Supporting information

Ultrathin MOF Coupling with Molecular Cobaloxime to Construct an Efficient Hybrid Hematite Photoanode for Photocatalytic Water Splitting

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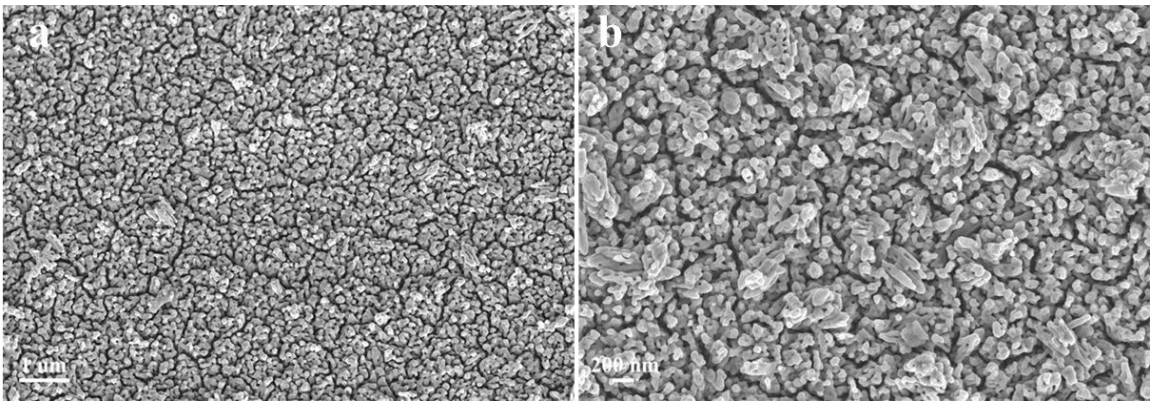

Figure S1. SEM images of Ti-PH.

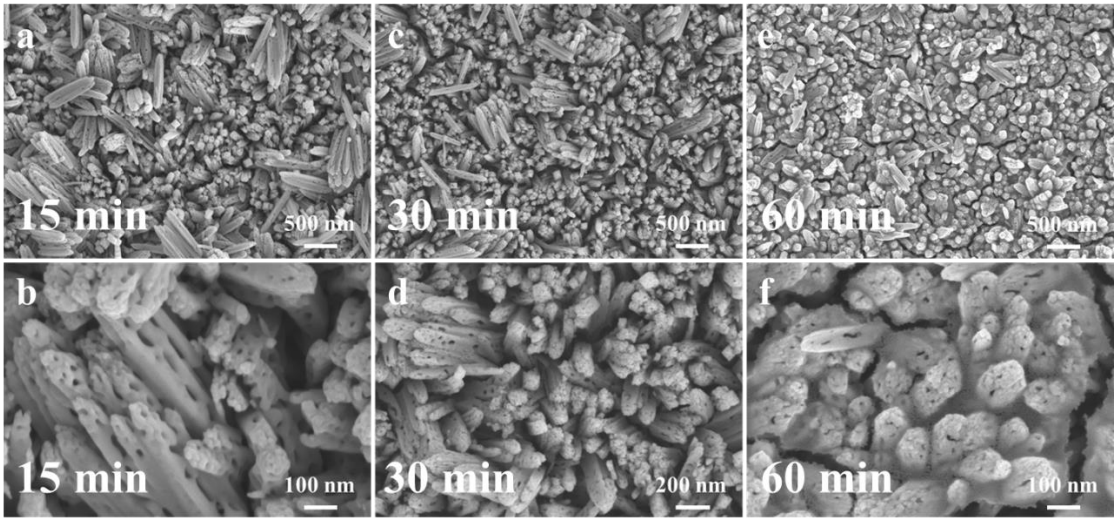

Figure S2. SEM images of Ti-PH/MOF with different reaction time.

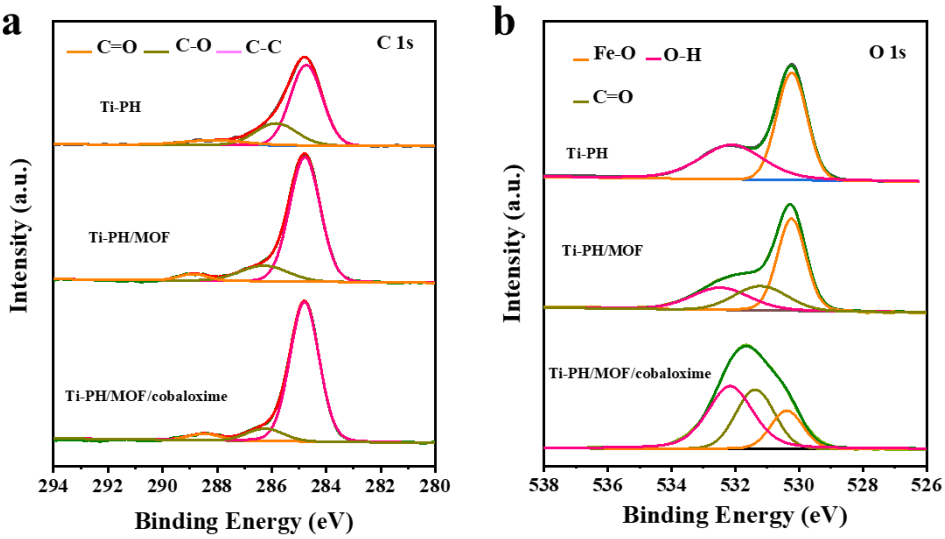

Figure S3. (a) C 1s, (b) O 1s XPS spectra of the three photoanodes.

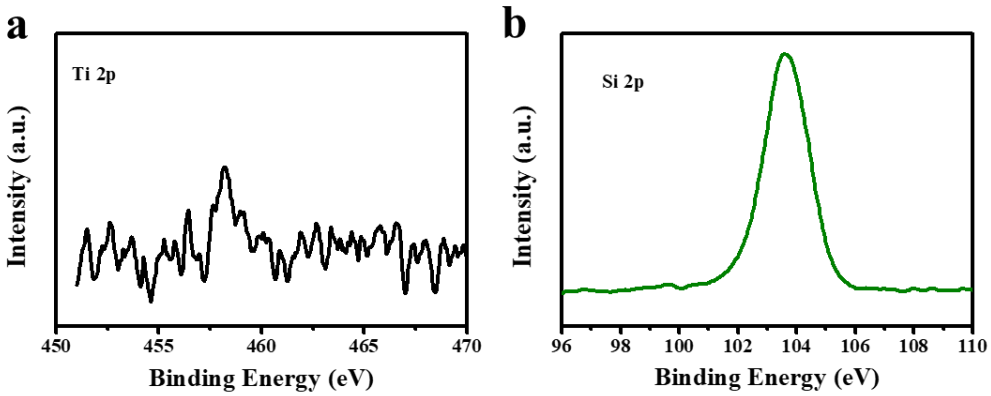

Figure S4. (a) Ti 2p, (b) Si 2p XPS spectra of Ti-PH

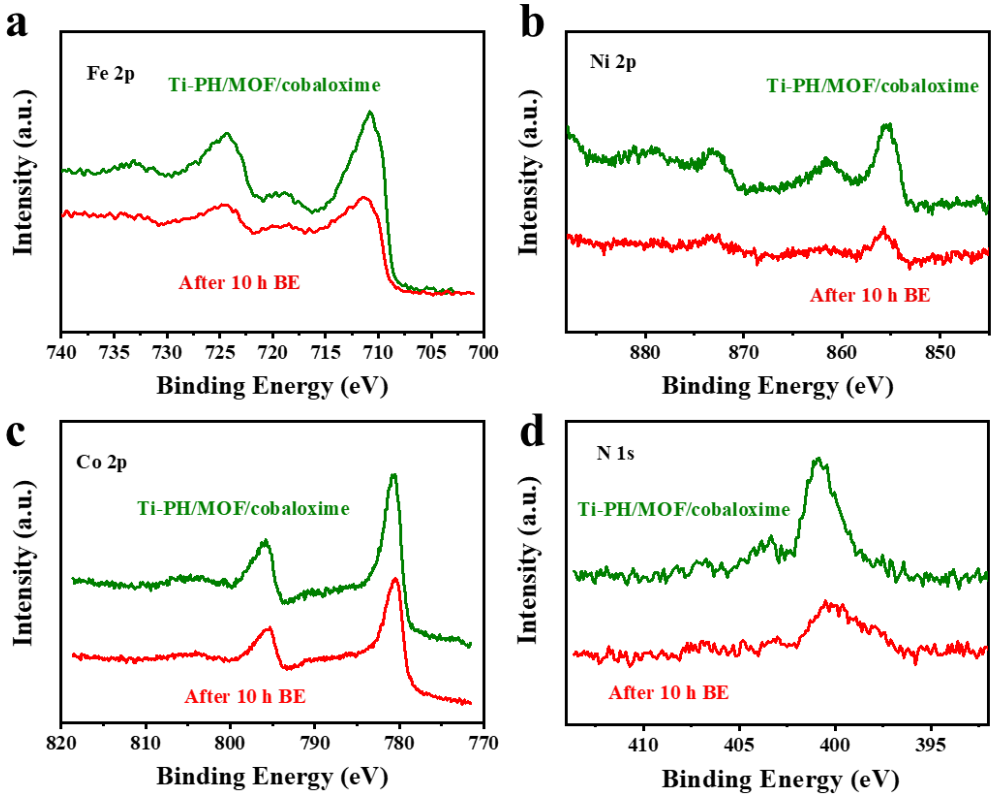

Figure S5. XPS data of Ti-PH/MOF/cobaloxime before and after 10 hours bulk electrolysis at 1.23 V vs. RHE under illumination.

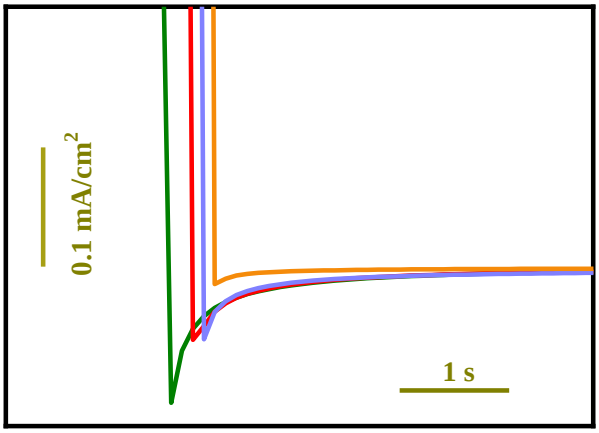

Figure S6. Current–time curves of the blue oval area in Figure 3c**.**

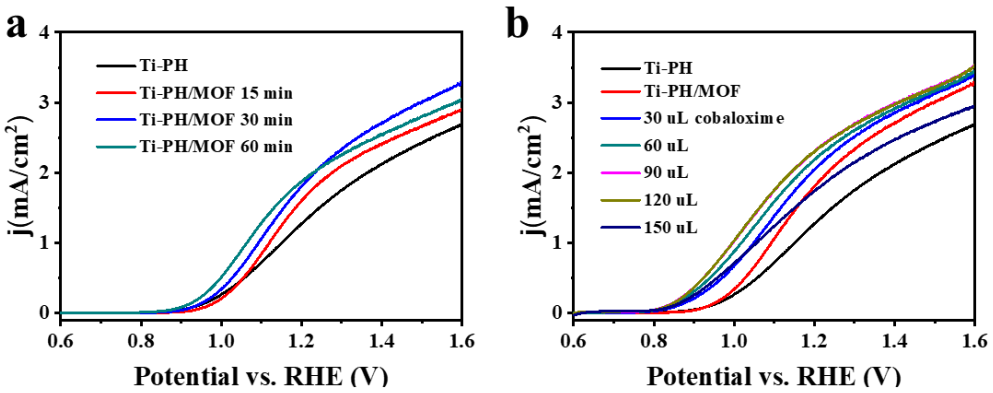

Figure S7. (a) Comparison of the LSV of Ti-PH/MOF with different reaction time, (b) comparison of the LSV of Ti-PH/MOF/cobaloxime with various drop volumes.

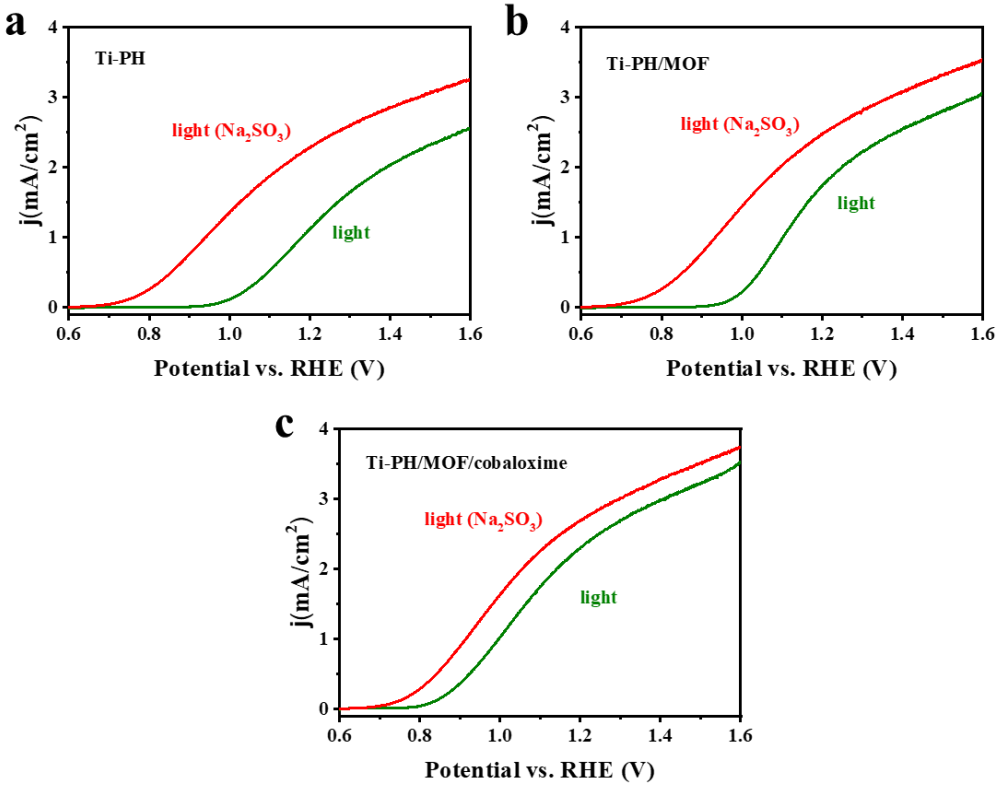

Figure S8. LSV curves of (a) Ti-PH, (b) Ti-PH/MOF, (c) Ti-PH/MOF/cobaloxime photoanode measured in 1.0 NaOH solution with (red line) and without (green line) 0.5 M Na_2SO_3 as a hole scavenger.

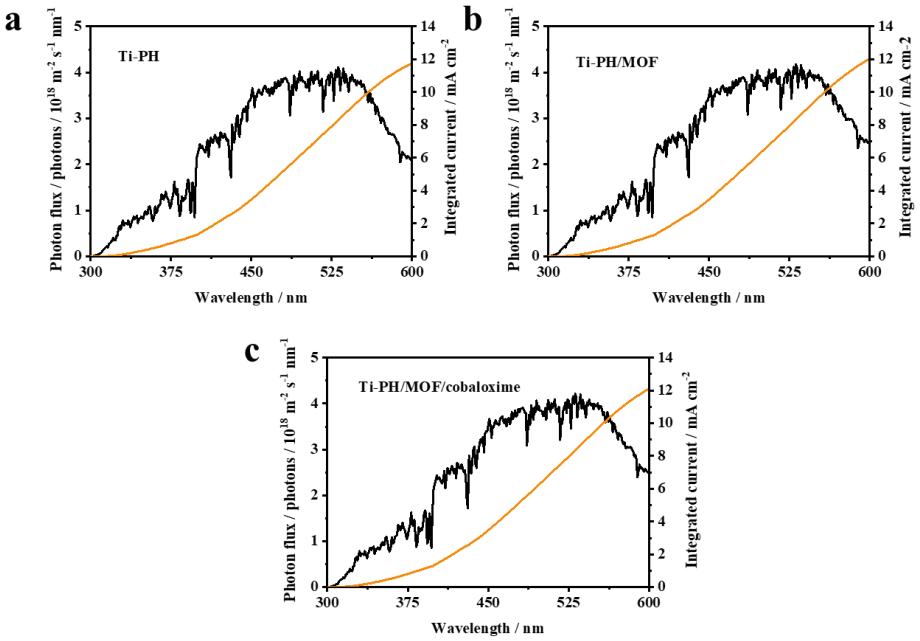

Figure S9. J_{abs} values of all composite photoanodes (APCE=100 %).

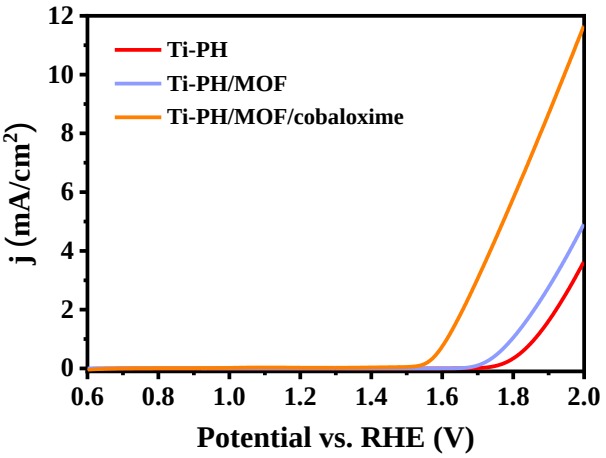

Figure S10. LSV curves of Ti-PH, Ti-PH/MOF, Ti-PH/MOF/cobaloxime in the dark.

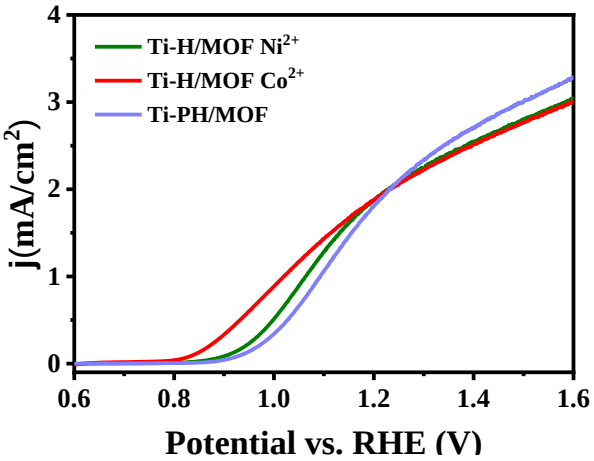

Figure S11. LSV curves of Ti-PH/NiFe MOF/50 nmol Ni^{2+}, Ti-PH/NiFe MOF/100 nmol Co^{2+}, and Ti-PH/NiFe MOF photoanodes.

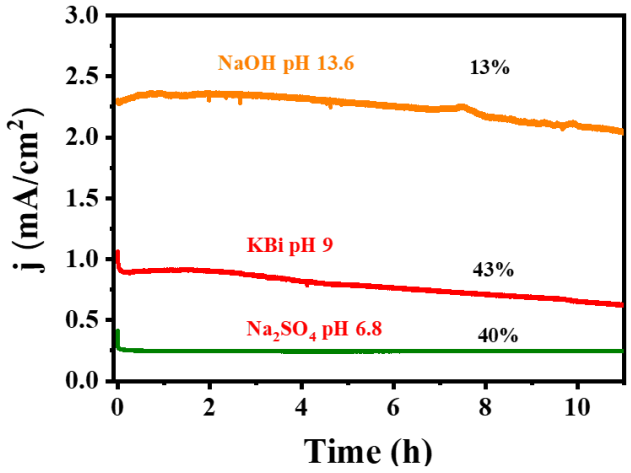

Figure S12. Long-term stability curves at 1.23 V vs. RHE of the Ti-PH/NiFe MOF/cobaloxime photoanode in 1 M NaOH (pH 13.6), 1 M KBi (pH 9), and 0.1 M Na_2SO_4 (pH 6.8).

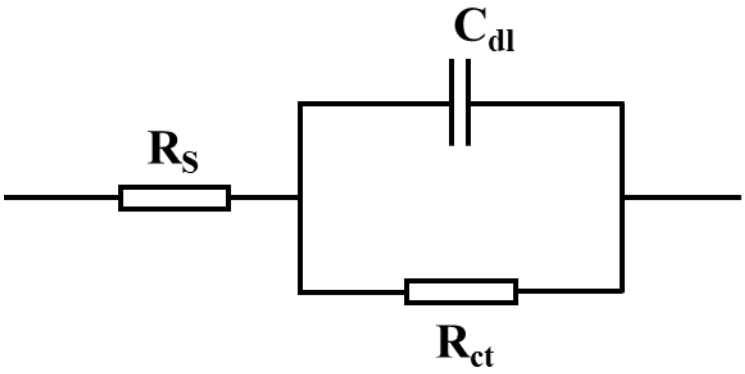

Figure S13. Equivalent electric circuit fitting from the EIS curve.

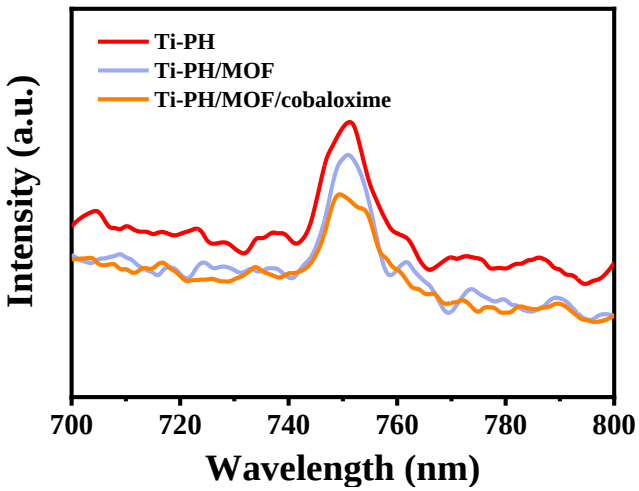

Figure S14. Photoluminescence spectra of all prepared samples.

Table S1. The fitting results using the equivalent model for EIS measurements.

Sample	RS	Rct
Ti-PH	33.18	3235
Ti-PH/NiFe MOF	35.38	2609
Ti-PH/NiFe MOF/cobaloxime	24.07	2029

Table S2. The Ni and Co contents in the Ti-PH/NiFe MOF/cobaloxime samples before and after 10 h bulk electrolysis at 1.23 V vs. RHE under illumination.

Sample	Ni (wt%)	Co (wt%)
Before test	0.85	3.4
After test	0.57	1.8

Table S3. The comparison of PEC performances between the Ti-PH/NiFe MOF/cobaloxime photoanode and other previously reported hematite photoanodes under AM 1.5 illumination (100 mW cm^{-2}).

Photoanodes	J (mA/cm^2) @1.23 V_{RHE}	Onset potential	IPCE	Stability	Reference
Fe_2O_3/FeB	1.65	~0.9	60% (at 380)	5 h (at 1.23 V_{RHE})	***J. Catal.*, 2017, 352, 113-119**
Ti-PH/CoP	2.1	0.77	~42% (at 360 nm)	3 h (8.9% decay at 1.23 V_{RHE})	***J. Catal.*, 2018, 366, 275-281**
Fe_2O_3/Fe_2TiO_5/FeNiOOH	2.2	~1.0	28.7% (at 350 nm)	2 h (at 1.23 V_{RHE})	***Energy Environ. Sci.*, 2017, 10, 2124-2136**
α-Fe_2O_3/Co-Mn Nanosheets	2.09	~0.7	39.1 (at 360 nm)	10 h (3% decay at 1.23 V_{RHE})	***Adv. Funct. Mater.*, 2019, 29, 1904622**
Fe_2O_3:Ti/NH_2-MIL-101(Fe)	2.27	0.92	42.3 (at 365 nm)	2500 s (13.3% decay at 1.23 V_{RHE})	***Appl. Catal. B: Environ.*, 2018, 237, 9–17**
F–Fe_2O_3/CoAl-LDH	2.46	~0.72	47.66% (at 300 nm)	6 h (10% decay at 1.23 V_{RHE})	***ACS Appl. Mater. Interfaces*, 2019, 11, 29799–29806**
α-Fe_2O_3/NiFe-phosphate	1.2	0.74	28.7% (at 340 nm)	5.5 h (at 1.23 V_{RHE})	***ACS Sustainable Chem. Eng.*, 2018, 6, 2353–2361**
Zn- Fe_2O_3/$FeNiO_x$	~0.68	~0.8	-	-	***ACS Catal.*, 2018, 8, 2754-2759**
Co-Pi/WRCN/Hematite	2.14	~0.9	~28 (380 nm)	2 h (at 1.23 V_{RHE})	***J. Phys. Chem. C*, 2021, 125, 13273–13282**
Zr-doped Hematite	~1.48	0.87	20.91% (at 350 nm)	1 h (at 1.23 V_{RHE})	***Chem. Eng. J.*, 2020, 390, 124504**
Ti-PH/NiFe MOF/cobaloxime	2.45	0.78	83.0% (at 365 nm)	6 h (2% decay at 1.23 V_{RHE})	This work