

Supporting Information for

Amorphous–Crystalline CoFeB/NiPS₃ Vertical Heterostructure with Built-in Electric Field for Robust Ampere-level Water Oxidation

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1. Calculation of turnover frequency (TOF):

$$TOF = \frac{1}{4F} \frac{1}{n} \quad (1)$$

In which I (A) represents the current at $\eta=300$ mV, F (~ 96485 C mol⁻¹) is the Faraday constant and n (mol) is the mole of metal atoms coated on the electrode during OER in 1M KOH. I can be evaluated with the following equation:

$$n = \frac{Q}{2F} \quad (2)$$

Here Q represent the integrated charge from the CV curve, which was obtained through electrochemical measurements in a 1M PBS electrolyte (pH=7), with a scanning rate of 50 mV s⁻¹ and a potential range of -0.2 to 0.6 V vs. RHE. As mentioned above, F is Faraday constant.

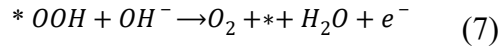
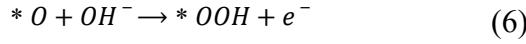
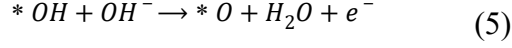
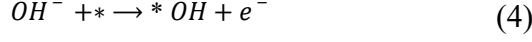
Calculation of mass activity (MA):

$$MA = \frac{j}{m} \quad (3)$$

In which j (mA cm⁻²) is the measured current density at $\eta=300$ mV, m (mg cm⁻²)=4 mg \times 10 μ L / 500 μ L=0.08 mg cm⁻².

2. DFT Calculations of OER Performance:

Under alkaline conditions, the process of OER reaction can be described by the following four steps,



in which * denotes the pure surface without adsorbed adsorbate, *O, *OH, *OOH denote the surface system with adsorbed intermediate O, OH, and OOH, respectively. The Gibbs free energy variations of the above four reaction steps are represented as ΔG_1 , ΔG_2 , ΔG_3 , and ΔG_4 , respectively.

$$\Delta G_1 = G_{*OH} + \left(\frac{G_{H_2}}{2} - 0.0592PH \right) + eU - G_* - G_{H_2O} \quad (8)$$

$$\Delta G_2 = G_{*O} + \left(\frac{G_{H_2}}{2} - 0.0592PH \right) + eU - G_{*OH} \quad (9)$$

$$\Delta G_3 = G_{*OOH} + \left(\frac{G_{H_2}}{2} - 0.0592PH \right) + eU - G_{*O} - G_{H_2O} \quad (10)$$

$$\Delta G_4 = G_{O_2} + G_* + \left(\frac{G_{H_2}}{2} - 0.0592PH \right) + eU - G_{*OOH} \quad (11)$$

U is the applied electrode potential. The standard electrode potential is 0.401 eV at pH = 14 under alkaline conditions, and the Gibbs free energy change of the determination step is $G^{OER} = \max(\Delta G_1, \Delta G_2, \Delta G_3, \Delta G_4)$, and the theoretical calculated overpotential is $\eta = G^{OER} - 0.401$. The adsorption sites are shown in Fig. S3 and the calculation results are shown in Table S1.

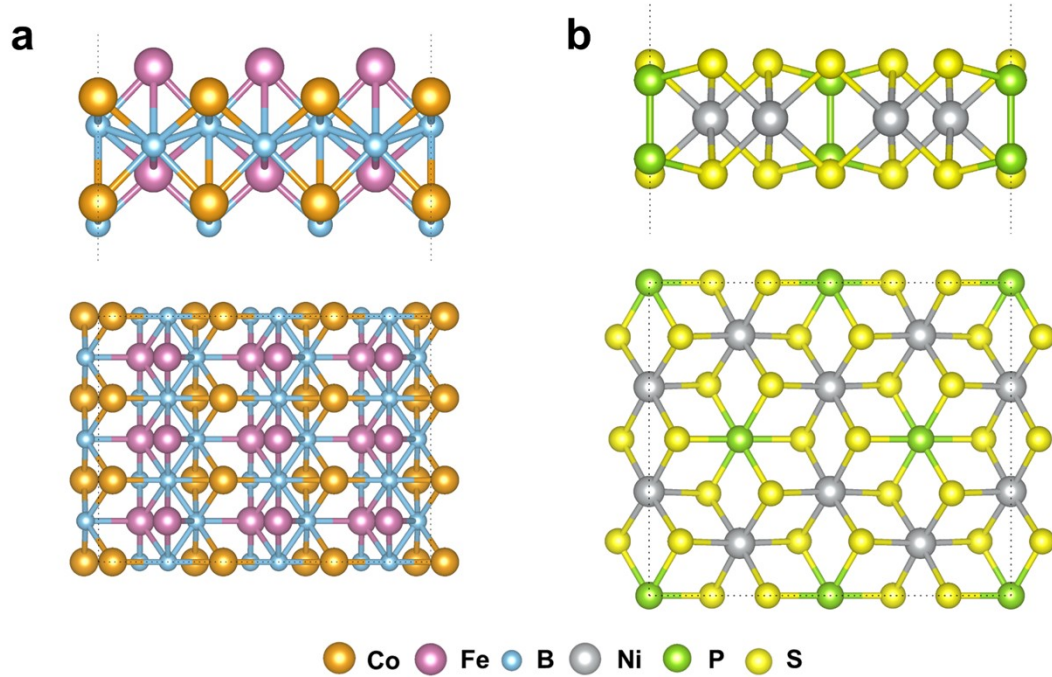


Fig. S1. Atomic models of (a) CoFeB and (b) NiPS₃.

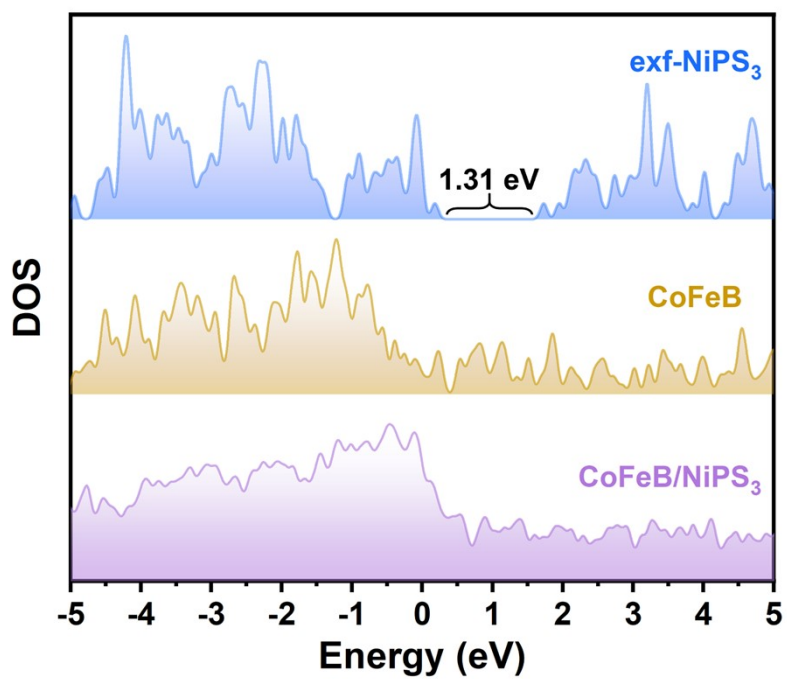


Fig. S2. DOS of CoFeB, NiPS₃, and CoFeB/NiPS₃.

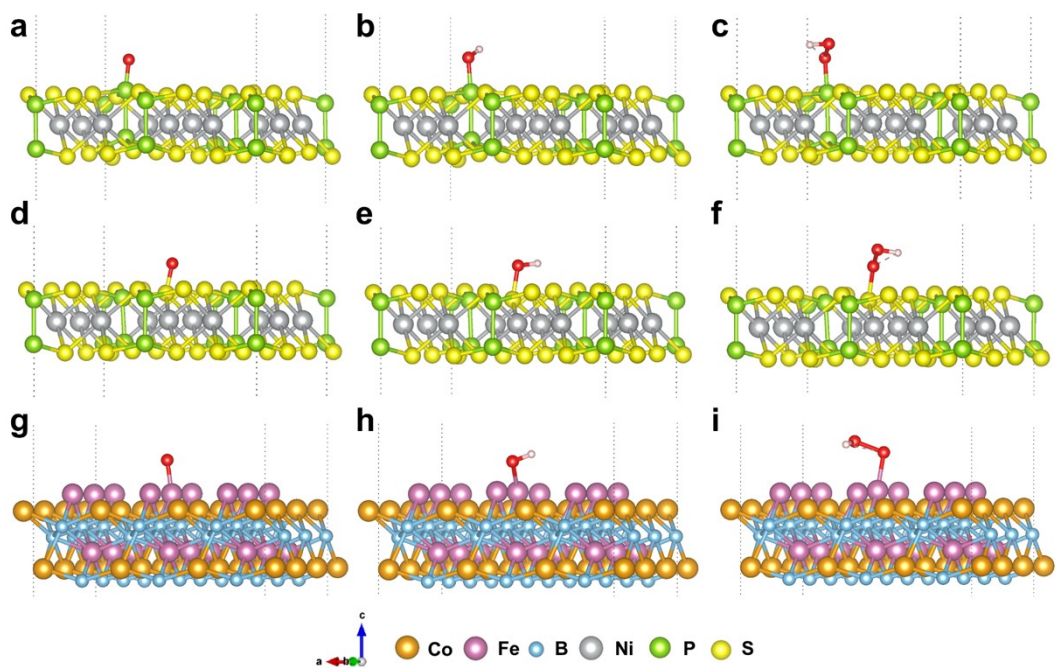


Fig. S3. The possible stable adsorption sites of $*O$, $*OH$, $*OOH$ on the (a-c) P edge of $NiPS_3$, (d-f) S edge of $NiPS_3$, (g-i) Fe edge of $CoFeB$.



Fig. S4. Electrochemical cathode exfoliation process from bulk $NiPS_3$ to exf- $NiPS_3$ nanosheets.

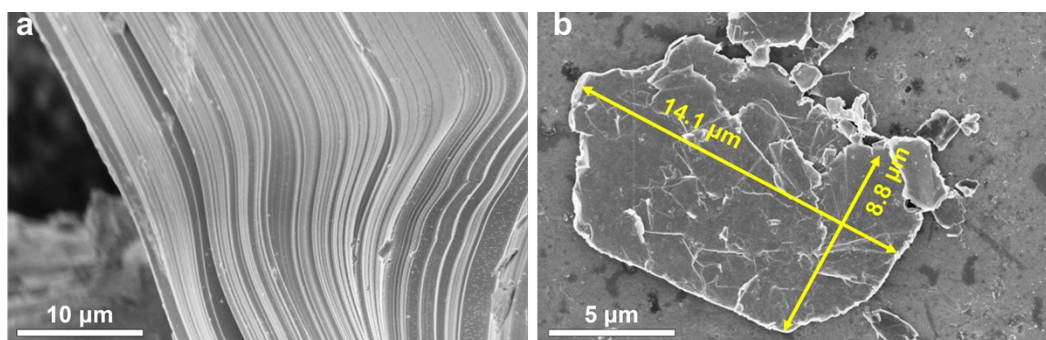


Fig. S5. SEM images of (a) the cross-section of bulk $NiPS_3$ and (b) exf- $NiPS_3$ nanosheets.

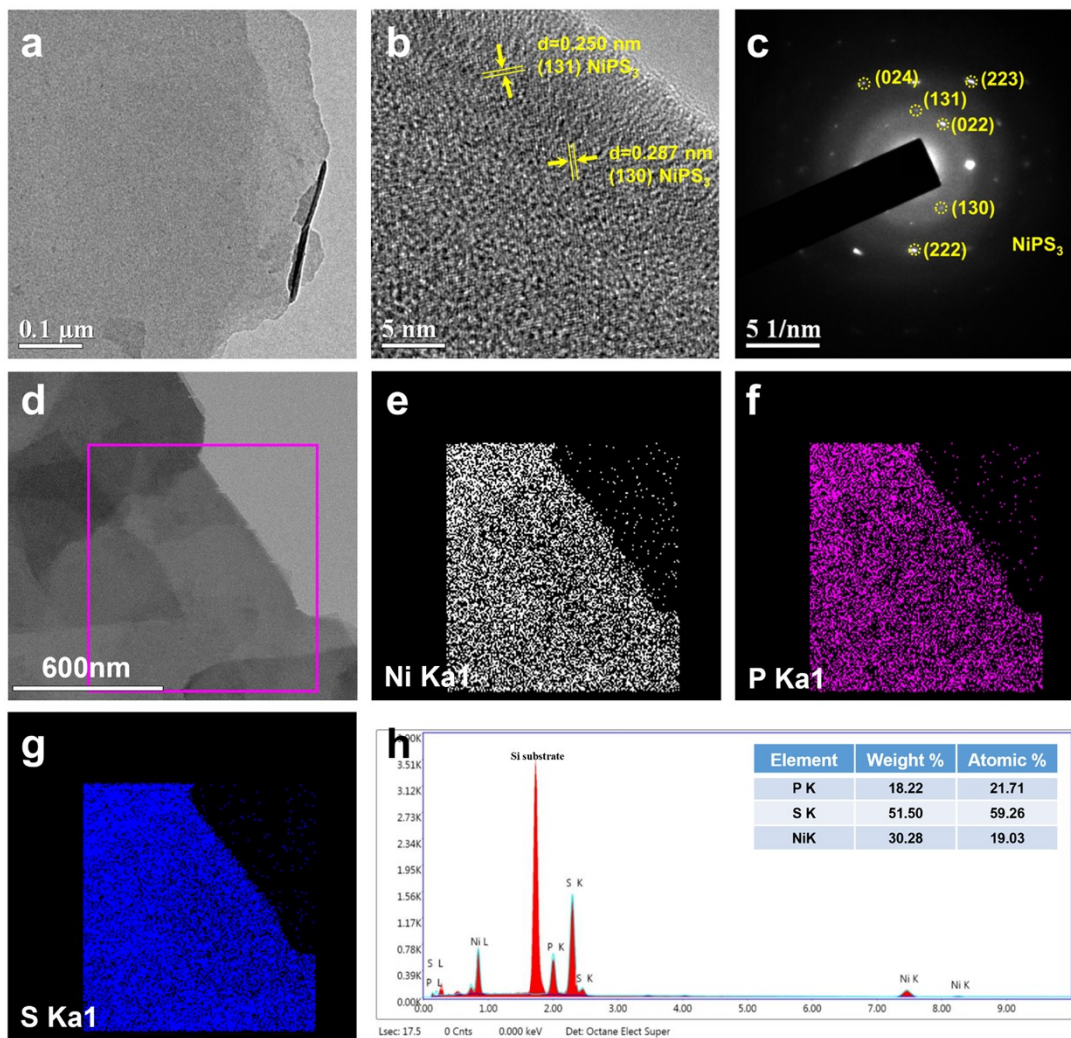


Fig. S6. (a) TEM image, (b) HRTEM image, (c) SAED pattern, (d-g) EDX mapping images, and (h) EDS spectrum of exf-NiPS₃.

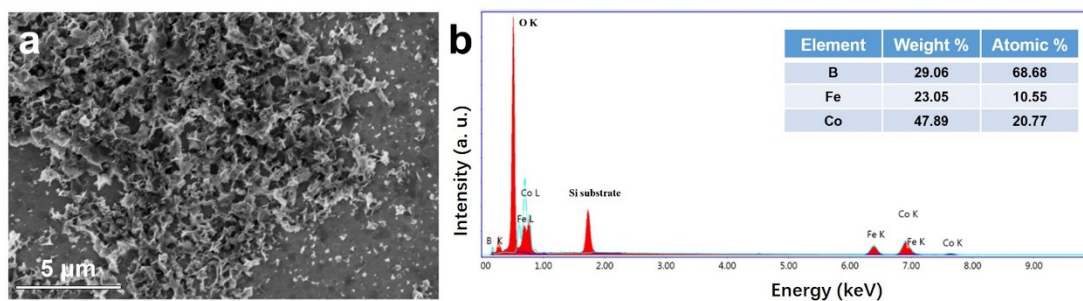


Fig. S7. (a) SEM image and (b) EDS spectrum of CoFeB.

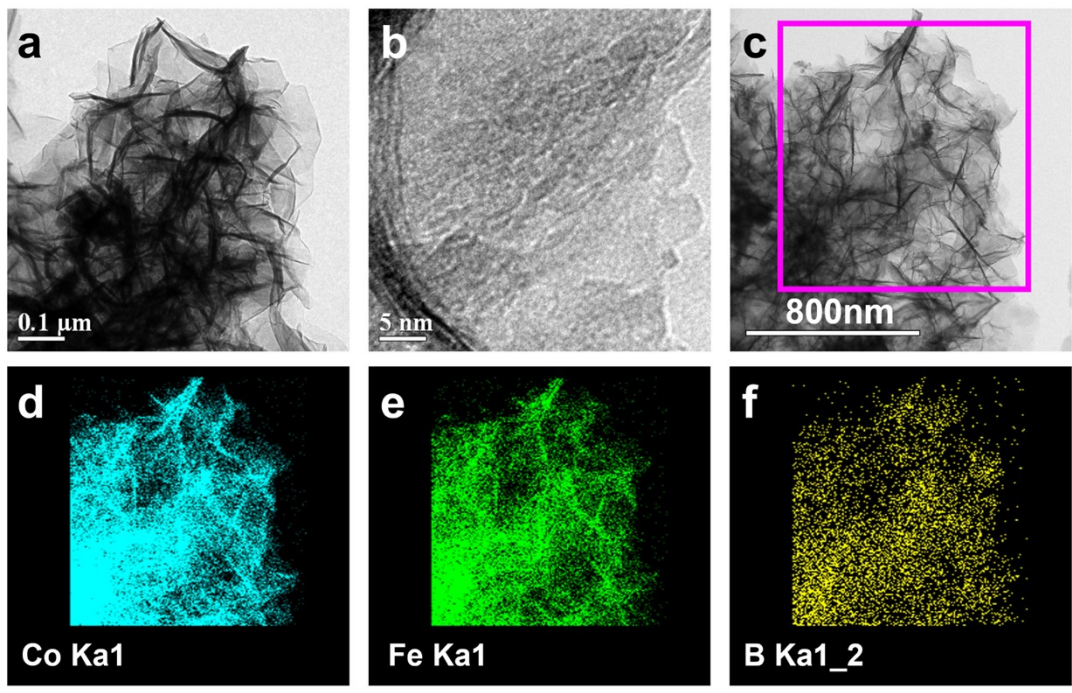


Fig. S8. (a) TEM image, (b) HRTEM image, and (c-f) EDX mapping images of CoFeB.

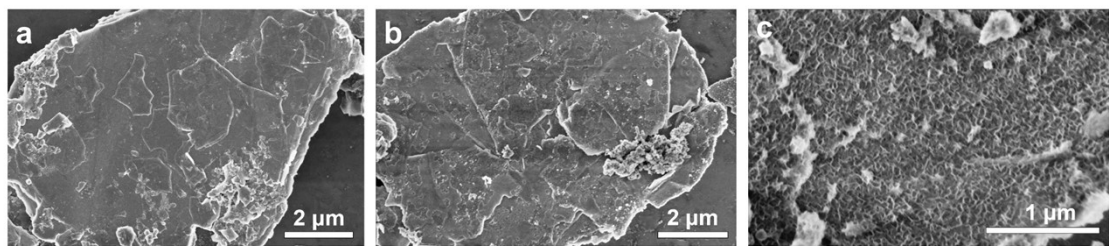


Fig. S9. SEM images of (a) CFBN-1, (b) CFBN-2 and (c) CFBN-4.

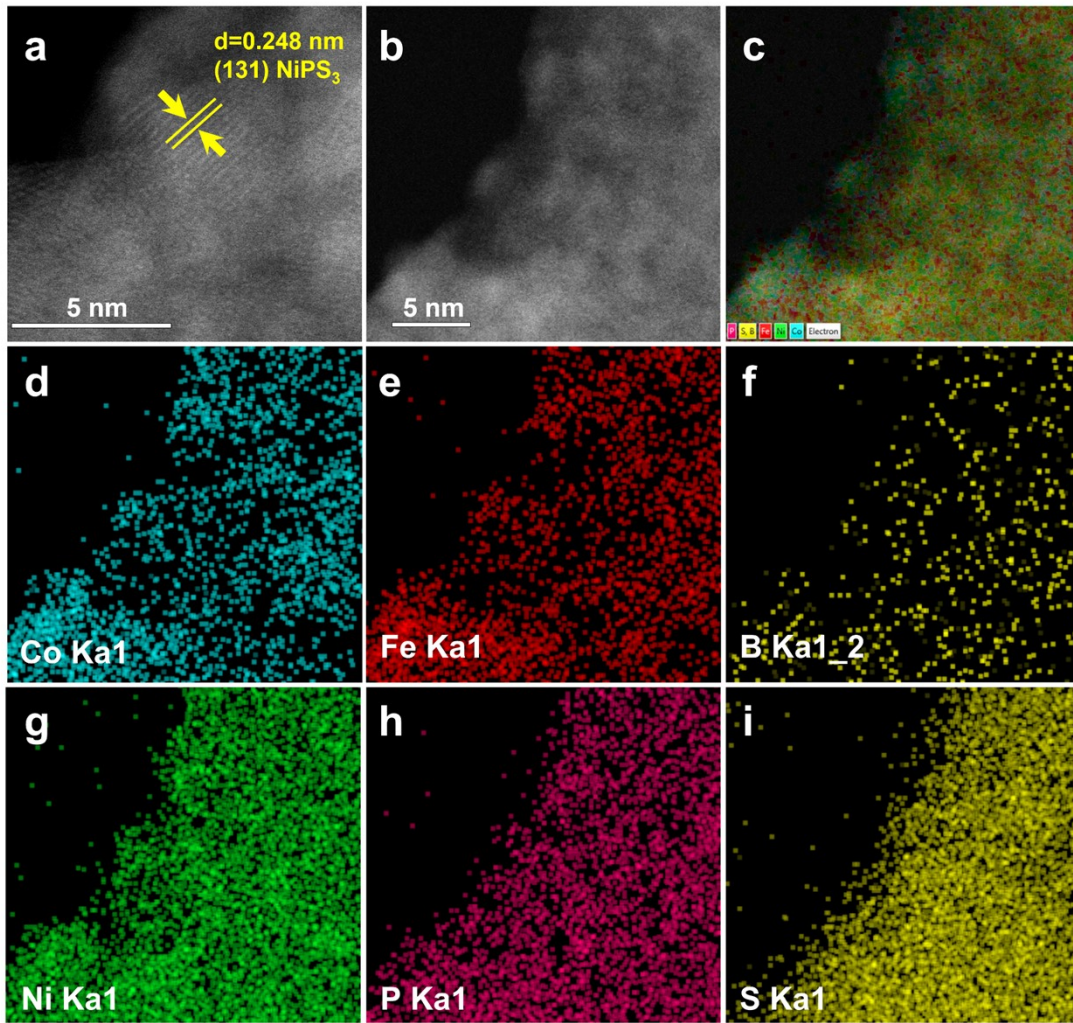


Fig. S10. (a) AC-TEM image, (b-i) EDX mapping images of CoFeB/NiPS₃.

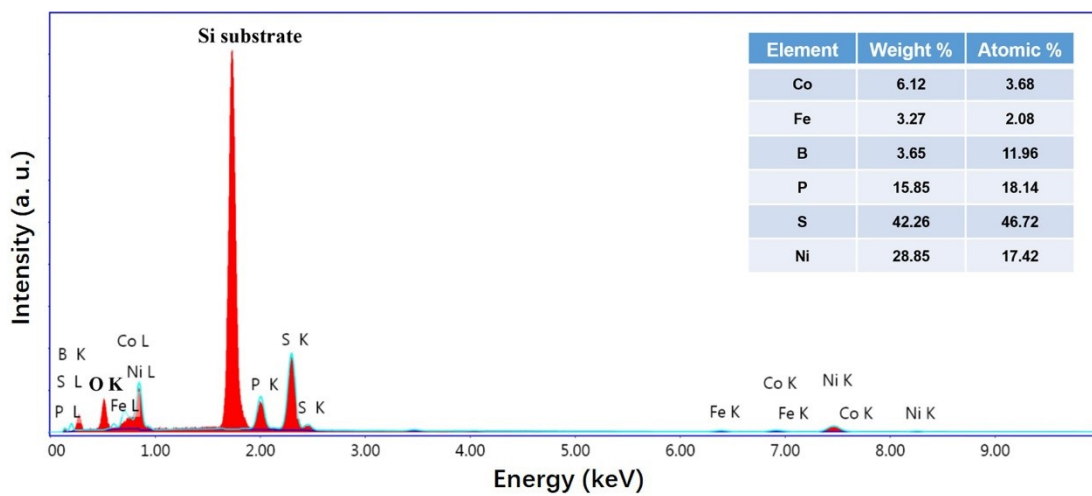


Fig. S11. EDS spectrum of CFBN-3.

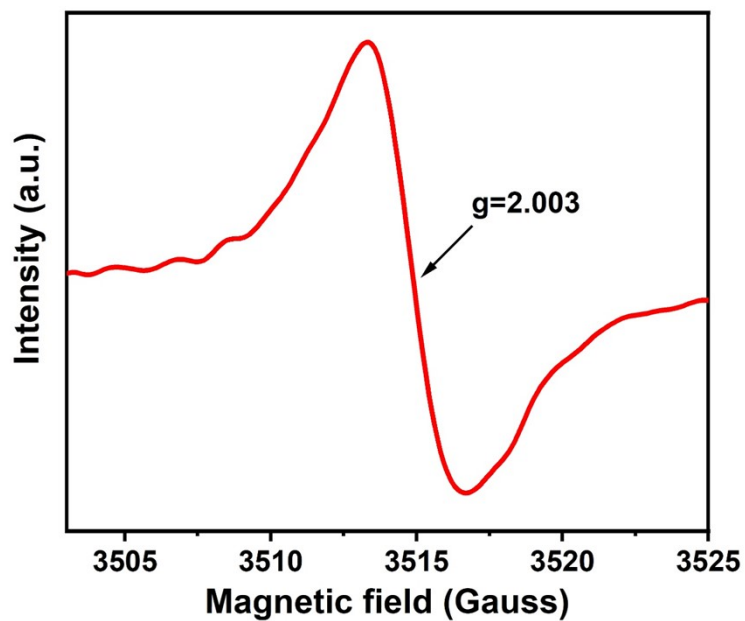


Fig. S12. The room-temperature EPR spectrum of CFBN-3 heterostructure.

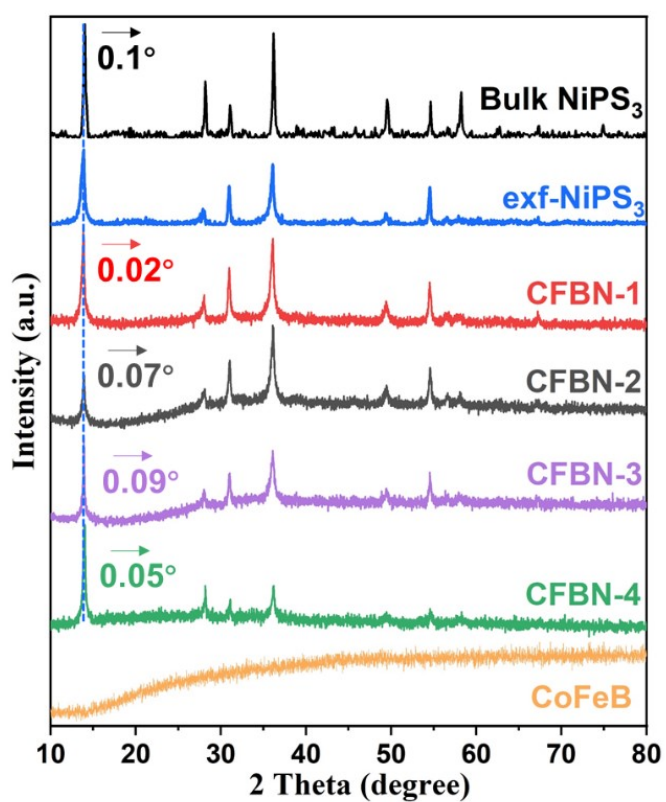


Fig. S13. XRD patterns of CFBN-1, CFBN-2, CFBN-3 and CFBN-4.

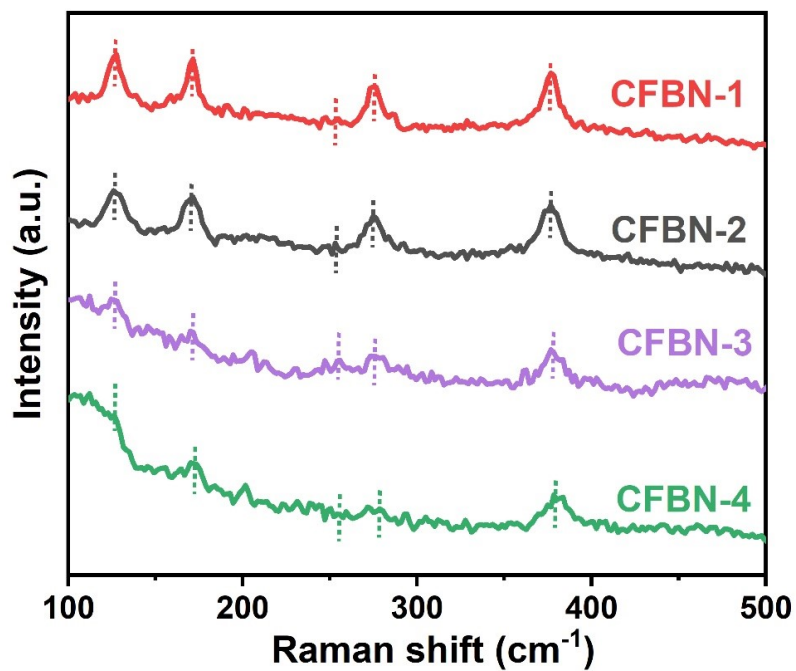


Fig. S14. Raman spectra of CFBN-1, CFBN-2, CFBN-3 and CFBN-4.

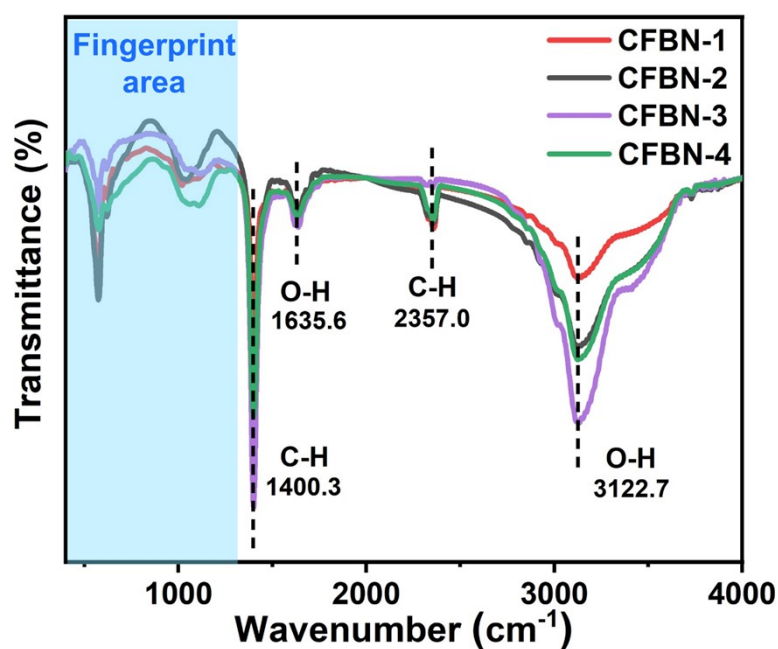


Fig. S15. FT-IR spectra of CFBN-1, CFBN-2, CFBN-3 and CFBN-4.

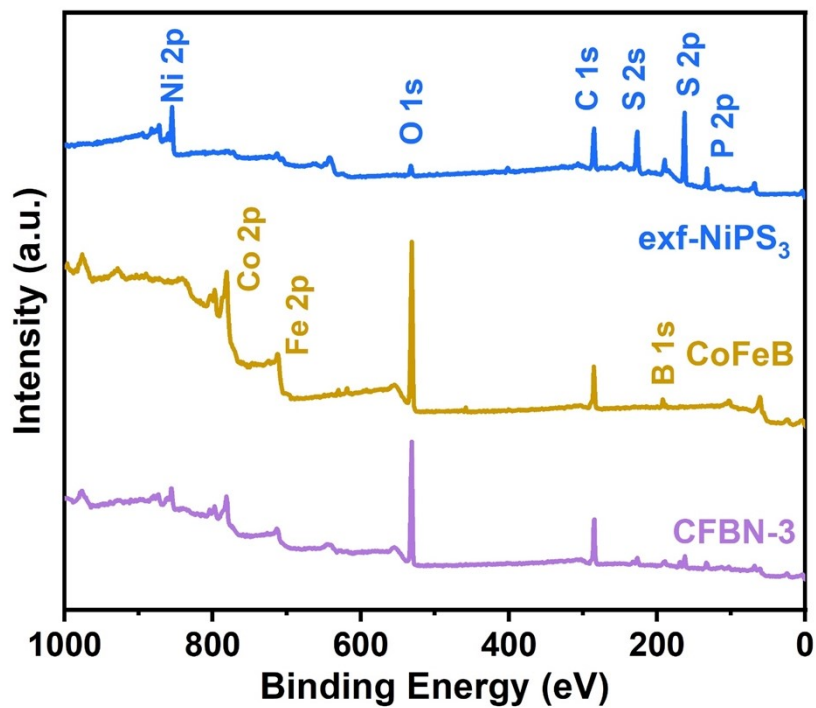


Fig. S16. XPS survey spectra of exf-NiPS₃, CoFeB and CFBN-3.

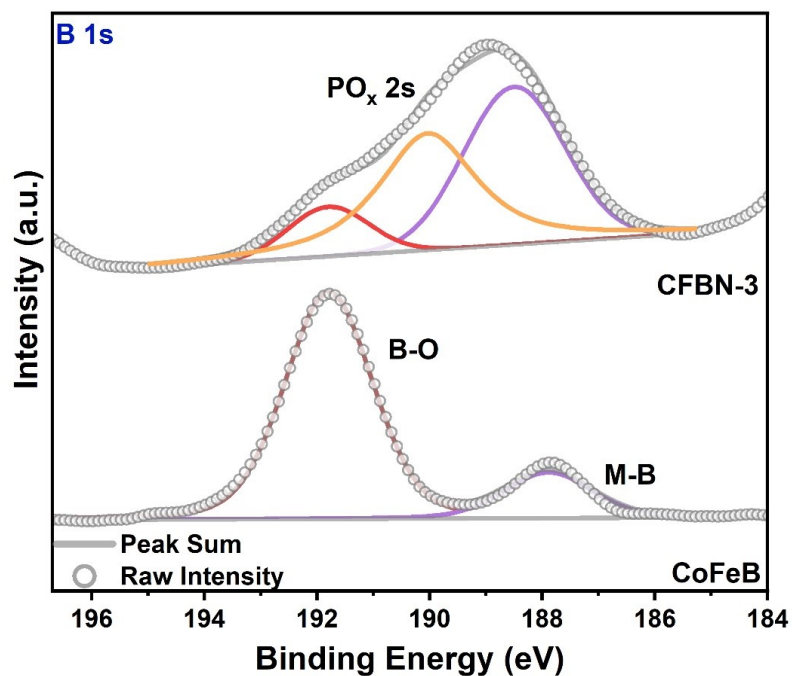


Fig. S17. B1s spectrum of CFBN-3 and CoFeB.

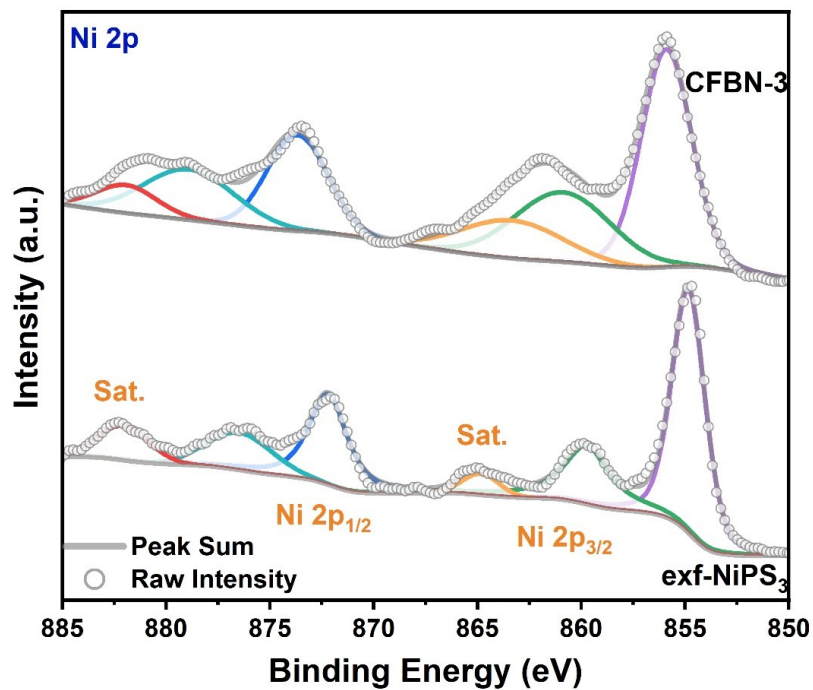


Fig. S18. Ni 2p spectra of CFBN-3 and exf-NiPS₃.

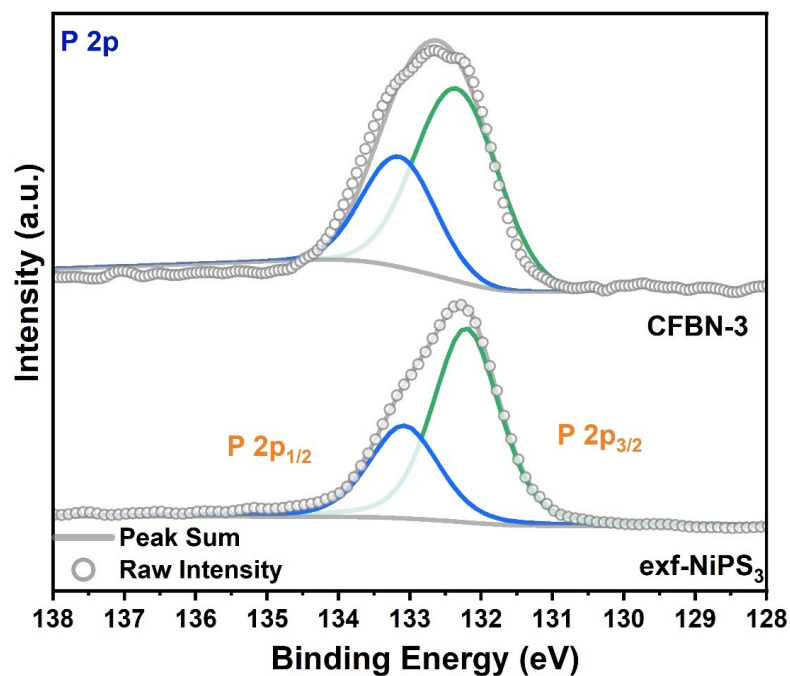


Fig. S19. P 2p spectra of CFBN-3 and exf-NiPS₃.

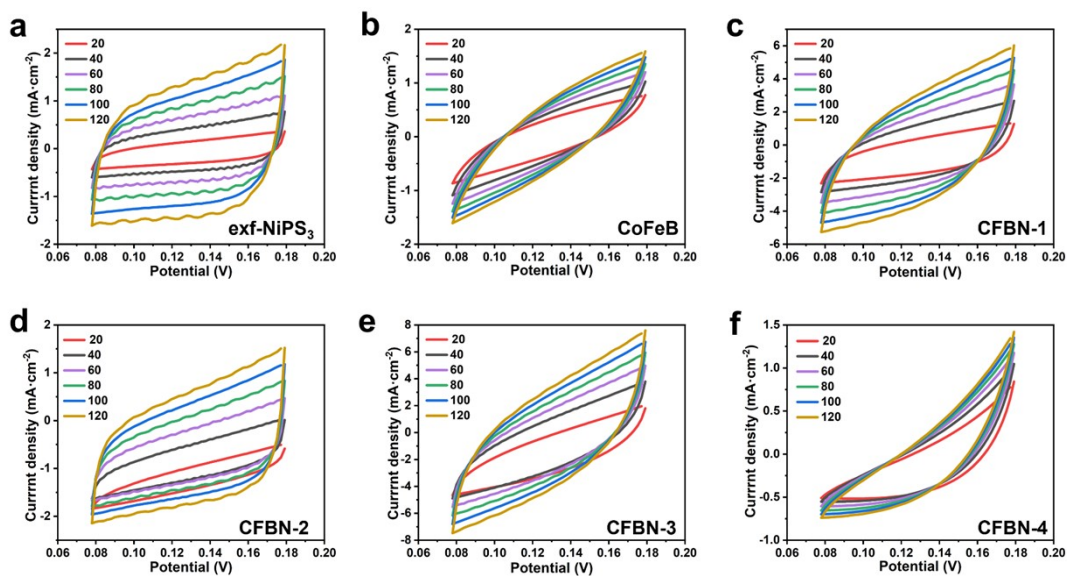


Fig. S20. The current-potential curves of samples with different scan rates for OER process in 1 M KOH: (a) exf-NiPS₃, (b) CoFeB, (c) CFBN-1, (d) CFBN-2, (e) CFBN-3, and (f) CFBN-4.

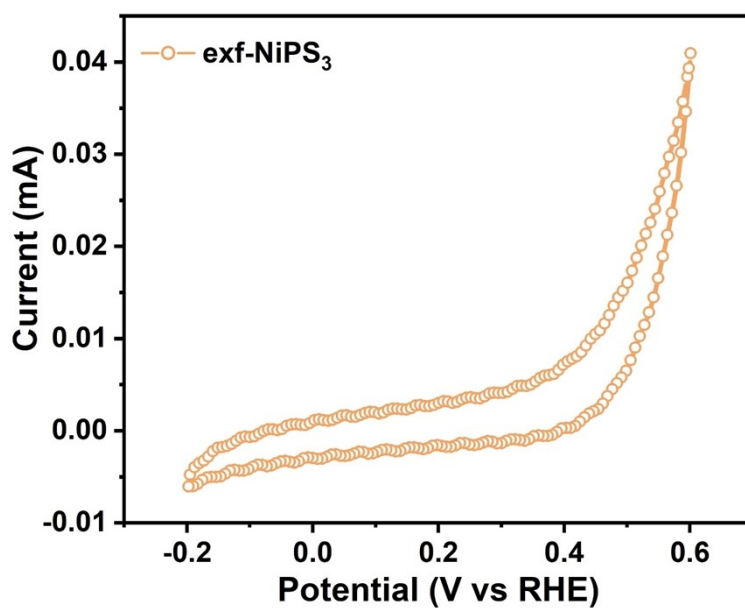


Fig. S21. CV curve for exf-NiPS₃ electrode in the potential ranges between -0.2 V vs. RHE and 0.6 V vs. RHE in 1 M PBS. The scan rate was 50 mV·s⁻¹.

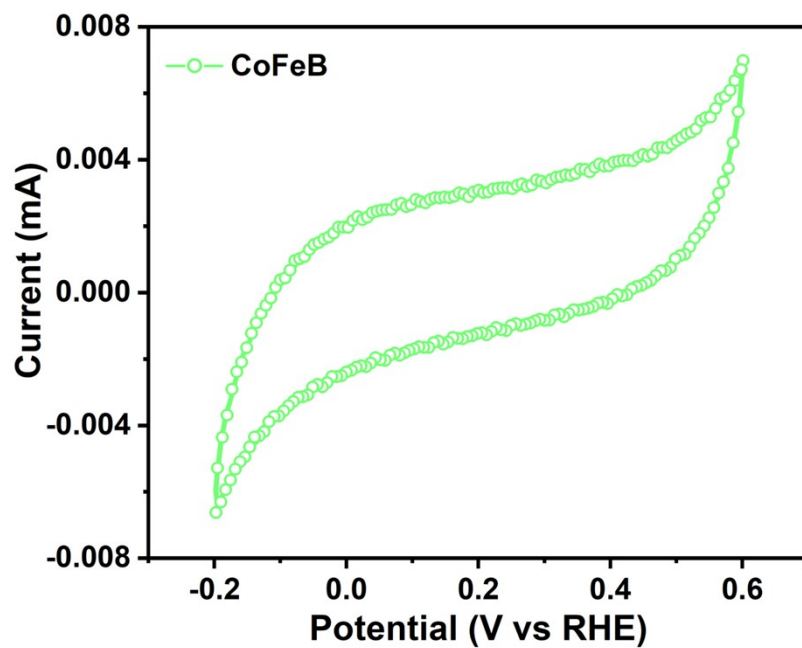


Fig. S22. CV curve for CoFeB electrode in the potential ranges between -0.2 V vs. RHE and 0.6 V vs. RHE in 1 M PBS. The scan rate was 50 $\text{mV}\cdot\text{s}^{-1}$.

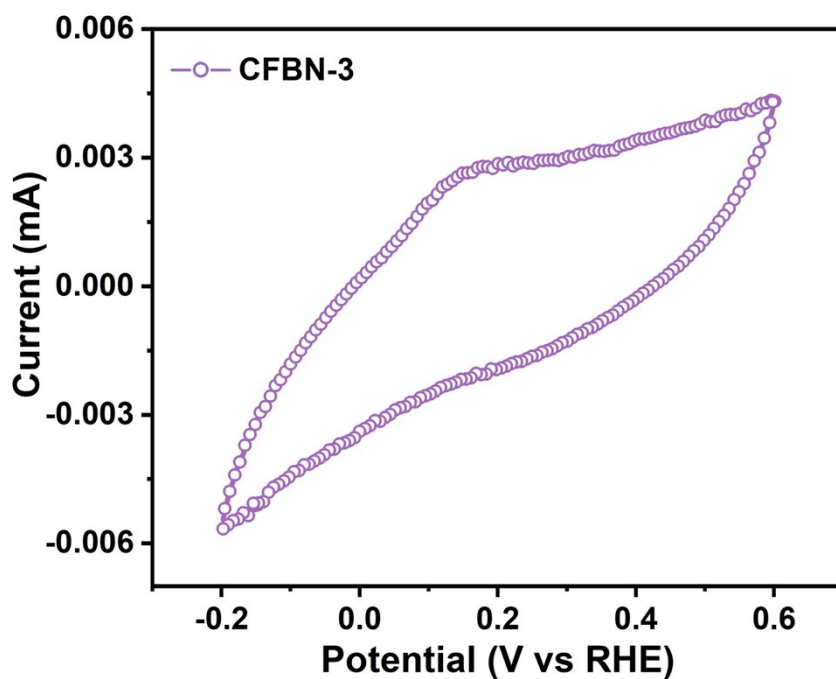


Fig. S23. CV curve for CFBN-3 electrode in the potential ranges between -0.2 V vs. RHE and 0.6 V vs. RHE in 1 M PBS. The scan rate was 50 $\text{mV}\cdot\text{s}^{-1}$.

Table S1. The calculation values of OER. (calibration at 298.15 K, the unit of the physical quantity is eV, U = 0 V).

System	Site	G*	G _{H2O}	G _{H2}	G _{O2}	ΔG ₁	ΔG ₂	ΔG ₃	ΔG ₄	η
CoFeB	Fe	-453.54	-14.22	-6.8	-9.91	-1.43	0.28	1.85	0.91	1.45
NiPS ₃	P	-205.59				-0.40	-0.41	2.02	0.40	1.62
	S					1.05	-1.12	2.82	-1.14	2.42
CoFeB/NiPS ₃	Fe	-669.76				-0.78	0.42	1.31	0.65	0.91

Table S2. Comparison of the oxygen evolution reaction (OER) performance of CoFeB/NiPS₃ with that of representative catalysts of similar elements or structures in 1 M KOH.

Catalysts	η _{OER@10 mA cm⁻²}	Tafel slope	References
	(mV)	(mV•decade ⁻¹)	
MoS ₂ /NiPS ₃	296	86	1
S-FeOOH/IF	244	59	2
ZnCo ₂ O _{4-x} F _x /CNTs	350	59.2	3
Ru/Co-N-C	247	27.8	4
P-Ce SAs@CoO	261	75	5
Co-N ₄ -pyridinic SACs	351	84	6
CoNiFeMoCr	310	/	7
Ru-Co/ELCO	247	49.1	8
a-NiCo/NC	252	49	9
Porous Ni ₃ S ₄	257	67	10
Co/Fe-SNC	240	47.92	11
F-CDs/CoP/NF,	250	96	12
CoFeB/NiPS₃	235	96(50)	This work

Supplemental References

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