

Supporting Information

Highly Effective Electrochemical Water Splitting with Enhanced Electron Transfer between Ni₂Co Layered Double Hydroxide Nanosheets Dispersed on Carbon Substrate

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2.5 FT-IR measured. Response to the reviewer: The mixed sample should be taken in an appropriate amount and thoroughly blended with KBr. Subsequently, the resulting mixture should be evenly distributed into a pristine tablet press mold and subjected to pressure using a hand press for approximately 30 seconds, yielding a visually stunning transparent sheet of sample. Following this, the sheet should be carefully positioned within the test chamber of the infrared spectrometer, starting with measuring the blank background before proceeding to capture the exquisite infrared spectrum of the sample by precisely aligning it along the optical path.

2.6 Overall water splitting tested. The overall water splitting test was conducted in a two-electrode electrolytic cell containing 1 M KOH. The anode and cathode consisted of $\text{Ni}_x\text{Co}_y\text{-LDH@C}$ and commercial Pt/C-modified glass carbon electrodes (diameter 0.196 cm^2), respectively. The catalyst load was set at 2 mg cm^{-2} . For the durability test of the assembled cell, $\text{Ni}_x\text{Co}_y\text{-LDH@C}$ served as the electrode base with a catalyst load of 2 mg cm^{-2} , while maintaining a battery voltage of 1.63 V. A comparison sample was tested under identical conditions using an electrolytic cell assembled with standard IrO_2 and Pt/C catalysts having the same load.

2.7 DFT calculation performed. The calculations were carried out using density functional theory with the PBE form of generalized gradient approximation functional (GGA). 1 The Vienna ab-initio simulation package (VASP) 2-5 was employed. The plane wave energy cutoff was set as 400 eV. 3 The Fermi scheme was employed for electron occupancy with an energy smearing of 0.1 eV. The first Brillouin zone was sampled in the Monkhorst–Pack grid. 6 The $3\times3\times1$ k-point mesh for the surface calculation. The energy (converged to $1.0 \times 10^{-6}\text{ eV/atom}$) and force (converged to 0.01 eV/\AA) were set as the convergence criterion for geometry optimization. The spin polarization was considered in all calculation.

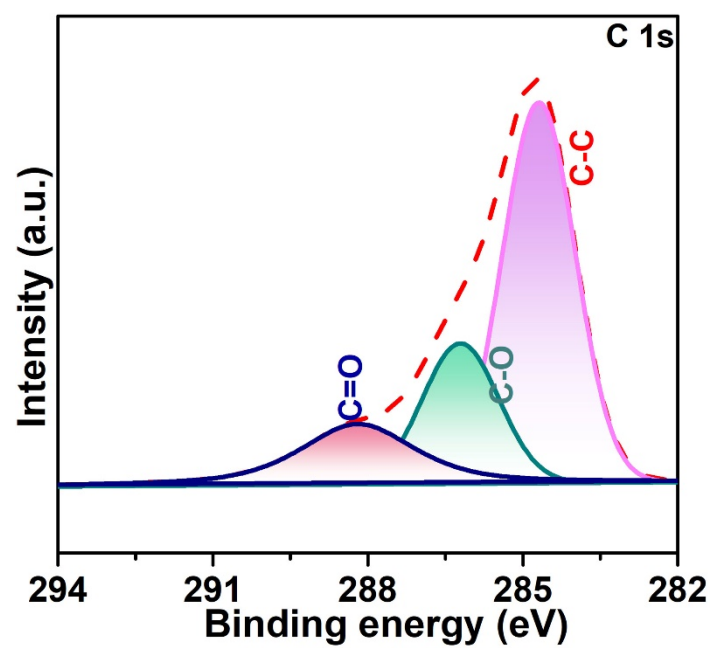


Figure S1 Fitted C_{1s} XPS spectra.

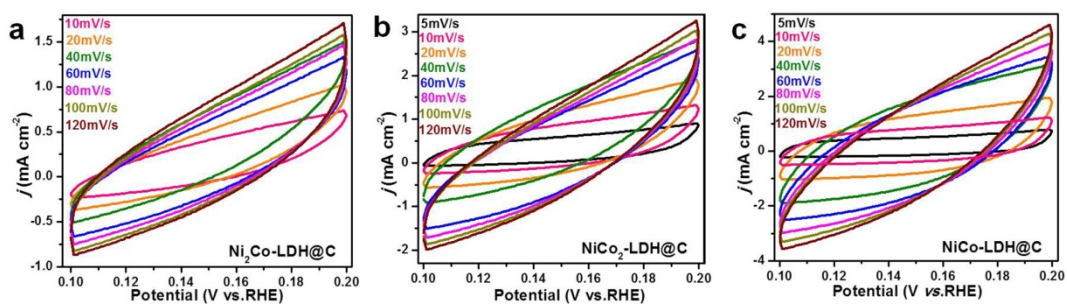


Figure S2 Cyclic voltammograms of (a) Ni₂Co-LDH@C; (b) NiCo₂-LDH@C and (c) NiCo-LDH@C, scan rates of 5, 10, 20, 40, 60, 80, 100 and 120 mV s⁻¹.

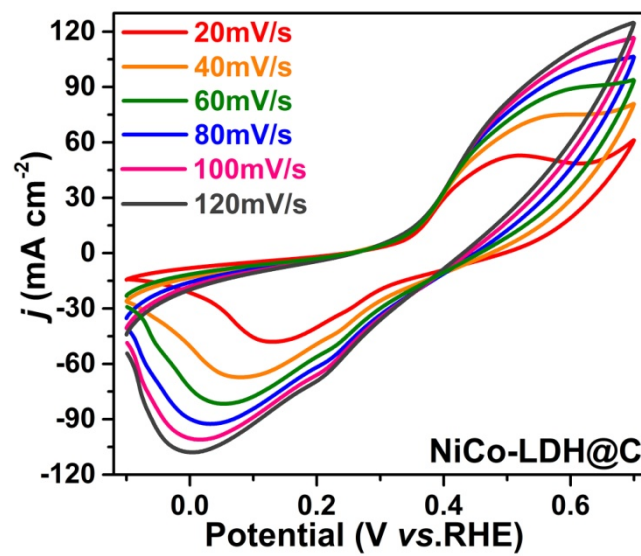


Figure S3 Cyclic voltammograms of NiCo-LDH@C at scan rates of 20, 40, 60, 80, 100 and 120 mV s^{-1} to calculate the active site density and TOF.

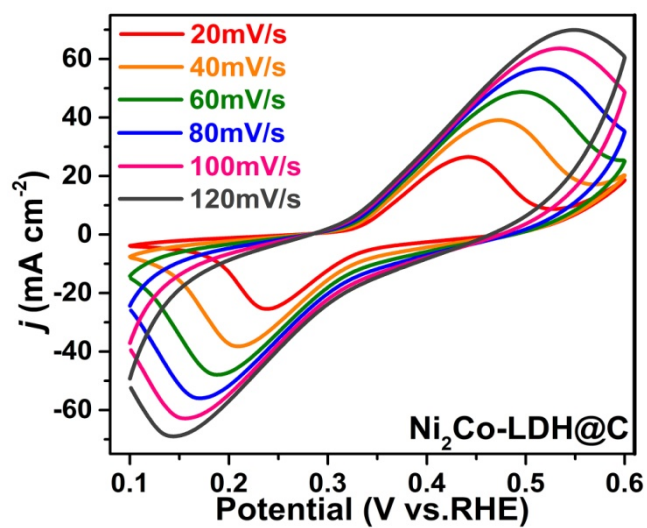


Figure S4 Cyclic voltammograms of $\text{Ni}_2\text{Co-LDH@C}$ at scan rates of 20, 40, 60, 80, 100 and 120 mV s^{-1} to calculate the active site density and TOF.

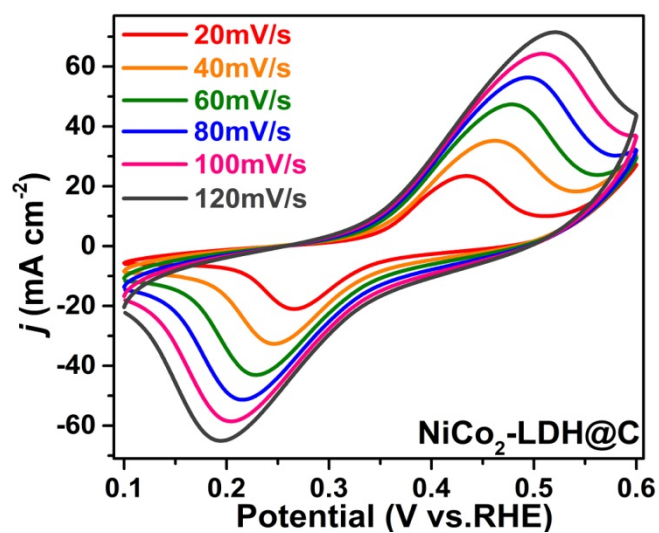


Figure S5 Cyclic voltammograms of NiCo₂-LDH@C at scan rates of 20, 40, 60, 80, 100 and 120 mV s⁻¹ to calculate the active site density and TOF.

Table. S1 Composition analysis on the Ni_xCo_y-LDH@C catalyst using XPS

Ni_xCo_y-LDH@C		Ni	Co	O	C
Atomic %	NiCo-LDH@C	2.14	1.54	46.78	49.54
	Ni ₂ Co-LDH@C	5.43	2.45	48.7	37.98
	NiCo ₂ -LDH@C	2.9	4.86	56.31	35.93

Table. S2 Composition analysis on the Ni_xCo_y-LDH@C catalyst using EDS

	Ni_xCo_y-LDH@C	Ni	Co	C
	NiCo-LDH@C	27.16	28.56	44.29
Wt %	Ni ₂ Co-LDH@C	40.98	18.37	40.65
	NiCo ₂ -LDH@C	15.98	33.17	50.85

Table.S3 The HER performances of the as-prepared electrode and other electrodes with electrocatalysts in 1.0 M KOH.

Catalyst	Electrolyte	j (mAcm ⁻²)	η (mV)	Tafel (mV·dec ⁻¹)	Ref.
Ni ₂ Co-LDH@C	1M KOH	10	167.6	113.9	This work
CoP/CNFs	1M KOH	10	225	100.8	[1]
Co ₉ S ₈ -Ni ₃ S ₂ -CNTs/NF	1M KOH	10	243	137	[2]
Co/N-CNF-800	1M KOH	10	241	132	[3]
Ni _{0.09} Co _{2.91} O ₄ /Ti ₃ C ₂ T _x -HT	1M KOH	10	210	106	[4]
Co ₂ P/Ni ₂ P/CNT-3	1M KOH	10	202	57.95	[5]

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Table.S4 The OER performances of the as-prepared electrode and other electrodes with electrocatalysts in 1.0 M KOH.

Catalyst	Electrolyte	j (mAcm ⁻²)	η (mV)	Tafel (mV·dec ⁻¹)	Ref.
Ni ₂ Co-LDH@C	1M KOH	10	183.4	108.27	This work
CeO _{2-x} /NiFe-LDH	1M KOH	10	216	74.1	[1]
CoNiRu _{0.5} SSe	1M KOH	10	261	52.2	[2]
Chestnut burr-A	1M KOH	10	235	55	[3]
C-RuO ₂ -RuSe-5	1M KOH	10	212	49.5	[4]
(Fe,Co)OOH/MI	1M KOH	10	230	73	[5]
MnSe@MWCNT	1M KOH	10	290	54.76	[6]
ENWs-FeNi-C ₂ O ₄	1M KOH	10	225	54.5	[7]
Co@NiFe-LDH	1M KOH	10	253	44	[8]
CFP-H3	1M KOH	10	251.9	39.3	[9]
R-CoSeO ₄	1M KOH	10	265	78.04	[10]

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Table.S5 OER-TOF determined at 183.4 mV (vs RHE).

Electrodes	OER-TOF ($\frac{O_2/s}{surface\ site}$)
NiCo-LDH@C	0.0103
Ni ₂ Co-LDH@C	0.0597
NiCo ₂ -LDH@C	0.0216

Table.S6 HER-TOF determined at 167.6 mV (vs RHE).

Electrodes	OER-TOF ($\frac{H_2/s}{surface\ site}$)
NiCo-LDH@C	0.0408
NiCo₂-LDH@C	0.0256
Ni₂Co-LDH@C	0.0615

Table S7. The overall performances of the as-prepared electrode and other electrodes with electrocatalysts in 1.0 M KOH.

Catalyst	Electrolyte	j (mA cm ⁻²)	η (V)	Ref.
Ni ₂ Co-LDH@C	1M KOH	10	1.63	This work
Co ₃ S ₄ /CeO ₂ -CF	1M KOH	10	1.64	[1]
Co ₉ S ₈ -Ni ₃ S ₂ -CNTs/NF	1M KOH	10	1.65	[2]
CFP-H3	1M KOH	10	1.75	[3]
Co/N-CNF-800	1M KOH	10	1.8	[4]
CoP/CNFs	1M KOH	10	1.65	[5]
v-NiS ₂ /CeO ₂ HSs	1 M KOH	10	1.64	[6]

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