Electronic Supplementary Information

Experimental section

Materials: All chemicals, including Hydrochloric acid (HCl), ethanol (C_2H_5OH), sodium hydroxide (NaOH), ammonium persulfate [$(NH_4)_2S_2O_8$], Cobalt(II) chloride hexahydrate ($Co(Cl)_2{\cdot}6H_2O$), Nickel nitrate hexahydrate ($Ni(NO)_2{\cdot}6H_2O$), potassium hydroxide (KOH) and Cu foil (CF) (thickness: 0.1mm, purity: Cu ≥ 99.99%) were purchased from commercial suppliers and used without further purification. Milli-Q ultrapure water of 18.25 MΩ.cm was applied in all experiments.

Preparation of the CuO/CF, CuO@NiCo LDH/CF, and NiCo LDH/CF: A piece of CF with a size of 1 × 1 cm^2 was cleaned in 2 M HCl and then cleaned ultrasonically in ethanol and deionized water, each for 15 min. The cleaned CF was then immersed into 50 mL of an aqueous solution (AOES) containing 5 g NaOH and 1.428 g $(NH_4)_2S_2O_8$ for 20 min. After that, CF with a light blue color was taken out from the solution, rinsed with deionized water, and dried in the air, followed by air annealing at 180 ^oC for 2 h at a speed of 2 ^oC min^{-1} to obtain CuO/CF. The electrodeposition of NiCo LDH was carried out in a three-electrode system with using CuO/CF 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 $Ni(NO_3)_2{\cdot}6H_2O$ (1.0905 g) and $CoCl_2{\cdot}6H_2O$ (0.89 g) in 25 mL water. The applied potential was -1.0 V vs. SCE and electrodeposition time with 50 s. The samples were then washed with deionized water and dried in the air. NiCo LDH/CF was similarly prepared on CF.

Preparation of the RuO_2/CF: to assemble RuO_2 on CF, 5mg commercial RuO_2 was diffused in a mixture solution encompassing 15 μL of 5 wt% Nafion, 85 μL DI water and 100 μL ethanol under increasing stirring for 20 min. Afterward, the as-prepared RuO_2 solution was painted onto CF. Then RuO_2/CF was dried in an oven at 60 ^oC for 2h.

Characterizations: XRD patterns were obtained from a Shimazu XRD-6100 diffractometer with Cu Kα radiation (40 kV, 30 mA) of wavelength 0.154 nm (Japan). TEM images were obtained from a Zeiss Libra 200FE transmission electron microscope operated at 200 kV. XPS measurements were performed on an ESCALABMK II X-ray photoelectron spectrometer using Mg as the exciting source. SEM measurements were carried out on an XL30 ESEM FEG scanning electron microscope at an accelerating voltage of 20 kV. Inductively coupled plasma mass 2 spectrometry (ICP-MS) measurement was carried out on Agilent 7700s.

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 OER 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 a 1 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.

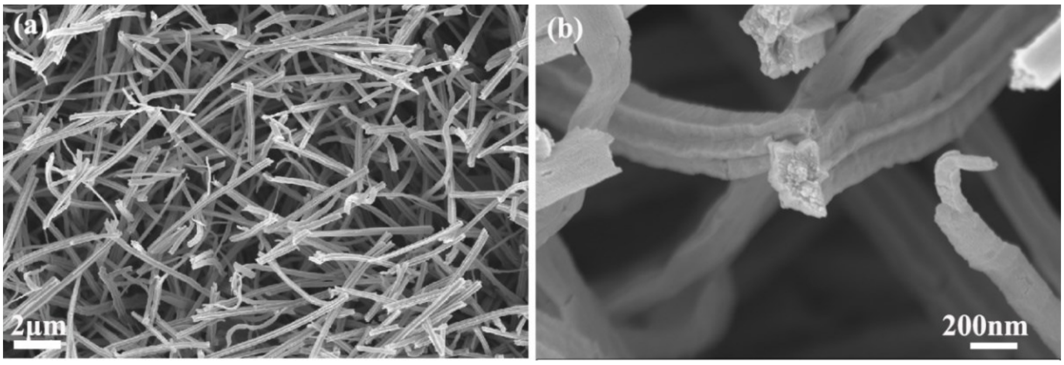

Fig. S1. SEM images for CuO /CF.

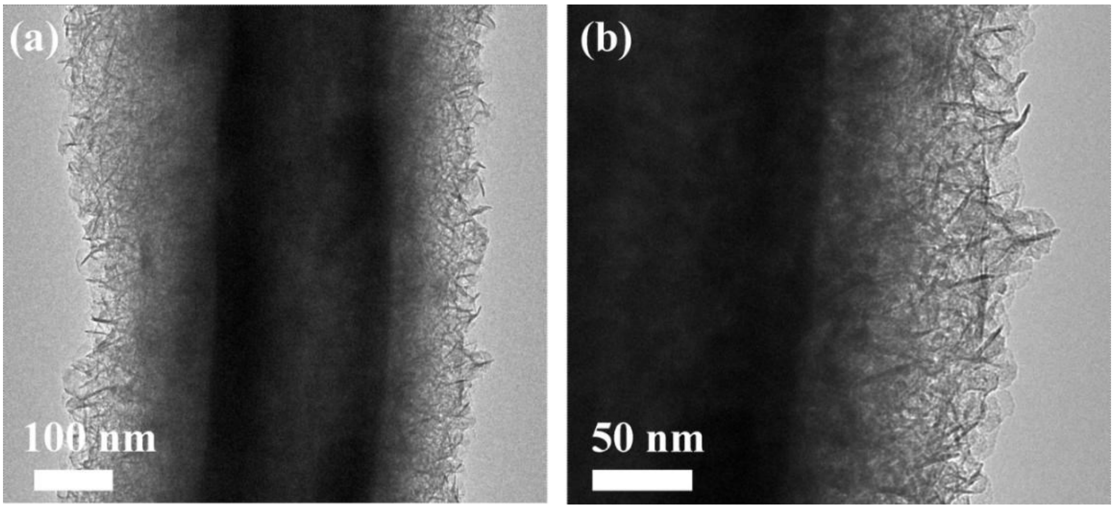

Fig. S2. HRTEM images of CuO@NiCo LDH nanowire.

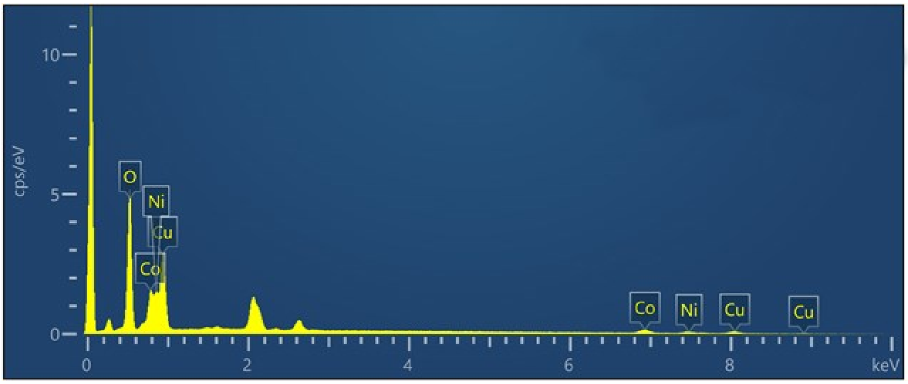

Fig. S3. EDX spectrum for CuO@ NiCo LDH/CF.

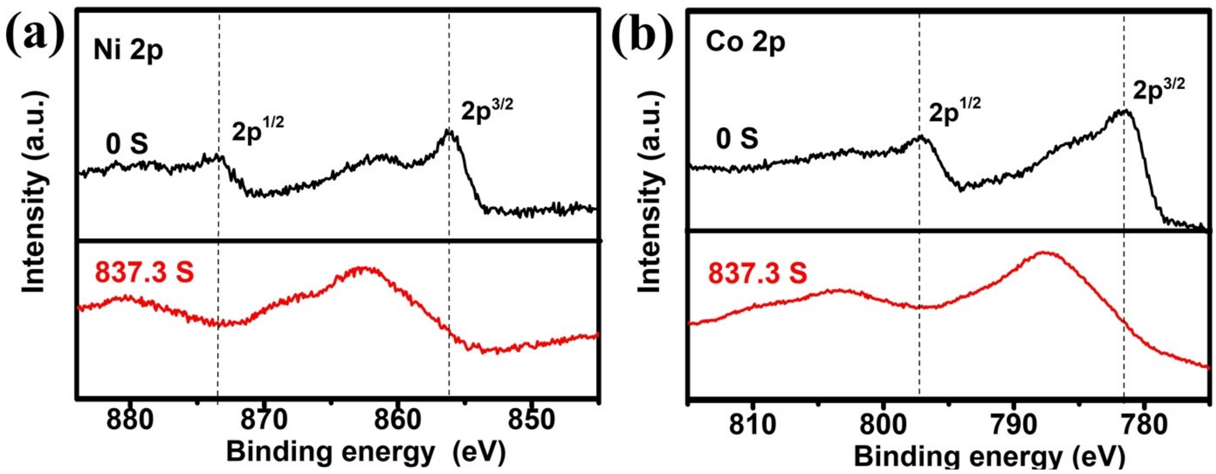

Fig. S4. Depth-resolved XPS analysis of CuO@NiCo LDH in the (a)Ni 2p; (b) Co 2p region. The times show the duration of a surface etching by the X-ray beam.

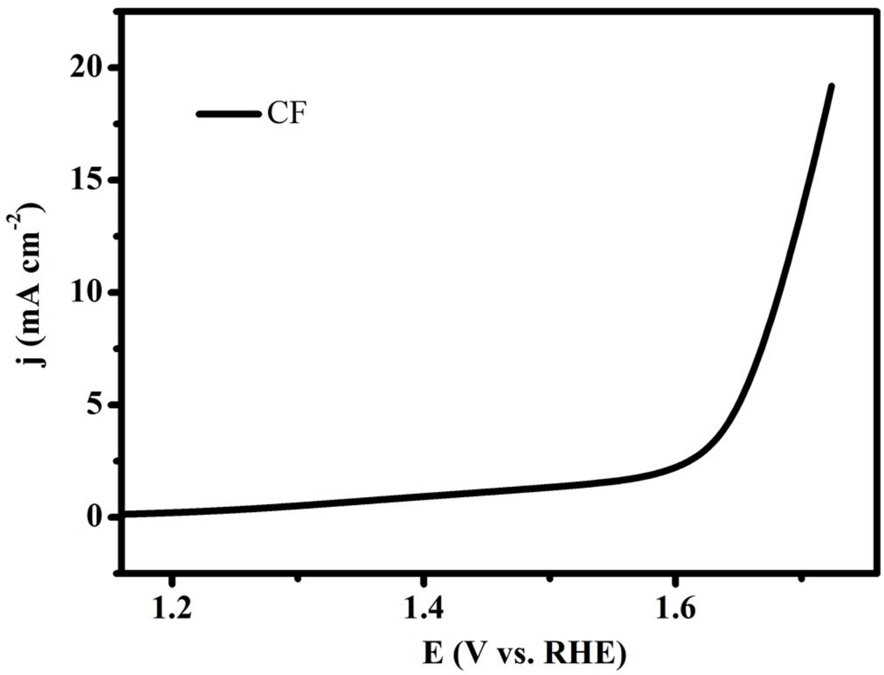

Fig. S5. Polarization curves of CF for OER with a scan rate of 5 mV s^{-1}.

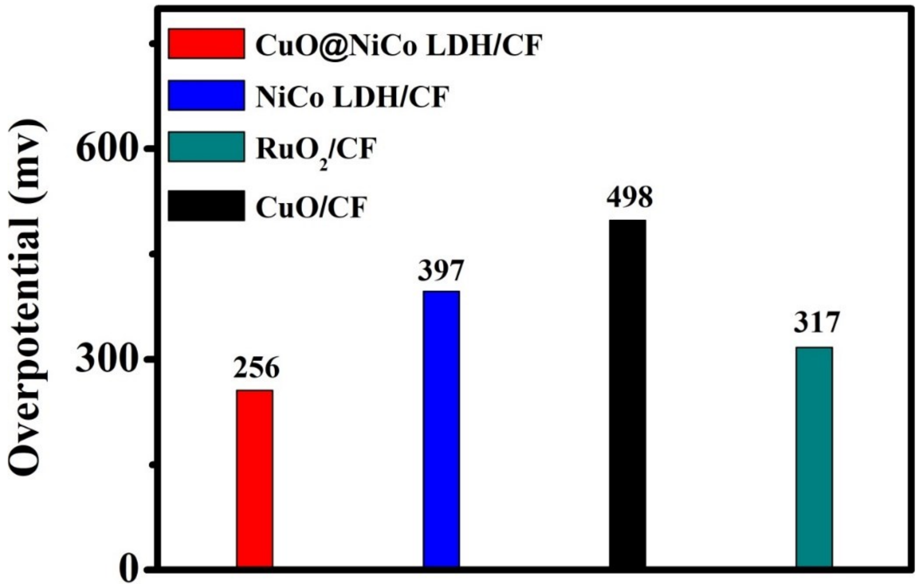

Fig. S6. Overpotential values needed for achieving fixed geometric catalytic current densities of 20 mA cm^{-2} for CuO@NiCo LDH/CF, NiCo LDH/CF, RuO_2/CF, and CuO/CF, respectively.

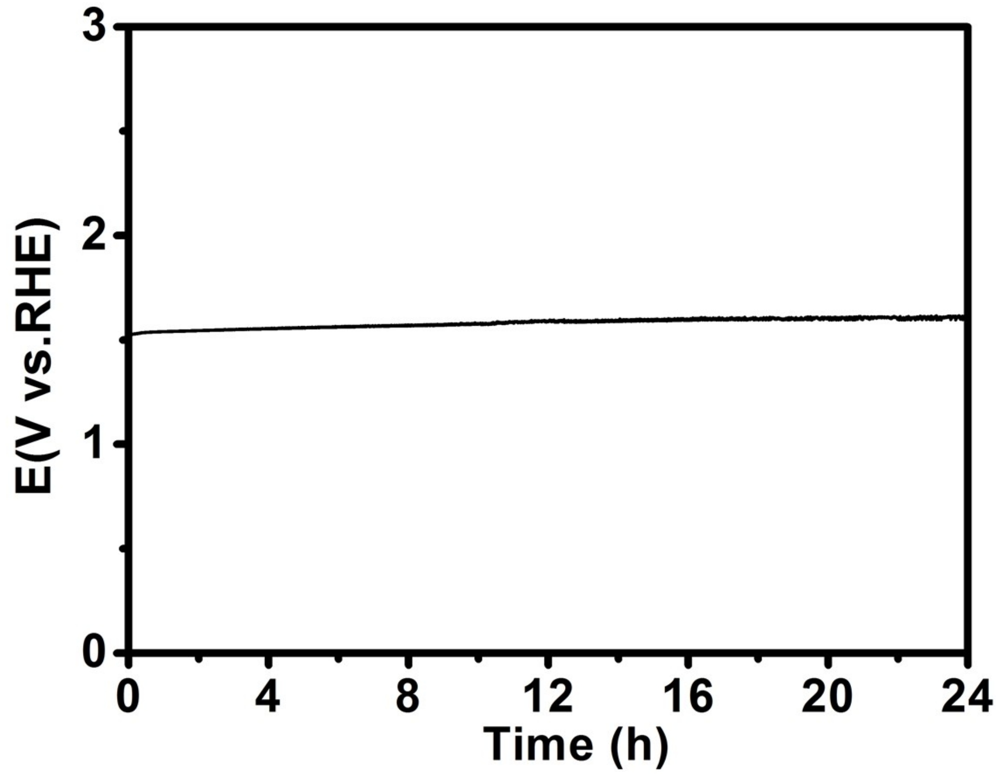

Fig. S7. Constant current chronopotentiometric stability measurements at a current density of 20 mA cm^{-2} (without iR correction).

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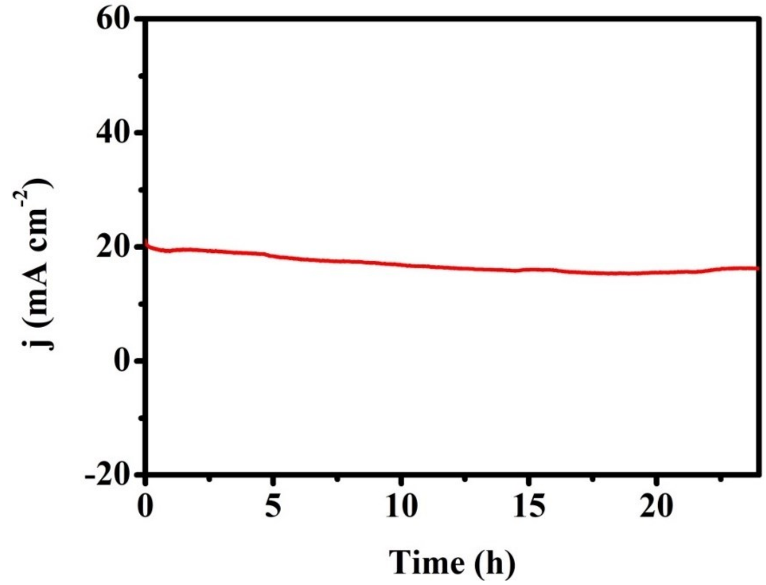

Fig. S8. Time-dependent current density curve for CuO@NiCo LDH/CF at 1.51 V (without iR correction).

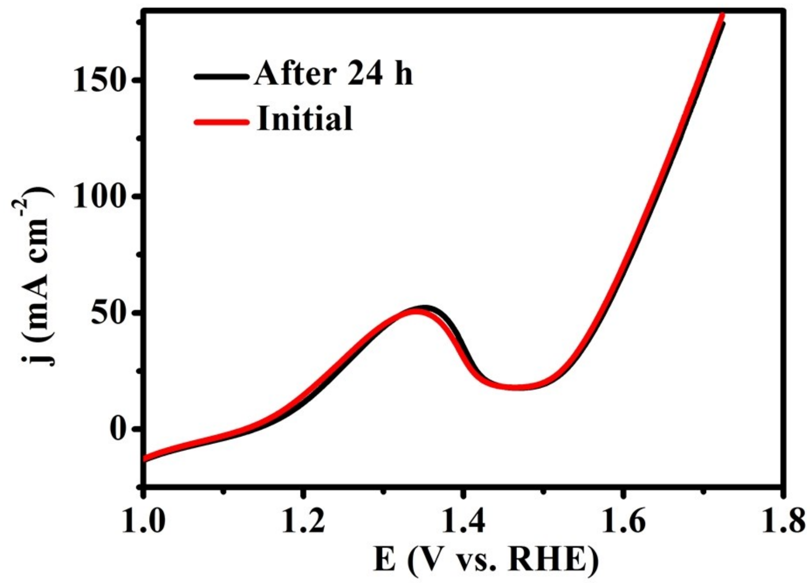

Fig. S9. LSV curves for CuO@NiCo LDH/CF before and after the time-dependent current density stability tests.

Table S1. Comparison of water oxidation performances of CuO@NiCo LDH/CF with other catalysts under alkaline conditions.

OER catalyst	Current density (mA cm^{-2})	Corresponding overpotential (mV)	Electrolyte	Ref.
CuO@NiCo LDH/CF	20	256	1 M KOH	**This work**
NiCo LDH/NF	10	271	1 M KOH	1
NiCo/NiCoOx@FeOOH	10	278	1 M KOH	2
$(Ni,Co)_{0.85}Se$	20	287	1 M KOH	3
$Co_{0.5}Ni_{0.5}$/rGO	10	288	1 M KOH	4
Calixarene intercalated NiCo LDH	10	290	1 M KOH	5
NiCo LDH/CF	10	307	1 M KOH	6
NiCo LDH monolayers	10	335	1 M KOH	7
NiCo LDH nanocages	10	350	1 M KOH	8
NiCo LDH nanosheets	10	367	1 M KOH	9
NiFe LDH on Cu mesh	10	292	1 M KOH	10
CuO-NiO on Ni foam	10	319	1 M KOH	11
NiCo@Ru core-shell nanoparticles	10	272	1 M KOH	12
NiFe LDH/CuO nanosheet	20	270	1 M KOH	13
$Co_1Ni_{0.3}$ LDHs	10	266	1 M KOH	14
CoNi LDH/CoO	10	300	1 M KOH	15

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