

Supporting Information

Multifunctional Electrocatalysis on Porous N-Doped NiCo₂O₄@C Nanonetwork

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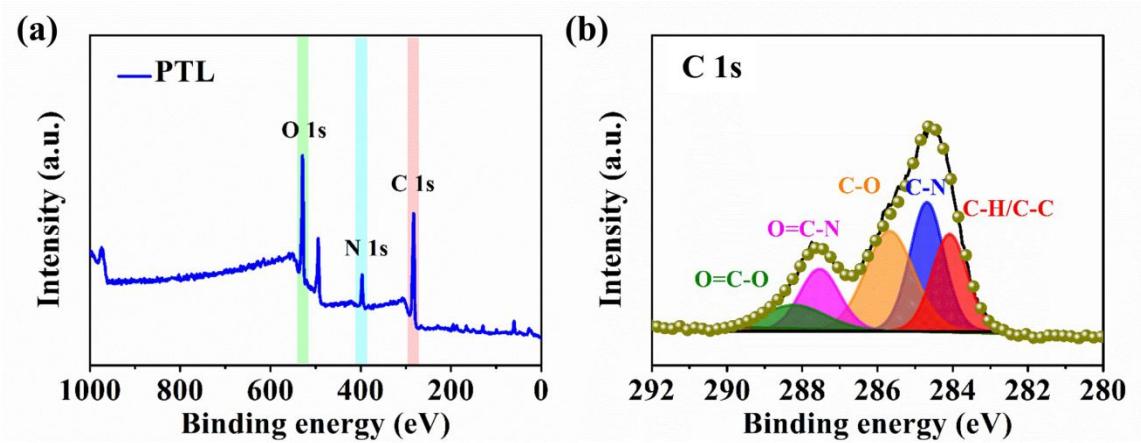


Figure S1. (a) XPS survey spectra of PTL; (b) corresponding high-resolution XPS spectra of the C 1s.^{S1, S2}

Note: the high-resolution XPS spectra in Figure S1b can be fitted into five peaks at 284.1, 284.7, 285.6, 287.5 and 288.3 eV corresponding to C-H/C-C, C-N, C-O, O=C-N and O=C-O, respectively, which is also similar to previous reports. ^{S1,S2}

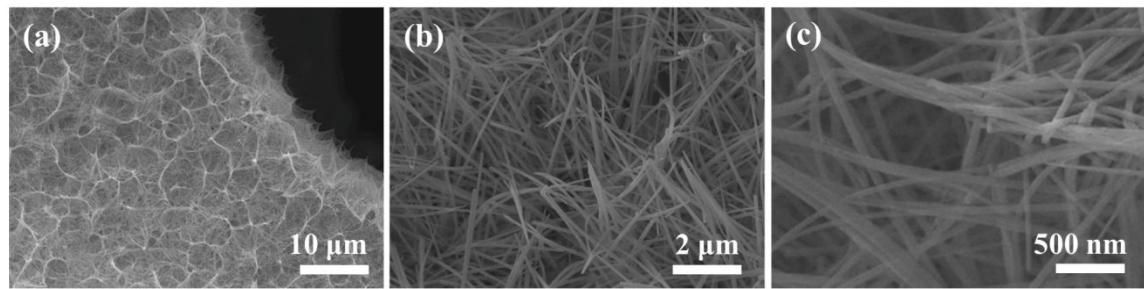


Figure S2. (a-c) FESEM images of $\text{NiCo}_2\text{O}_4@\text{PTL}@\text{NF}$ precursor.

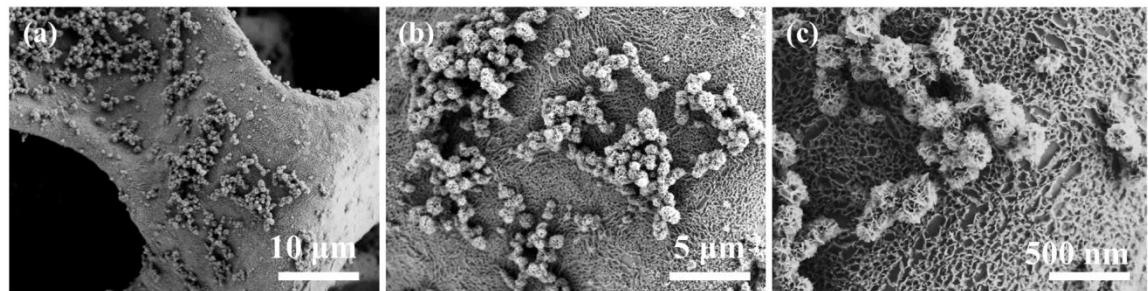


Figure S3. (a-c) FESEM images of NiCo_2O_4 structures directly grown on nickel foam without PTL nanofilm ($\text{NiCo}_2\text{O}_4@\text{NF}$).

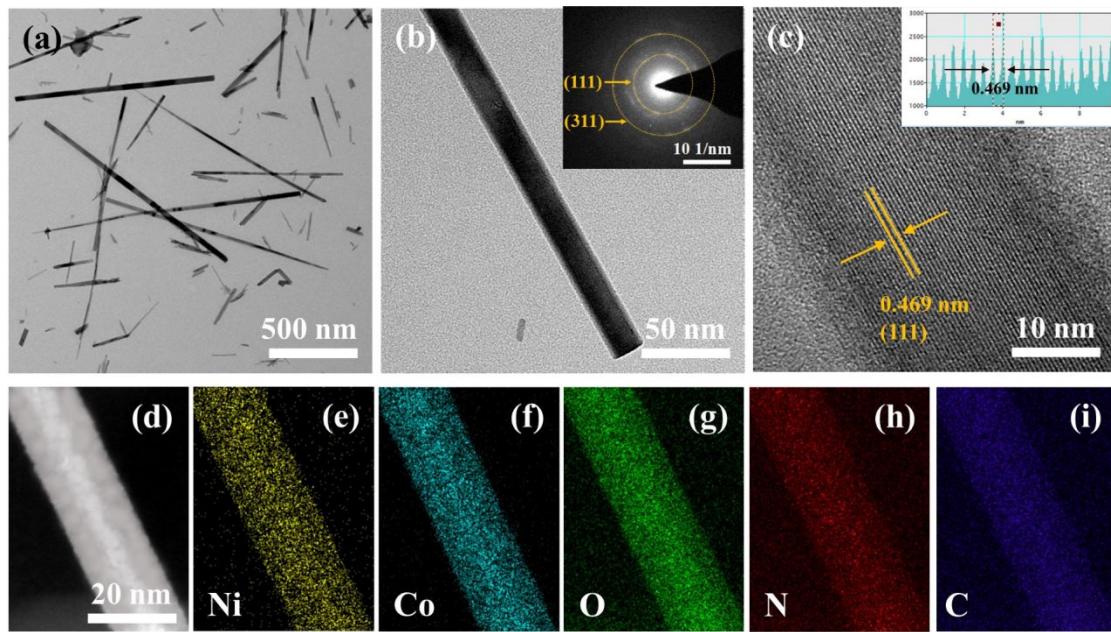


Figure S4. $\text{NiCo}_2\text{O}_4@\text{PTL}$ precursor: (a) TEM image; (b) magnified TEM image; (c) HRTEM image, the inset in Figure S4b showing the corresponding SAED and the lattice spacing corresponding to the selected areas in panel (Figure S4c); (d) HAADF-STEM image and (e-i) EDS elemental mapping images of $\text{NiCo}_2\text{O}_4@\text{PTL}$ precursor.

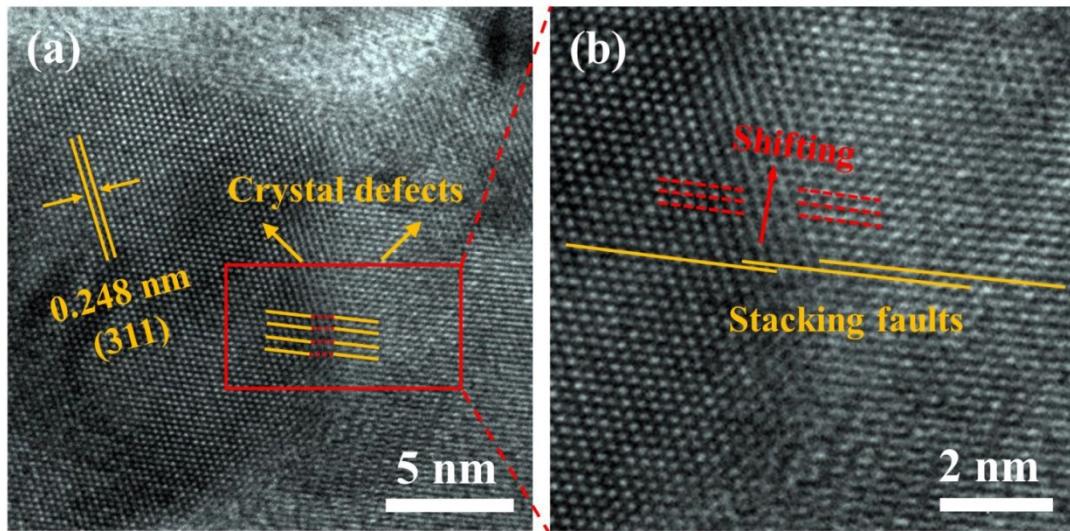


Figure S5. N-NiCo₂O₄@C: (a) HRTEM image (b) magnified HRTEM image recorded from red square in (a).

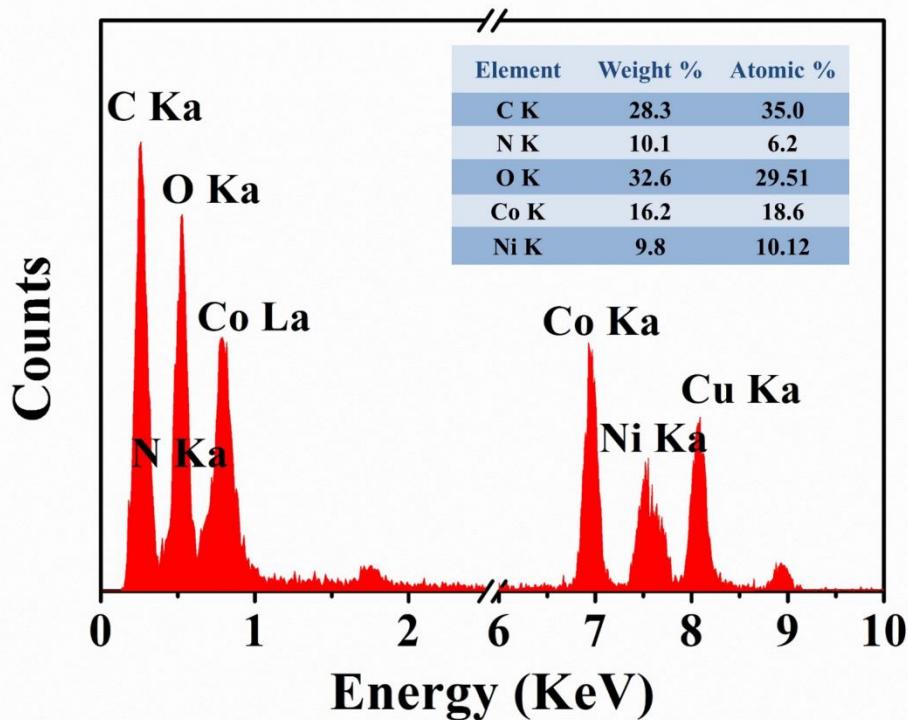


Figure S6. EDS results of N-NiCo₂O₄@C. The EDS elemental results confirm that the corresponding Co/Ni ratio of synthesized N-NiCo₂O₄@C are close to the chemical formula for spinal NiCo₂O₄.

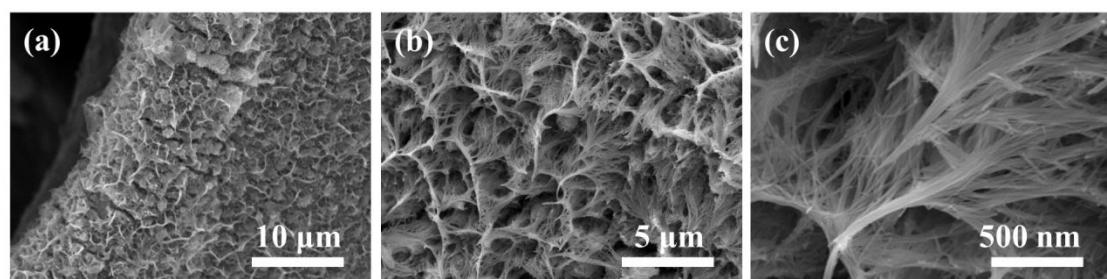


Figure S7. (a-c) FESEM images of N-NiCo₂O₄@C@NF after 15 min high-powered ultrasonication.

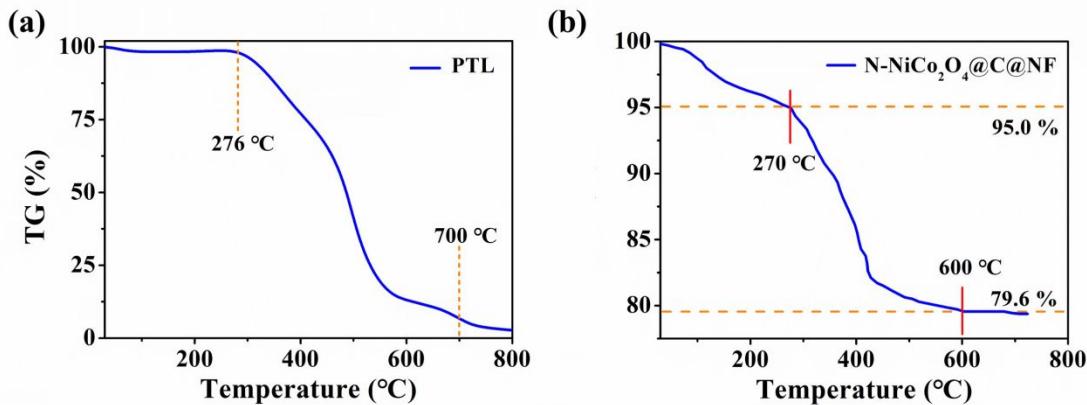


Figure S8. TGA curves of (a) PTL and (b) N-NiCo₂O₄@C@NF under air atmosphere at a heating rate of 10 °C min⁻¹.

The carbon content in the N-NiCo₂O₄@C composites can be evaluated from the TGA results as following (Noted that the products were shaved from NF before TGA testing):

Firstly, the TG curve of PTL shows a sharp mass decrease from 276 to 700°C, corresponding to the decomposition of PTL, whereas the N-NiCo₂O₄@C exhibits a slow mass loss before 270 °C, caused by the removal of water and hydroxy groups probably adsorbed on the surface of the composite. The continuous weight loss from 270 °C to 600 °C is ascribed to the burning of PTL-carbon since the temperature of thermal decomposition. As for the N-NiCo₂O₄@C, a weight loss of 15.4 % was found after heating 232 °C, which could be ascribed to the combustion of PTL.

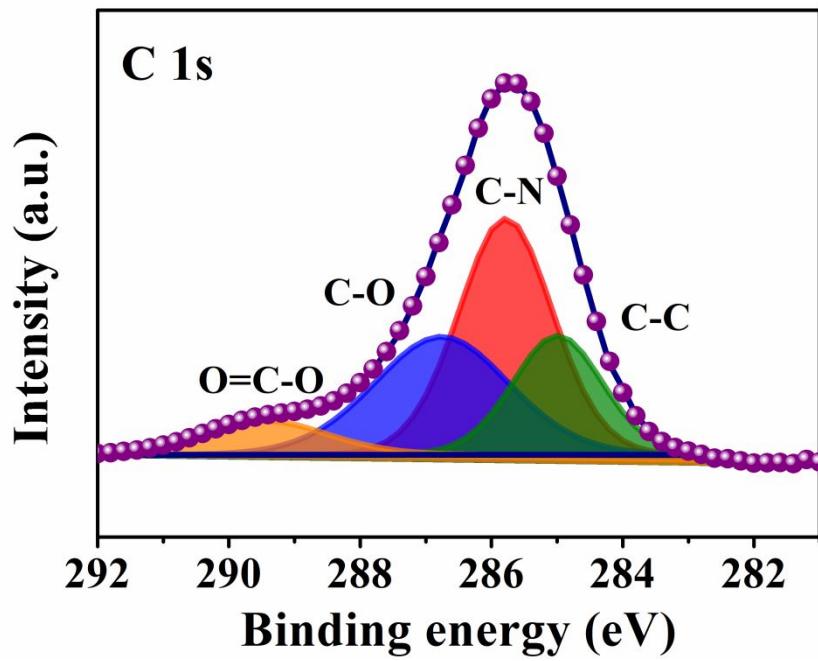


Figure S9. High-resolution XPS spectra of C 1s in N-NiCo₂O₄@C.

Element	Position (eV)	Atomic %
C 1s	284.6	7.35
N 1s	400.2	4.8
O 1s	529.8	45.22
Co 2p	779.6	16.68
Ni 2p	854.3	25.95

Figure S10. Quantitative analysis of the N-NiCo₂O₄@C based on the high-resolution XPS spectra.

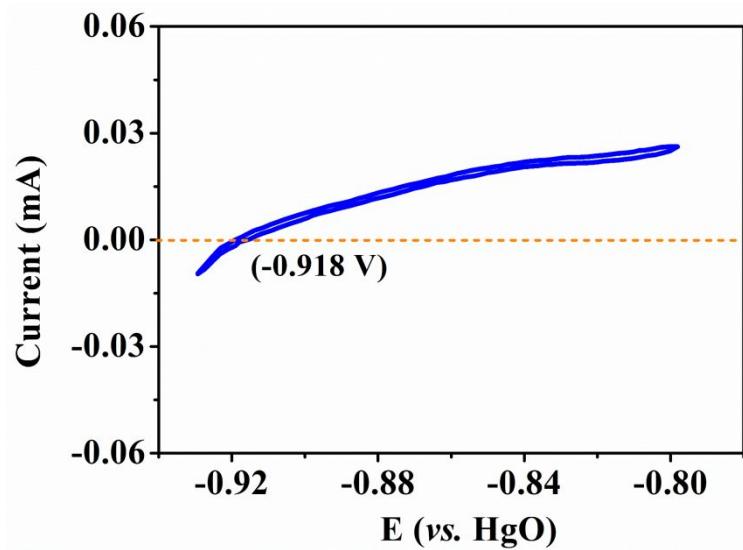


Figure S11. The CV curve at a scan rates of 1 mV s^{-1} in hydrogen-saturated 1.0 M KOH.

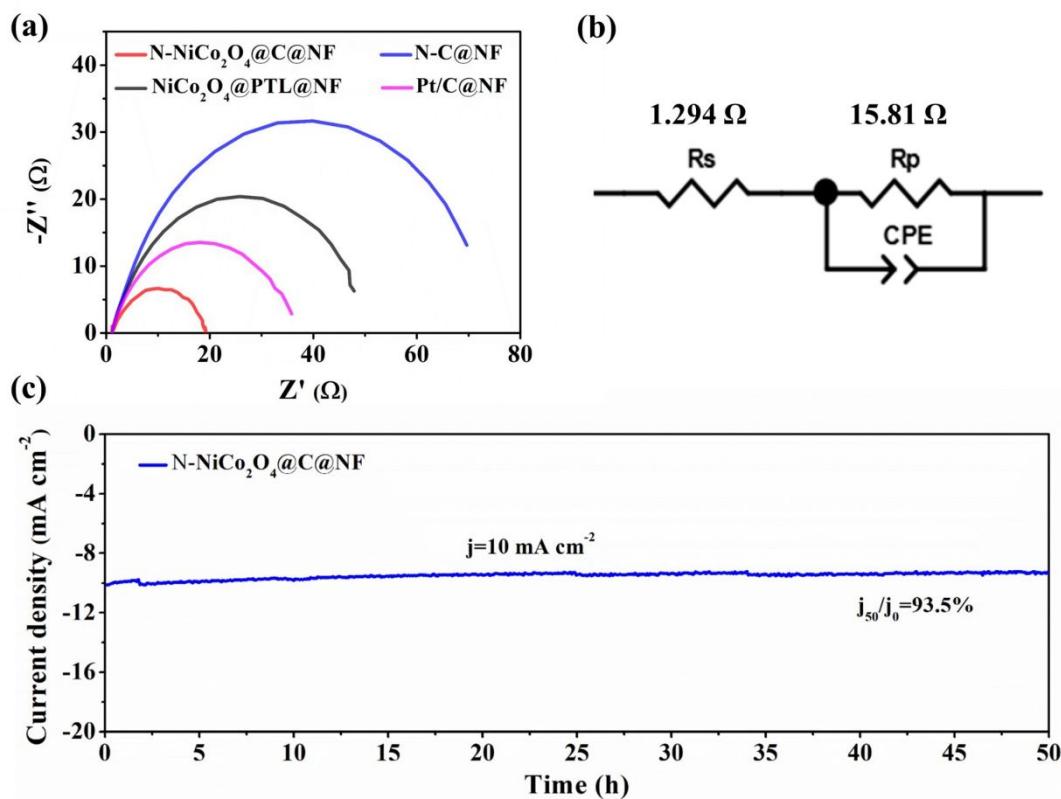


Figure S12. (a) EIS spectra of N- NiCo_2O_4 @C@NF, NiCo_2O_4 @PTL@NF precursor, N-C@NF and Pt/C@NF for HER; (b) corresponding equivalent circuit diagram; (c) stability test of N- NiCo_2O_4 @C@NF under high current density for 50 h at a constant potential of -0.046 V vs. RHE.

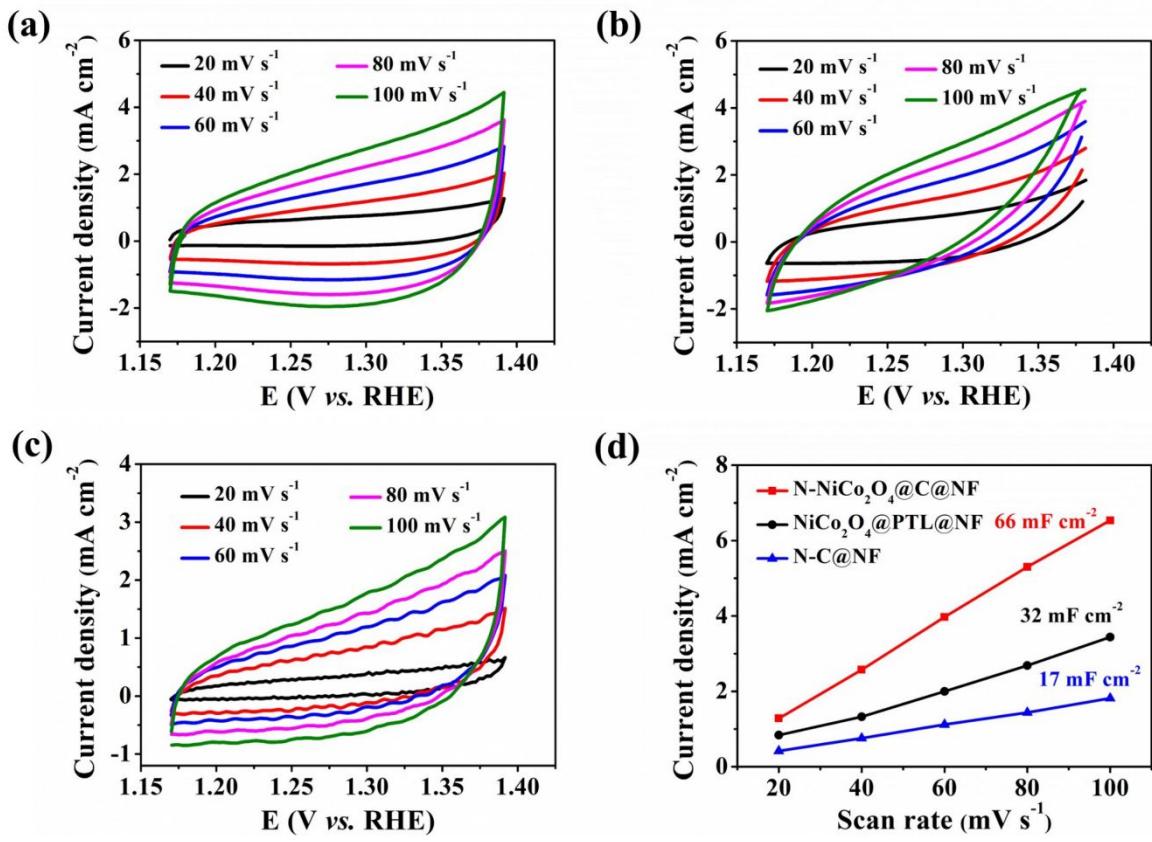


Figure S13. CV curves of (a) N-NiCo₂O₄@C@NF, (b) NiCo₂O₄@PTL@NF precursor and (c) N-C@NF at different scan rates, and (d) corresponding plots of the capacitive currents as a function of scan rate.

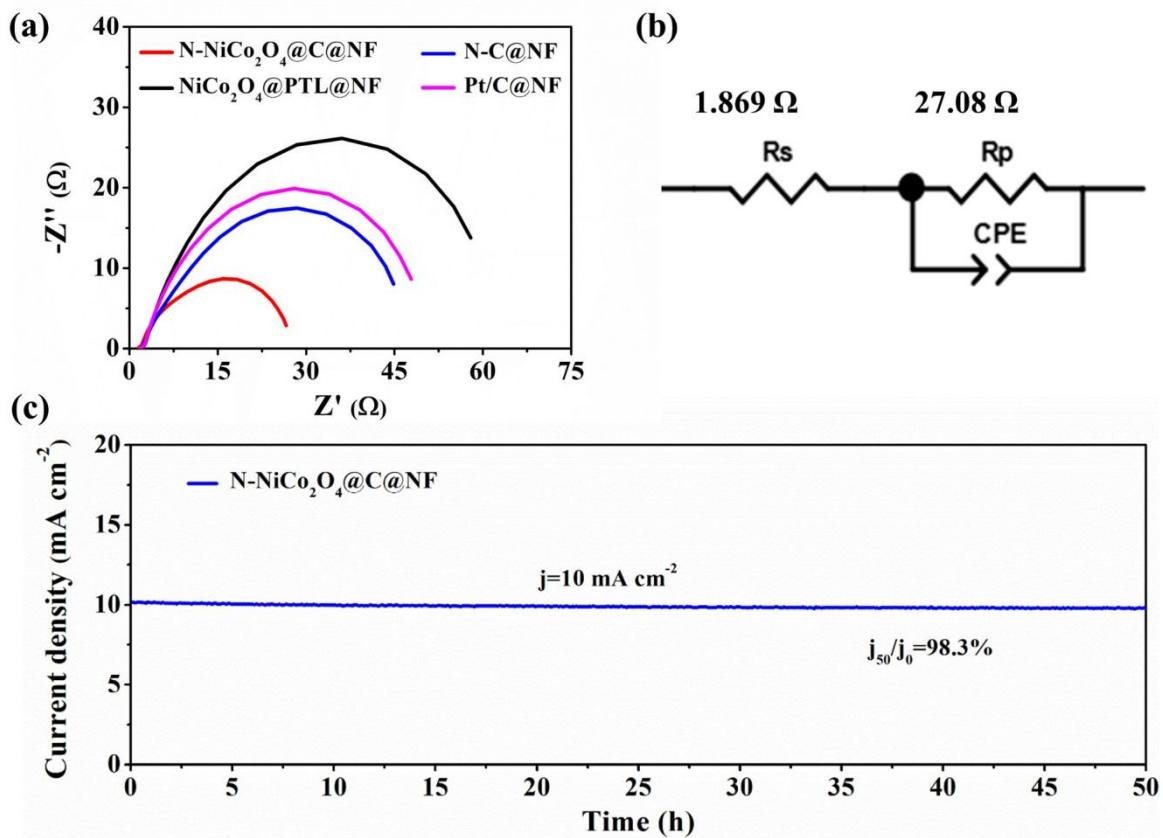


Figure S14. (a) EIS spectra of N- NiCo_2O_4 @C@NF, NiCo_2O_4 @PTL@NF precursor, N-C@NF and Pt/C@NF for OER; (b) corresponding equivalent circuit diagram; (c) stability test of N- NiCo_2O_4 @C@NF at a constant potential of -1.46 V vs. RHE for 50 h.

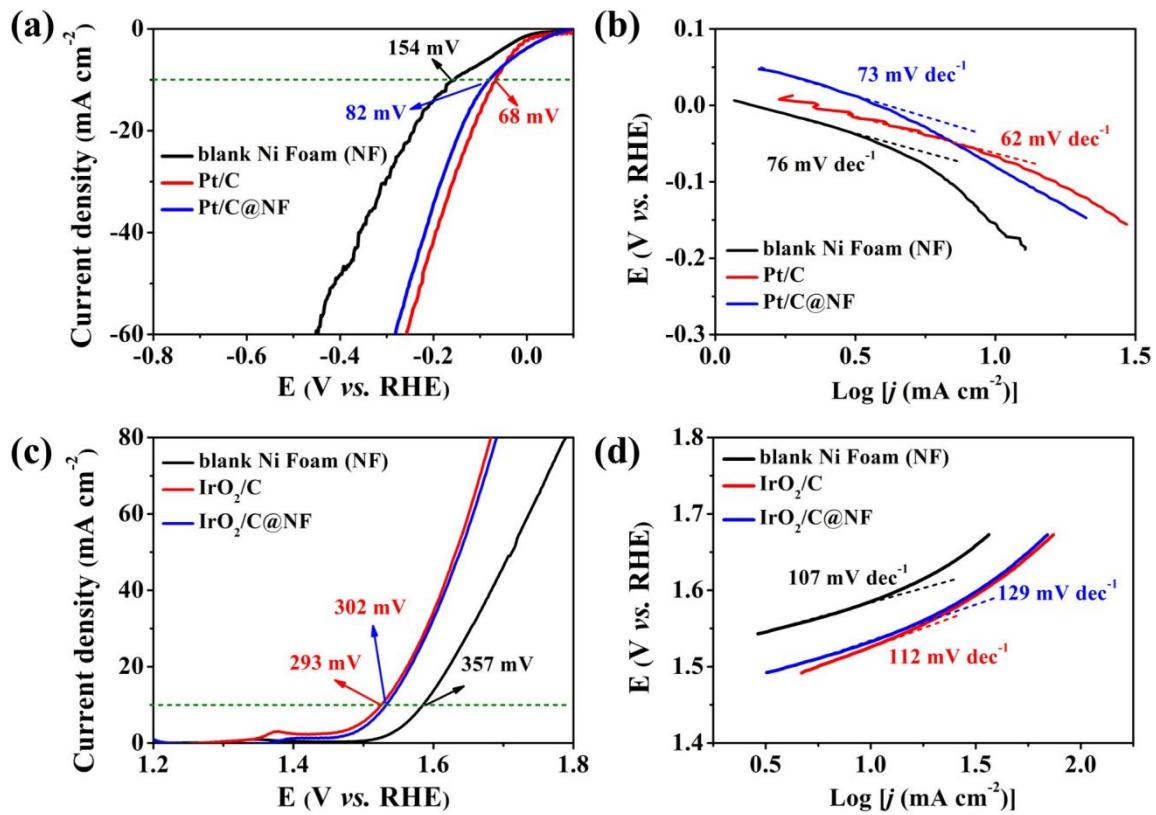


Figure S15. HER and OER performance of blank Ni foam, pure Pt/C and Pt/C@NF or pure IrO₂/C and IrO₂/C@NF. (a) HER LSV curves, (b) HER Tafel plots, and (c) OER LSV curves, (d) OER Tafel plots.

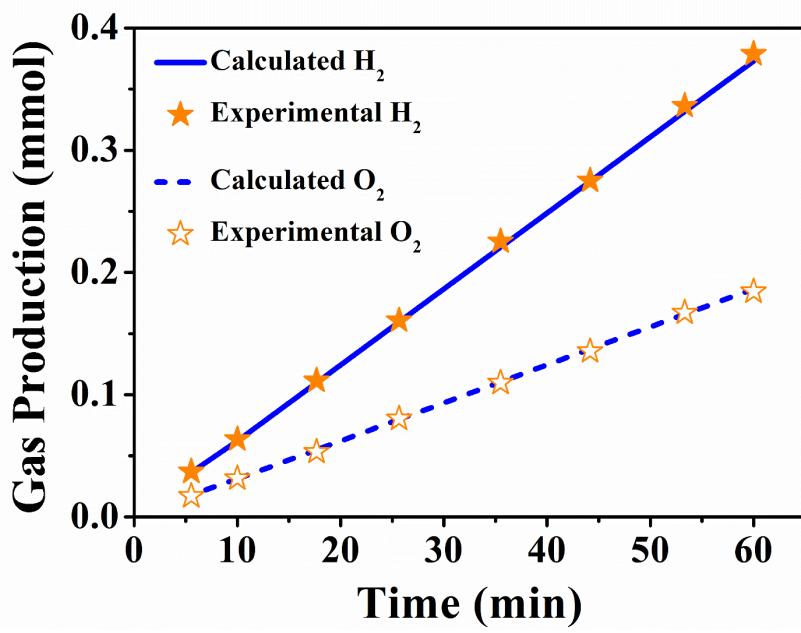


Figure S16. The measured and theoretical yields of O₂ and H₂ over time during electrolysis of N-NiCo₂O₄@C@NF at the current density of 10 mA cm⁻².

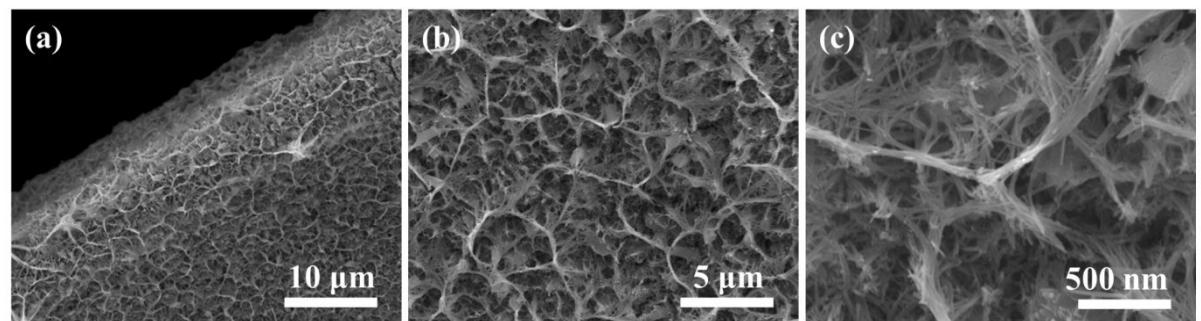


Figure S17. (a-c) FESEM images of the N-NiCo₂O₄@C@NF after cycling test for water splitting.

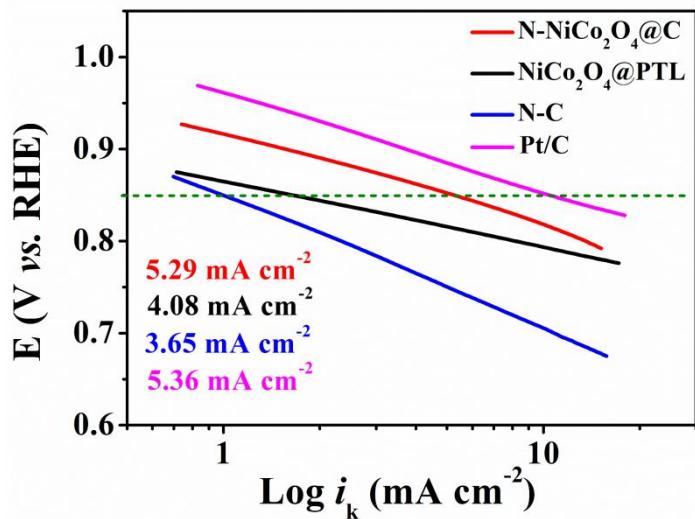


Figure S18. The ORR Tafel plots of N-NiCo₂O₄@C, NiCo₂O₄@PTL, N-C, and Pt/C catalysts in 0.1 M KOH.

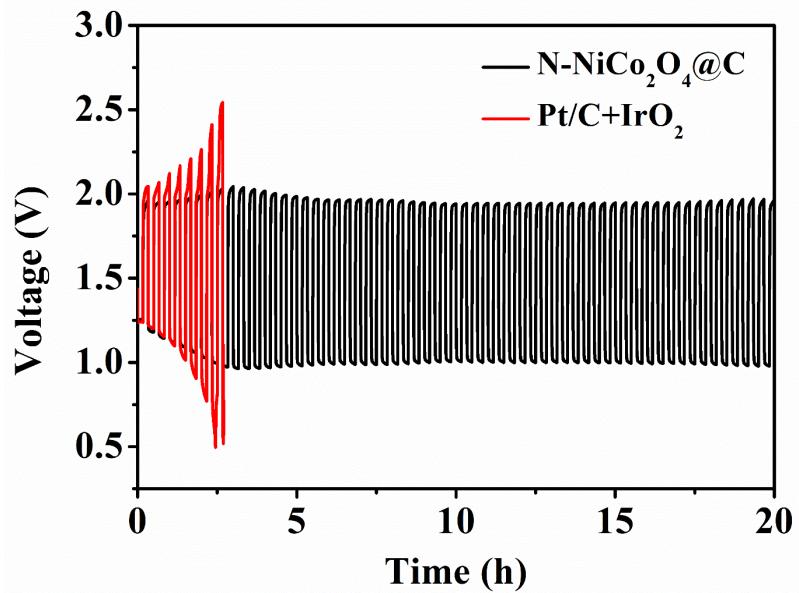


Figure S19. Galvanostatic charge and discharge test of Zn-air battery based on N-NiCo₂O₄@C and Pt/C+IrO₂ at current density of 10 mA cm⁻².

Table S1. Comparison of the electrocatalytic activity of N-NiCo₂O₄@C@NF and the reported catalysts for HER in 1.0 M KOH.

Catalyst	Current density (j)	overpotential	Ref.
N-NiCo₂O₄@C@NF	10 mA cm⁻²	42 mV	This work
NiCo ₂ O ₄ hollow microcuboids	10 mA cm ⁻²	110 mV	(S3)
NiCo/NiCoO _x /NF	10 mA cm ⁻²	155 mV	(S4)
Fe _{11%} -NiO/NF	10 mA cm ⁻²	88 mV	(S5)
Ni _x Co _{3-x} S ₄ /Ni ₃ S ₂ /NF	10 mA cm ⁻²	136 mV	(S6)
Co ₄ Ni ₁ P NTS	10 mA cm ⁻²	192 mV	(S7)
NiCo ₂ O ₄ NA/CC	10 mA cm ⁻²	175 mV	(S8)
Ni _{0.82} Co _{0.18} O@C /NF	10 mA cm ⁻²	194 mV	(S9)
NiCoFe LTHs/CFC	10 mA cm ⁻²	200 mV	(S10)
NiCo-PBA/NF	10 mA cm ⁻²	62 mV	(S11)
NiCo ₂ S ₄	10 mA cm ⁻²	80 mV	(S12)
3D Se-(NiCo)S _x /(OH) _x nanosheets	10 mA cm ⁻²	103 mV	(S13)
NiCo ₂ O ₄ @CoMoO ₄ /NF	10 mA cm ⁻²	121 mV	(S14)
NiCoP/CC	10 mA cm ⁻²	62 mV	(S15)
Ni _{0.5} Co _{0.5} nanowire	10 mA cm ⁻²	36 mV	(S16)

Table S2. Comparison of the electrocatalytic activity of N-NiCo₂O₄@C@NF and the reported catalysts for OER in 1.0 M KOH.

Catalyst	Current density (j)	overpotential	Ref.
N-NiCo₂O₄@C@NF	10 mA cm⁻²	242 mV	This work
NiCo ₂ O ₄ hollow microcuboids	10 mA cm ⁻²	270 mV	(S3)
Co ₄ Ni ₁ P NTS	10 mA cm ⁻²	245 mV	(S7)
Ni _{0.82} Co _{0.18} O@C /NF	10 mA cm ⁻²	330 mV	(S9)
NiCoFe LTHs/CFC	10 mA cm ⁻²	239 mV	(S10)
NiCo-PBA/NF	10 mA cm ⁻²	200 mV	(S11)
NiCo ₂ S ₄	10 mA cm ⁻²	243 mV	(S12)
3D Se-(NiCo)Sx/(OH)x nanosheets	10 mA cm ⁻²	155 mV	(S13)
MnFe ₂ O ₄ /NiCo ₂ O ₄	10 mA cm ⁻²	330 mV	(S17)
NiCo/NLG-270	10 mA cm ⁻²	340 mV	(S18)
NiCo ₂ S ₄ /N-CNT	10 mA cm ⁻²	370 mV	(S19)
NiCo/PFC aerogels	10 mA cm ⁻²	400 mV	(S20)
NiCo ₂ O ₄ /Graphene	10 mA cm ⁻²	440 mV	(S21)
NiCoP/C nanoboxes	10 mA cm ⁻²	330 mV	(S22)
Ni _x Co _{3-x} O ₄	10 mA cm ⁻²	337 mV	(S23)

Table S3. The two-electrode system for overall water electrolysis in 1.0 M KOH.

Catalyst	Current density (j)	Potential	Ref.
N-NiCo₂O₄@C@NF	10 mA cm⁻²	1.43 V	This work
NiCo ₂ O ₄ hollow microcuboids	10 mA cm ⁻²	1.65 V	(S3)
Ni _x Co _{3-x} S ₄ /Ni ₃ S ₂ /NF	10 mA cm ⁻²	1.53 V	(S6)
Co ₄ Ni ₁ P nanotubes	10 mA cm ⁻²	1.59 V	(S7)
NiCo ₂ O ₄ NA/CC	10 mA cm ⁻²	1.68 V	(S8)
Ni _{0.82} Co _{0.18} O@C /NF	10 mA cm ⁻²	1.42 V	(S9)
NiCoFe LTHs/CFC	10 mA cm ⁻²	1.55 V	(S10)
NiCo-PBA/NF	10 mA cm ⁻²	1.49 V	(S11)
NiCo ₂ S ₄	10 mA cm ⁻²	1.58 V	(S12)
3D Se-(NiCo)S _x /(OH) _x nanosheets	10 mA cm ⁻²	1.60 V	(S13)
NiCo ₂ O ₄ @CoMoO ₄ /NF	10 mA cm ⁻²	1.55 V	(S14)
NiCo ₂ P _x	10 mA cm ⁻²	1.61 V	(S24)
NiCo ₂ O ₄ /Ni ₂ P/N	10 mA cm ⁻²	1.59 V	(S25)
NiCoP/NF	10 mA cm ⁻²	1.58 V	(S26)
NiFe/NiCo ₂ O ₄ /Ni	10 mA cm ⁻²	1.67 V	(S27)

Table S4. Summary of reported half-wave potential ($E_{1/2}$) and diffusion-limited current density (J_L) values at 1600 rpm for NiCo-based Electrocatalysts. Noted all data are obtained in 0.1 M in an O₂ saturated 0.1 M KOH solution at a rotation rate of 1600 rpm.

Catalyst	E_{onset} (vs. RHE)	$E_{1/2}$ (vs. RHE)	J_L (mA cm ⁻²)	Ref.
N-NiCo₂O₄@C@NF	0.91 V	0.81 V	5.2	This work
NiCo ₂ S ₄ /N-CNT	0.93 V	0.80 V	3.4	(S19)
NiCo ₂ S ₄ .CNT	0.84 V	0.69 V	2.7	(S19)
NiCo ₂ S ₄	0.77 V	0.64 V	2.2	(S19)
NiCo/PFC aerogels	0.92 V	0.79 V	5.8	(S20)
NiCo ₂ O ₄ /Graphene	0.87 V	0.69 V	3.4	(S21)
NiCo ₂ O ₄ NWs	0.80 V	0.76 V	6.0	(S28)
macroporous NiCo ₂ O ₄	0.83 V	0.78 V	1.6	(S29)
NiCo ₂ O ₄ /rGO	0.91 V	0.78 V	1.6	(S30)
NiCo ₂ O ₄ /graphene foam	0.9 V	0.86 V	6.2	(S31)
1D-spinel NiCo ₂ O ₄	0.84 V	0.78 V	6.0	(S32)
NiCo@N-C	0.96 V	0.81 V	6.5	(S33)
NiCo ₂ S ₄ @N/S-rGO	0.88 V	0.80 V	4.3	(S34)
NiCo ₂ O ₄ /HCS	0.90 V	0.78 V	5.8	(S35)
NiCo ₂ O ₄ /Co,N-CNTs NCs	0.92 V	0.86 V	5.2	(S36)

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