Electronic Supporting Information

Ammonia-Assisted Synthesis of low-crystalline FeCo Hydroxides for Efficient Electrochemical Overall Water Splitting

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Experimental Procedures

Materials.

$CoCl_2$, $FeCl_3$, ammonia (37%) and KOH were purchased from Sigma-Aldrich. All of these chemicals used in this experiment without further purification. The Milli-Q water used in all experiments with a resistivity of 18.2 MΩ cm was prepared using a Milli-Q water system.

Preparation of low-crystalline FeCo Hydroxides.

In a typical experiment, 1.0 mol of $CoCl_2$, 1.0 mol of $FeCl_3$ was dissolved in 500 ml of H_2O with magnetic stirring, and then 30 mL ammonia (37%) was added in the solution stirring for 30 min. The final product was collected by centrifugation, washed with water for three times.

Preparation of high-crystalline FeCo Hydroxides.

In a typical experiment, 500 mg of low-crystalline FeCo Hydroxides dissolved in 2 mL of water and 20 mL of DMF to form a homogeneous solution. After the mixture was sealed in a 30-ml pressure vessel, it was heated from room temperature to 140°C in 30 min and then kept at 140°C for 10 h in oven. The product was collected and rinsed three times with Milli-Q water.

Preparation of Fe Hydroxides.

I In a typical experiment, 1.0 mol of $FeCl_3$ was dissolved in 500 ml of H_2O with magnetic stirring, and then 15 mL ammonia (37%) was added in the solution stirring for 30 min. The final product was collected by centrifugation, washed with water for three times.

Preparation of Co Hydroxides.

I In a typical experiment, 1.0 mol of $CoCl_2$ was dissolved in 300 ml of H_2O with magnetic stirring, and then 15 mL ammonia (37%) was added in the solution stirring for 30 min. The final product was collected by centrifugation, washed with water for three times.

Characterizations.

TEM was carried out on JEOL JEM-2100F microscope and JEM-2010. All samples were dispersed in ethanol and then dropped on Cu grids. X-ray diffraction (XRD) patterns were recorded with a Shimadazu XRD-6000. XPS data were acquired using a Kratos Axis Ultra XPS. All electrochemical measurements were carried out on a CHI 760e electrochemical workstation in a three-electrode configuration cell using as-prepared sample loading on carbon paper as the working electrode, where the active area was precisely controlled at 1×1 cm^2, platinum plate (1×1 cm^2) as the counter electrode, and saturated calomel electrode (SCE) as the reference electrode. In 1.0 M KOH, $E(RHE) = E(SCE) + 0.2412 + pH \times 0.0592 = E(SCE) + 1.046$. As for HER, overpotential = 0 V – E(RHE). As for OER, overpotential = E(RHE) – 1.23 V. All the electrolysis was conducted at room temperature and 80% IR correction was applied in all measurements.

Results and Discussion

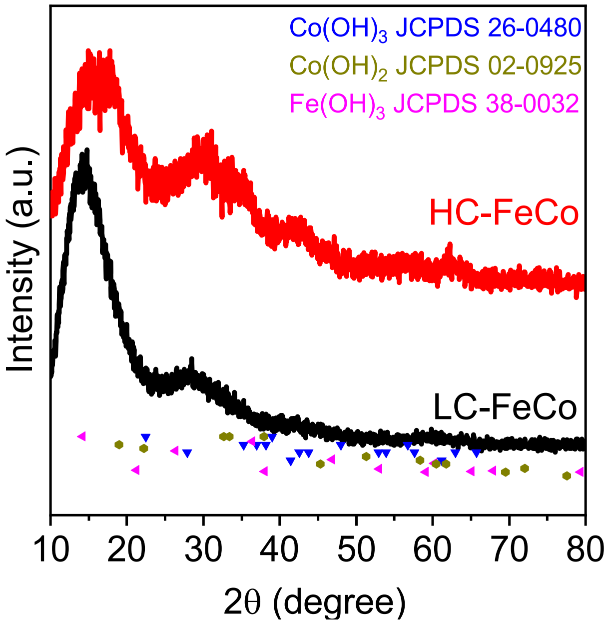

Fig. S1 XRD patterns of the LC-FeCo and HC-FeCo.

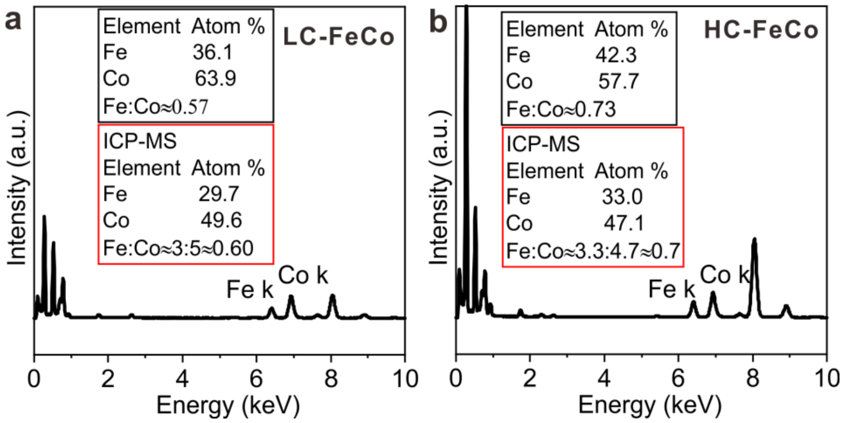

Fig. S2 EDX spectrum of the (a) LC-FeCo and (b) HC-FeCo.

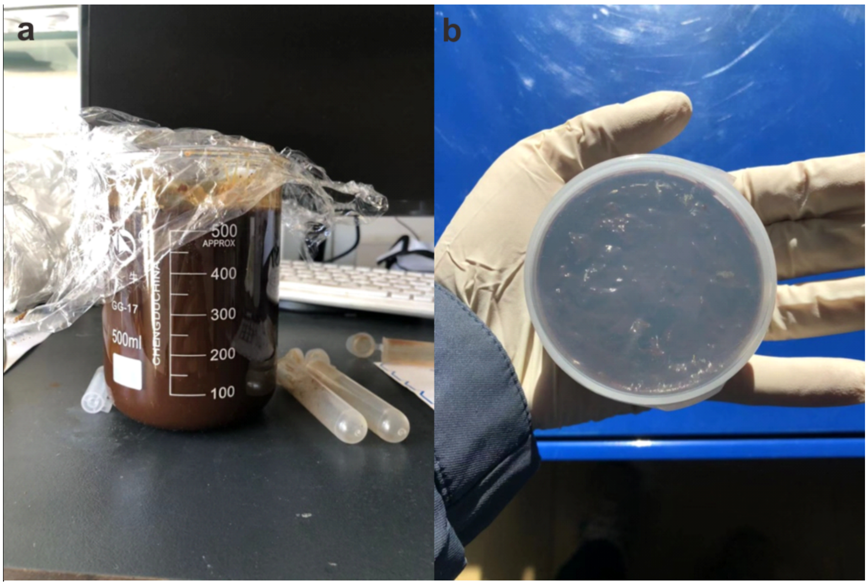

Fig. S3 (a) Pictures of the reaction solution during the large-scale preparation and (b) pictures of the product after cleaning and centrifugation.

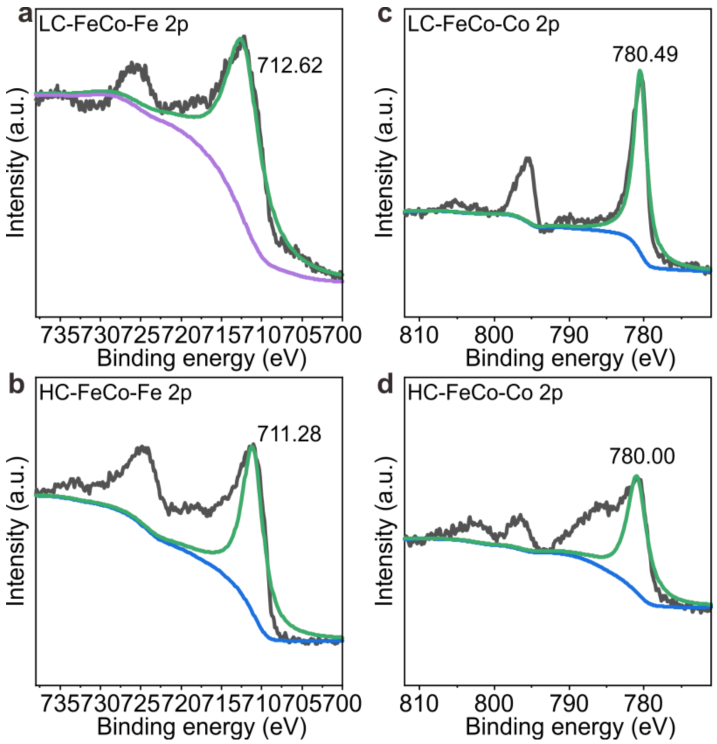

Fig. S4 XPS spectra of the LC-FeCo and HC-FeCo. Fe 2p spectra in (a) LC-FeCo and (b) HC-FeCo, Co 2p spectra in (c) LC-FeCo and (d) HC-FeCo.

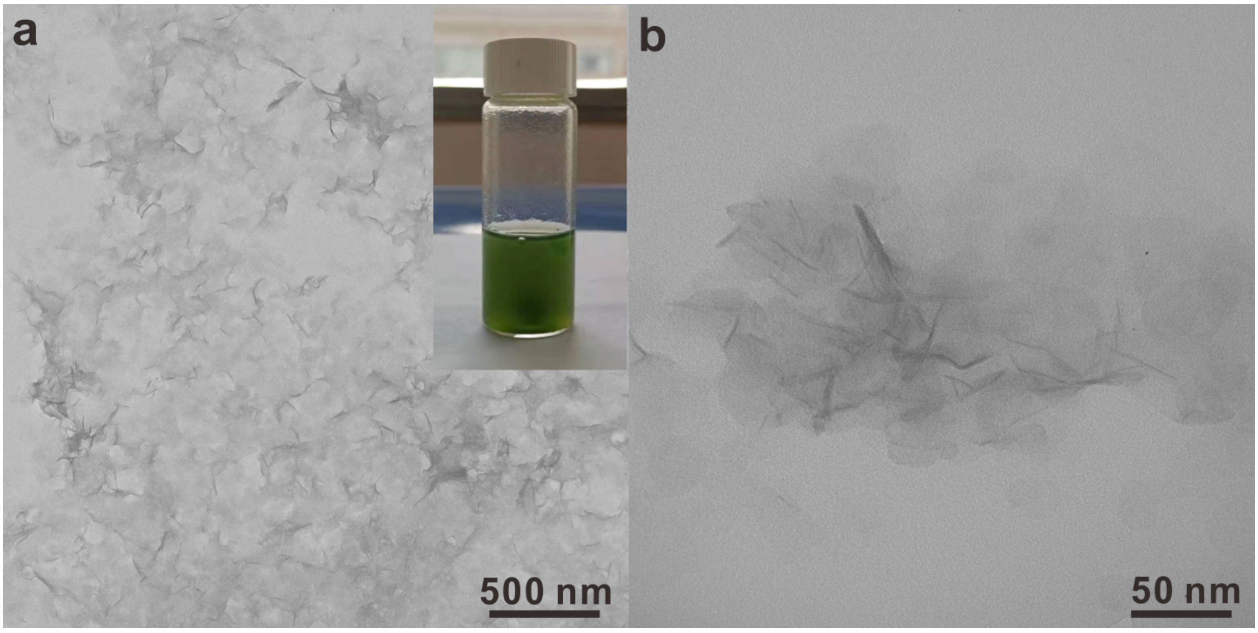

Fig. S5 Low-magnification TEM images of Co nanosheets

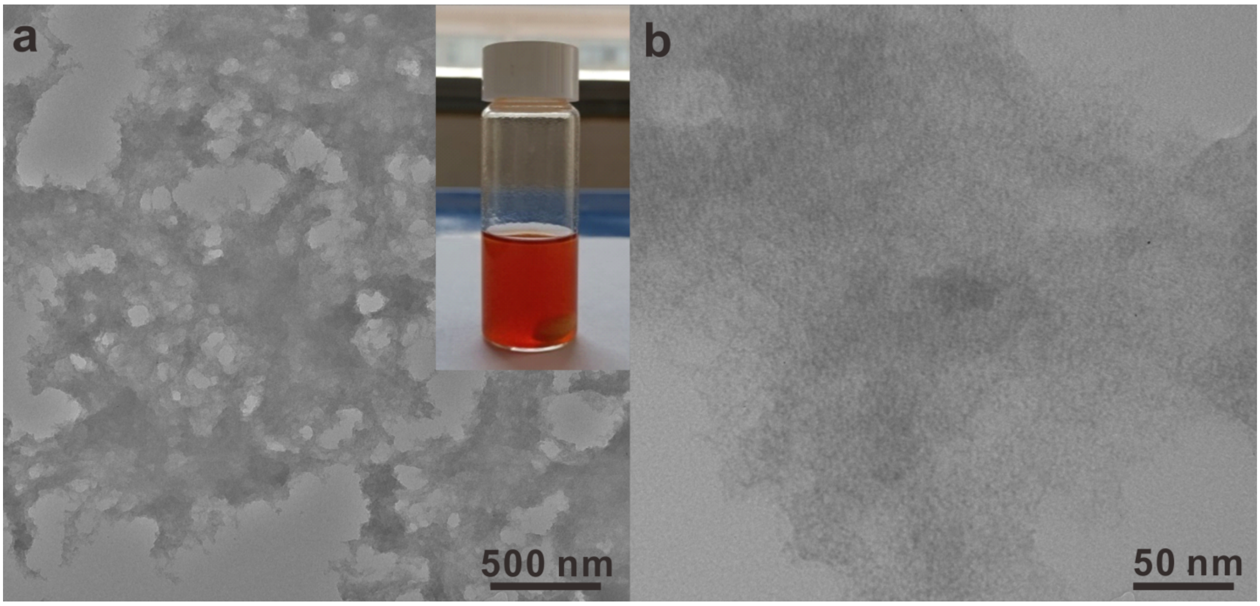

Fig. S6 Low-magnification TEM images of Fe nanosheets.

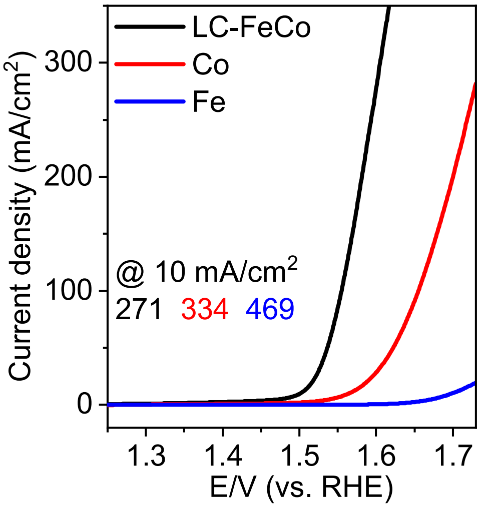

Fig. S7 OER performances of LC-FeCo, Co and Fe in KOH. The polarization curves normalized by electrode surface area. The loading amounts of all catalysts on carbon paper are 1.0 mg/cm^2.

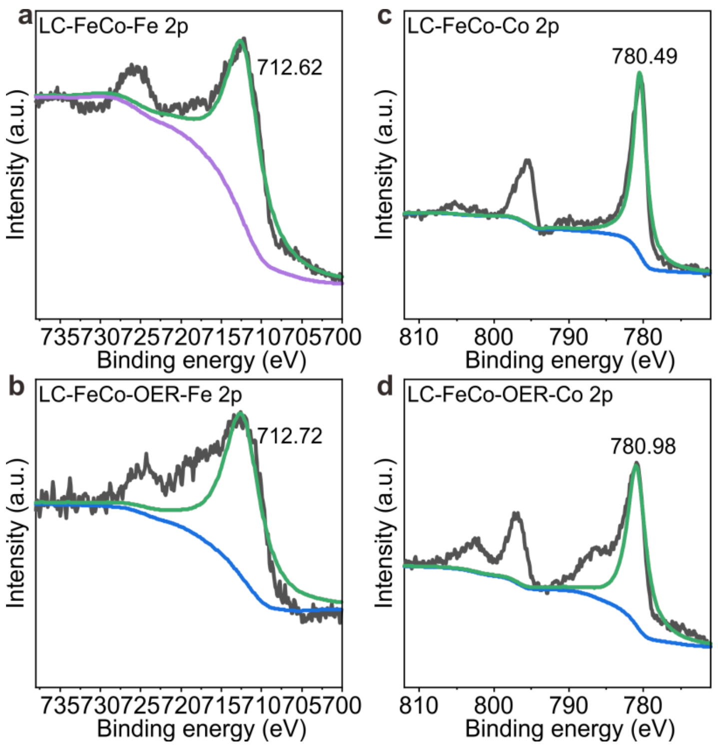

Fig. S8 XPS spectra of the LC-FeCo. Fe 2p spectra in LC-FeCo (a) before and (b) after OER, Co 2p spectra in LC-FeCo (c) before and (d) after OER.

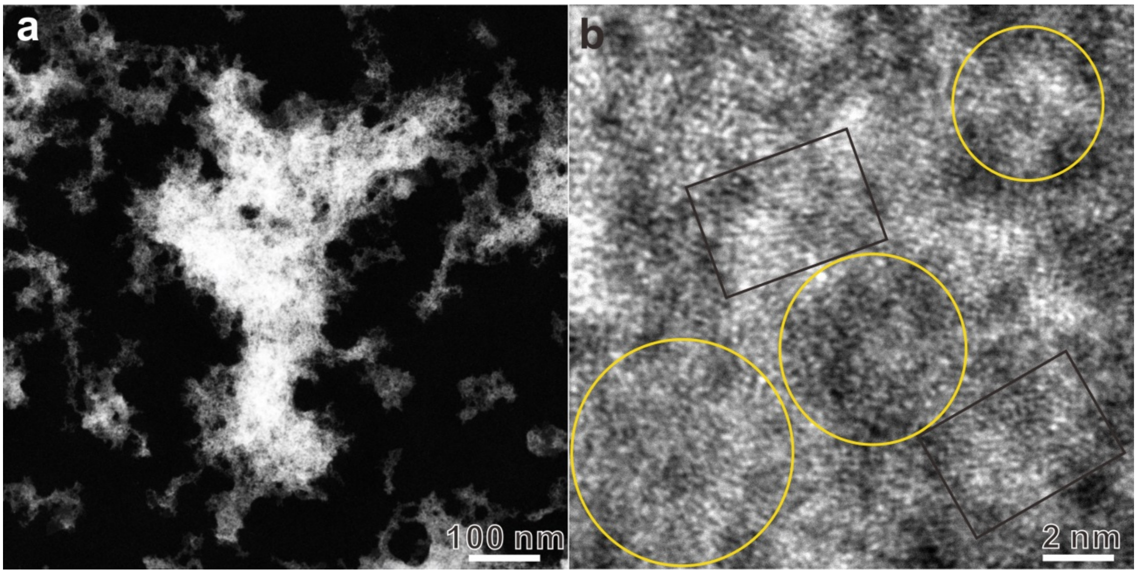

Fig.S9 (a) STEM and (b) HR-TEM images of LC-FeCo after OER The circles and quadrilaterals in Figure B highlight the amorphous and crystalline regions of the material, respectively.

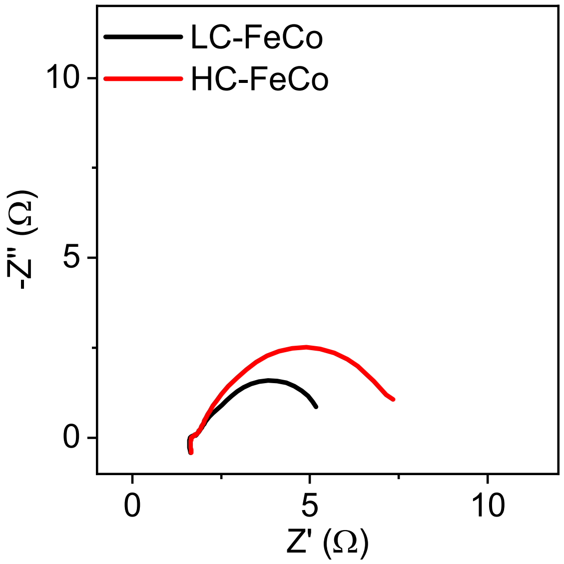

Fig. S10 Nyquist plots of LC-FeCo and HC-FeCo under the potential of 0.4 V (vs. reversible hydrogen electrode (RHE)). Z′ is the real impedance, and -Z″ is the imaginary impedance. LC-FeCo exhibited superior electrical conductivity and ion transport kinetics over HC-FeCo. The electrochemical interface was modeled by an equivalent circuit containing total Ohmic resistance of the cell (R_Ω), charge-transfer resistance (R_{ct}) and the double-layer capacitance (C_{dl}). The loading amounts of all catalysts on carbon papers were 1.0 mg/cm^2.

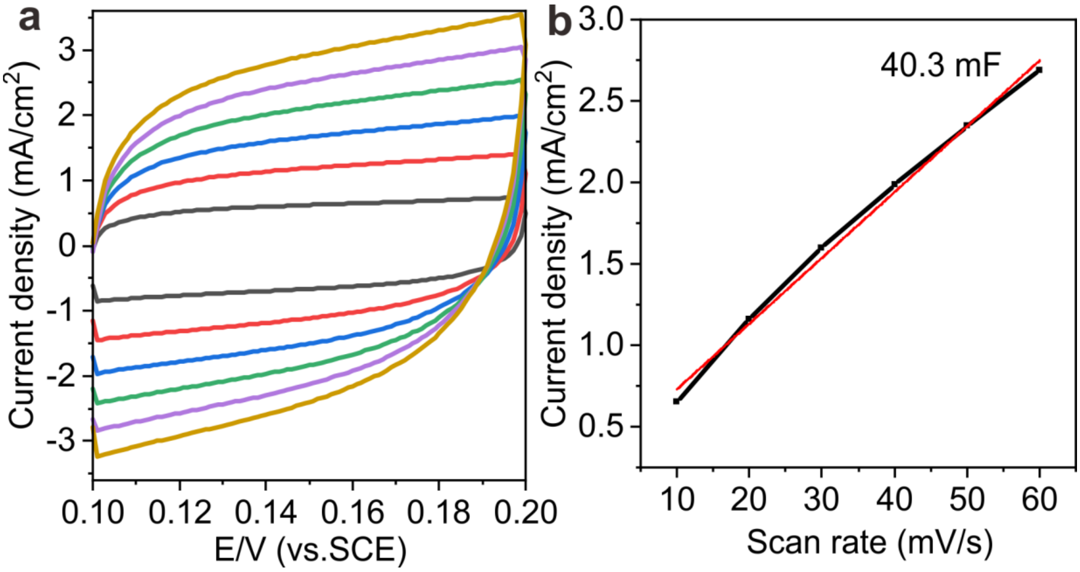

Fig. S11 (a) Cyclic voltammetry at different rates (10–60 mV/s) between 0.1 to 0.2 V (vs. SCE) and (b) the electrochemical double-layer capacitance (C_{dl}) value of LC-FeCo.

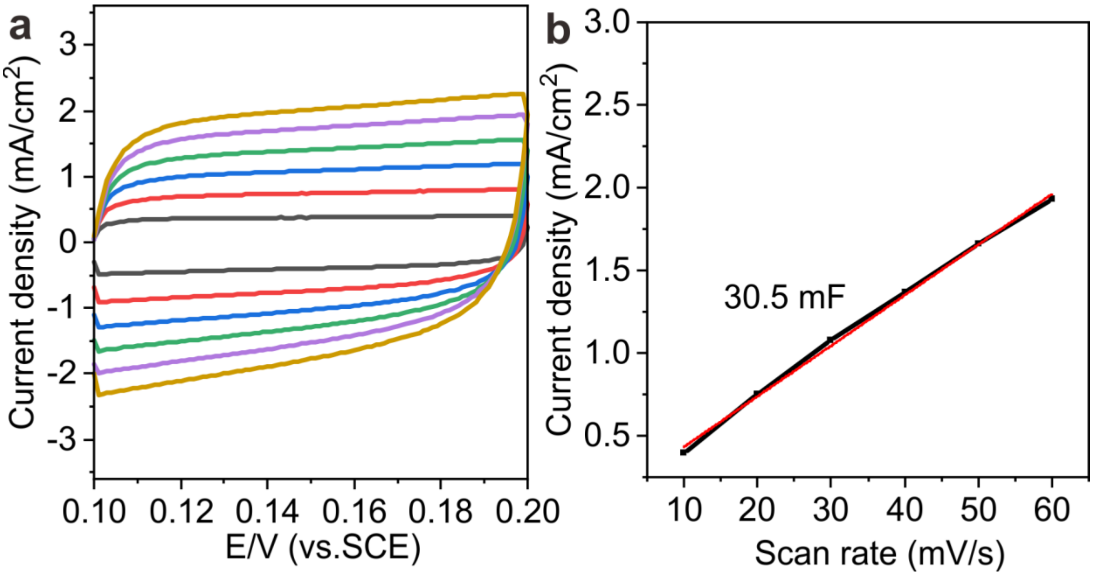

Fig. S12 (a) Cyclic voltammetry at different rates (10–60 mV/s) between 0.1 to 0.2 V (vs. SCE) and (b) the electrochemical double-layer capacitance (C_{dl}) value of HC-FeCo.

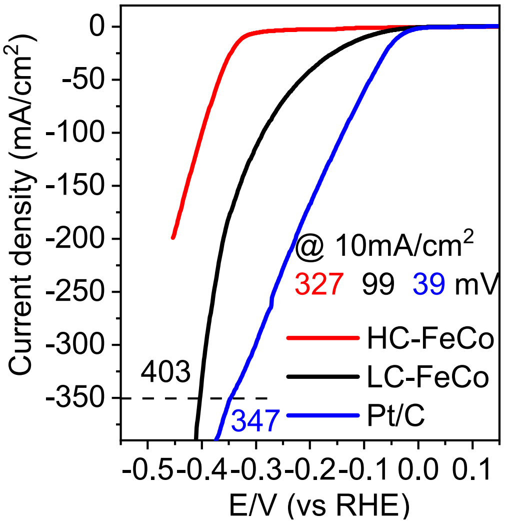

Fig. S13 HER performances of LC-FeCo, HC-FeCo and Pt/C in KOH. The polarization curves normalized by electrode surface area. The loading amounts of all catalysts on carbon paper are 1.0 mg/cm^2.

Tab. S1 Comparison of recently reported FeCo-based nanomaterials used as electrocatalysts for OER.

Catalysts	**Electrolyte**	**Overpotential (mV) @ 10 mA/cm^2**	**Tafel plots (mV/dec)**	**Ref.**
LC-FeCo	1.0 KOH	**271**	**51.6**	**This work**
$Fe_{0.33}Co_{0.67}OOH$ PNSAs/CFC	1.0 KOH	266	30	Angew. Chem. Int. Ed. 2018, 57, 2672
Fe-CoOOH/G	1.0 KOH	330	37	Adv. Energy Mater. 2017, 1602148.
CoFe LDHs-Ar	1.0 KOH	266	37.85	Angew. Chem., Int. Ed. 2017, 56, 5867.
Co_5/Fe-LDHs	1.0 KOH	285	44.6	Inorg. Chem. 2021, 60, 5252
DH-CoFe LDHs	1.0 KOH	276	40.5	Sci. China Chem., 2019, 62, 1365
Fe_1Co_1 LDHs	1.0 KOH	278	27	Nanoscale, 2019, 11, 10348–10357.
CoFe-H	1.0 KOH	280	28	Adv. Funct. Mater. 2017, 27, 1603904
Co-FeOOH/CFP	1.0 KOH	250	36	Small 2019, 15: 1901015.
FeCo-MOF-EH/NF	1.0 KOH	231	42	Angew. Chem. Int. Ed. 2020, 59, 13101–13108
α -$Co_4Fe(OH)_x$	1.0 KOH	295	52	J. Mater. Chem. A 2017, 5: 1078
CoFe LDH-F	1.0 KOH	300	40	ACS Appl. Mater. Interfaces 2016, 8, 34474−34481

Tab. S2 Detailed catalytic performance of the catalysts in our work.

Catalytic Performances		Catalyst		
		LC-FeCo	HC-FeCo	RuO_2 (Pt/C)
OER	10 mA/cm^2	271 mV	312 mV	342 mV
	200 mA/cm^2	352 mV	434 mV	--
	Tafe splos	51.6 mV/dec	75.5 mV/dec	145.8 mV/dec
HER	10 mA/cm^2	99 mV	327 mV	(39 mV)
	350 mA/cm^2	403 mV	--	(347 mV)
Water Splitting	10 mA/cm^2	1.582 V	--	1.622 V
	200 mA/cm^2	1.951 V	--	2.126 V