<div align="center">**Electronic Supplementary Information**</div>

**Experimental section**

*Materials*: All chemicals, including Nitric acid ($HNO_3$), ethanol ($C_2H_5OH$), Nickel nitrate hexahydrate ($Ni(NO_3)_2 \cdot 6H_2O$), Cobalt(II) chloride hexahydrate ($CoCl_2 \cdot 6H_2O$), ammonium chloride ($NH_4Cl$), and potassium hydroxide (KOH) were purchased from commercial suppliers and used without further purification. Graphite felt was purchased from Beijing Jinglong Tetan Technology Co., Ltd. Graphite paper was provided by Hongshan District, Wuhan Instrument Surgical Instruments business. Milli-Q ultrapure water of 18.25 MΩ.cm was applied in all experiments.

*Preparation of the NiCo LDHs/GF and NiCo LDHs/GP:* A piece of GF with a size of $1 \times 1$ cm$^2$ was cleaned in ethanol for 15 min, cleaned in $HNO_3$ and boil for two hours. After that, cleaned ultrasonically in deionized water for 15 min. Place it in a 60 ℃ oven to dry for 4 h. The electrodeposition of NiCo LDHs was carried out in a three-electrode system using GF as the working electrode, a saturated calomel electrode (SCE) as the reference electrode, and Pt slice as the counter electrode. The electrolyte was obtained by dissolving $Ni(NO_3)_2 \cdot 6H_2O$ (1.45 g), $NH_4Cl$ (0.178 g), and $CoCl_2 \cdot 6H_2O$ (2.38 g) in 25 mL ultrapure water. The applied potential was -1.0 V vs. SCE and electrodeposition time is 5 min. The samples were then washed with ultrapure water and dried in air. NiCo LDHs/GP was similarly prepared.

*Characterizations:* X-ray diffraction (XRD) data were collected by using a LabX XRD-6100 X-ray diffractometer with a Cu Kα radiation (40 kV, 30 mA) of wavelength 0.154 nm (SHIMADZU, Japan). X-ray photoelectron spectra (XPS) spectra were acquired with an ESCALABMK II X-ray photoelectron spectrometer using a nonmonochromatized A1 Kα X-ray source (1486.6 eV). Scanning electron microscopy (SEM) images were collected on a Helios G4 UC scanning electron microscope (Thermo Fisher Scientific). Transmission electron microscopy (TEM) images were obtained by using a HITACHI H-8100 electron microscopy (Hitachi, Tokyo, Japan) operated at 200 kV.

*Electrochemical measurements:* Electrochemical measurements were performed with a CHI 660E potentiostat (CH Instruments, China) in a standard three-electrode setup

with the prepared samples as the working electrode, a graphite rod as the counter electrode, the Hg/HgO electrode as the reference electrode. The HER activity was evaluated using linear sweep voltammetry (LSV) with a sweep rate of 5 mV s$^{-1}$. Electrochemical impedance spectroscopy (EIS) was measured at a frequency between 0.1 Hz and $10^6$ Hz. The tests were performed in 1.0 M KOH solution. All the potentials were displayed versus reversible hydrogen electrode (RHE) by: E (RHE) = E (Hg/HgO) + 0.098 + 0.059 × pH. The 100% iR-corrected was applied in the LSV and chronopotentiometry experiments.
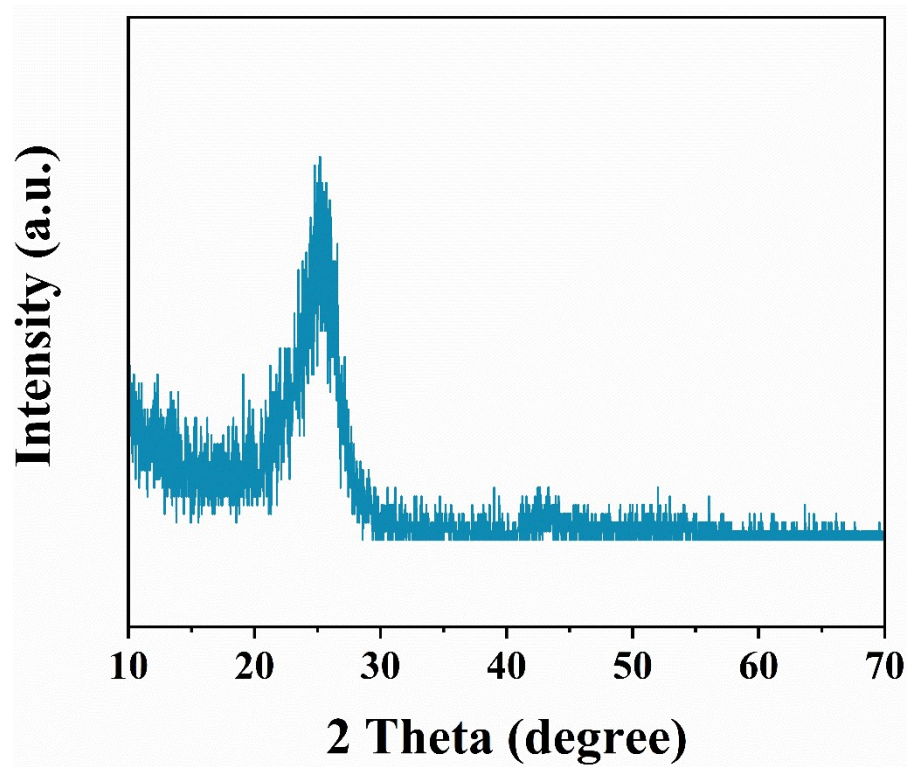
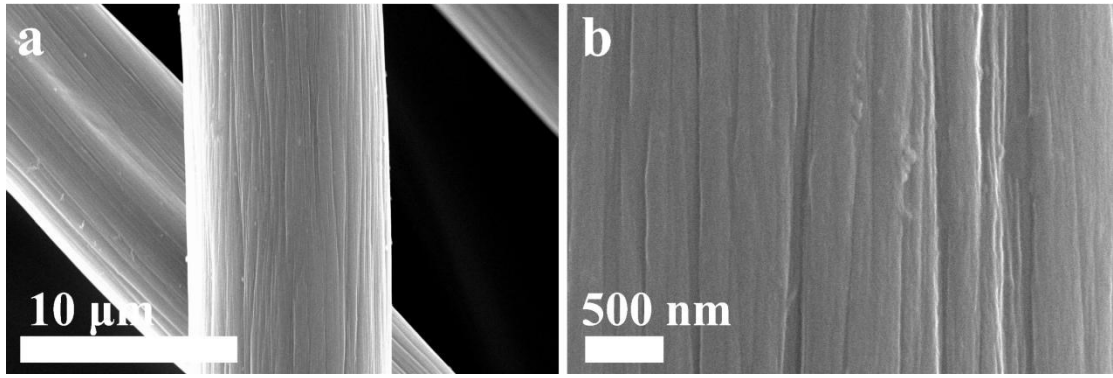**Fig. S1** XRD pattern of bare GF.

**Fig. S2** (a) Low- and (b) high-magnification SEM images for bare GF.
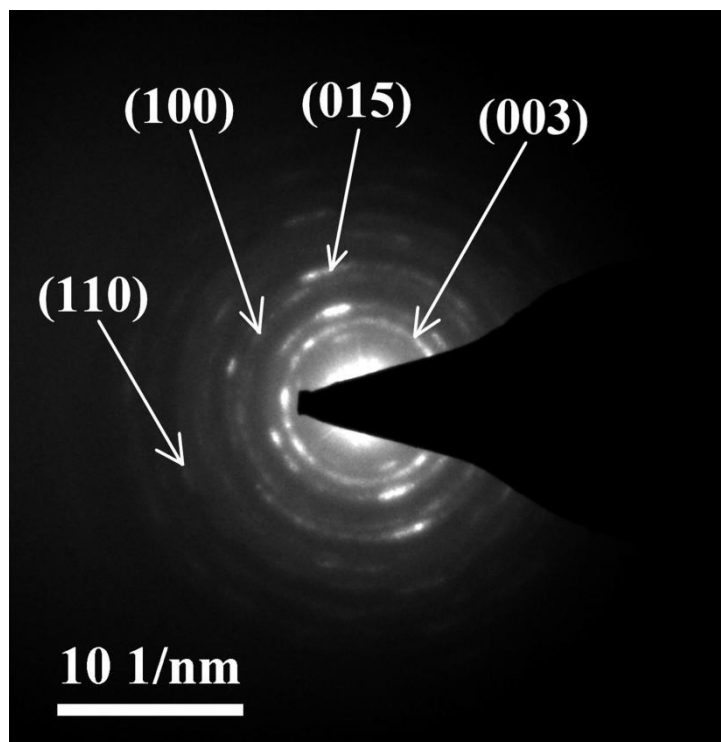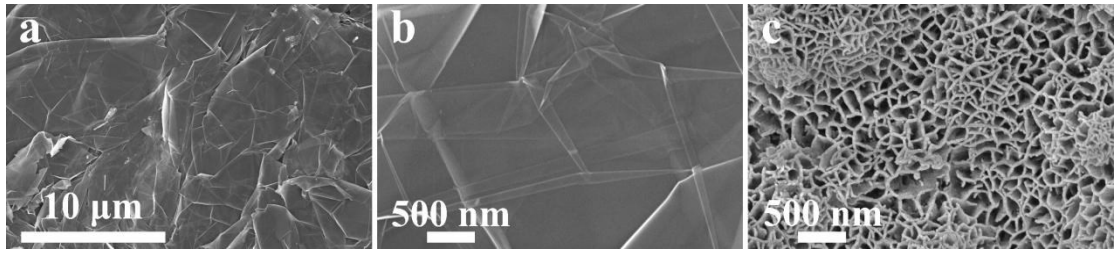
**Fig. S3** SAED pattern of NiCo LDHs.

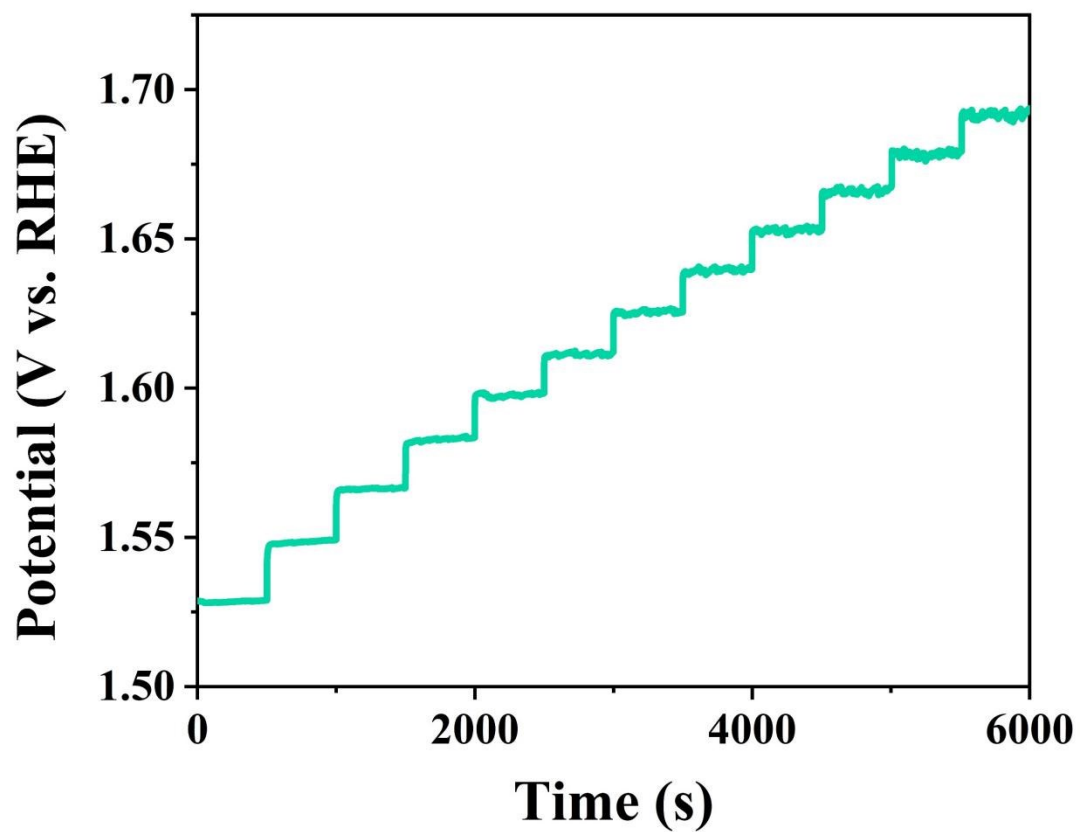**Fig. S4** SEM images for (a,b) GP and (c) NiCo LDHs/GP.

**Fig. S5** Multi-step chronopotentiometric curve for NiCo LDHs/GF with the current being started from 20 to 130 mA cm$^{-2}$ (10 mA cm$^{-2}$ per 500 s).
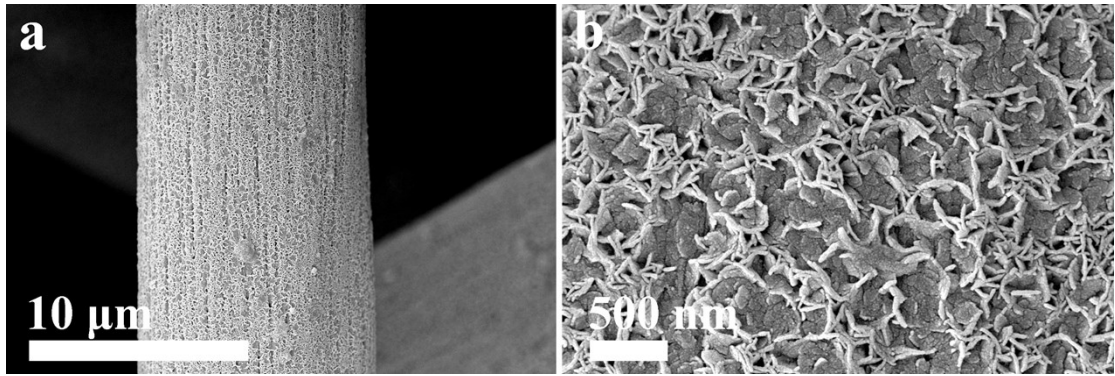
**Fig. S6** (a) Low- and (b) high-magnification SEM images for NiCo LDHs/GF after the stability test.
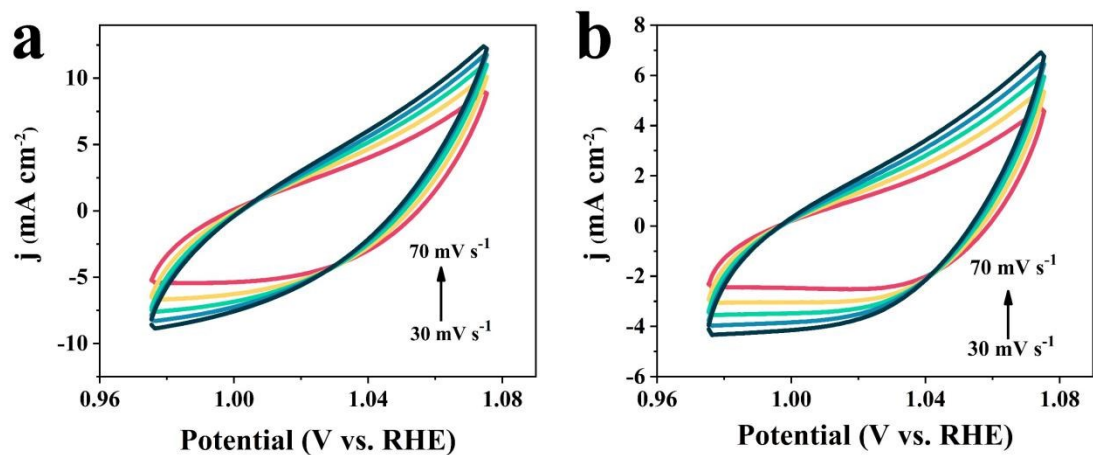
**Fig. S7** Cyclic voltammograms of (a) NiCo LDHs/GF and (b) NiCo LDHs/GP in the non-Faradaic capacitance current range at scan rates of 30, 40, 50, 60, and 70 mV s$^{-1}$.

**Fig. S8** LSV curves of NiCo LDHs/GF with different electrodeposition time (3 min, 5 min, and 10 min).

**Fig. S9** Cyclic voltammograms of (a) NiCo LDHs/GF-5 min and (b) NiCo LDHs/GF-10 min in the non-Faradaic capacitance current 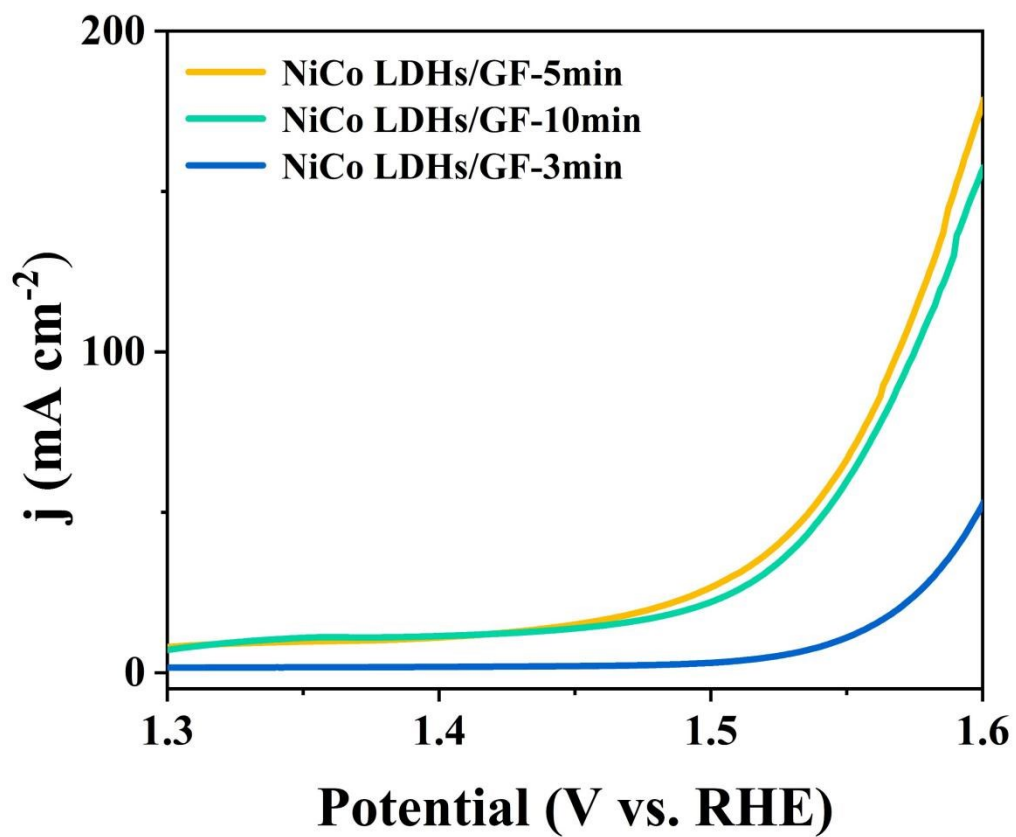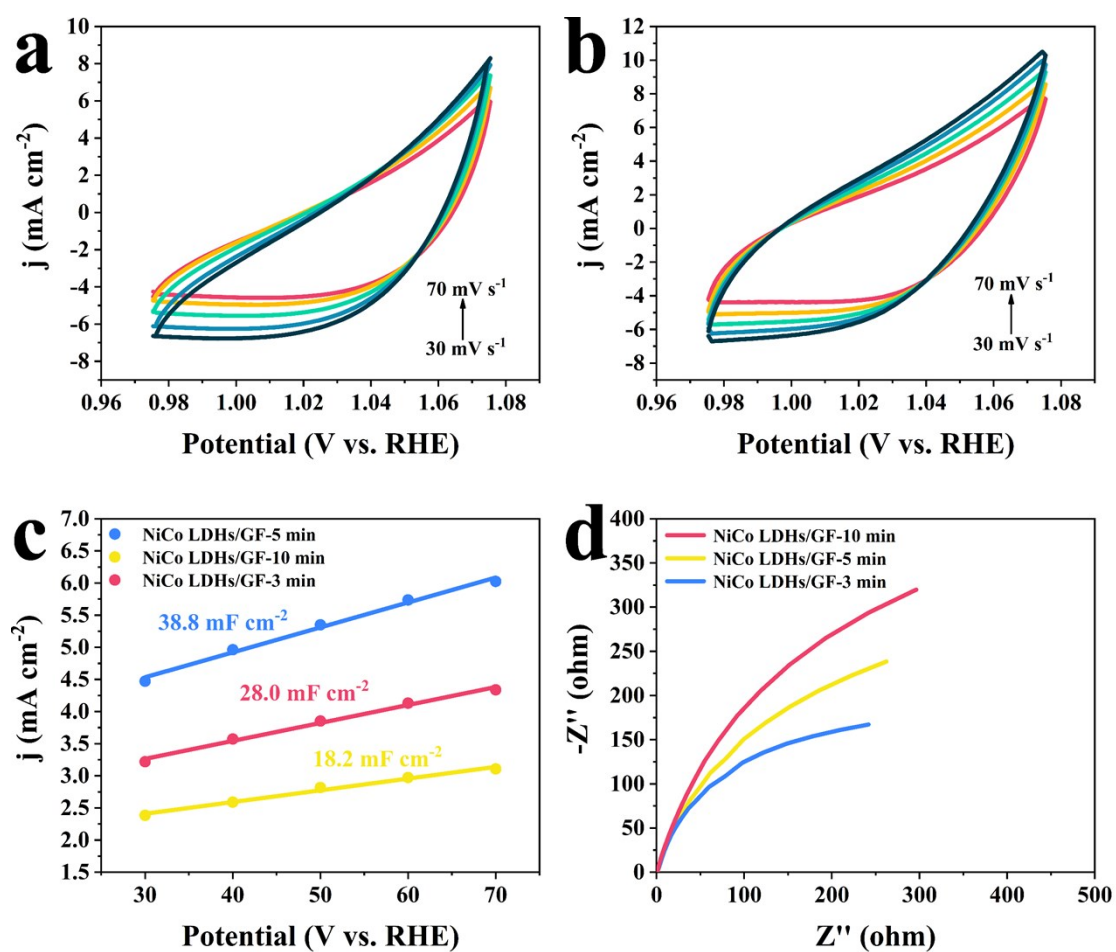range at scan rates of 30, 40, 50, 60, and 70 mV s$^{-1}$. (c) Capacitive currents at 1.02 V vs. RHE as a function of scan rate and (d) Nyquist plots for NiCo LDHs/GF-3 min, NiCo LDHs/GF-5 min, and NiCo LDHs/GF-10 min.

**Table S1.** Comparison of water oxidation performances of NiCo LDHs/GF with

| OER catalyst | η (mA cm$^{-2}$) | Overpotential (mV) | ECSA (mF cm$^{-2}$) | Electrolyte | Ref. |
|---|---|---|---|---|---|
| NiCo LDHs/GF | 20 | 249 | 38.8 | 1.0 M KOH | This work |
| NiCo-NP-7.50 | 10 | 314 | 59.82 | 1.0 M KOH | 1 |
| NiCo-LDH@HOS | 10 | 293 | 5.38 | 0.1M KOH | 2 |
| NiCo-LDH nanosheets | 10 | 346 | 3.24 | 0.1M KOH | 2 |
| Co$_{0.5}$Ni$_{0.5}$/rGO | 10 | 288 | 7.69 | 1.0 M KOH | 3 |
| NiCo-LDH/Ar | 10 | 299 | 2.58 | 1.0 M KOH | 4 |
| CoNi-LDH@PCPs | 10 | 350 | - | 1.0 M KOH | 5 |
| NiCo-LDHs | 20 | 310 | 5.3 | 1.0 M KOH | 6 |
| NiCo/NiCoOx@FeOOH | 10 | 278 | - | 1.0 M KOH | 7 |
| NiCo LDH nanosheets | 10 | 367 | 0.0041 | 1.0 M KOH | 8 |
| NiCo-NS | 10 | 335 | 0.0093 | 1.0 M KOH | 9 |
| CoNi LDH/CoO | 10 | 300 | - | 1.0 M KOH | 10 |
| NiCo@Ru core-shell nanoparticles | 10 | 272 | - | 1.0 M KOH | 11 |
| Ni$_3$Co$_3$@Ru HNS | 10 | 300 | 2.9 | 1.0 M KOH | 12 |

other catalysts under alkaline conditions.

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