Supplementary Information

Charge Optimization Induces Reconstruction via Compounding $Ni(OH)_2$ and CoP: A Novel Route to Construct Electrocatalysts for Overall water splitting

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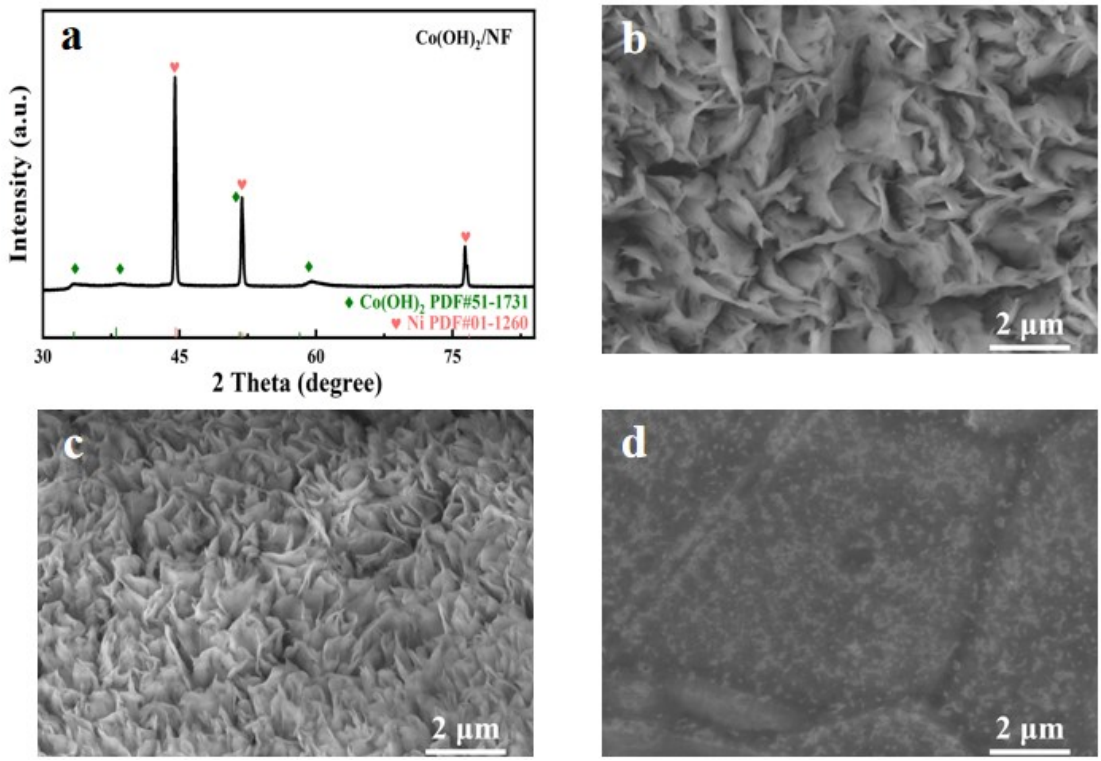

Figure S1. (a) XRD patterns of $Co(OH)_2$/NF, (b-d) SEM images of $Co(OH)_2$/NF with the deposition time of 5 minutes, 10 minutes and 20 minutes, respectively.

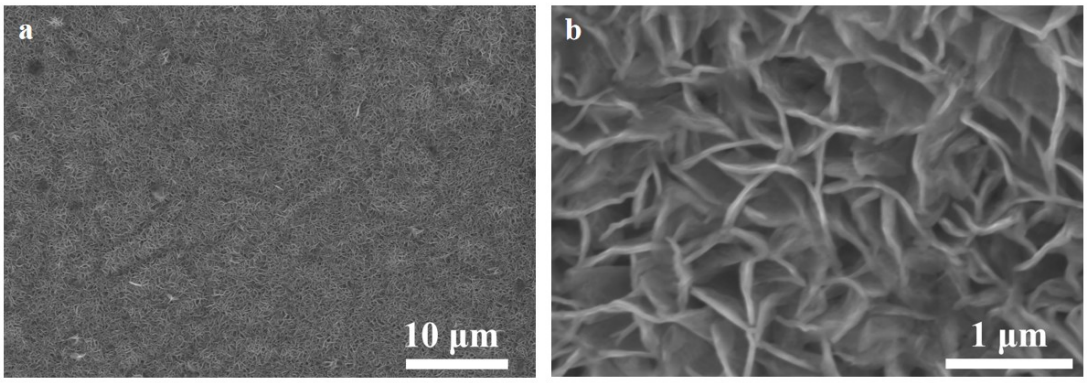

Figure S2. (a) Low and (b) high-resolution SEM images of CoP.

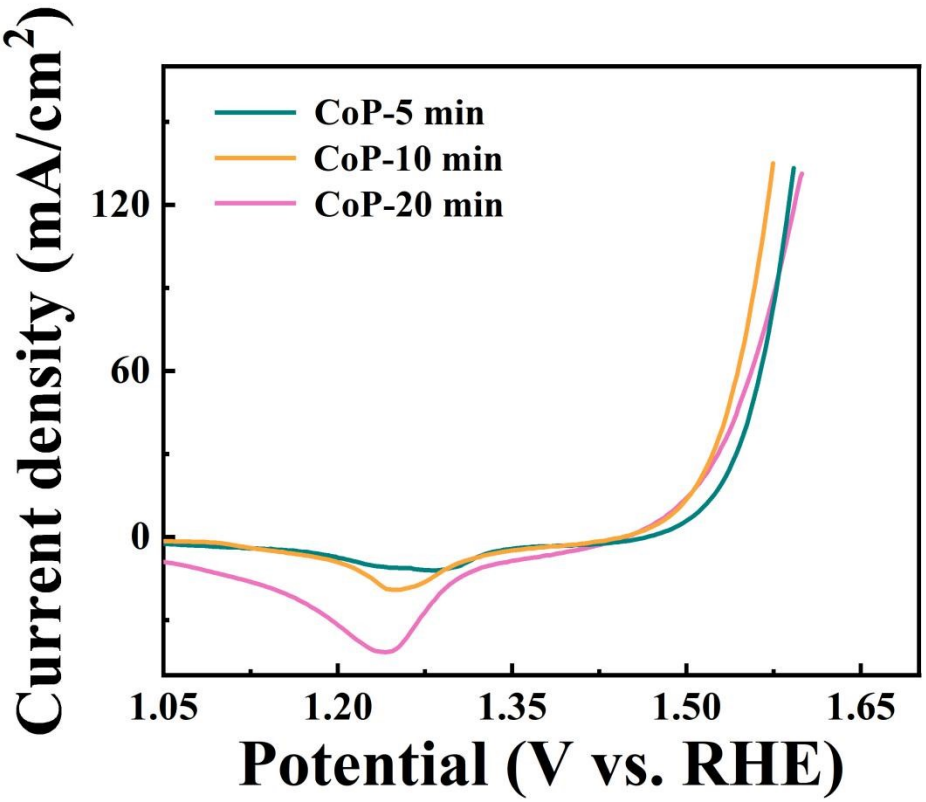

Figure S3. OER LSV curves of CoP with different description time.

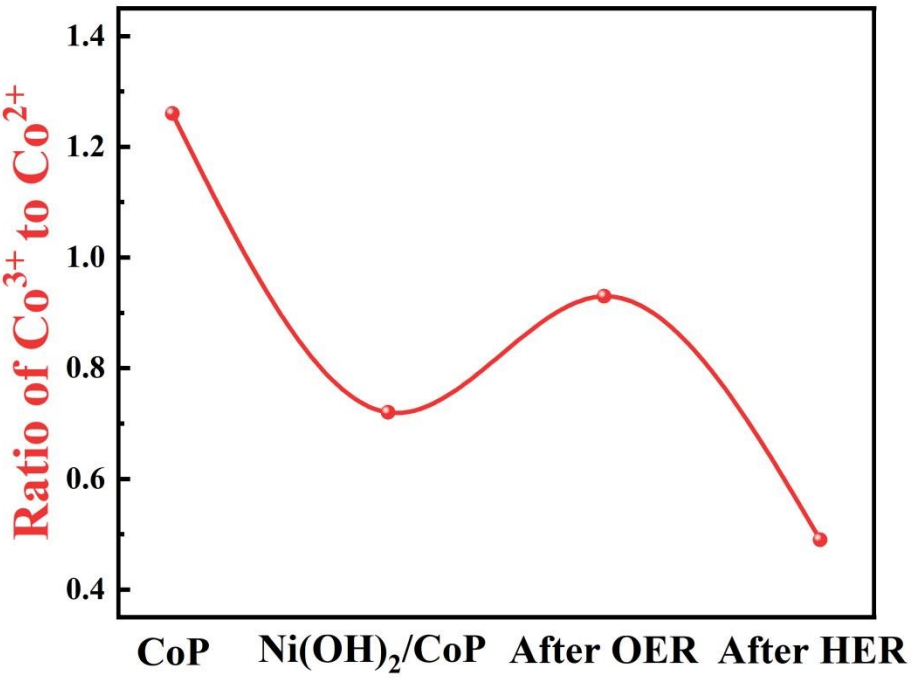

Figure S4. The ratio of Co^{3+} to Co^{2+} in CoP, $Ni(OH)_2$/CoP, $Ni(OH)_2$/CoP after OER and $Ni(OH)_2$/CoP after HER.

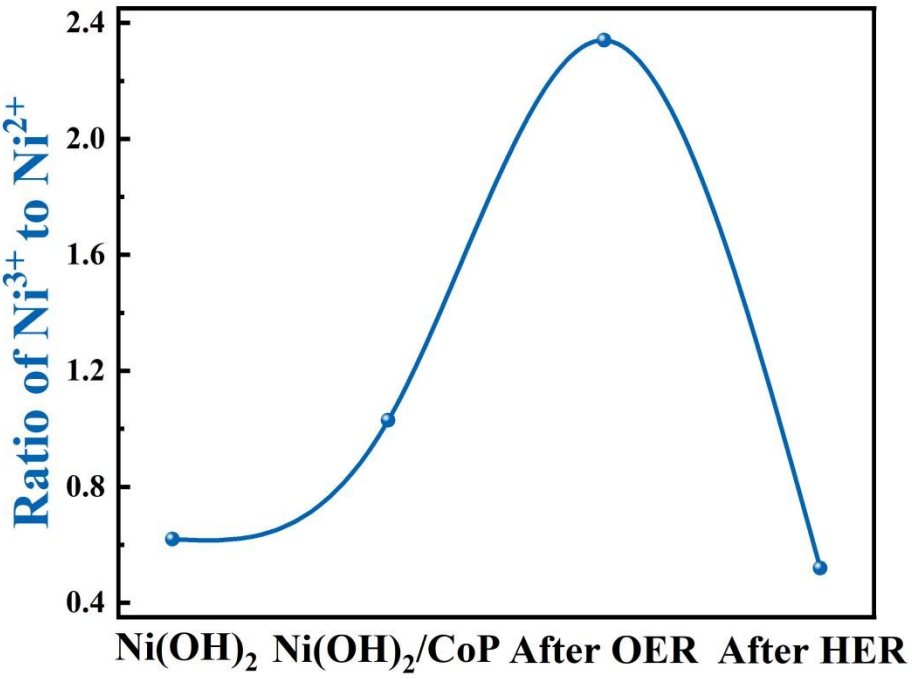

Figure S5. The ratio of Ni^{3+} to Ni^{2+} in $Ni(OH)_2$, $Ni(OH)_2$/CoP, $Ni(OH)_2$/CoP after OER and $Ni(OH)_2$/CoP after HER.

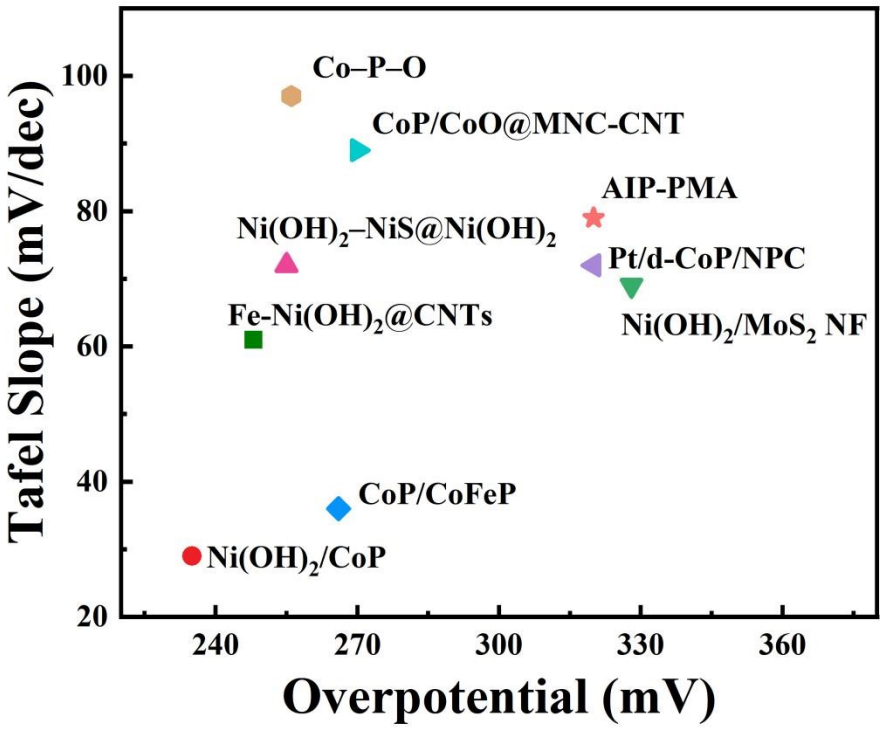

Figure S6. Overpotential and Tafel slope of $Ni(OH)_2$/CoP and the reported noble-free electrocatalysts for OER, respectively.

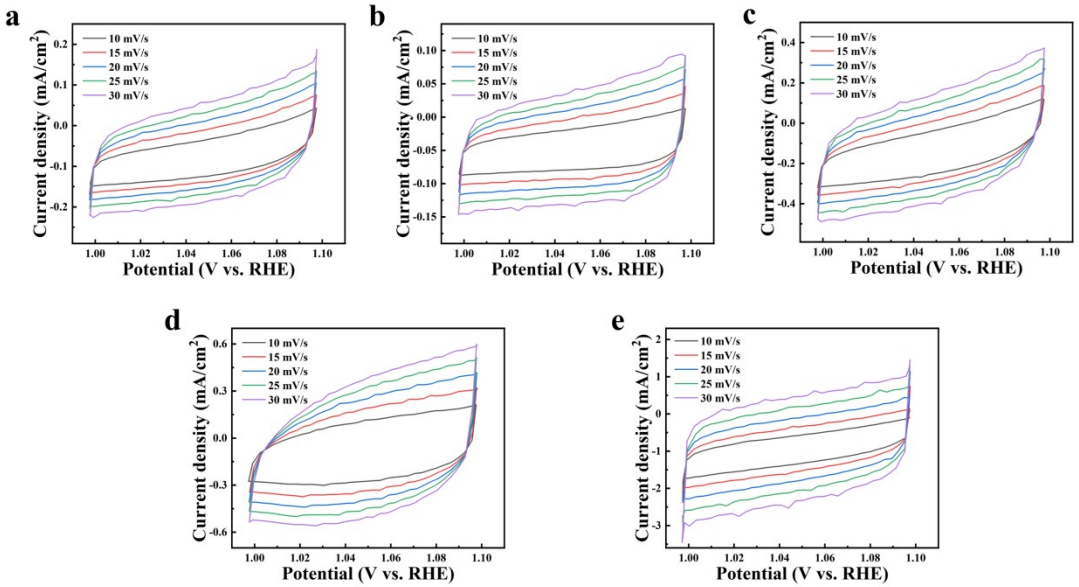

Figure S7. CV curves at different scan rates between 1.00 and 1.10 V vs. RHE for (a) CoP, (b) $Ni(OH)_2$, (c) $Ni(OH)_2$/CoP-5, (d) $Ni(OH)_2$/CoP-10 and (e) $Ni(OH)_2$/CoP-20, respectively.

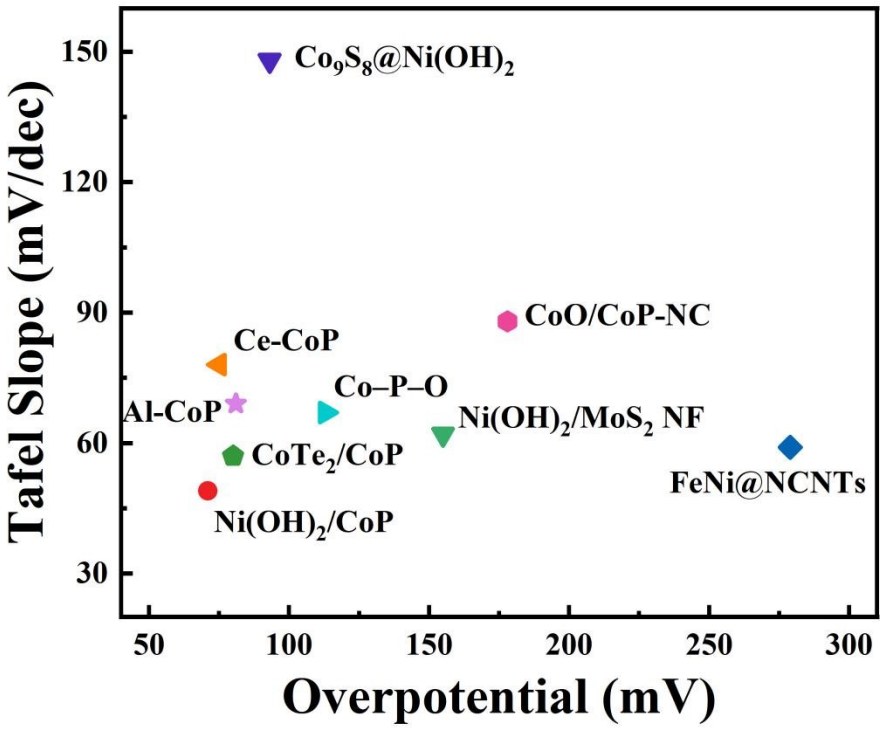

Figure S8. Overpotential and Tafel slope of $Ni(OH)_2$/CoP and the reported noble-free electrocatalysts for HER, respectively.

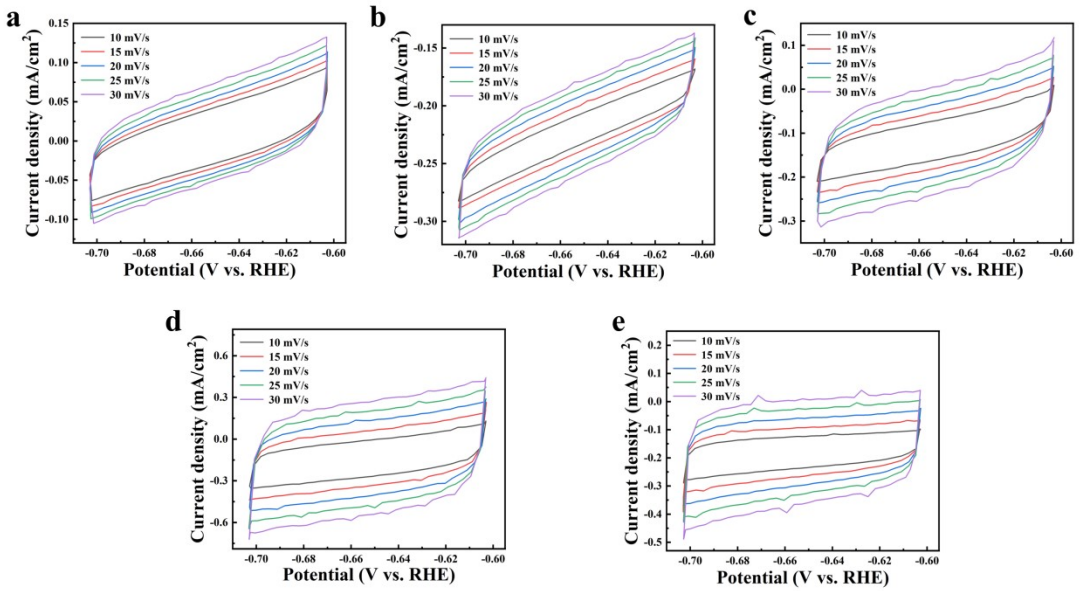

Figure S9. CV curves at different scan rates between -0.70 and -0.60 V vs. RHE for (a) CoP, (b) $Ni(OH)_2$, (c) $Ni(OH)_2$/CoP-5, (d) $Ni(OH)_2$/CoP-10 and (e) $Ni(OH)_2$/CoP-20,respectively.

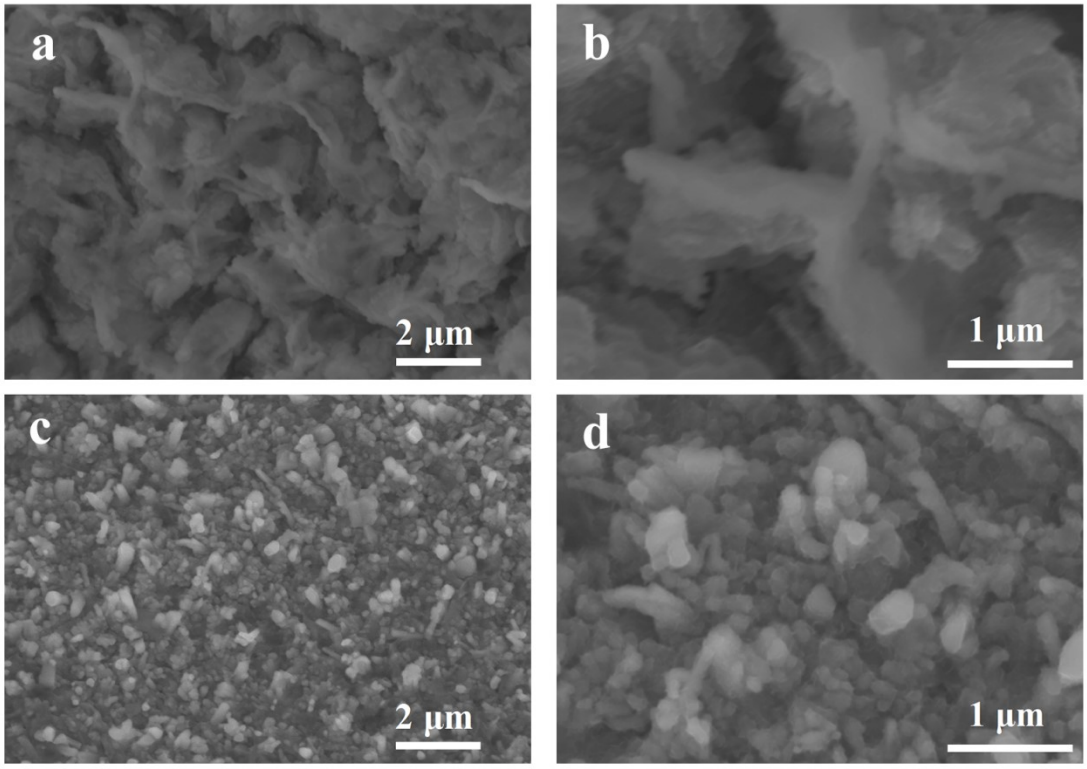

Figure S10. Low and high-resolution SEM images of (a,b) $Ni(OH)_2$/CoP-10 after OER and (c,d) $Ni(OH)_2$/CoP-10 after HER.

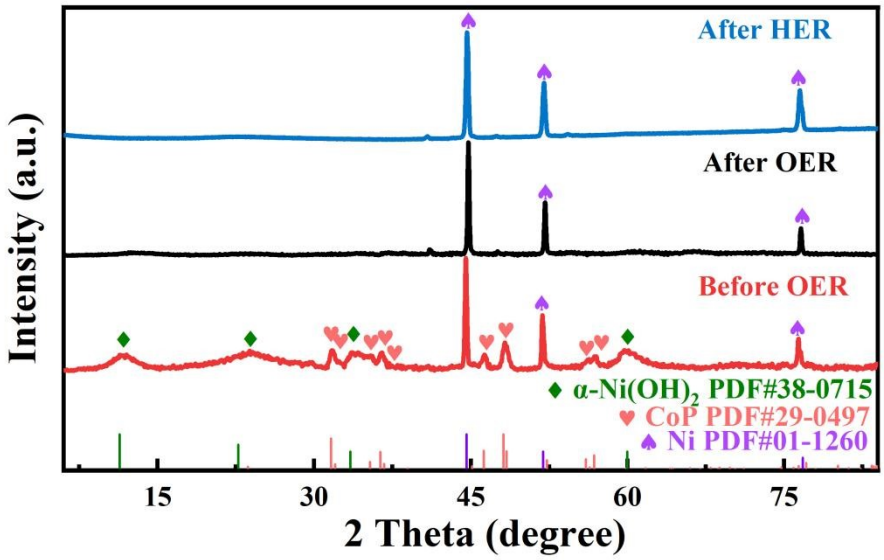

Figure S11. XRD patterns of $Ni(OH)_2$/CoP-10 before OER, after OER and after HER.

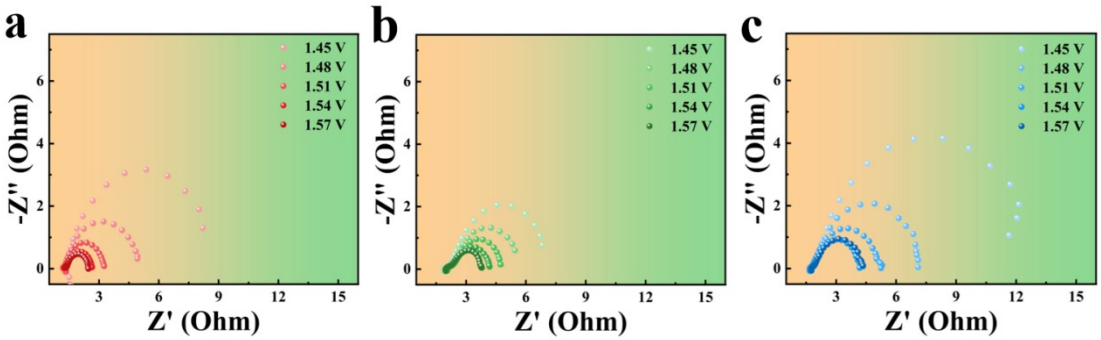

Figure S12. Situ EIS plots of (a) $Ni(OH)_2$/CoP, (b) CoP and (c) $Ni(OH)_2$.

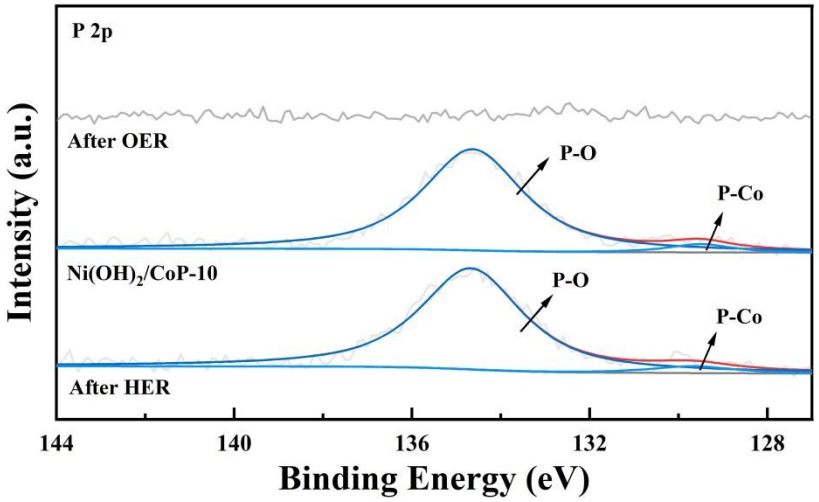

Figure S13. P 2p XPS spectra of $Ni(OH)_2$/CoP-10, $Ni(OH)_2$/CoP-10 after OER and $Ni(OH)_2$/CoP-10 after HER.

Table S1. Overpotential and Tafel slope of the reported noble-free electrocatalysts for OER (η_{10}, η_{50} and η_{100}: overpotential at 10, 50 and 100 mA/cm^2, respectively).

Catalysts	Electrolyte	η (mV)	Tafel Slope (mV/dec)	References
$Ni(OH)_2$/CoP	**1M KOH**	**η_{10}=235 η_{50}=266 η_{100}=283**	**29**	**This work**
Fe-$Ni(OH)_2$@CNTs	1M KOH	η_{10}=248	61	1
$Ni(OH)_2$–NiS@$Ni(OH)_2$	1M KOH	η_{10}=255	72	2
Se-NiS_2	1M KOH	η_{50}=343	140	3
$Ni(OH)_2$/MoS_2 NF	1M KOH	η_{10}=328	69	4
Ni/$Ni(OH)_2$@NM	1M KOH	η_{100}=337	47	5
CoP/CoFeP	1M KOH	η_{10}=266	36	6
Pt/d-CoP/NPC	1M KOH	η_{10}=320	72	7
CoP/CoO@MNC-CNT	1M KOH	η_{10}=270	89	8
Co–P–O	1M KOH	η_{10}=256	97	9
AIP-PMA	1M KOH	η_{10}=320	79	10

Table S2. Overpotential and Tafel slope of the reported noble-free electrocatalysts for HER (η_{10}, η_{50} and η_{100}: overpotential at 10, 50 and 100 mA/cm^2, respectively).

Catalysts	Electrolyte	η (mV)	Tafel Slope (mV/dec)	References
$Ni(OH)_2$/CoP	**1M KOH**	**η_{10}=71 η_{50}=113 η_{100}=145**	**49**	**This work**
$Ni(OH)_2/MoS_2$ NF	1M KOH	η_{10}=155	62	4
$Ni/Ni(OH)_2$@NM	1M KOH	η_{100}=164	90	5
Co–P–O	1M KOH	η_{10}=113	67	9
Co_9S_8@$Ni(OH)_2$	1M KOH	η_{10}=93	148	11
FeNi@NCNTs	1M KOH	η_{10}=279	59	12
Al-CoP	1M KOH	η_{10}=75	78	13
$CoTe_2$/CoP	1M KOH	η_{10}=80	57	14
NiZn@C-CoP	1M KOH	η_{10}=78	57	15
CoO/CoP-NC	1M KOH	η_{10}=178	88	16
Ce-CoP	1M KOH	η_{10}=81	69	17

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