Electronic Supplementary Material (ESI) for Materials Advances.

Surface Engineering Assisted CoNiP Nanosheet Arrays for Electrochemical Overall Water Splitting

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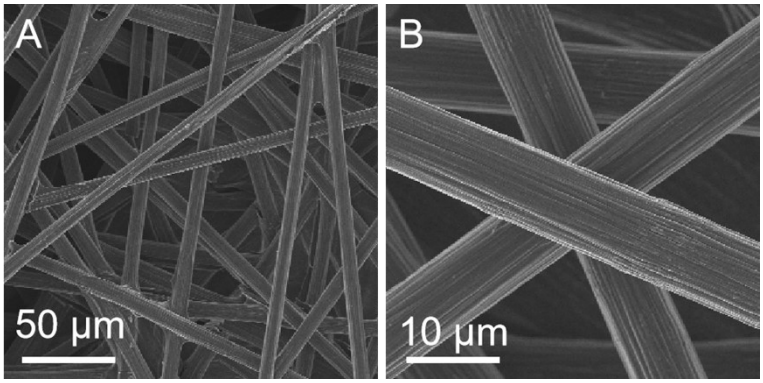

Fig. S1 SEM image shows the morphology of bare CFP with width up to tens of micrometer.

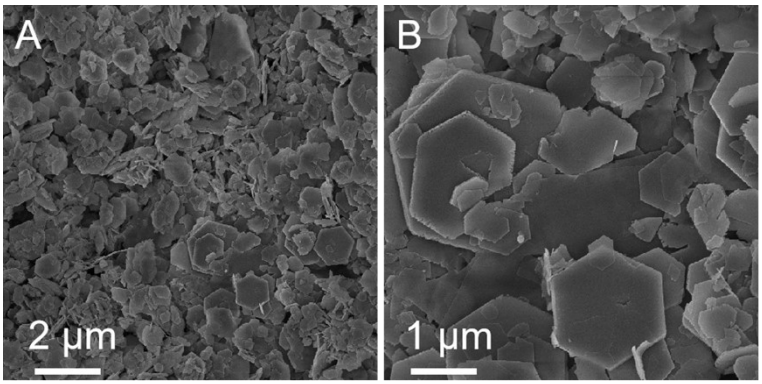

Fig S2 (A, B) the low magnification SEM images of the $Co(OH)_2$ hexagonal nanoflakes without CFP.

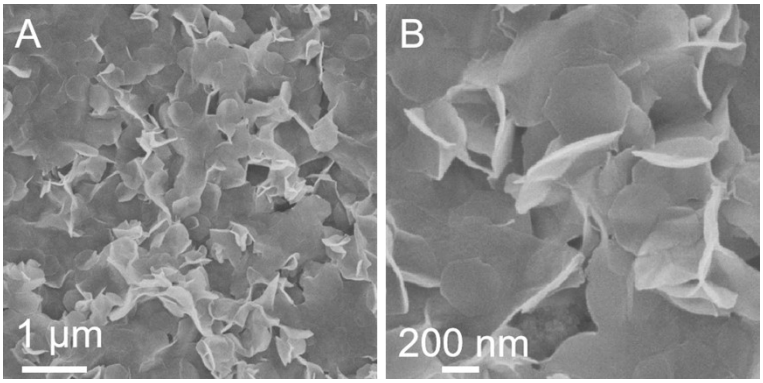

Fig. S3 SEM images of $CoNi(OH)_2$ layered double hydroxides (CoNi-LDH) without CFP.

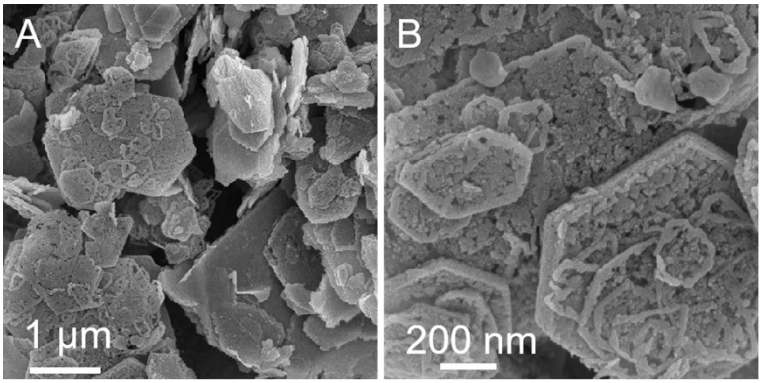

Fig. S4 SEM images of porous hexagonal CoP nanoflakes without CFP.

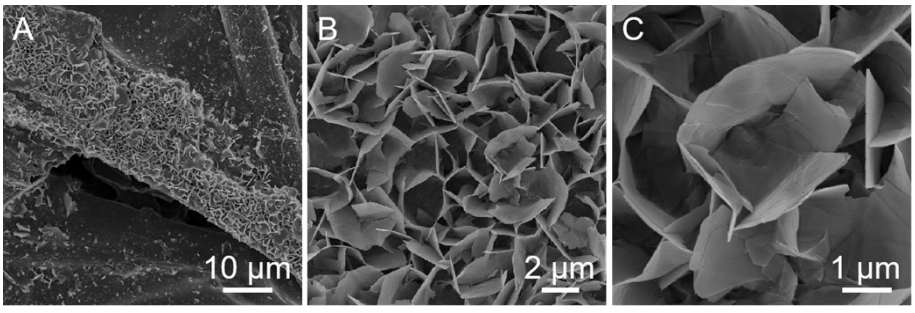

Fig. S5 SEM images of $Co(OH)_2$ nanosheets electrodeposited on carbon fibers paper.

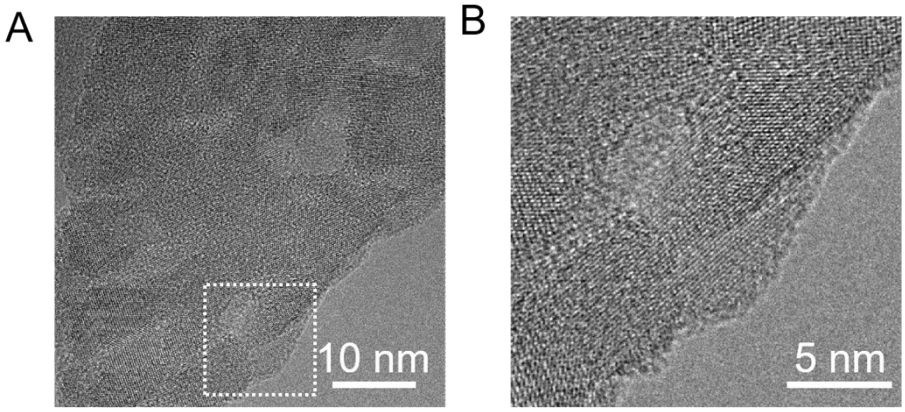

Fig. S6 HRTEM images of porous CoNiP nanosheets on the surface of CFP.

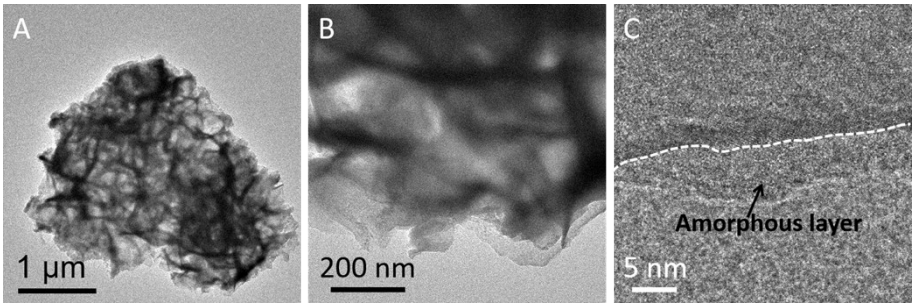

Fig. S7 TEM and HRTEM images of porous CoNiP/CoOOH nanosheets on the surface of CFP.

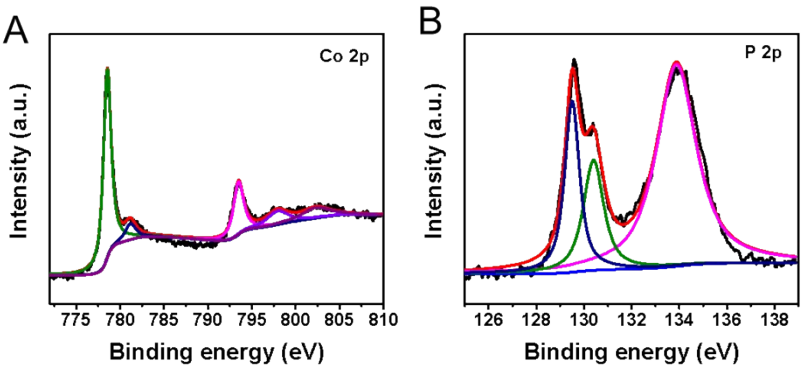

Fig. S8 High-resolution XPS spectra of the as-prepared CoP/CoOOH-CFP. (A) Co 2p, (B) P 2p. The red curve is the sum, the purple curve is the baseline, and the other curves are the fitted.

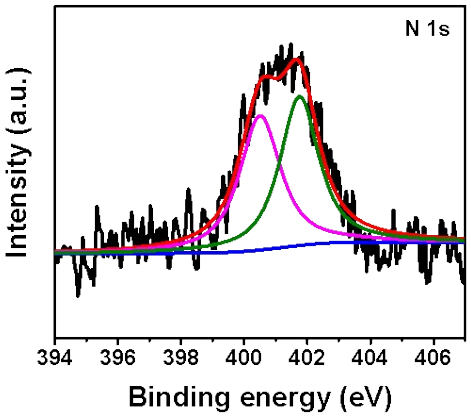

Fig. S9 N 1s spectrum of High-resolution XPS spectra of the as-prepared CoNiP/ CoOOH-CFP.

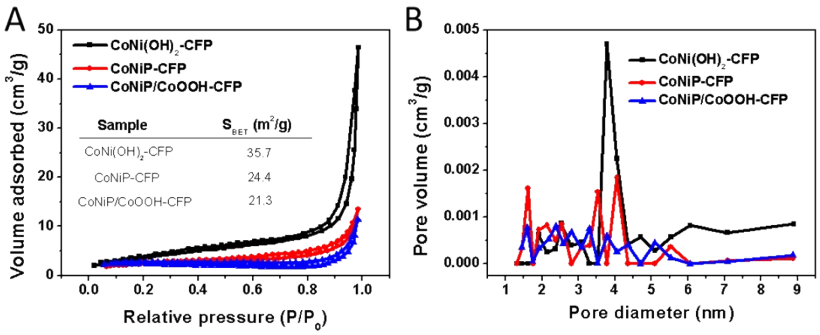

Fig. S10 (A) N_2 adsorption-desorption isothermal plots of $CoNi(OH)_2$-CFP, CoNiP-CFP, and CoNiP/CoOOH-CFP. (B) The related pore-size distribution curves for those examined samples.

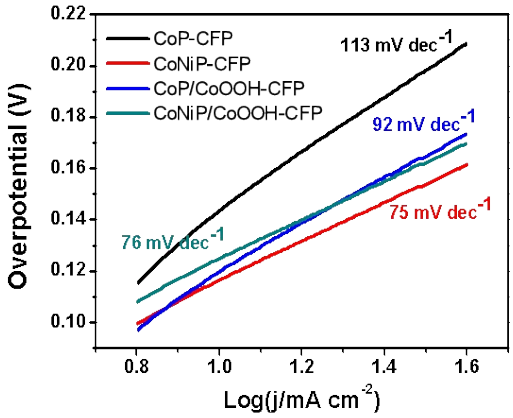

Fig. S11 LSV polarization curves derived Tafel plots of different catalysts in 1.0 M KOH electrolyte for electrocatalytic HER.

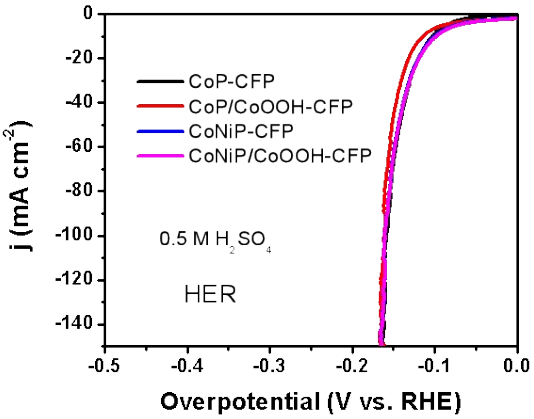

Fig. S12 LSV polarization curves of different catalysts in 0.5 M H_2SO_4 electrolyte for HER.

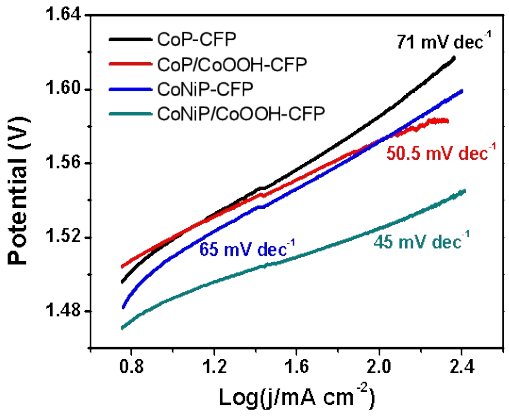

Fig. S13 LSV polarization curves derived Tafel plots of different catalysts in 1.0 M KOH electrolyte for electrocatalytic OER.

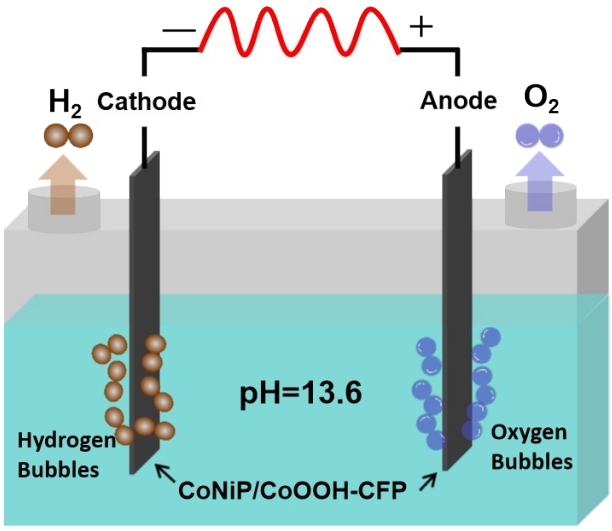

Fig. S14 Schematic diagram of self-made alkaline electrolyzer for overall water splitting.

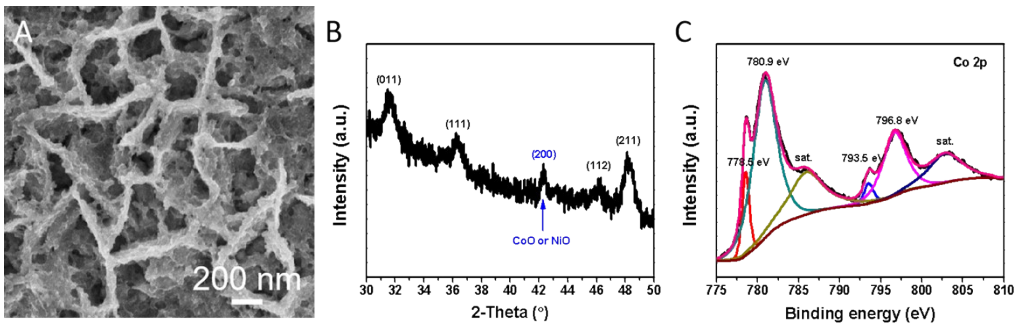

Fig. S15 The SEM image, XRD patterner, and Co 2p spectrum of CoNiP/CoOOH-CFP after 50 hours.

Table S1. Electrochemical activities of CFP-supported CoNiP-CoOOH and other cobalt-based HER electrocatalysts in alkaline solution.

Samples	η_{10} for HER	Tafel slope (HER)	References
CoNiP-CoOOH	0.120 V	76	This work
$CoNi_4P_2$	0.430 V	NG	*Energy Environ. Sci.*, 2014, **7**, 329
CoO-$CoSe_2$	0.337 V	131	*Adv. Sci.*, 2016, **3**, 1500426
CoS_2-Ti	0.260 V	129	*Electro. Acta*, 2014, **148**, 170
CoP/PCNF	0.191 V	111	*Nano Res.* , 2018, **11**, 1274
CoN_x/C	0.247 V	NG	*Nat. Commun.*, 2015, **6**, 7992
NiCoP/rGO	0.124 V	91	*Adv. Funct. Mater.*, 2016, **26**, 6785
NiFeCoP/NM	0.330 V	130	*Electro. Acta*, 2019, **306**, 651
Co_3O_4@Ni	0.225 V	68	*J. Power Sources*, 2017, **341**, 250
Ni_2P/CoP@C-NSG	0.144 V	61	*Mater. Today Sustain.* ,

Table S2. Electrochemical activities of CFP-supported CoNiP-CoOOH and other cobalt-based OER electrocatalysts in alkaline solution.

Samples	η_{10} for HER	Tafel slope (HER)	References
CoNiP-CoOOH	1.488 V	45	This work
Co_3O_4@Ni	1.500 V	84	*J. Power Sources*, 2017, **341**, 250
NiP Plates	1.530 V	64	*Energy Environ. Sci*., 2016, **9**, 1246.
CoP-TiO_x	1.567 V	72.1	*Small*, 2020, **16**, 1905075.
Cu-CoP	1.482 V	89.1	*Chem. Eng. J*., 2022, **432**, 134303.
CeO_x-CoP/C	1.543 V	69	*J. Mater. Sci*., 2018, **53**, 12123.
O-CoP	1.543 V	91.3	*Adv. Funct. Mater*., 2020, **30**, 1905252.
FeP-CoP	1.506 V	37.71	*RSC Adv*., 2023, **13**, 15031.
Mn-CoP	1.547 V	65.1	*J. Mater. Res*., 2018, **33**, 1258.
CoP-CoFeP	1.496 V	35.6	*ACS Nano*, 2023, **17**, 22744.

CoP-NCNHP	1.540 V	70	*J. Am. Chem. Soc.*, 2018, **140**, 2610.
CoP/Co-N-C	1.550 V	115	*Small*, 2023, **19**, 2207474.
CoFeP@C	1.492 V	44.8	*Adv. Energy Mater.*, 2022, **12**, 2202394.