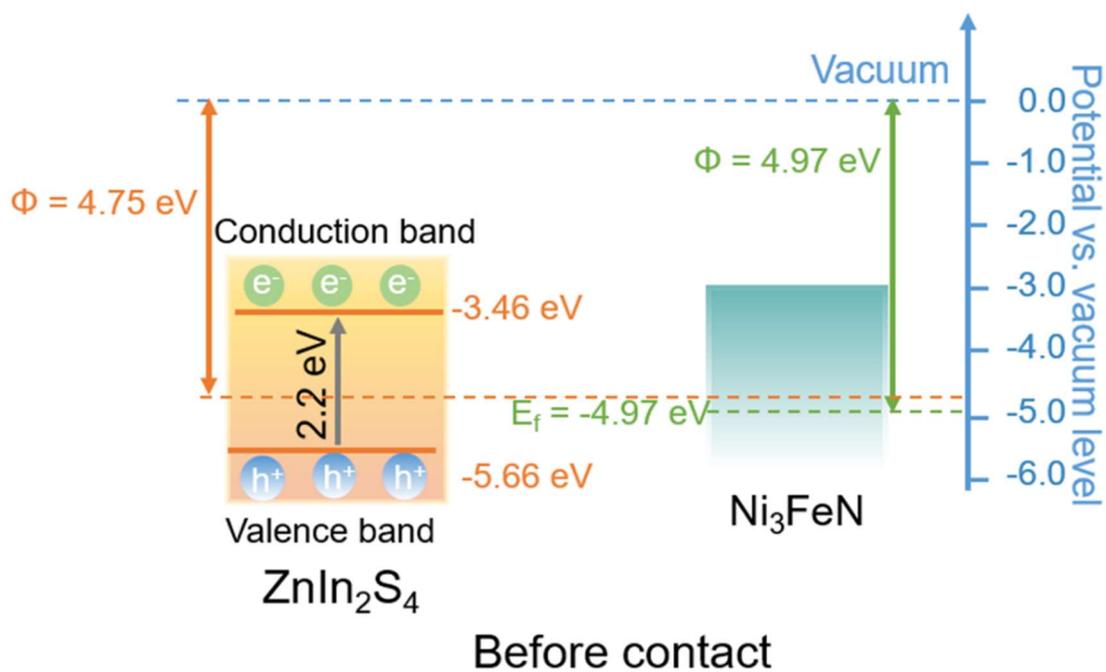


Figure S25. Energy band diagram of ZIS and Ni<sub>3</sub>FeN before contact.

**Table 1.** Physicochemical Properties of Bare ZIS, ZIS/P-Ni<sub>3</sub>FeN and ZIS/M-Ni<sub>3</sub>FeN

Samples	S <sub>BET</sub> (m <sup>2</sup> g <sup>-1</sup> )	Total pore volume (cm <sup>3</sup> g <sup>-1</sup> )	Mean pore diameter (nm)
ZIS	11.59	0.04	14.24
ZIS/P-Ni <sub>3</sub> FeN	27.39	0.08	12.16
ZIS/M-Ni <sub>3</sub> FeN	35.34	0.08	9.51

**Table 2.** Comparison of Performance towards Photocatalytic Coupled Selective Alcohol Oxidation and H<sub>2</sub> Evolution between ZnIn<sub>2</sub>S<sub>4</sub>/Ni<sub>3</sub>FeN and Some Recently Developed Photocatalysts

Photocatalyst	Solvent	Light source	Reaction	Selectivity (%)	H <sub>2</sub> (μmol g <sup>-1</sup> h <sup>-1</sup> )	Ref.
ZnIn <sub>2</sub> S <sub>4</sub> /Ni <sub>3</sub> FeN	CH <sub>3</sub> CN	λ > 420 nm	BA to BAD	99	2427.9	This work
ZnS-Ni <sub>x</sub> S <sub>y</sub>	H <sub>2</sub> O	λ > 200 nm	BA to BAD	90.5	2943	(1)
Pt-g-C <sub>3</sub> N <sub>4</sub>	H <sub>2</sub> O	λ > 400 nm	BA to BAD	90	255	(2)
ZnIn <sub>2</sub> S <sub>4</sub> /CeO <sub>2</sub>	CH <sub>3</sub> CN	AM 1.5G	BA to BAD	96	1496.6	(3)
CdS(ZB)/CdS (WZ)/Ni-BTC	H <sub>2</sub> O	λ > 420 nm	BA to BAD	90.2	2891	(4)
Pd/HNb <sub>3</sub> O <sub>8</sub> -H <sub>2</sub>	H <sub>2</sub> O	Xe lamp	BA to BAD	99	3170	(5)
VN-UP-CN	H <sub>2</sub> O	λ > 420 nm	BA to BAD	98	196.08	(6)
W <sub>SA</sub> -CN-PUNS	H <sub>2</sub> O	λ > 420 nm	BA to BAD	93.6	298.7	(7)
MoS <sub>2</sub> /ZnIn <sub>2</sub> S <sub>4</sub>	H <sub>2</sub> O	λ > 420 nm	BA to BAD	-	3880	(8)
WO <sub>3</sub> /ZnIn <sub>2</sub> S <sub>4</sub>	H <sub>2</sub> O	UV-vis	BA to BAD	94.8	4280	(9)
Co-ZnIn <sub>2</sub> S <sub>4</sub>	H <sub>2</sub> O	λ > 420 nm	BA to BAD	-	2823.7	(10)
CdS/MIL-53(Fe)	CH <sub>3</sub> CN	λ > 420 nm	BA to BAD	99	2334	(11)
O-ZnIn <sub>2</sub> S <sub>4</sub> /TiO <sub>2-x</sub>	H <sub>2</sub> O	λ > 420 nm	BA to BAD	-	2584.9	(12)

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