

Electronic Supplementary Information

Experimental section

Materials: Ni foam was purchased from Hangxu filter flagship store. HMT, $\text{Co}(\text{NO}_3)_2 \cdot 6\text{H}_2\text{O}$ and SDS were purchased from Beijing Chemical Works. Nafion (5 wt%) and $\text{RuCl}_3 \cdot 3\text{H}_2\text{O}$ were bought from Sigma-Aldrich Chemical Reagent Co., Ltd. The water used throughout all experiments was purified through a Millipore system. All the reagents and chemicals were used as received without further purification.

Synthesis of DS-Co(OH)₂/NF: DS-Co(OH)₂/NF was prepared using a traditional solvothermal method. A piece of NF (2 cm × 4 cm) was first cleaned with 3 M HCL, ethanol and deionized water by sonication sequentially to obtain a clean surface before use. First, $\text{Co}(\text{NO}_3)_2 \cdot 6\text{H}_2\text{O}$ (1.164g), SDS (1.153 g), and HMT (1.680 g) were dissolved in 40 mL distilled water. After gently stirring for 20 min at room temperature, the clear solution was transferred to a 50 mL Teflon-lined stainless-steel autoclave and then a piece of Ni foam was immersed into the solution. The autoclave was sealed and maintained at 95 °C for 48 h in an electric oven. After the autoclave cooled down at room temperature, the resulting green production was taken out and washed with distilled water and ethanol several times, followed by drying at 60 °C for 2 h to obtain DS-Co(OH)₂/NF.

Synthesis of P-DS-Co(OH)₂/NF: P-DS-Co(OH)₂/NF was prepared as follows. The DS-Co(OH)₂/NF was into a quartz boat and pyrolyzed at 300 °C for 2 h with a rising rate of 2 °C min⁻¹ to be transformed into P-DS-Co(OH)₂/NF.

Synthesis of Co(OH)₂/NF: $\text{Co}(\text{NO}_3)_2 \cdot 6\text{H}_2\text{O}$ (1.45 g) and HMT (1.4 g) were dissolved in 36 mL of water under vigorous stirring for 30 min. Then, the solution was transferred to a 50 mL Teflon-lined stainless-steel autoclave in which a piece of NF with a clean surface was immersed into the solution. The autoclave was sealed and maintained at 100 °C for 10 h in an electric oven to obtain the Co(OH)₂/NF product.

Synthesis of Co₄O₃/NF: The Co₄O₃/NF was obtained after pyrolyzed Co(OH)₂/NF at 300 °C for 2 h with a rising rate of 2 °C min⁻¹.

Synthesis of RuO₂: RuO₂ was prepared according to previous publication.¹ Briefly,

2.61 g of $\text{RuCl}_3 \cdot 3\text{H}_2\text{O}$ and 30 mL KOH (1.0 M) were added into 100 mL distilled water and stirred for 45 min at 100 °C. Then the above solution was centrifuged for 10 minutes and filtered. The precipitates were collected and washed with water several times. Finally, the product was dried at 80 °C overnight and then annealed at 350 °C in air atmosphere for 1 h.

Characterizations: X-ray diffraction (XRD) measurements were operated on a Panalytical/Empryan diffractometer with Cu $K\alpha$ radiation ($\lambda=1.5418 \text{ \AA}$). Scanning electron microscope (SEM) measurements were conducted on a XL30 ESEM FEG scanning electron microscope with an accelerating voltage of 20 kV. Transmission electron microscopy (TEM) images were recorded on a HITACHI H-8100 electron microscopy (Hitachi, Tokyo, Japan) at an accelerating voltage of 200 kV. X-ray photoelectron spectroscopy (XPS) data of the samples was collected on an ESCALABMK II X-ray photoelectron spectrometer using Al as the exciting source.

Electrochemical measurements: Electrochemical measurements were performed on an RST electrochemical workstation (Zhengzhou Shiruisi Technology Co., Ltd.) in a standard three-electrode setup using a P-DS- $\text{Co}(\text{OH})_2/\text{NF}$ as the working electrode. A platinum wire and an Hg/HgO were used as the counter electrode and the reference electrode, respectively. The temperature of solution was kept at 25 °C for all the measurements via the adjustment of air condition and heating support, which ensured the variation of diffusion coefficient below 1%. All potentials reported in this work were calibrated to RHE unless especially illustrated, using the following equation: $E(\text{RHE}) = E(\text{Hg/HgO}) + (0.098 + 0.059 \text{ pH}) \text{ V}$.

Turnover frequency (TOF) calculations: For TOF calculations, the surface concentration of active sites associated with the redox Co species should be first calculated, and the linear relationship between the oxidation peak current and scan rate is extracted from the electrochemical cyclic voltammetry scans. The slope of the line can be calculated based on the following equation:

$$\text{Slope} = n^2 F^2 A \Gamma_0 / 4RT$$

Where n is the number of electrons transferred; F is Faraday's constant; A is the surface area of the electrode; Γ_0 is the surface concentration of active sites (mol cm^{-2}),

and R and T are the ideal gas constant and the absolute temperature, respectively.²

TOF values can be finally calculated from the formula:

$$\text{TOF} = JA/4Fm$$

Where J is the current density at a certain overpotential, A is the area of the electrode, 4 indicates the mole of electrons consumed for evolving one mole of O_2 from water, F is Faraday's constant and m is the number of moles of active sites.³

Faradaic efficiency (FE) calculations: The evolved O_2 was confirmed by gas chromatography (GC) analysis and its total amount (n) was measured quantitatively using a calibrated pressure sensor to monitor the pressure change in the anode compartment of an H-type electrolytic cell. The total amount of charge (Q) passed through the cell was obtained from the current–time curve and the FE can be calculated from $\text{FE} = n/(Q/4F)$, where F is the Faraday constant.⁴

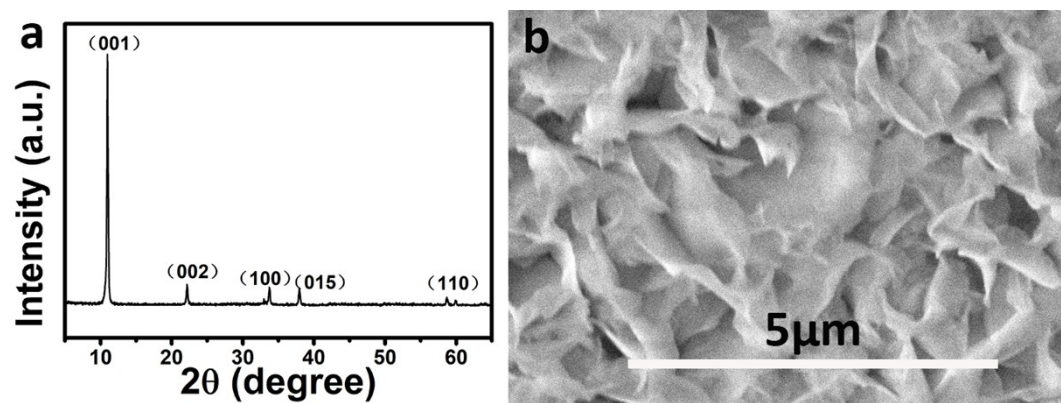


Fig. S1. (a) XRD pattern for Co(OH)_2 . (b) SEM image for $\text{Co(OH)}_2/\text{NF}$.

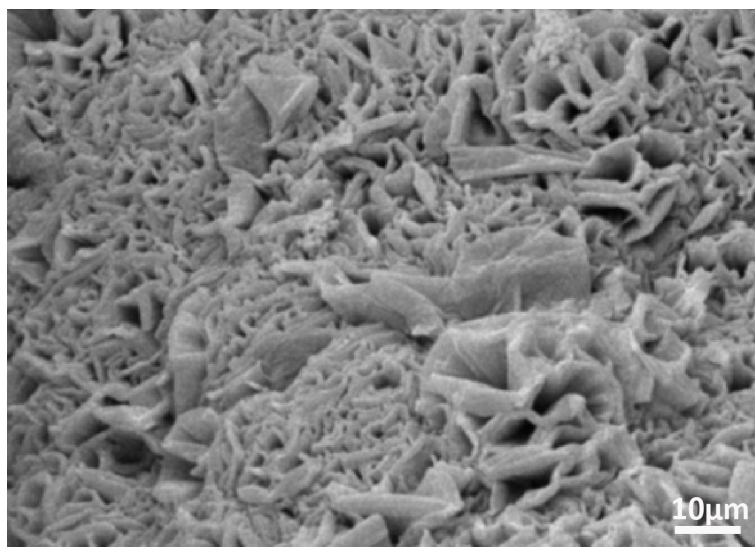


Fig. S2. SEM image for DS-Co(OH)₂/NF.

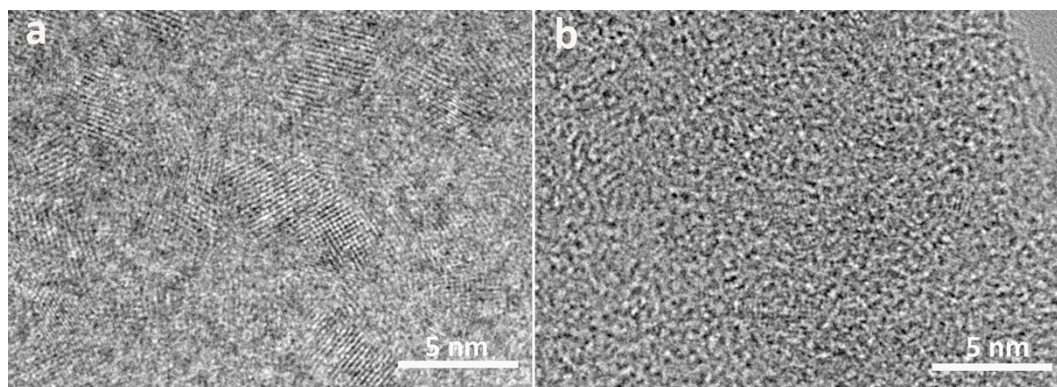


Fig. S3. HRTEM images for (a) DS-Co(OH)₂ and (b) P-DS-Co(OH)₂.

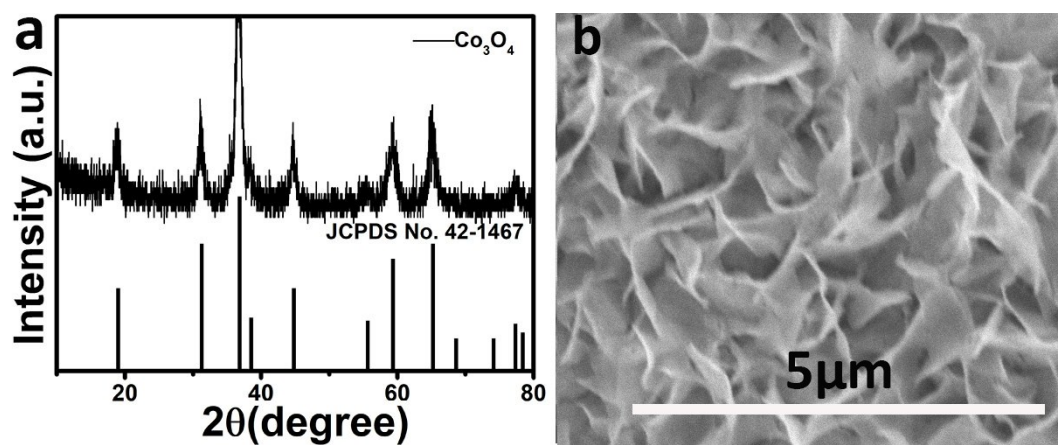


Fig. S4. (a) XRD pattern for Co_3O_4 . (b) SEM image for $\text{Co}_3\text{O}_4/\text{NF}$.

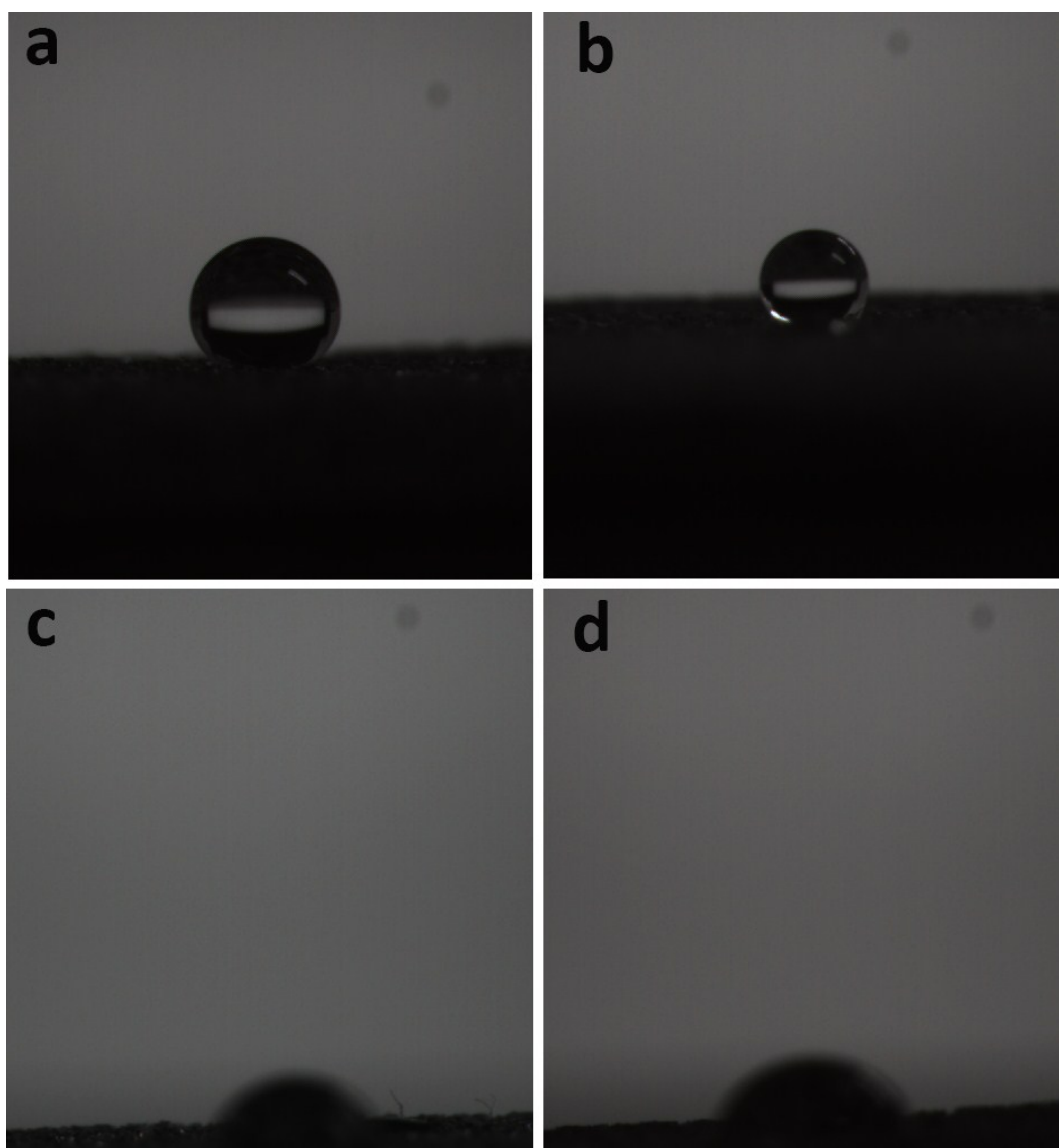


Fig. S5. Optical images of the water contact angles for (a) P-DS-Co(OH)₂/NF and (b) DS-Co(OH)₂/NF and 1M KOH contact angles for (c) P-DS-Co(OH)₂/NF and (d) DS-Co(OH)₂/NF.

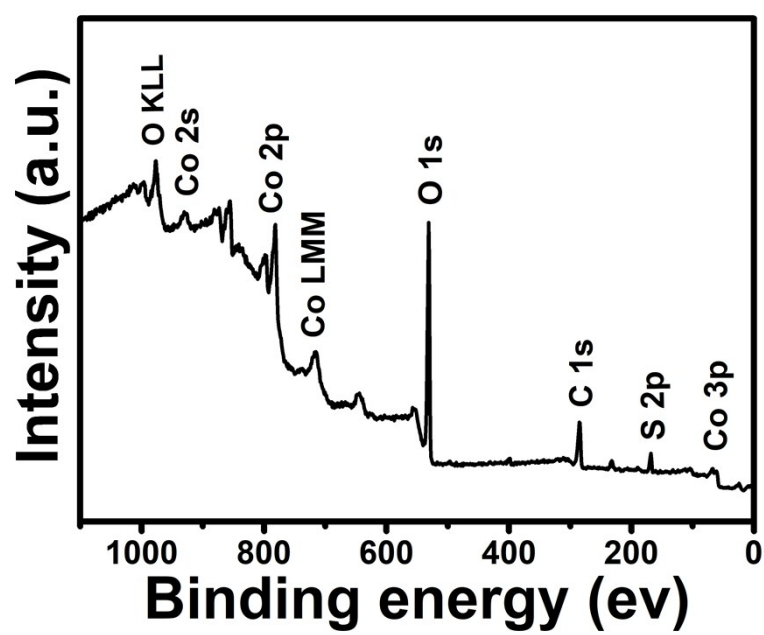


Fig. S6. XPS survey spectrum for P-DS-Co(OH)₂.

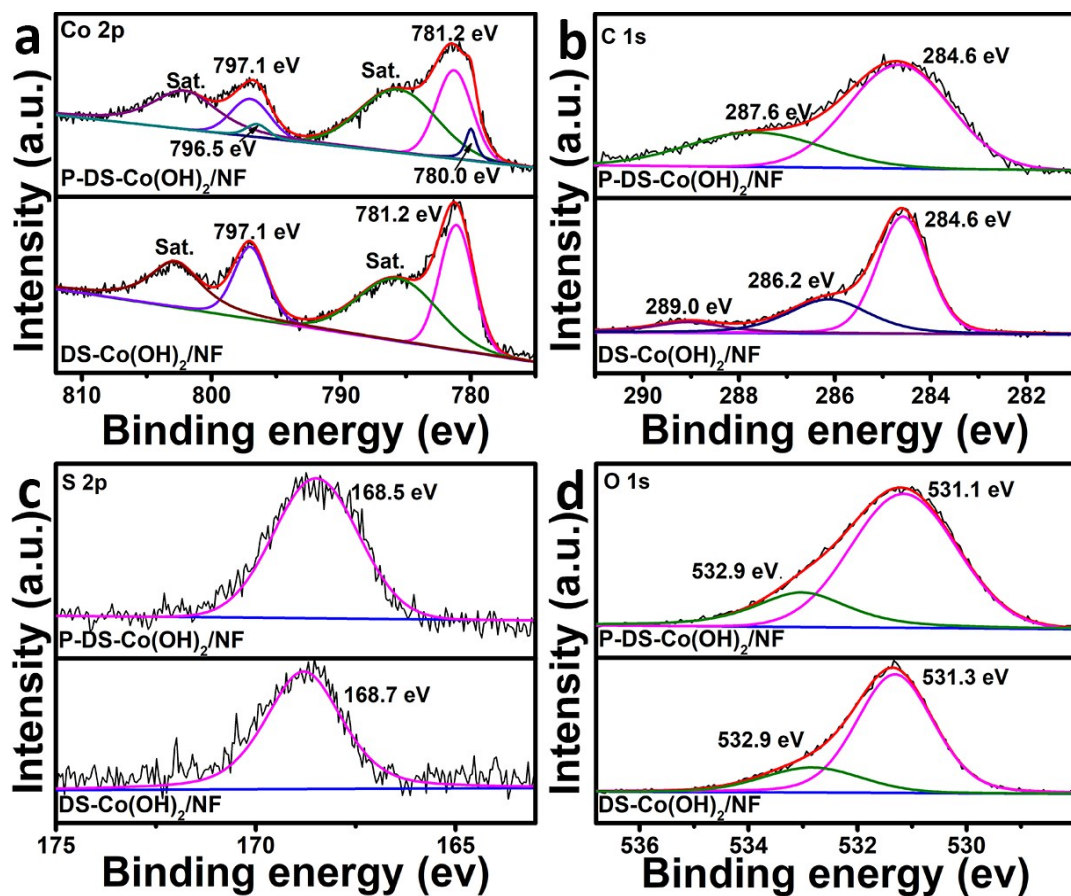


Fig. S7. XPS spectra for P-DS-Co(OH)₂/NF and DS-Co(OH)₂/NF in the (a) Co 2p (b) C 1s (c) S 2p and (d) O 1s regions.

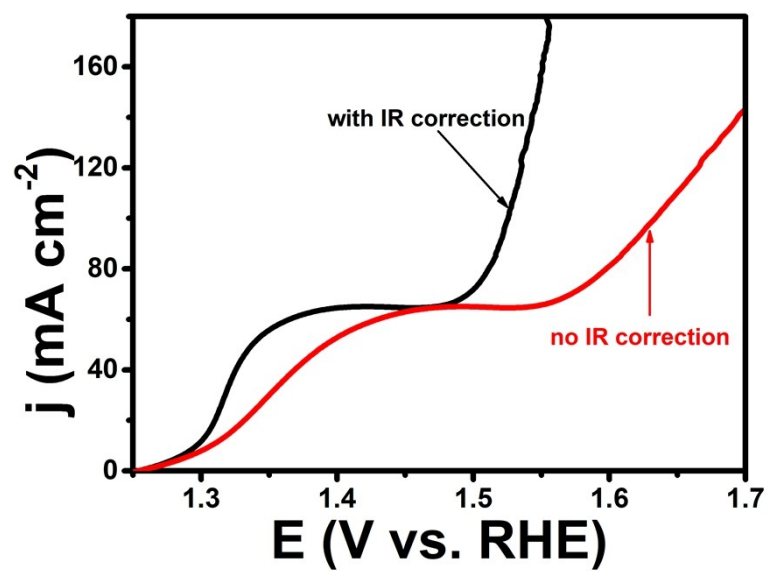


Fig. S8. LSV curves of P-DS-Co(OH)₂ for OER before and after iR correction.

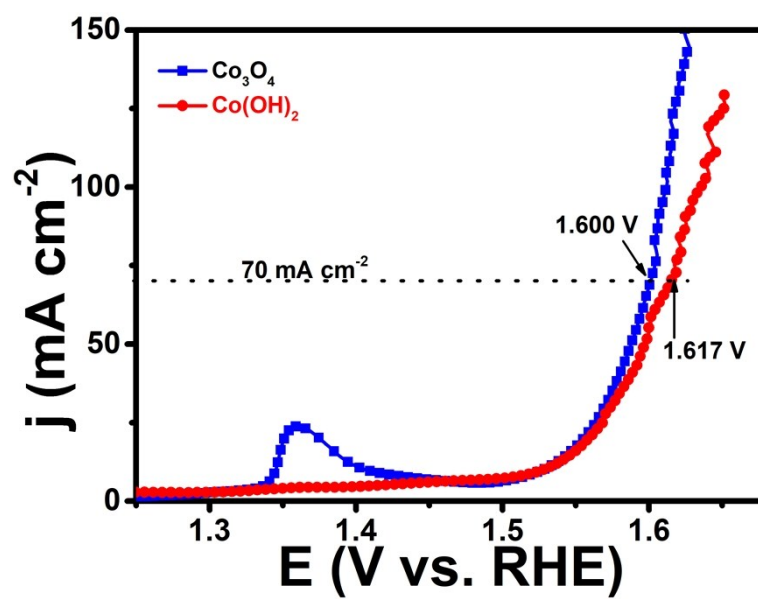


Fig. S9. LSV curve for $\text{Co(OH)}_2/\text{NF}$ and $\text{Co}_3\text{O}_4/\text{NF}$.

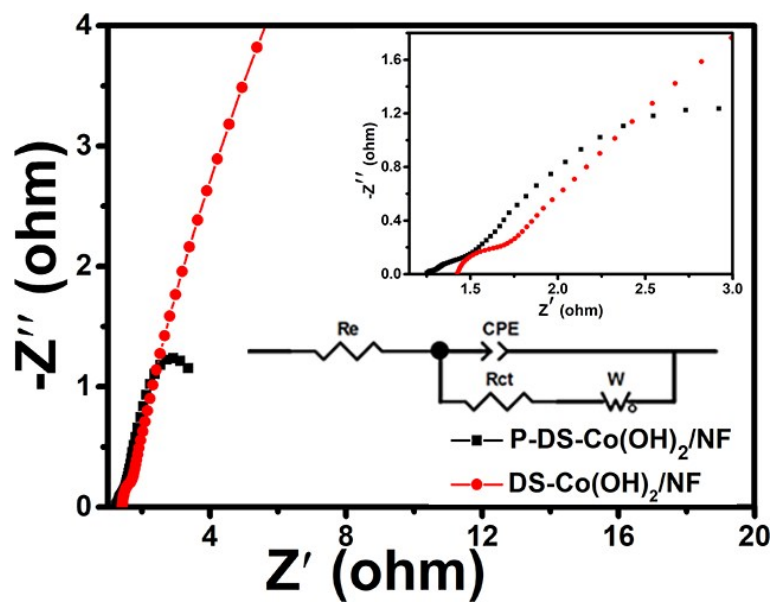


Fig. S10. Nyquist plots for P-DS-Co(OH)₂/NF and DS-Co(OH)₂/NF recorded in 1.0 M KOH. The insets show the zoom in view at high-frequency and equivalent circuit, respectively.

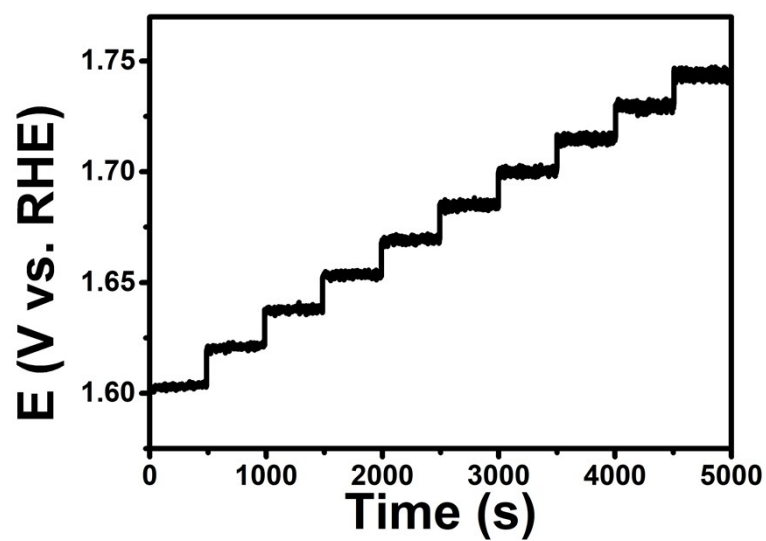


Fig. S11. Multi-current process for P-DS-Co(OH)₂/NF in 1.0 M KOH. The current density started at 70 mA cm⁻² and finished at 160 mA cm⁻², with an increment of 10 mA cm⁻² per 500 s without *iR* correction.

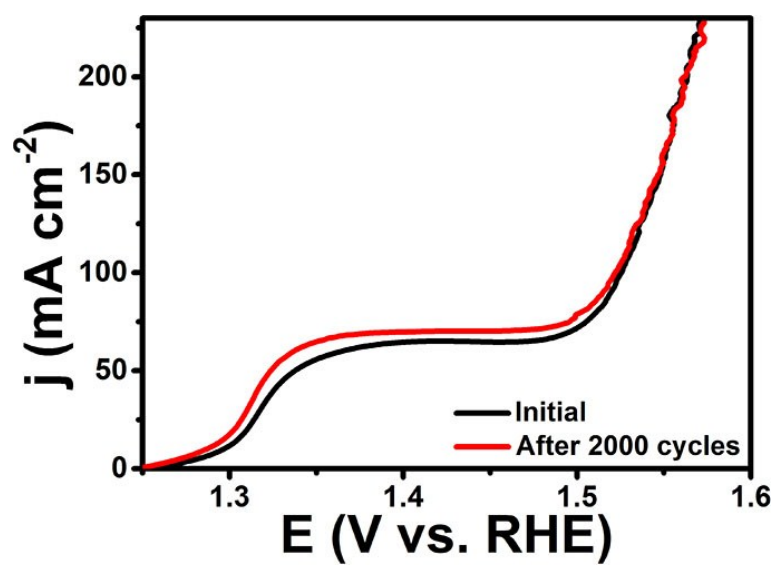


Fig. S12. LSV curves for P-DS-Co(OH)₂/NF before and after 2000 cyclic voltammetry cycles.

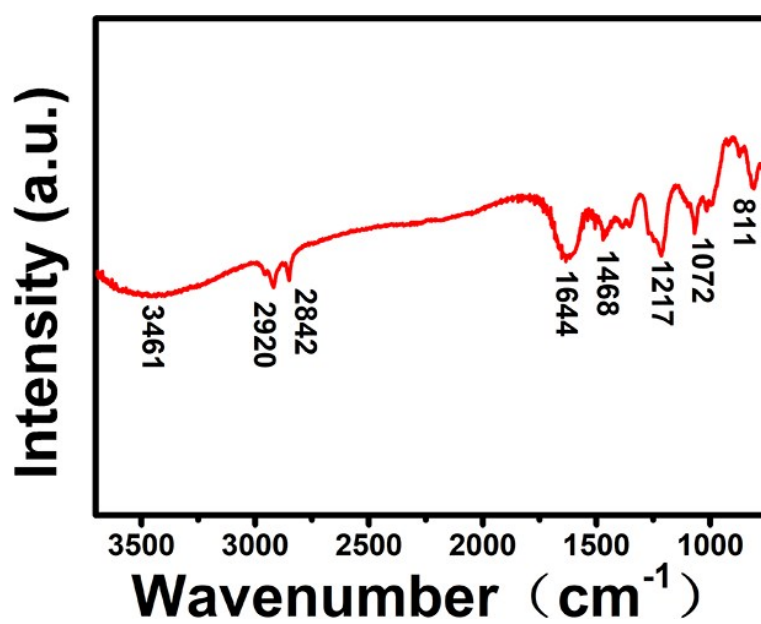


Fig. S13. FTIR spectra for DS- Co(OH)_2 /NF after 500 cyclic voltammetry cycles from 1.7 to 1.8 V.

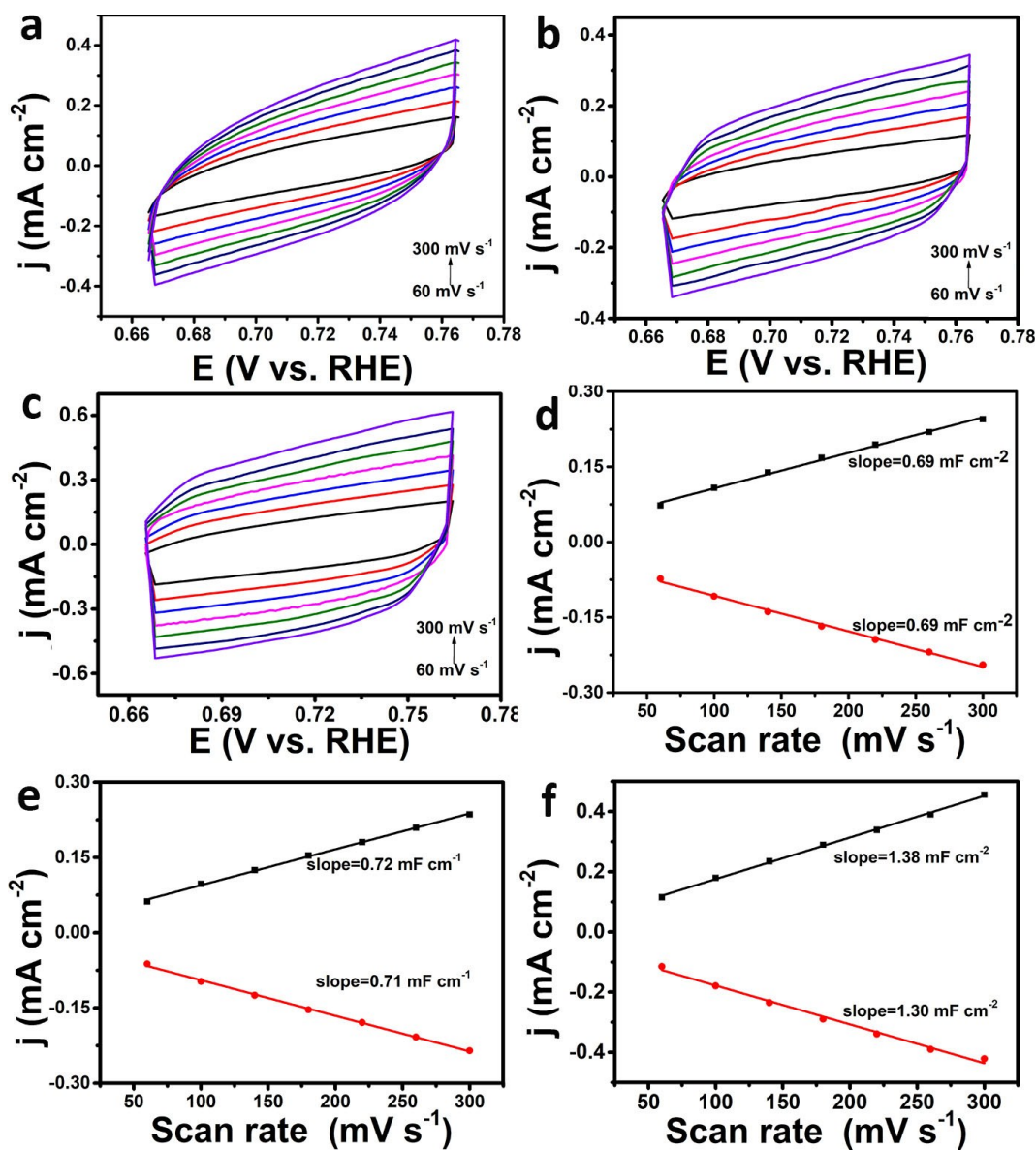


Fig. S14. CVs for (a) $\text{Co(OH)}_2/\text{NF}$, (b) $\text{DS-Co(OH)}_2/\text{NF}$ and (c) $\text{P-DS-Co(OH)}_2/\text{NF}$ in the non-faradaic capacitance current range at scan rates of 60, 100, 140, 180, 220, 260, and 300 mV s^{-1} in 1 M KOH. Corresponding capacitive currents at 0.71 V as a function of scan rate for (d) $\text{Co(OH)}_2/\text{NF}$, (e) $\text{DS-Co(OH)}_2/\text{NF}$ and (f) $\text{P-DS-Co(OH)}_2/\text{NF}$.

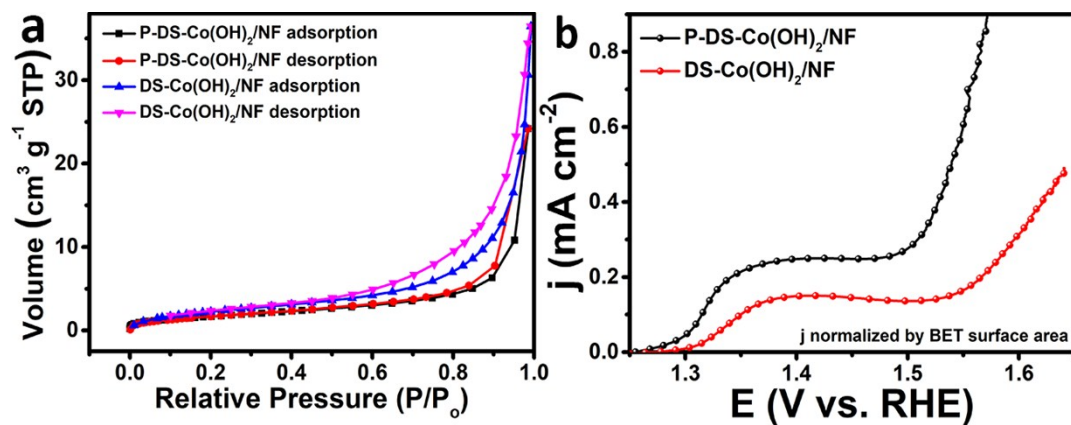


Fig. S15. (a) Nitrogen adsorption/desorption isotherm curves and (b) the LSV plots normalized by BET surface area of P-DS-Co(OH)₂/NF and DS-Co(OH)₂/NF with IR correction.

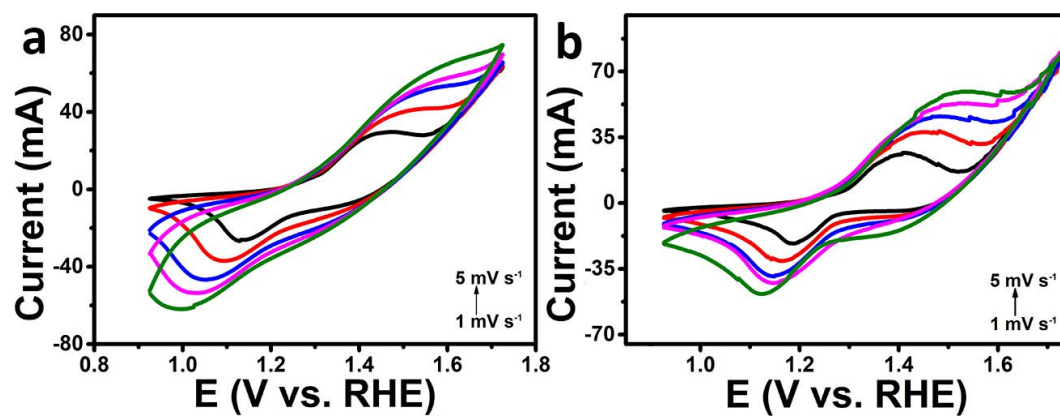


Fig. S16. CVs for (a) P-DS-Co(OH)₂/NF and (b) DS-Co(OH)₂/NF in the faradic capacitance current range at scan rates from 1 to 5 mV s⁻¹ in 1.0 M KOH.

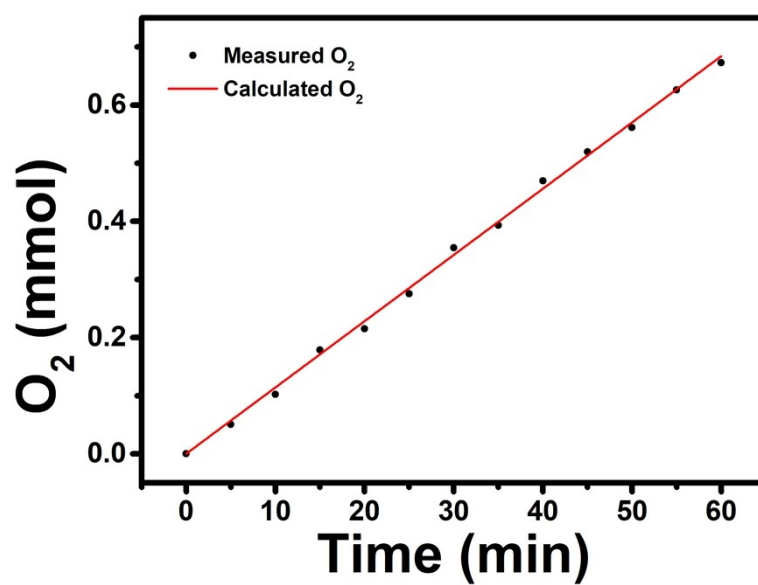


Fig. S17. The amount of theoretically calculated oxygen and experimentally measured oxygen versus time for P-DS-Co(OH)₂/NF at 1.50 V for 60 min in 1.0 M KOH.

Table S1. Comparison of water oxidation performance for P-DS-Co(OH)₂/NF with other non-noble-metal electrocatalysts under alkaline conditions.

Catalyst	j (mA cm ⁻²)	η (mV)	Electrolyte	Ref.
P-DS-Co(OH) ₂ /NF	70	266	1.0 M KOH	This work
	100	295		
DS-Co(OH) ₂ /NF	70	340		
benzoate-Co(OH) ₂ /NF	50	291	1.0 M KOH	5
Co(OH) ₂	15.5	300	0.1 M KOH	6
Micron-sized Co(OH) ₂	19.4	300	1.0 M KOH	7
CoO	10	312	1.0 M KOH	8
NiCo LDH	10	334	1.0 M KOH	9
CoCo LDH	10	393	1.0 M KOH	
ZnCo LDH	10	512-570	0.1 M KOH	10
Cl-intercalated Co(OH) ₂	10	320	1.0 M OH	11
NiCo LDH nanosheet	10	367	1.0 M KOH	12
Co(OH) ₂	10	390	1.0 M KOH	13
Ni Co(OH) _x	10	410	1.0 M KOH	
γ -CoOOH bulk	10	374	1.0 M KOH	14
γ -CoOOH nanosheet	10	300	1.0 M KOH	
CoFe LDH/NF	10	300	1.0 M KOH	15
Ni ₇₆ Co ₂₄ -LDHs	10	296	1.0 M KOH	16
CoMn-LDH/CNT	10	335	1.0 M KOH	17
Zn-Co-LDH	15.06	540	1.0 M KOH	18
NiCoFe-LDH nanoarray	80	257	1.0 M KOH	19
Fe incorporated α -Co(OH) ₂	10	295	1.0 M KOH	20
CQD/NiFe-LDH	10	235	1.0 M KOH	21

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