Supporting Information (SI):

Electronic modulation of CoP nanosheets array by Zn doping as efficient electrocatalysts for overall water splitting

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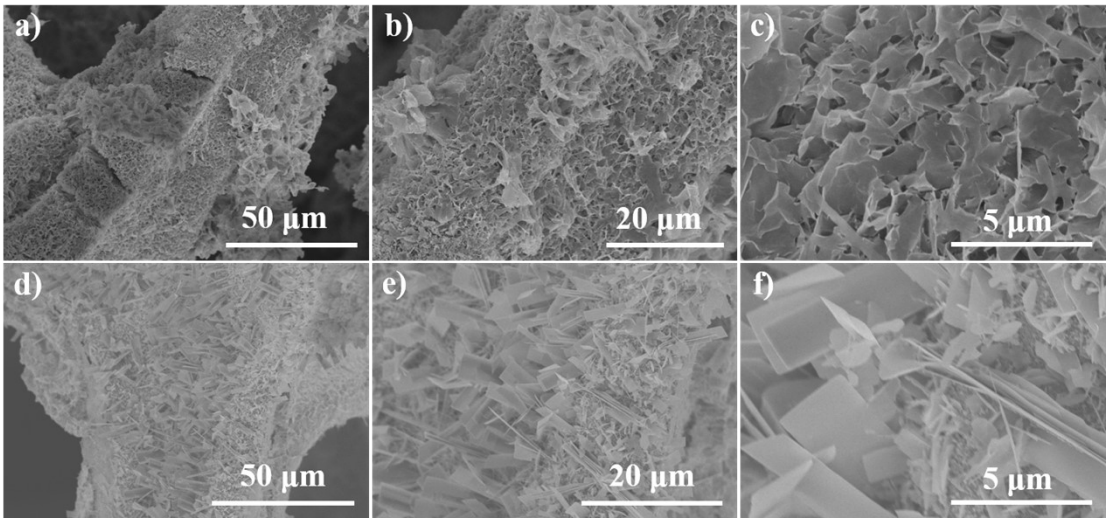


Fig. S1 SEM images of (a-c) $CoZn(OH)_x$/CF and (d-f) $Co(OH)_x$/CF.

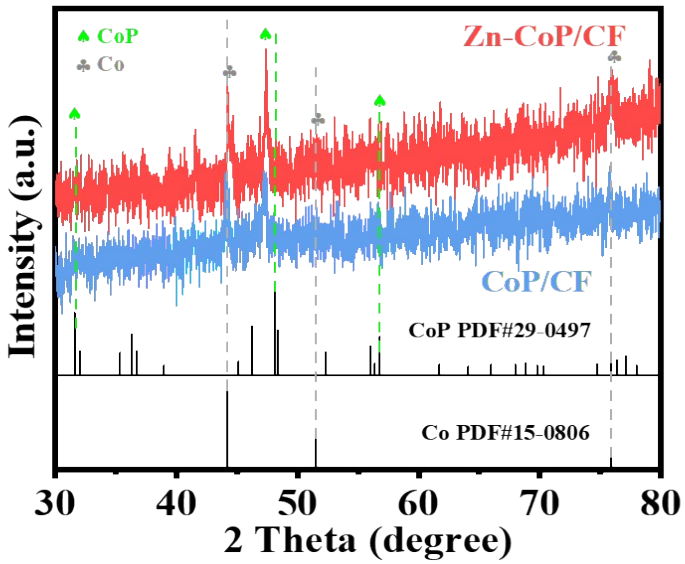


Fig. S2 XRD pattern of Zn-CoP/CF and CoP/CF.

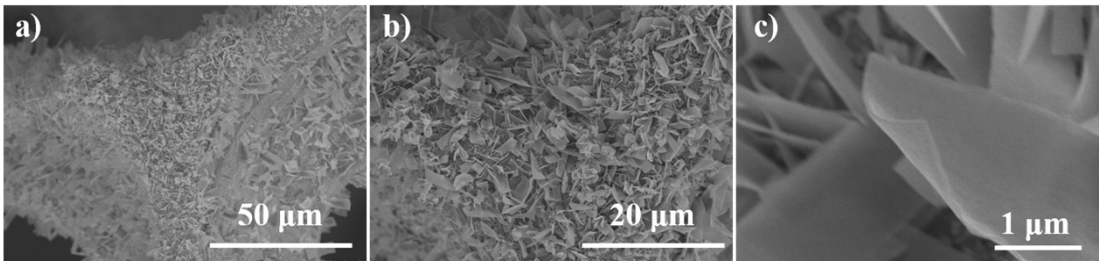
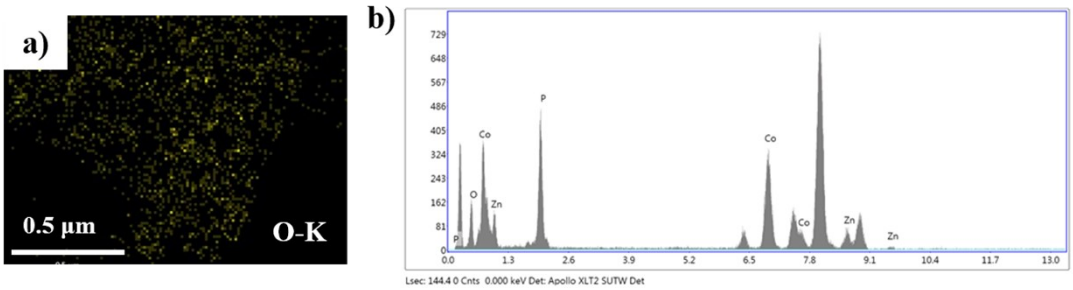


Fig. S3 SEM images of (a-c) CoP/CF.



c)

Element	Weight %	Atomic %	Net Int.	Error %	Kratio
O K	59.58	75.59	14.7	9.78	0.21
P K	33.95	22.25	56.6	7.30	0.14
CoK	4.57	1.58	70.7	1.63	0.04
ZnK	1.90	0.59	36.3	3.03	0.02

Fig. S4 (a) EDX element mapping of O element in Scanning TEM image of Zn-CoP/CF; (b-c) EDX results of Zn-CoP/CF.

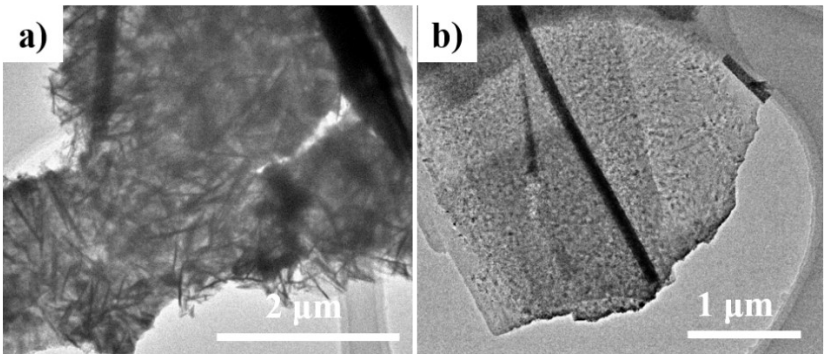


Fig. S5 (a-b) TEM images at different magnifications of CoP/CF.

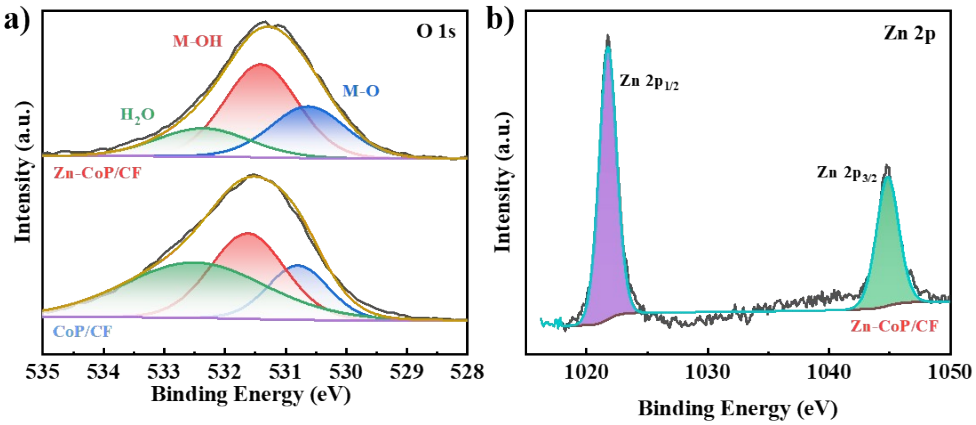


Fig. S6 (a) High-resolution XPS spectra of O 1s of Zn-CoP/CF and CoP/CF. (b) High-resolution XPS spectra of Zn 2p of Zn-CoP/CF.

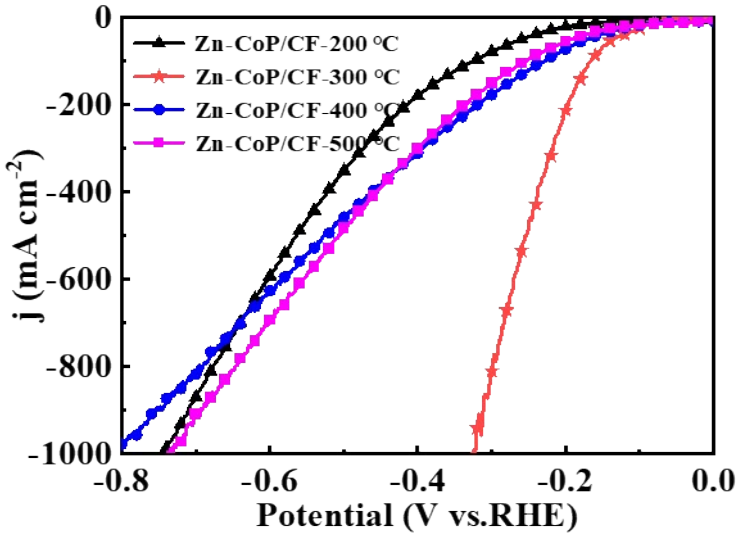


Fig. S7 Polarization curves for HER of catalysts at different phosphating temperatures.

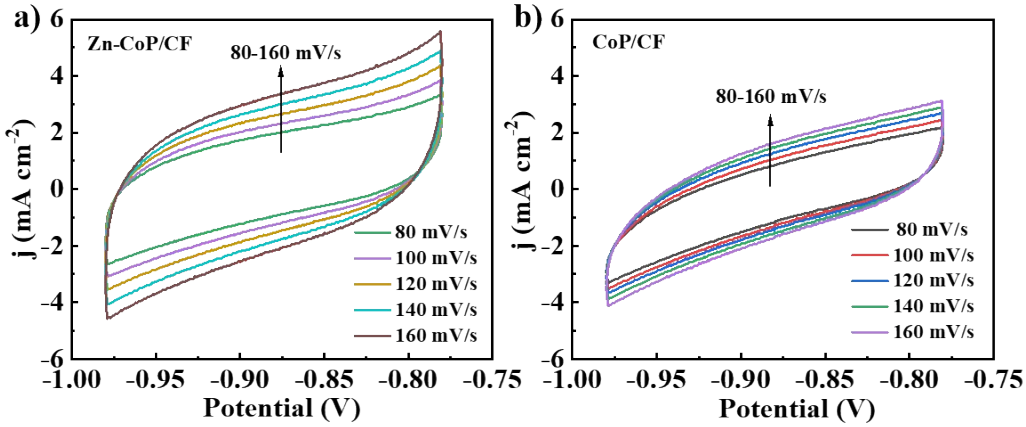


Fig. S8 For HER: CV curves of (a) Zn-CoP/CF and (b) CoP/CF in 1 M KOH solution.

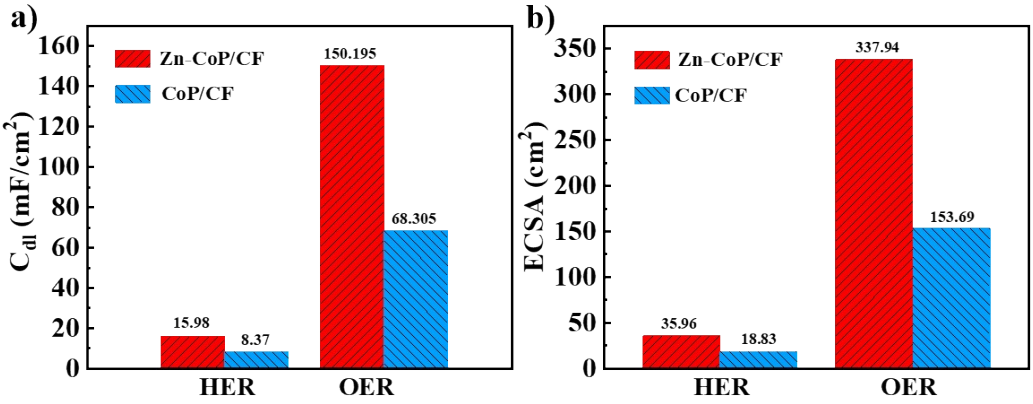


Fig. S9 For HER and OER: (a) C_{dl} values and (b) ECSA values of different samples for comparison. C_{dl} is 1/2 of the slope of capacitive current density vs. scan rate in linear plots.

According to the previously reported work, the ECSA was calculated based on the values of C_{dl}.

$$\mathrm{ECSA} = \frac{\mathrm{Cdl}}{\mathrm{Cs}}$$

The specific capacitance (C_s) is used as 40 μF/cm^2 as reported in 1 M KOH. In our work, C_{dl} is multiplied by the geometric area of the electrode first.

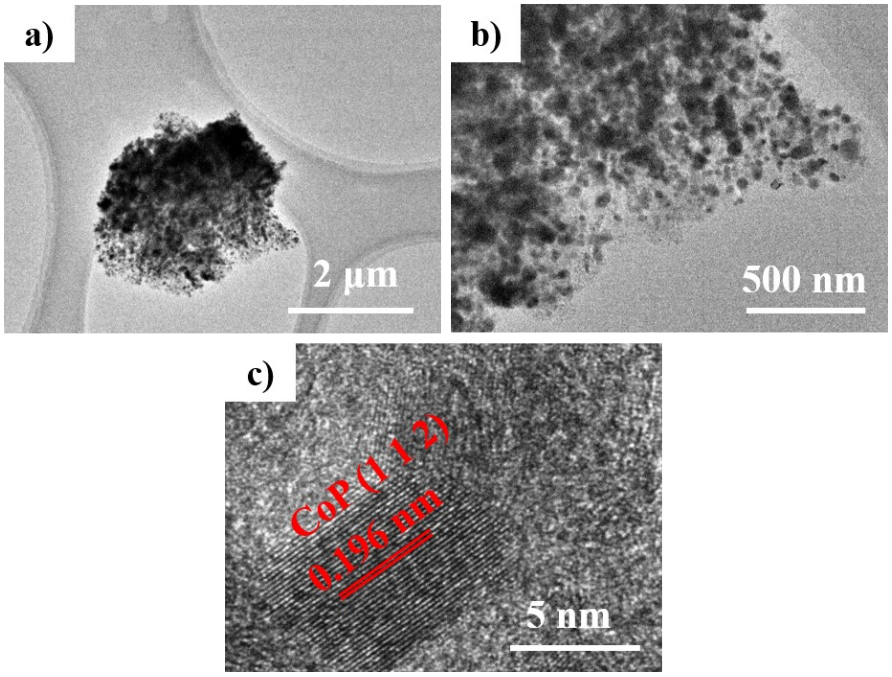


Fig. S10 (a-b) TEM images and (c) high-resolution TEM image of Zn-CoP/CF after HER stability test.

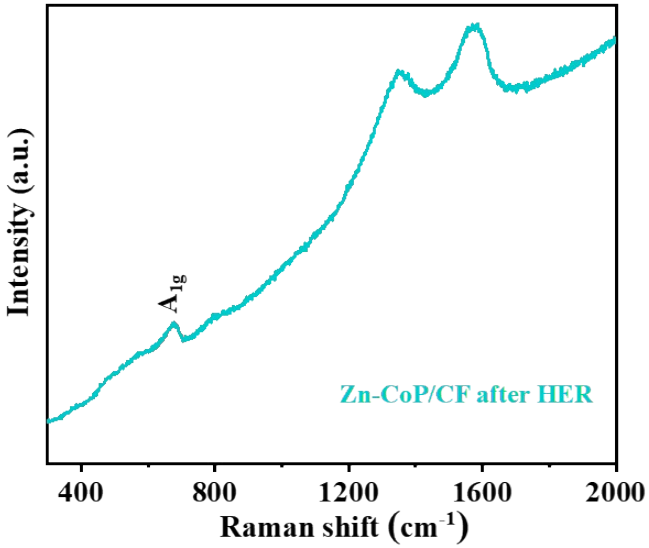


Fig. S11 Raman spectra of Zn-CoP/CF after HER stability test.

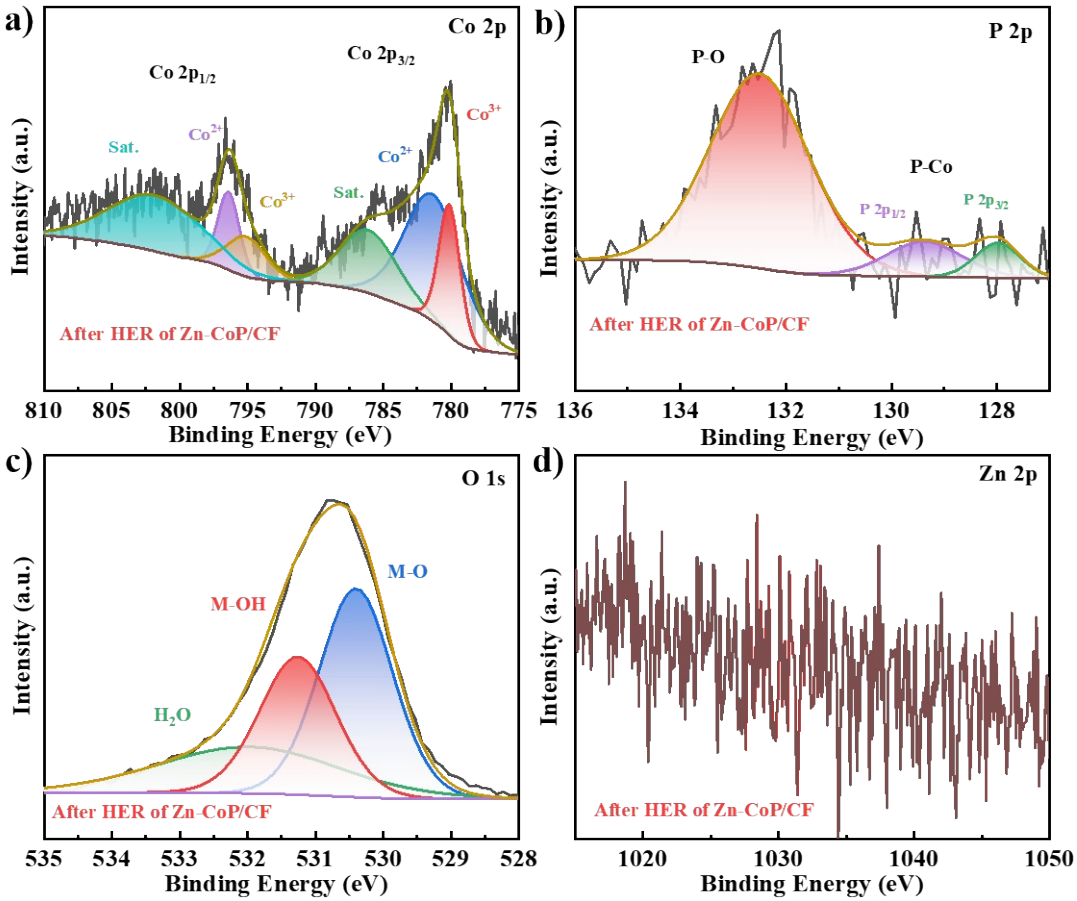


Fig. S12 XPS spectrum of Zn-CoP/CF after HER electrochemical test: (a) Co 2p; (b) P 2p; (c) O 1s; (d) Zn 2p.

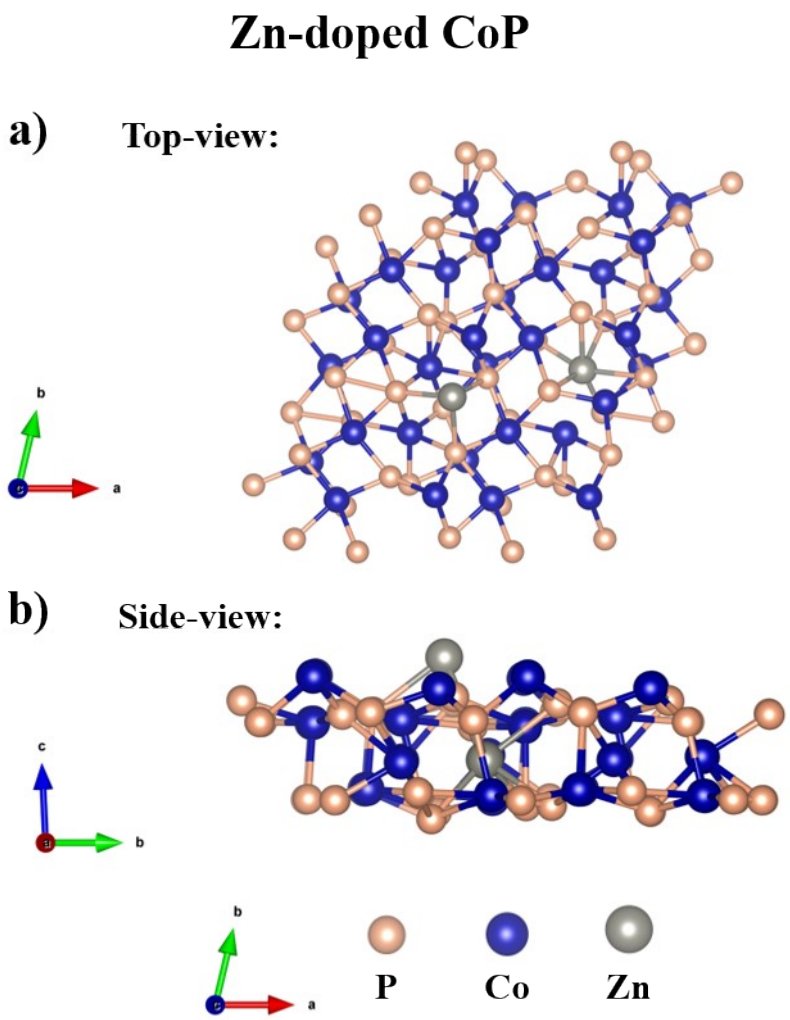


Fig. S13 (a) Top view and (b) side view of Zn-doped CoP. Orange, blue, and gray balls represent P, Co, and Zn atoms, respectively.

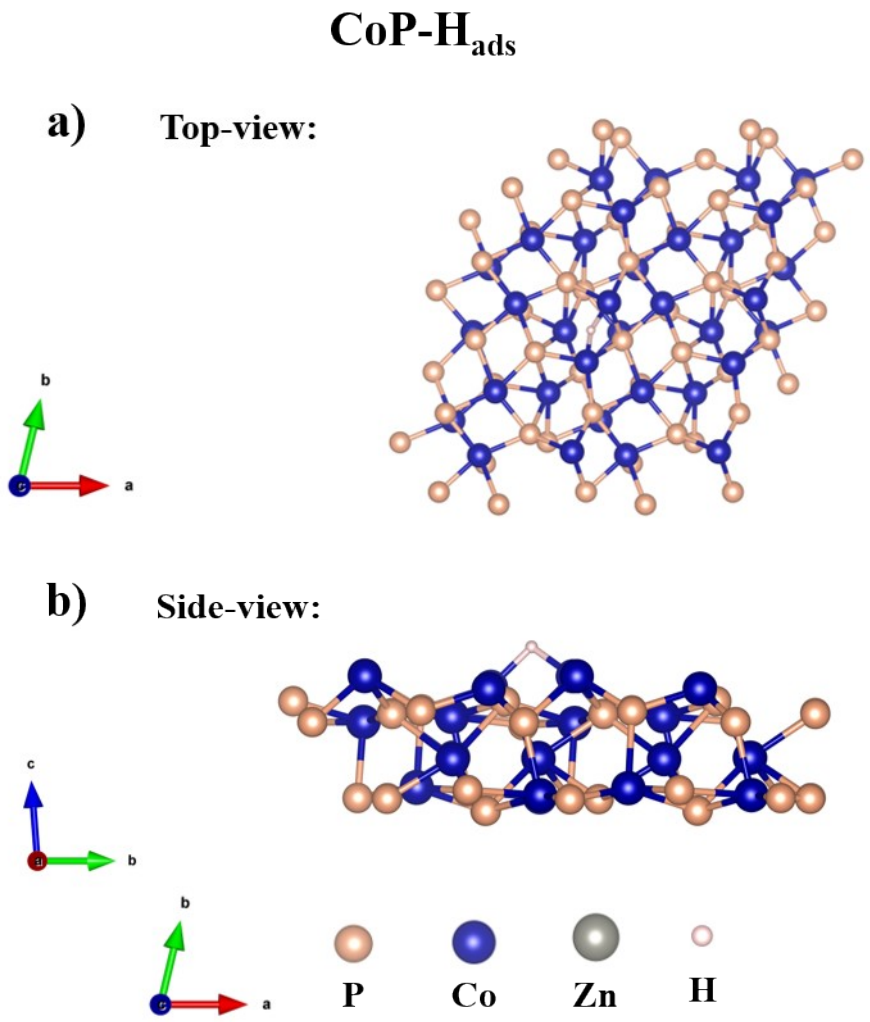


Fig. S14 (a) Top and (b) side view of optimized models for hydrogen adsorption on CoP (111) surface.

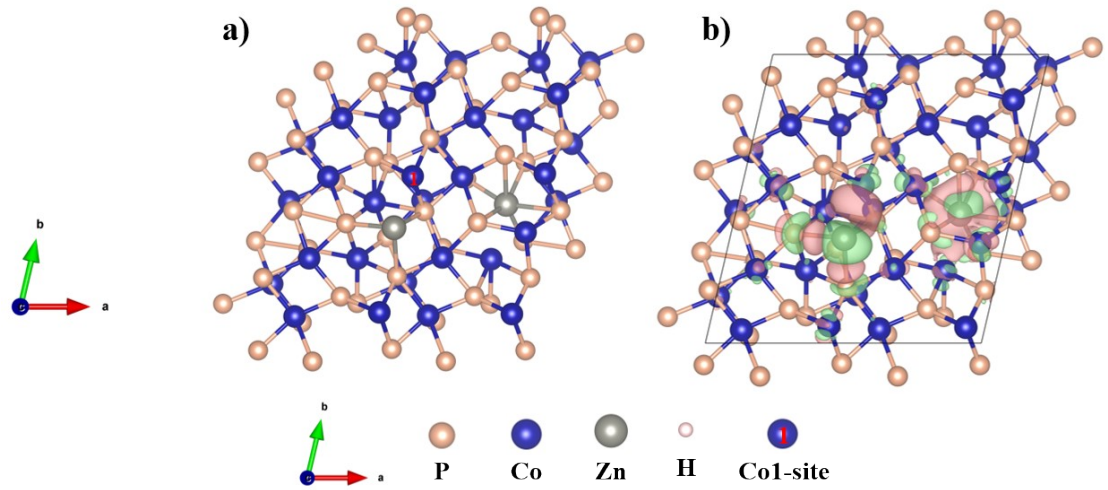


Fig. S15 (a) Location of Co1-site in the Zn-doped CoP structure. (b) The charge density difference at the Zn-doped CoP with an isosurface value of 0.002 e $Å^{-3}$. Green and pink colors represent charge depletion and accumulation, respectively.

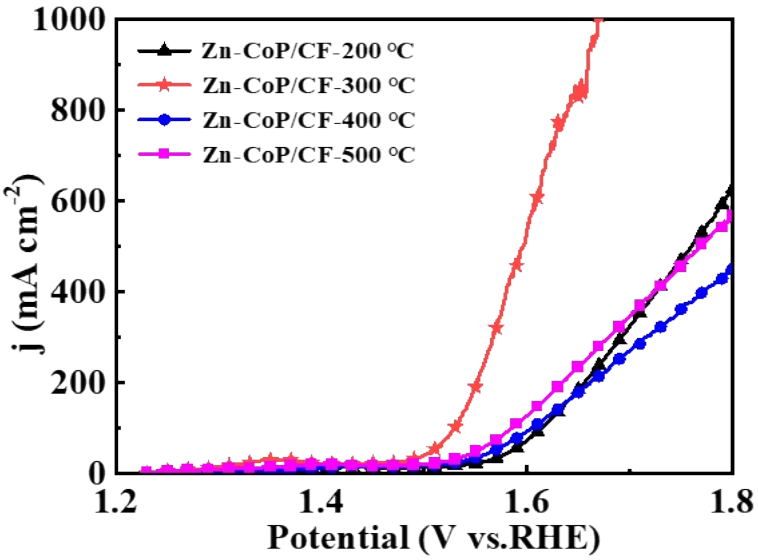


Fig. S16 Polarization curves for OER of catalysts at different phosphating temperatures.

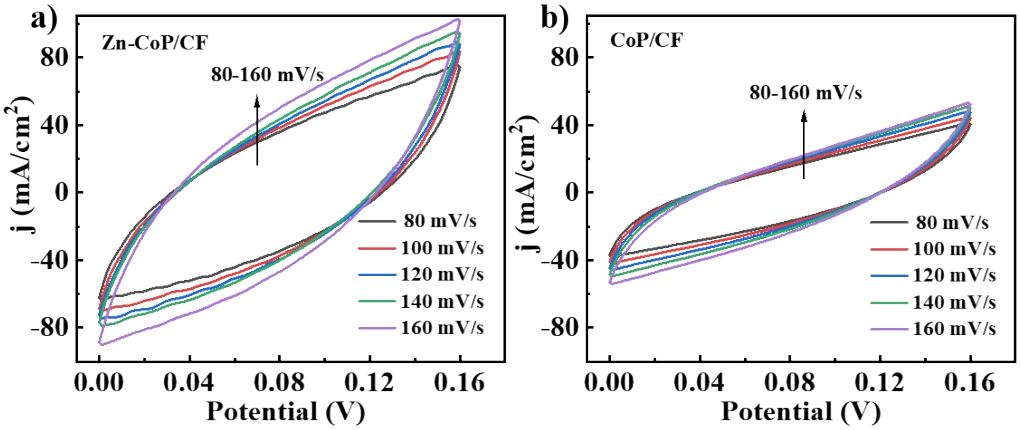


Fig. S17 For OER: CV curves of (a) Zn-CoP/CF and (b) CoP/CF in 1 M KOH solution.

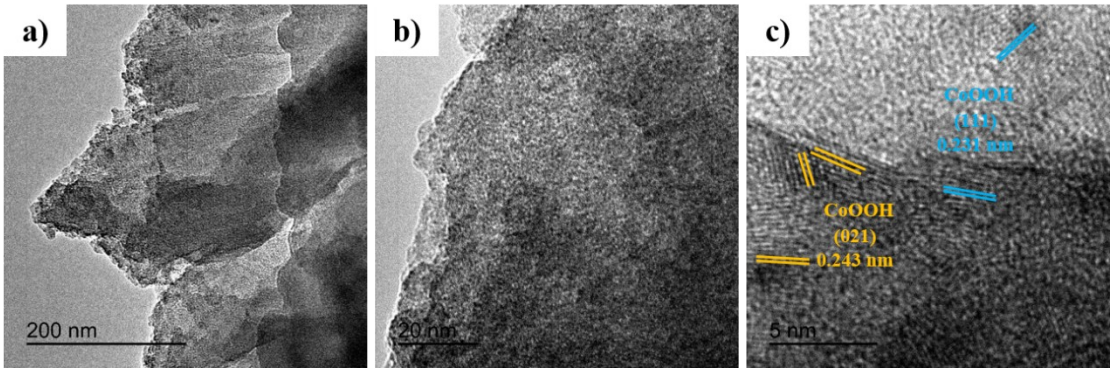


Fig. S18 (a-b) TEM images and (c) high-resolution TEM image of Zn-CoP/CF after OER stability test.

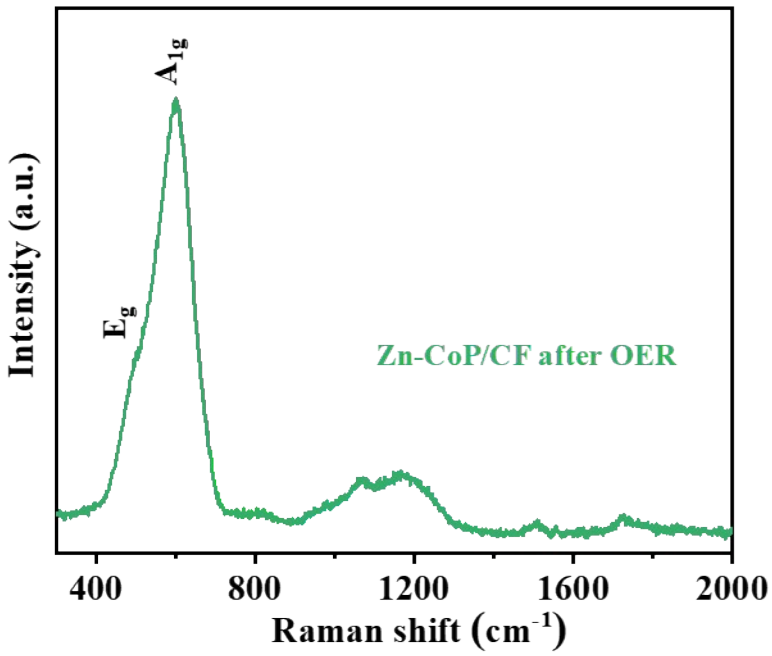


Fig. S19 Raman spectra of Zn-CoP/CF after OER stability test.

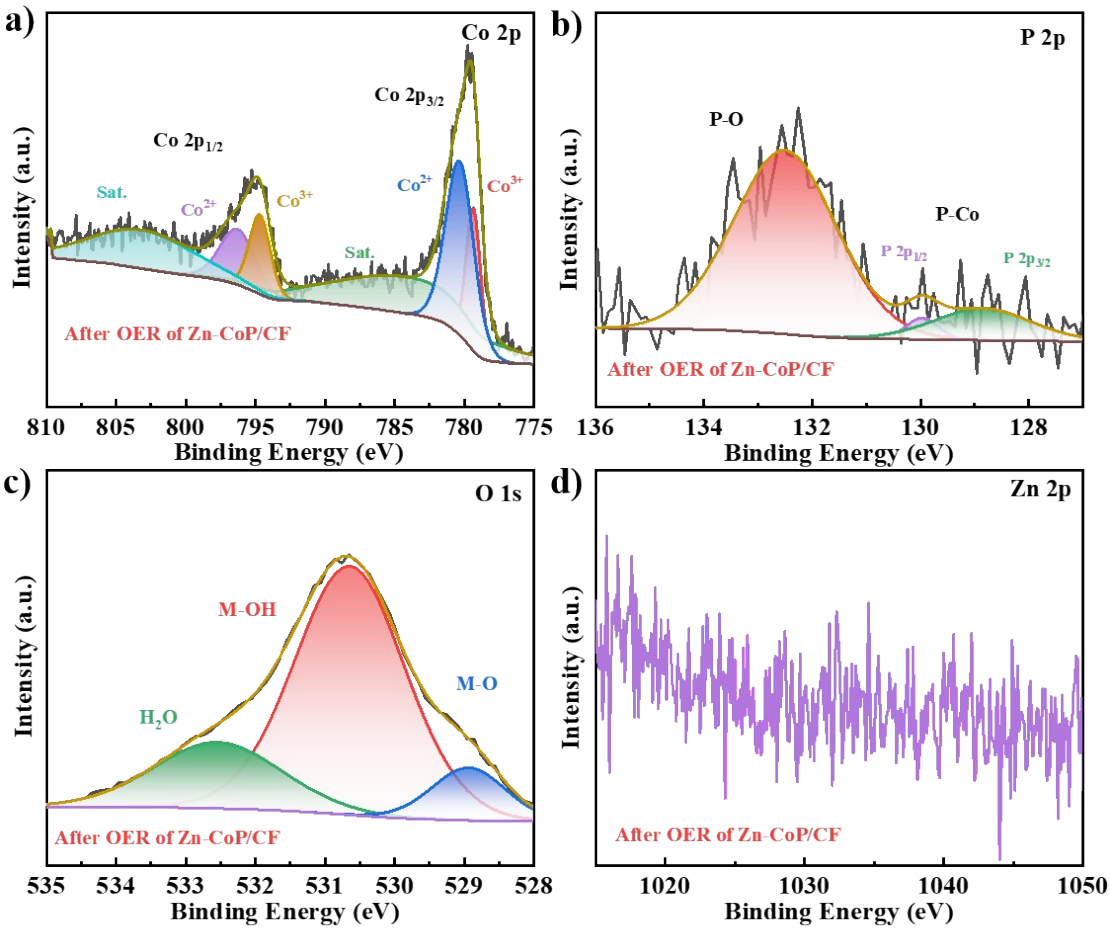


Fig. S20 XPS spectrum of Zn-CoP/CF after OER electrochemical test: (a) Co 2p; (b) P 2p; (c) O 1s; (d) Zn 2p.

Table S1. Fitted data from Nyquist plots of as-synthesized samples in alkaline conditions.

Catalysts	Electrolyte	HER		OER	
		R_s (ohm)	R_{ct} (ohm)	R_s (ohm)	R_{ct} (ohm)
Zn-CoP/CF	1 M KOH	3.596	231.1	3.543	21.81
CoP/CF	1 M KOH	3.557	312.8	3.879	77.72

Table S2. The recently reported Co-based bifunctional electrocatalysts for HER and OER in alkaline media.

Catalysts	Electrolyte	HER		OER		References
		j (mA/cm^2)	Overpotential (mV)	j (mA/cm^2)	Overpotential (mV)	
Zn-CoP/CF	1 M KOH	100 500 1000	166 255 327	100 500 1000	300 367 440	This work
Fe-CoP HNSs	1 M KOH	10	79	10	220	Int. J. Hydrogen Energy, **2021**, 46(52), 26391-26401
Cr-CoP/CP	1 M KOH	10	67	10	251	Chem. Eng. J., **2021**,425, 130651
V-CoP@a-CeO_2	1 M KOH	100	140	100	340	Adv. Funct. Mater. **2020**, 30(14), 1909618
CoFeP/CF	1 M KOH	50	152.6	50	277.9	J. Colloid Interface Sci., **2020**, 569, 140-149
CoP/SPNF	1 M KOH	100	160	100	266	Inorg. Chem. **2020** 59 (12), 8522-8531
Fe_3Co_7@PCNSs	1 M KOH	10	205	10	300	J. Colloid Interface Sci., **2019**, 537, 280-294
CoNi-OOH-30(40)	1 M KOH	10	210	10	279	Electrochim. Acta, **2019**, 301, 449-457
Fe-CoP HTPs/NF	1 M KOH	10	98	10	230	Small **2018**, 14 (14),

						1704233
Ni-Co-S/Ni-Co-P	1 M KOH	20	110	50	240	J. Mater. Chem. A **2018**, 6 (41), 20297-20303
NiCoP NWAs/NF	1 M KOH	100	197	100	370	J. Mater. Chem. A **2017**, 5 (28), 14828-14837

Table S3. The recently reported Co-based bifunctional electrocatalysts for overall water splitting in alkaline media.

Catalysts	Electrolyte	Overall water splitting		References
		j ($mA\ cm^{-2}$)	Potential (mV)	
Zn-CoP/CF	1 M KOH	100 1000	1.71 2.01	This work
Fe-CoP HNSs	1 M KOH	20	1.60	Int. J. Hydrogen Energy, **2021**, 46(52), 26391-26401
Cr-CoP/CP	1 M KOH	100	1.73	Chem. Eng. J., **2021**,425, 130651
V-CoP@a-CeO_2	1 M KOH	100	1.71	Adv. Funct. Mater. **2020**, 30(14), 1909618
CoFeP/CF	1 M KOH	10	1.495	J. Colloid Interface Sci., **2020**, 569, 140-149
CoP/SPNF	1 M KOH	50	1.621	Inorg. Chem., **2020**, 59 (12), 8522-8531
Fe_3Co_7@PCNSs	1 M KOH	100	1.794	J. Colloid Interface Sci., **2019**, 537, 280-294
CoNi-OOH-30(40)	1 M KOH	10	1.76	Electrochim. Acta, **2019**, 301, 449-457
Fe-CoP HTPs/NF	1 M KOH	10	1.59	Small, **2018**, 14 (14), 1704233
NiCoP NWAs/NF	1 M KOH	20	1.64	J. Mater. Chem. A, **2017**, 5 (28), 14828-14837