Plasma-engineered bifunctional Cobalt-metal organic framework derivatives for high-performance complete water electrolysis

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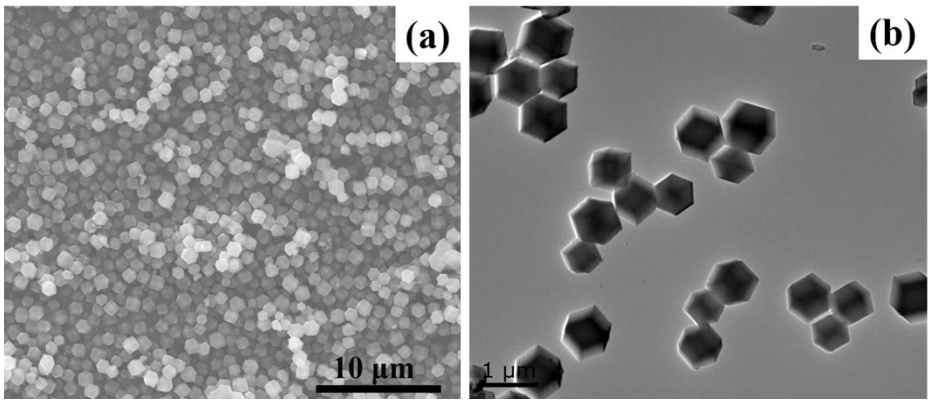

Fig. S1 (a) SEM images of the ZIF-67 and corresponding (b) TEM images.

Fig. S2 3D ZIF-67 polyhedron is disposed via P dopants and pyrolysis procedures simultaneously under Ar-N_2 RF plasma discharge process.

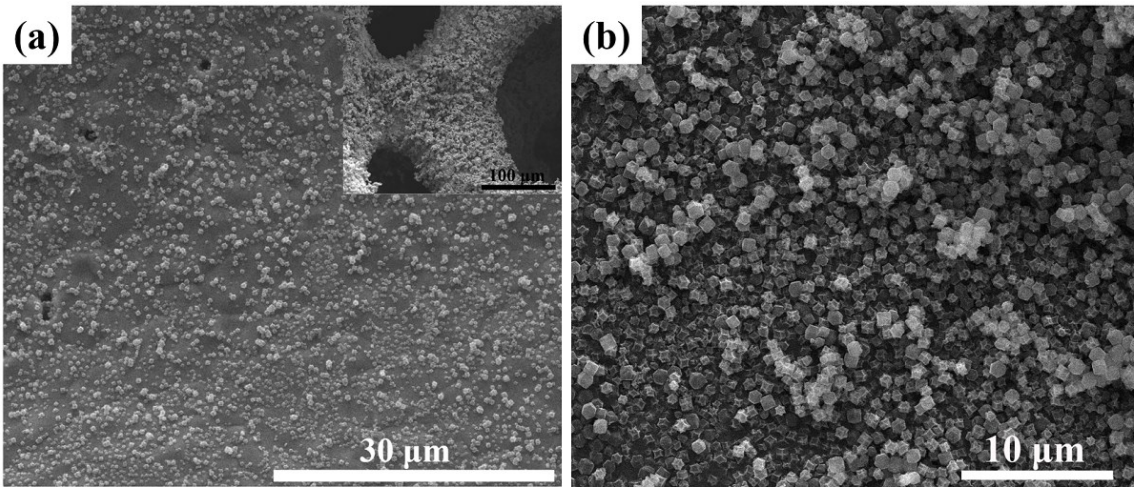

Fig. S3 The SEM images of the MOFs-derived CoPO on NF.

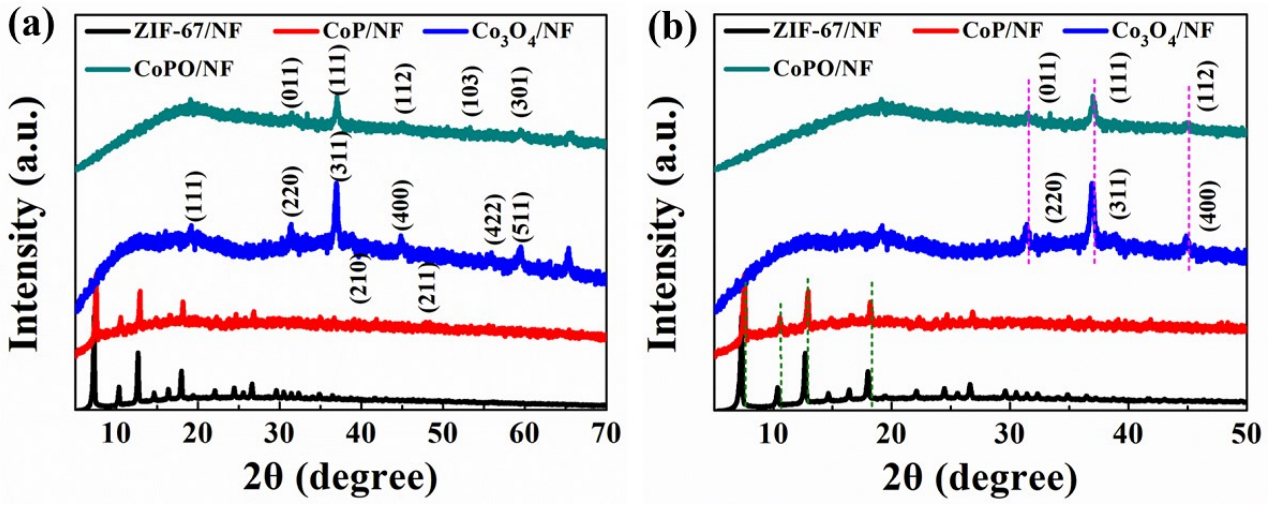

Fig. S4 (a) The XRD patterns of these samples and (b) the enlarged view.

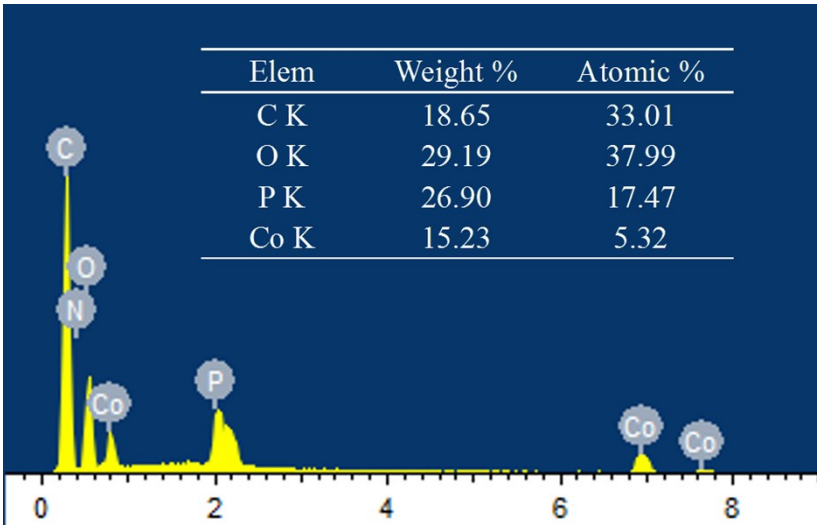

Fig. S5 The EDX analysis of the CoPO/NF.

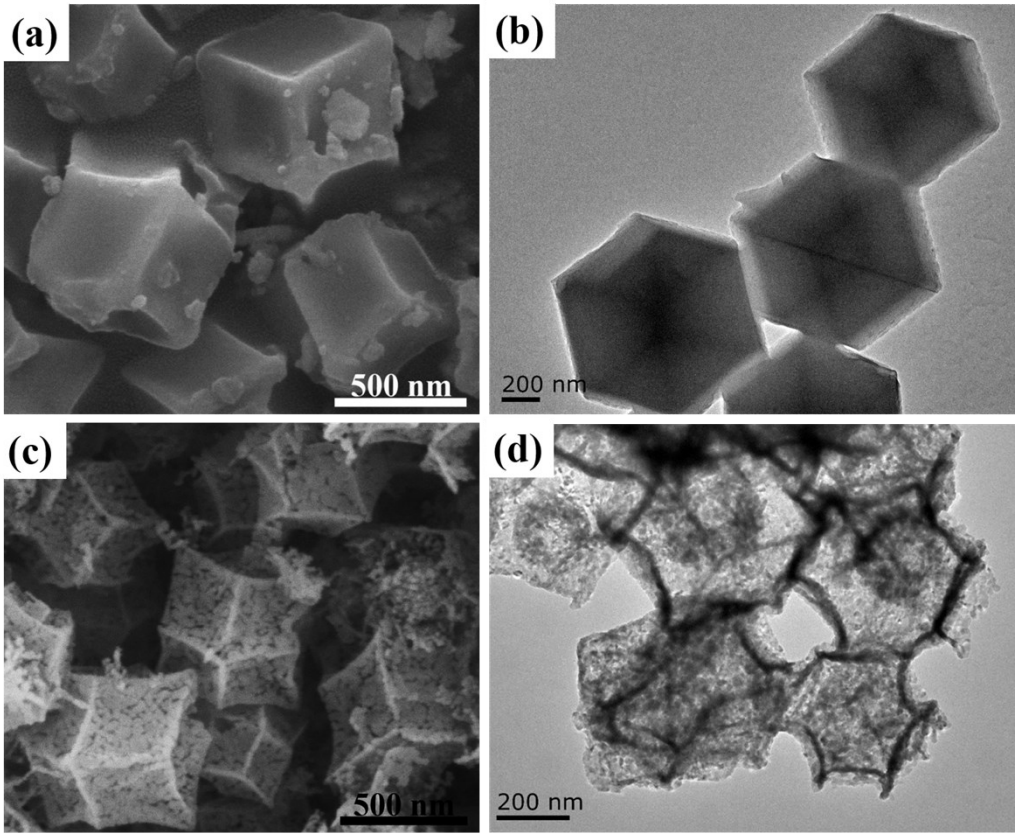

Fig. S6 SEM and TEM images of the CoP and Co_3O_4.

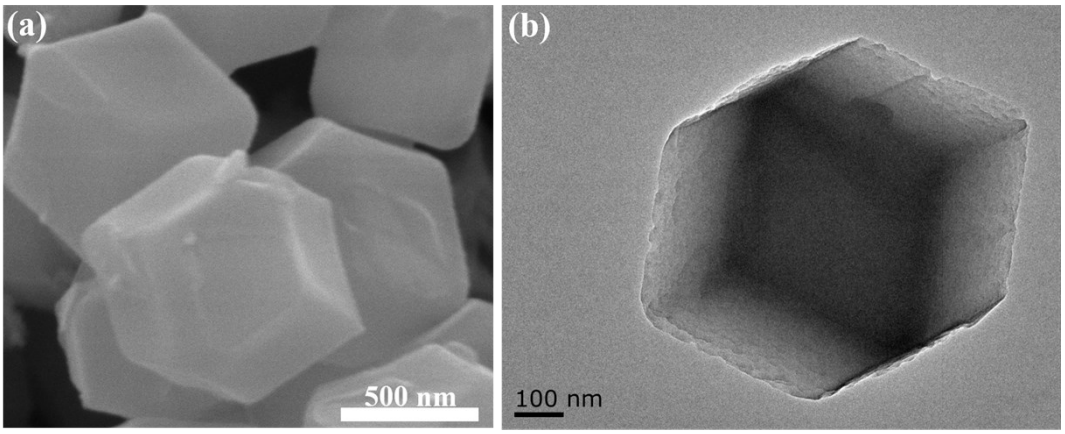

Fig. S7 (a) SEM and (b) TEM images of ZIF-67 treated by Ar-N_2 RF plasma.

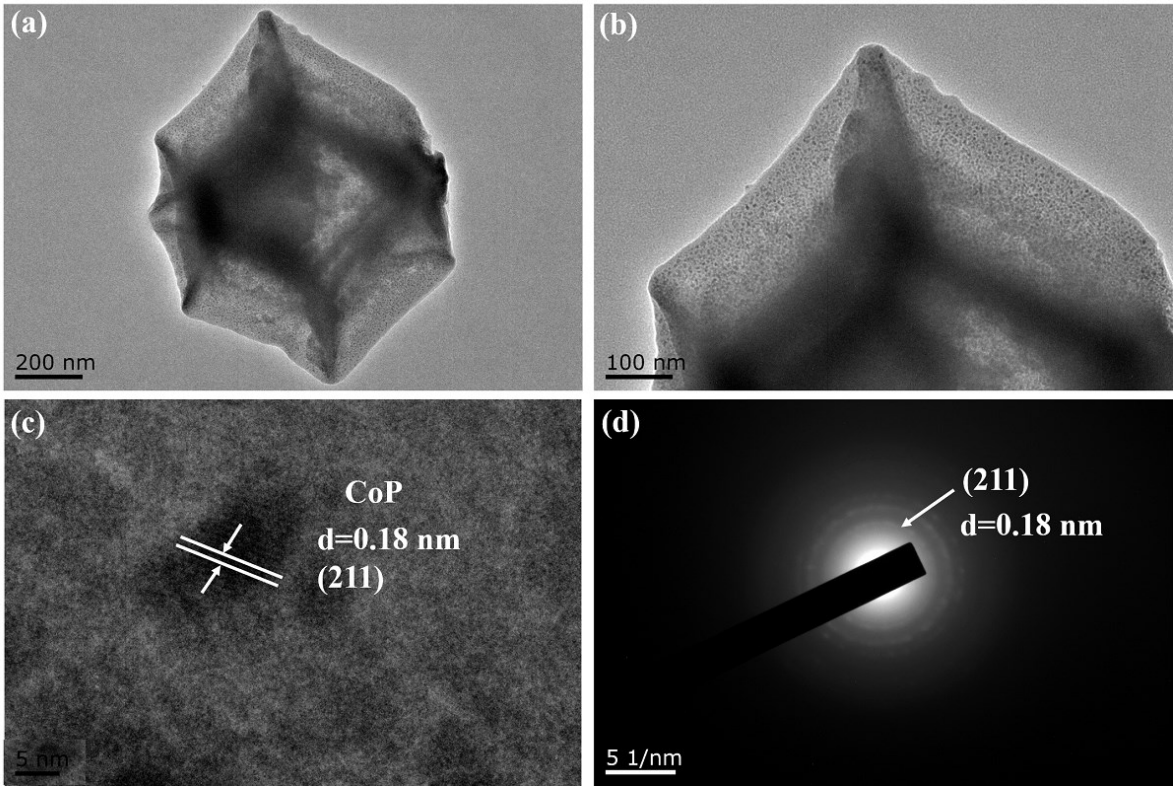

Fig. S8 (a) and (b) TEM, (c) HRTEM, and (d) the corresponding SAED pattern of the CoP samples.

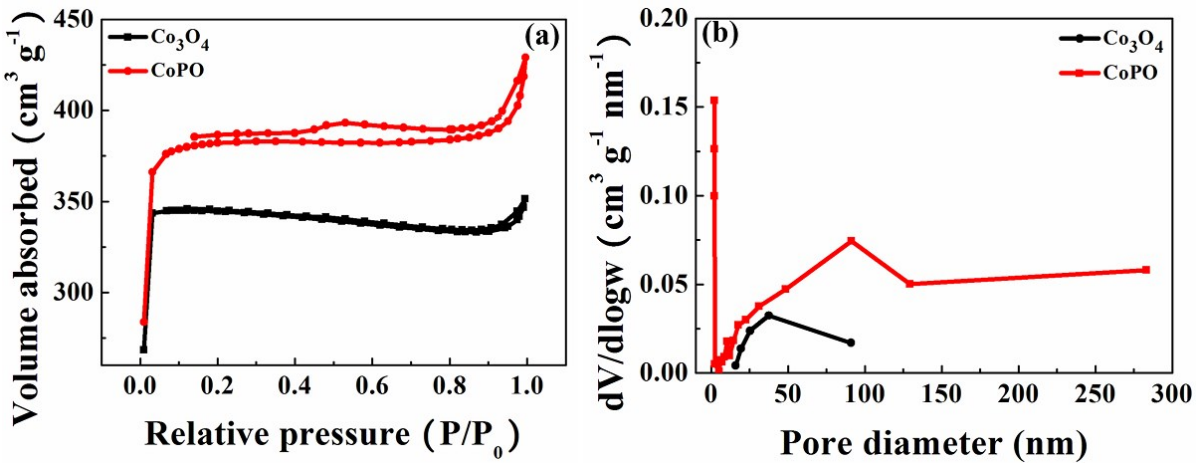

Fig. S9 The N_2 adsorption-desorption isotherm and pore size distribution of the CoPO and ZIF-67.

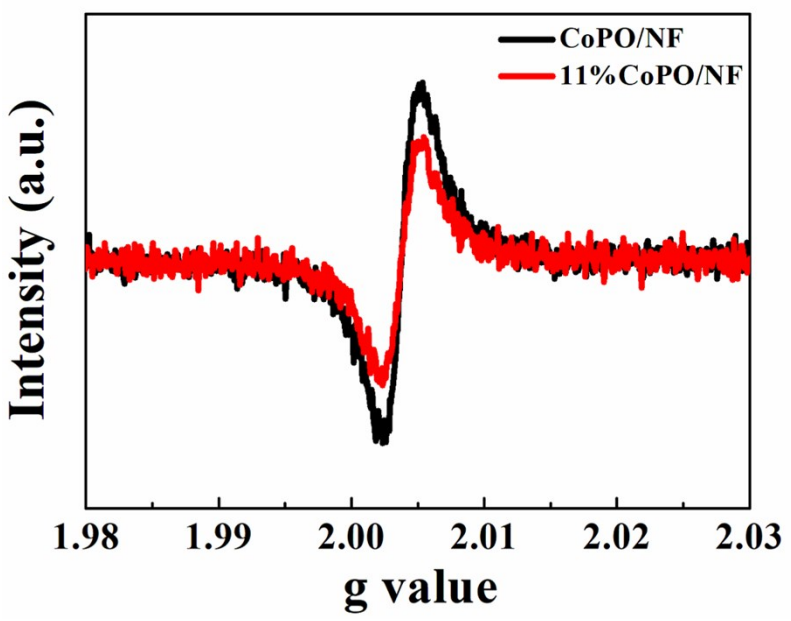

Fig. S10 The EPR analysis of the prepared samples.

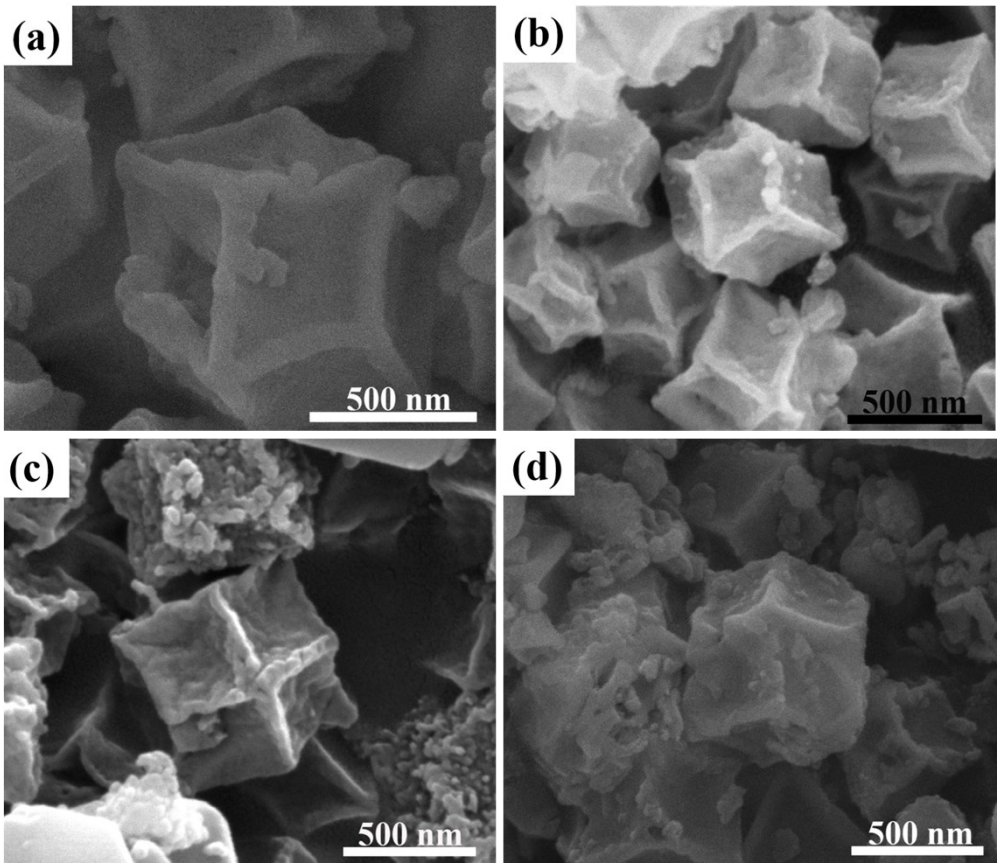

Fig. S11 SEM images of CoPO with different amounts of P.

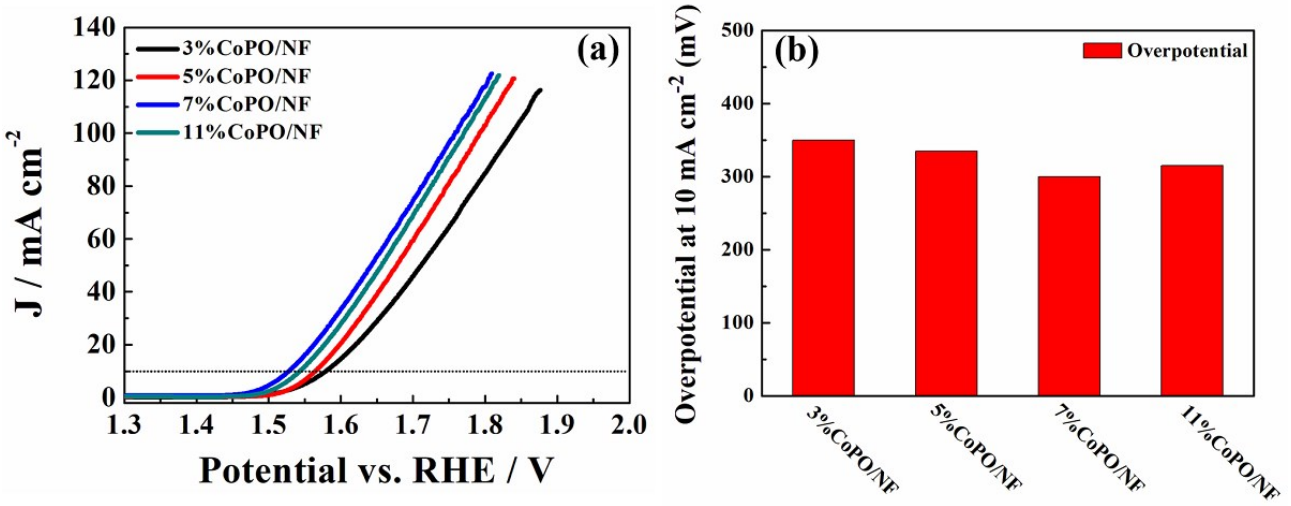

Fig. S12 LSV curves of (a) 3%CoPO/NF, 5%CoPO/NF, 7%CoPO/NF, 11%CoPO/NF for the OER, and (d) the corresponding overpotential at 10 mA cm^{-2}.

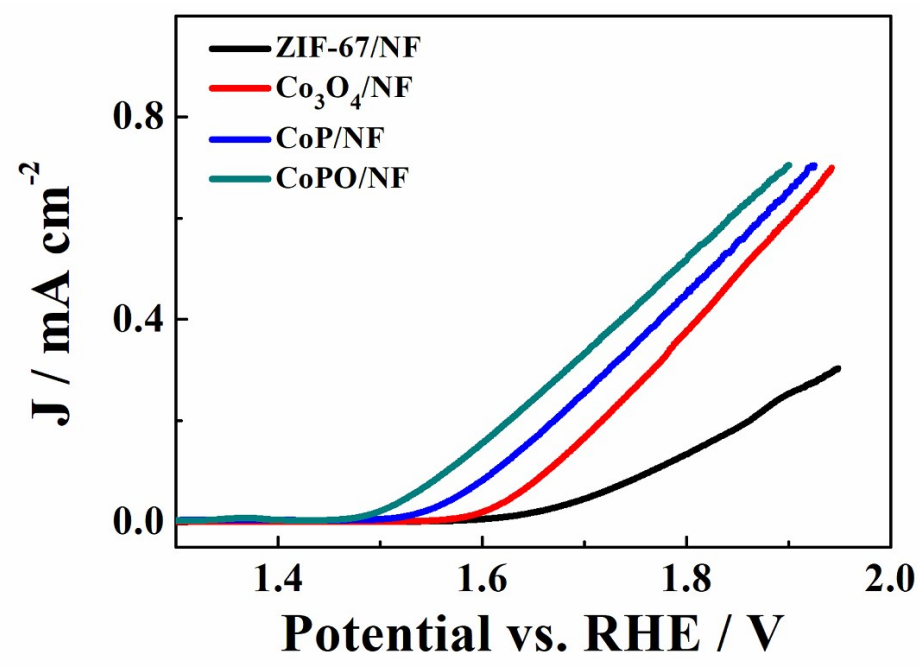

Fig. S13 The LSV curves normalized by ECSA for the prepared samples.

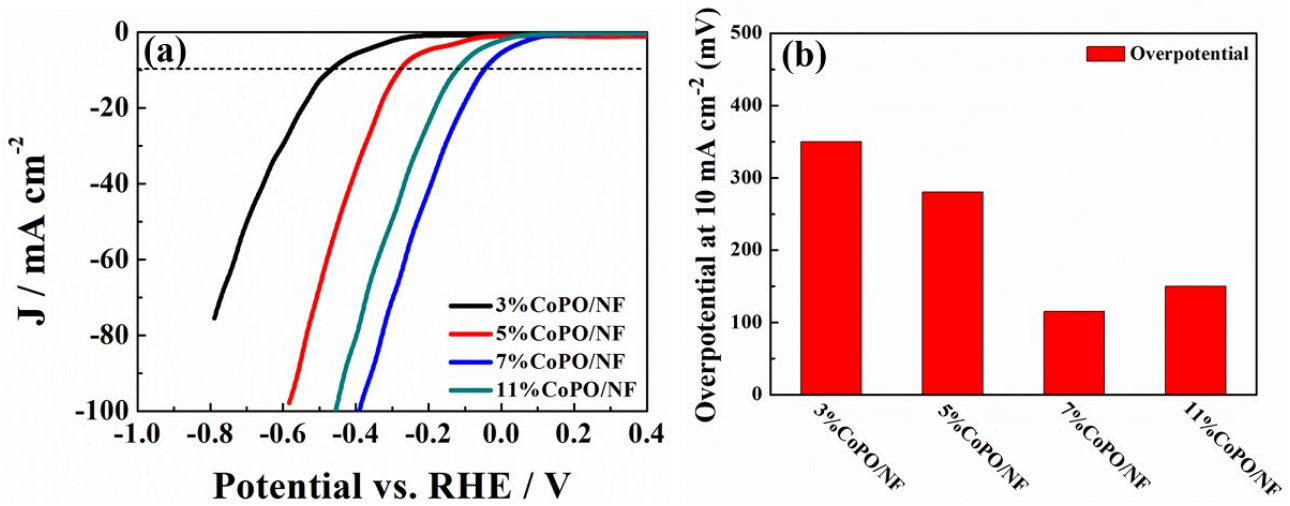

Fig. S14 LSV curves of (a) 3%CoPO/NF, 5%CoPO/NF, 7%CoPO/NF, 11%CoPO/NF for the HER, and (d) the corresponding overpotential at 10 mA cm^{-2}.

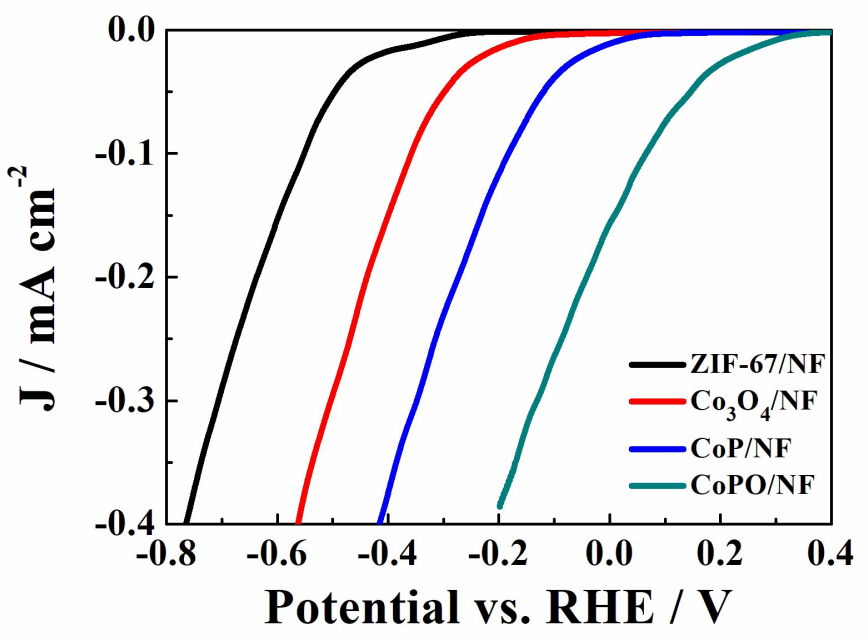

Fig. S15 The LSV curves normalized by ECSA for the prepared samples.

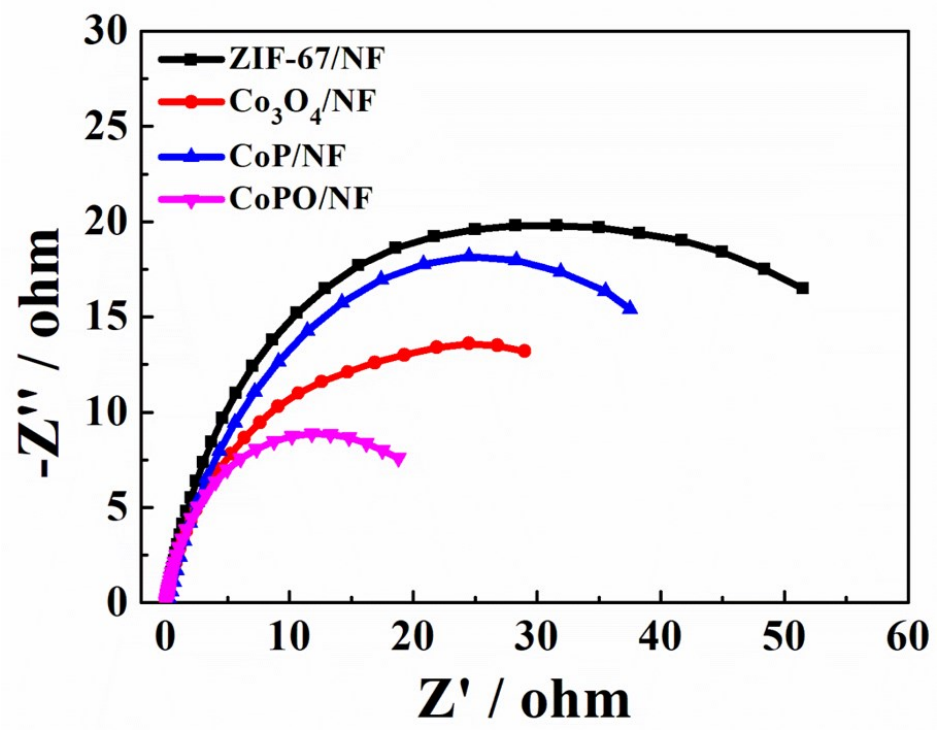

Fig. S16 The electrochemical impedance spectroscopy (EIS) spectra.

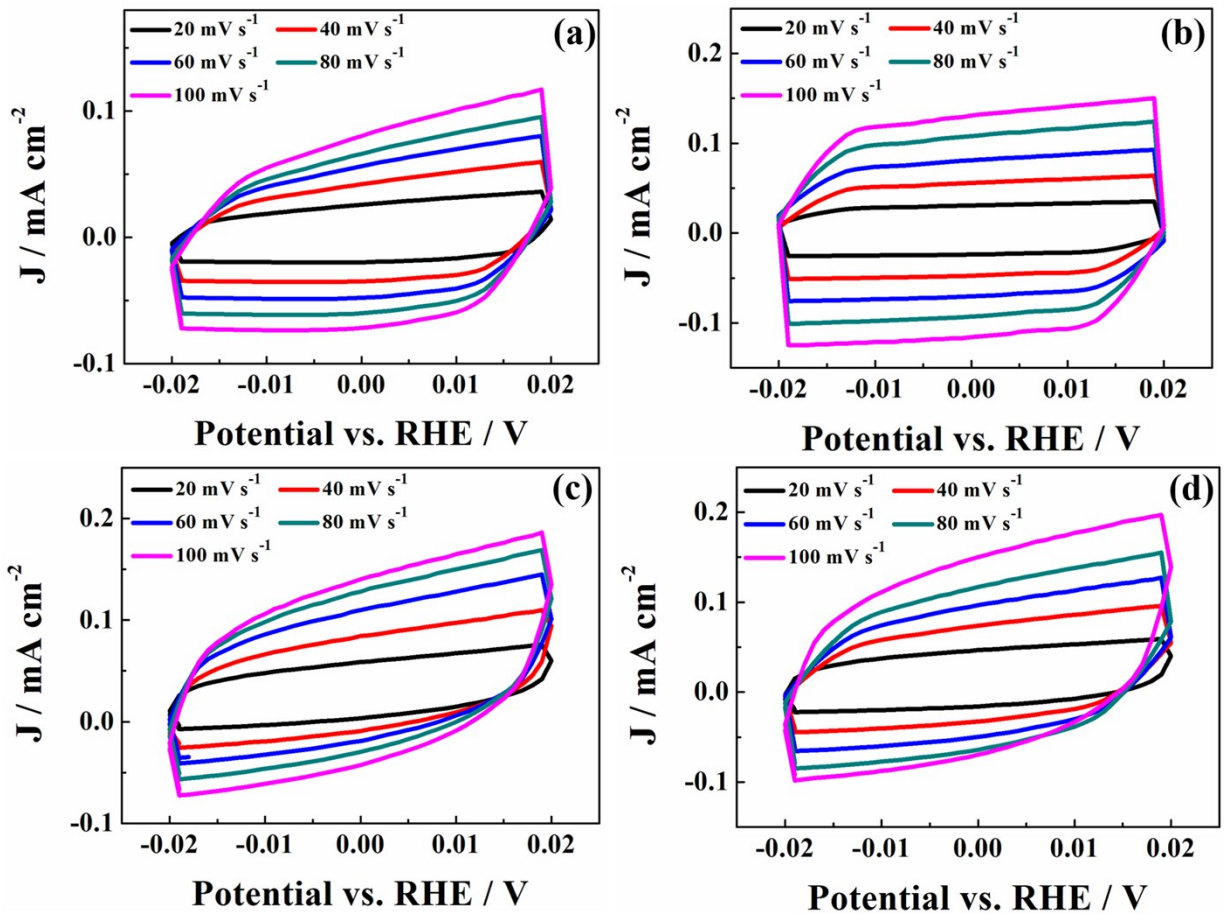

Fig. S17 CV curves at various scan rates in the potential range -0.02~0.02 V vs. RHE for (a) ZIF-67/NF, (b) Co_3O_4/NF, (c) CoP/NF and (d) CoPO/NF, respectively.

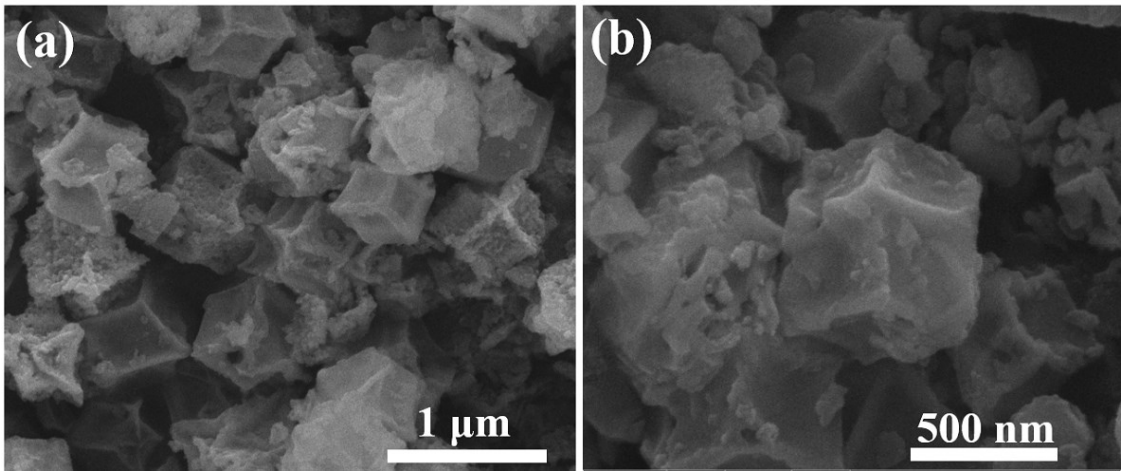

Fig. S18 The SEM image of the CoPO/NF after the OER and HER tests.

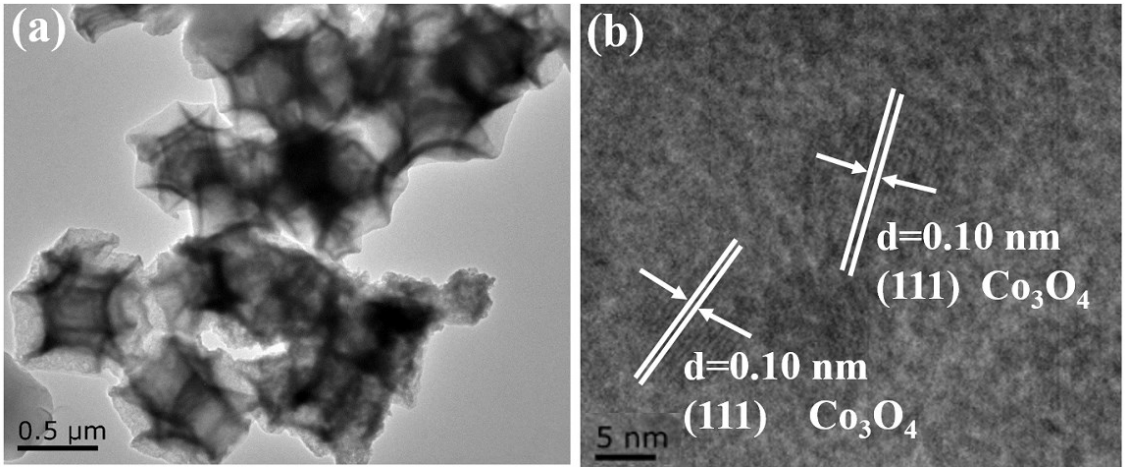

Fig. S19 The TEM image of the CoPO/NF after the OER and HER tests.

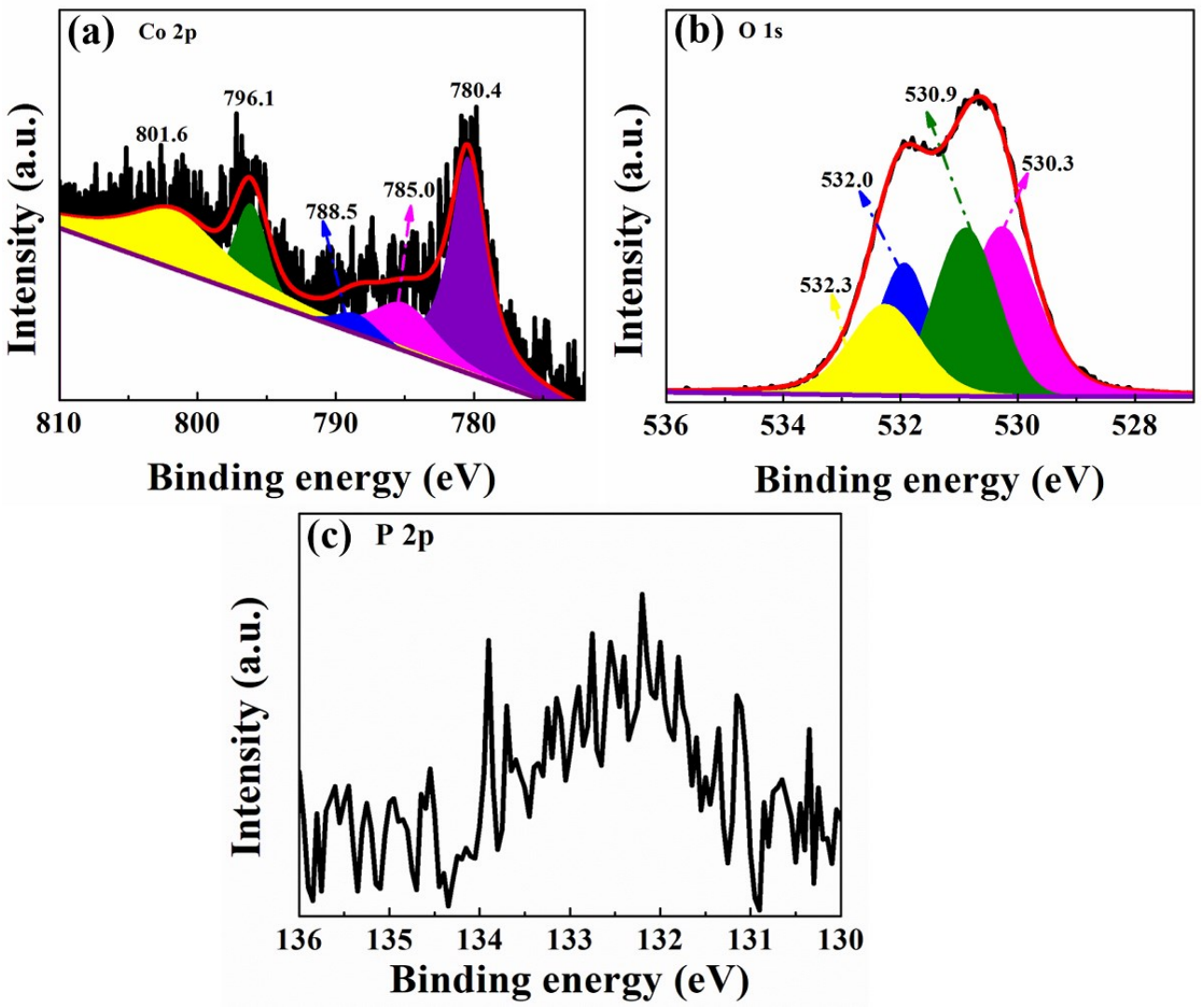

Fig. S20 XPS spectra of (a) Co 2p, (b) O 1s and (c) P 2p after electrochemistry test.

Table S1. The comparison of OER performance with state-of-the-art electrocatalysts.

Materials	**Supports**	**Electrolytes**	$\eta_{J=10\ mA\ cm^{-2}}$ **(mV)**	**References**
CoPO/NF	Ni foam	1 M KOH	275	This work
CoP/NCNHP	-	1 M KOH	310	1
CoP-2	-	1 M KOH	310	2
Co 2 P NCs	-	1 M KOH	280	3
Co-P film	-	1 M KOH	345	4
Co/CoP	-	1 M KOH	340	5
NiFe/$NiCo_2O_4$/NF	Ni Foam	1M KOH	340	6
NiCoP/C	-	1M KOH	330	7

Table S2 TOF of the as-prepared catalysts at overpotential of 200, 250 and 300 mV corresponding to OER.

Samples / TOF s^{-1}(mV)	ZIF-67/NF	Co_3O_4/NF	CoP/NF	CoPO/NF
η=200	7.410×10^{-7}	1.523×10^{-6}	4.244×10^{-6}	4.954×10^{-6}
250	8.809×10^{-7}	1.860×10^{-6}	4.617×10^{-6}	1.467×10^{-5}
300	1.471×10^{-7}	2.539×10^{-6}	1.581×10^{-5}	7.721×10^{-5}

Table S3. The comparison of HER performance with state-of-the-art electrocatalysts.

Materials	Supports	Electrolytes	$\eta_{J=10\ mA\ cm^{-2}}$ (mV)	References
CoPO/NF	Ni foam	1 M KOH	156	This work
Co1Mn1CH	-	1 M KOH	180	8
Co-NC/CNT	NF	1 M KOH	203	9
Co-Zn/PNC	NF	1 M KOH	180	10
Co/β-Mo_2C@N-CNTs	-	1 M KOH	170	11
$Co(OH)_2$@NCNT	NF	1 M KOH	170	12
O-Co_2P-3	-	1M KOH	160	13
CoP@C-NPs/GA-5		1M KOH	225	14
Co/CoP	-	1M KOH	253	15
CoPS@NPS-C	-	1M KOH	191	16

Table S4. TOF of the as-prepared catalysts at overpotential of 200, 250 and 300 mV corresponding to HER.

Samples / TOF s^{-1}(mV)	ZIF-67/NF	Co_3O_4/NF	CoP/NF	CoPO/NF
η=200	6.950×10^{-7}	1.498×10^{-6}	3.991×10^{-6}	3.995×10^{-6}
250	7.998×10^{-7}	1.796×10^{-6}	3.895×10^{-6}	1.051×10^{-5}
300	1.501×10^{-7}	2.241×10^{-6}	1.052×10^{-5}	6.154×10^{-5}

Table S5. Comparison of the full water-splitting performances of CoPO/NF with other state-of-the-art electrocatalysts in 1.0 M KOH.

Materials	**Cell voltages (V) at J = 10 mA cm^{-2}**	**References**
CoPO/NF	1.62	This work
$NiCo_2O_4$	1.65	17
Co_1Mn_1CH	1.68	8
Ni-P/CP	1.63	18
CoP/NCNHP	1.64	1
$NiCo_2N$/NF	1.70	19
BP/Co_2P	1.92	20
$(Co\text{-}NMC)_1$/NC/GCE	1.78	21

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