# **Supporting Information**

# Interfacial synergistic regulation of MXene-composited nickel-cobalt double hydroxide for high-performance supercapacitors

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## Experimental

1.1 Preparation of negative electrode materials

Mix activated carbon (AC), carbon black (CB), and polytetrafluoroethylene (PTFE) in ethanol at a weight ratio of 8:1:1. Then, press the mixture into thin sheets on a glass plate and cut them into  $1 \times 1$  cm<sup>2</sup> square pieces. Stack these square pieces with stainless steel mesh and press them together under 8 MPa pressure to form the negative electrode sheets. The mass of the activated carbon electrode material on each prepared negative electrode sheet is approximately 3.0 mg cm<sup>-2</sup>.

### 1.2 Material characterization

The phase structure of the samples was determined using X-ray diffraction (XRD) with Cu K $\alpha$  radiation ( $\lambda$  = 1.54178 Å) on a Rigaku diffractometer operating at 50 kV and 200 mA. The XRD scans were performed in the 2 $\theta$  range from 10° to 80°, at a scanning rate of 5° min<sup>-1</sup>. The morphology and structure of the samples were examined using

a HITACHI SU020 scanning electron microscope (SEM) at an accelerating voltage of 3 kV. Transmission electron microscopy (TEM) was conducted using a Philips FEI Tecnai G2S-Twin microscope equipped with a 200 kV field emission gun to further investigate the material's morphology. The elemental composition and oxidation states of the elements were analyzed using X-ray photoelectron spectroscopy (XPS) on a Thermo Fisher Scientific ESCALAB 250Xi system, with an Al-K $\alpha$  (1486.6 eV) X-ray source. The pore size distribution of the samples was measured using nitrogen adsorption-desorption isotherms at 77 K on a Quantachrome Autosorb-IQ-Mplx system.

#### 1.3 Electrochemical testing

The electrochemical performance was evaluated using a CHI 660E electrochemical workstation (CHI Instruments Inc., Shanghai, China). The prepared positive electrode materials were used as the working electrode, with a platinum electrode as the counter electrode and a Hg/HgO electrode as the reference, in a 6 M KOH electrolyte, forming a three-electrode system. Cyclic voltammetry (CV) was performed over a potential range of 0 to 0.65 V. Galvanostatic charge-discharge (GCD) measurements were conducted within a potential range of 0 to 0.5 V. Electrochemical impedance spectroscopy (EIS) was performed at open-circuit potential, with an amplitude of 5 mV over a frequency range from 0.01 Hz to 100 kHz. For further electrochemical performance evaluation, the prepared positive electrode materials and activated carbon (AC) were assembled into asymmetric supercapacitors.

#### 1.4 Electrochemical evaluation

The electrochemical performance of the materials was assessed using a threeelectrode system, comprising a platinum counter electrode, a Hg/HgO reference electrode, and the prepared materials as the working electrode. A 6 M KOH solution was used as the electrolyte. All samples were tested under identical conditions to ensure comparability, with the material composition being the only variable.

To evaluate the practical application of the Ni<sub>1</sub>Co<sub>2</sub>-LDH/MXene composite, it was paired with activated carbon (AC) to form an asymmetric supercapacitor (ASC) for further electrochemical testing. Ni<sub>1</sub>Co<sub>2</sub>-LDH/MXene served as the positive electrode and AC as the negative electrode, with 6 M KOH as the electrolyte, creating the device configuration  $Ni_1Co_2$ -LDH/MXene//AC. The same assembly process was applied to Ni1Co2-LDH, which was designated as  $Ni_1Co_2$ -LDH//AC.

The specific capacitance can be calculated using the following formula:

$$C_m = \frac{I * t}{\Delta V * m} \tag{S1}$$

where  $C_m$  is the specific capacitance of the electrode material, I is the discharge current, t is the discharge time,  $\Delta V$  is the potential window, and m is the mass of the active substance on the electrode material<sup>1</sup>.

The investigation of charge storage mechanisms in materials can be achieved by analyzing the relationship between peak current and scan rate. This relationship is expressed as:

$$Log(i) = blog(v) + log(a)$$
(S2)

where i represents the peak current of the anode or cathode, v is the scan rate, and a and b are constants. A b value of 0.5 indicates that the electrochemical process is predominantly controlled by diffusion, whereas a b value of 1 signifies control by capacitive processes<sup>2</sup>.

$$i(v) = {k_1 v} + {k_2 v}^{\frac{1}{2}}$$
(S3)

$$i(v) v^{2} = k_{1}v^{2} + k_{2}$$
 (S4)

where v represents the scan rate, and  $k_1v$  and  $k_2v^2$  denote capacitive and  $\frac{1}{\sqrt{2}} \sqrt{\frac{1}{2}}$  diffusion-controlled processes, respectively. By plotting linear fits of  $v^2$  versus  $i(v)v^2$  at different potentials, the values of  $k_1$  and  $k_2$  can be determined, revealing the contributions of each mechanism<sup>3</sup>.

#### 1.5 DFT simulation details

All spin-polarized density functional theory (DFT) calculations were performed using the

Vienna Ab-initio Simulation Package (VASP). The generalized gradient approximation following the scheme of the Perdew, Burke and Ernzerrof functional was chosen to describe the exchange-correlation interactions, while the projector augmented-wave4 method was used to represent core-valence electron interactions. Gaussian smearing was used for the electronic states of our models and the width was set to 0.05 eV. The Brillouin zone was sampled using Monkhorst-Pack meshes of 2 \* 2 \* 1. The DFT-D3 method of Grimme was used to describe van der Waals interactions to obtain a better description of the interactions between molecules. The kinetic cut-off energy was set to 500 eV. The atomic geometric structures were fully relaxed, and the energy and force convergence tolerance were set to below 10<sup>-0.6</sup> eV per cell and 0.02 eV A<sup>-1</sup>.



Fig.S1. TEM images of (a) (b) Ni<sub>1</sub>Co<sub>2</sub>-LDH/MXene, (c) (d) Ni<sub>1</sub>Co<sub>2</sub>-LDH.



**Fig. S2.** The nitrogen adsorption-desorption isotherm of (a) Ni<sub>1</sub>Co<sub>2</sub>-LDH/MXene, (b) Ni<sub>1</sub>Co<sub>2</sub>-LDH, (c)MXene. The pore size distribution curves of (d) Ni<sub>1</sub>Co<sub>2</sub>-LDH/MXene, (e) Ni<sub>1</sub>Co<sub>2</sub>-LDH, (f)MXene.

Note: To further assess whether MXene reduces the aggregation of Ni<sub>1</sub>Co<sub>2</sub>-LDH and enhances the material's specific surface area, we conducted N<sub>2</sub> adsorptiondesorption tests to measure the specific surface area and pore size distribution. Fig. S2(a) presents the N<sub>2</sub> adsorption-desorption isotherms of Ni<sub>1</sub>Co<sub>2</sub>-LDH/MXene. The calculated specific surface area of Ni<sub>1</sub>Co<sub>2</sub>-LDH/MXene is 141.95 m<sup>2</sup> g<sup>-1</sup>, reflecting a 44.8% increase compared to the specific surface area of Ni<sub>1</sub>Co<sub>2</sub>-LDH (98.06 m<sup>2</sup> g<sup>-1</sup>), as shown in Fig. S2(b). According to literature, the optimal pore size distribution for supercapacitor electrode materials ranges between 2 and 5 nm. The data indicate that the pore size of Ni<sub>1</sub>Co<sub>2</sub>-LDH/MXene is concentrated between 3.4 and 5 nm (Fig. S2(df)).



Fig. S3. The XRD patterns of MXene,  $Ni_1Co_2$ -LDH and  $Ni_1Co_2$ -LDH/MXene.

Note: MXene displays distinct characteristic peaks at  $6.9^{\circ}, 18.9^{\circ}, 30.5^{\circ}, 35.6^{\circ}$  and  $62.3^{\circ}$ , corresponding to the (002), (006), (008), (111) and (110) planes, respectively. The characteristic peaks of Ni<sub>1</sub>Co<sub>2</sub>-LDH are observed at 11.7^{\circ}, 23.6^{\circ}, 34.1^{\circ}, 38.7^{\circ}, and 62.3^{\circ}, corresponding to the (003), (006), (012), (015), and (110) planes.



Fig.S4. The respective high resolution XPS spectra of Ni<sub>1</sub>Co<sub>2</sub>-LDH /MXene. (a) XPS survey scan spectrum (b) Ti 2p XPS spectrum; (c) Co 2p XPS spectrum; (d) Ni 2p XPS spectrum.



Fig. S5. The respective high resolution XPS spectra of MXene. (a) XPS survey scan spectrum (b) Ti 2p XPS spectrum; (c) C 1s XPS spectrum; (d) C 1s XPS spectrum.

Note: The XPS spectra of the main elements in MXene are shown in Fig. S5. Fig. S5(a) identifies the primary elements in MXene as F (684 eV), O (530 eV), Ti (458 eV), and C (285 eV). Fig. S5(b) presents the Ti 2p spectrum, displaying two distinct pairs of peaks corresponding to Ti  $2p_{3/2}$  and Ti  $2p_{1/2}$ . Detailed peak fitting reveals eight peaks, including Ti-C peaks at 454.88 eV and 461.68 eV, and Ti-O characteristic peaks at 459.68 eV and 466.28 eV. Peaks associated with Ti<sup>2+</sup> and Ti<sup>3+</sup> are located at 455.98/463.58 eV and 458.78/464.98 eV, respectively. Fig. S5(c) shows that the C 1s spectrum is fitted to four peaks at 287.85 eV, 285.80 eV, 284.35 eV, and 281.05 eV, corresponding to O=C-O, C-O, C-C, and C-Ti, respectively. Similarly, the O 1s spectrum is deconvoluted into four peaks at 533.10 eV, 531.65 eV, 530.95 eV, and 529.25 eV, which correspond to C-OH, Ti-O, Ti-O-Ti, and adsorbed O species (Fig. S5(d)).



**Fig. S6.** respective high resolution XPS spectra of Ni<sub>1</sub>Co<sub>2</sub>-LDH. (a) XPS survey scan spectrum (b) Co 2p XPS spectrum; (c)Ni 2p XPS spectrum; (d) O 1s XPS spectrum.

Note: The XPS spectra of the main elements in Ni<sub>1</sub>Co<sub>2</sub>-LDH are presented in Fig. S6. Fig. S6(a) shows the primary elements in Ni<sub>1</sub>Co<sub>2</sub>-LDH as Ni (856 eV), Co (781 eV), O (531 eV), and C (285 eV). Fig. S6(b) depicts the Co 2p spectrum, which includes four distinct peaks, two of which are satellite peaks at 786.01 eV and 801.90 eV, along with the Co  $2p_{3/2}$  and Co  $2p_{1/2}$  peaks. These peaks can be further deconvoluted into Co<sup>2+</sup> (781.90/797.69 eV) and Co<sup>3+</sup> (779.91/795.89 eV). Similarly, the Ni 2p spectrum reveals two satellite peaks (861.89/879.30 eV), and the Ni  $2p_{3/2}$  and Ni  $2p_{1/2}$  peaks, which can be further resolved into Ni<sup>2+</sup> (855.10/872.71 eV) and Ni<sup>3+</sup> (856.51/873.90 eV), resulting in four main peaks (Fig. S6(c)). The O 1s spectrum in Fig. S6(d) is fitted into three peaks at 531.85 eV, 530.85 eV, and 530.25 eV, corresponding to O<sub>L</sub>, O<sub>V</sub>, and O<sub>W</sub>, respectively.



Fig. S7. EPR spectra of Ni<sub>1</sub>Co<sub>2</sub>-LDH/MXene and Ni<sub>1</sub>Co<sub>2</sub>-LDH.



**Fig. S8.** Electrochemical test results of Ni<sub>1</sub>Co<sub>2</sub>-LDH using a three-electrode system in a 6 M KOH electrolyte. (a) CV curves at different scan rates from 5 to 100 mV s<sup>-1</sup>. (b) GCD curves of Ni<sub>1</sub>Co<sub>2</sub>-LDH electrode at different current densities (c) Percent of capacitive and diffusive contribution at different scan rates of Ni<sub>1</sub>Co<sub>2</sub>-LDH (d) Bar plot of the gravimetric capacitance at different current densities from 1 to 15 A  $g^{-1}$ .

Note: The electrochemical performance of Ni<sub>1</sub>Co<sub>2</sub>-LDH is illustrated in Fig. S8. Fig. S8(a) presents the CV curves of Ni<sub>1</sub>Co<sub>2</sub>-LDH at scan rates between 5 and 100 mV s<sup>-1</sup>, where distinct redox peaks are observed, indicating its inherent pseudocapacitive behavior. Fig. S8(b) shows the GCD curves at current densities ranging from 1 to 15 A g<sup>-1</sup>. At 1 A g<sup>-1</sup>, Ni<sub>1</sub>Co<sub>2</sub>-LDH exhibits a specific capacitance of 984 F g<sup>-1</sup>, 92.99% lower than that of Ni<sub>1</sub>Co<sub>2</sub>-LDH /MXene. Fig. S8(c) illustrates the changes in capacitive and diffusive contributions across different scan rates, revealing that Ni<sub>1</sub>Co<sub>2</sub>-LDH has a lower pseudocapacitive contribution compared to Ni<sub>1</sub>Co<sub>2</sub>-LDH /MXene. As shown in Fig. S8(d), the specific capacitance of Ni<sub>1</sub>Co<sub>2</sub>-LDH drops from 984 F g<sup>-1</sup> at 1 A g<sup>-1</sup> to 645 F g<sup>-1</sup> at 15 A g<sup>-1</sup>, with a capacitance retention rate of just 65.54%, which is 110.23% lower than that of Ni<sub>1</sub>Co<sub>2</sub>-LDH /MXene under identical conditions.



Fig. S9. EIS fitting curves for six materials

Table S1. Impedance comparison of six materials

Electrode material	R <sub>s</sub> (Ω)	R <sub>ct</sub> (Ω)
Ni <sub>1</sub> Co <sub>2</sub> -LDH/MXene	0.60	0.41
Ni <sub>1.5</sub> Co <sub>1.5</sub> -LDH/MXene	0.65	0.47
Ni <sub>2</sub> Co <sub>1</sub> -LDH/MXene	0.67	0.44

Co(OH) <sub>2</sub> /MXene	0.76	0.57
Ni(OH) <sub>2</sub> /MXene	0.78	0.52
Ni <sub>1</sub> Co <sub>2</sub> -LDH	1.76	1.32



Fig. S10. Optimized structure model of (a) Ni(OH)<sub>2</sub>/MXene, (b) Ni<sub>2</sub>Co<sub>1</sub>-LDH/MXene and Ni<sub>1.5</sub>Co<sub>1.5</sub>-LDH/MXene



Fig. S11. The density of states of (a) Ni(OH)\_2/MXene  $\sim$  (b) Ni\_2Co\_1-LDH/MXene and Ni\_1.5Co\_1.5-LDH/MXene



**Fig. S12.** Electrochemical performance of Ni<sub>1</sub>Co<sub>2</sub> composites in asymmetric supercapacitor. (a) CV curves of the ASC at various scan rates. (b) GCD curves at diverse current densities for the ASC. (c) comparison of CV curves for Ni<sub>1</sub>Co<sub>2</sub>-LDH/MXene//AC and Ni<sub>1</sub>Co<sub>2</sub>-LDH//AC at a scan rate of 50 mV s<sup>-1</sup>. (d) comparison GCD curves of Ni<sub>1</sub>Co<sub>2</sub>-LDH/MXene//AC and Ni<sub>1</sub>Co<sub>2</sub>-LDH//AC at 1 A g<sup>-1</sup>. (e) Percent of capacitive and diffusive contribution at different scan rates of Ni<sub>1</sub>Co<sub>2</sub>-LDH. (f) capacitive contributions to the total charge storage at 100 mV s<sup>-1</sup>.

**Note:** Fig. S12(a) illustrates the variation in the CV curves of Ni<sub>1</sub>Co<sub>2</sub>-LDH//AC at scan rates ranging from 1 mV s<sup>-1</sup> to 100 mV s<sup>-1</sup>, with redox peaks evident in all curves, indicating the pseudocapacitive behavior of Ni<sub>1</sub>Co<sub>2</sub>-LDH. Fig. S12(b) presents the GCD curves of the Ni<sub>1</sub>Co<sub>2</sub>-LDH//AC device, from which a specific capacitance of 209.4 F g<sup>-1</sup> is calculated, representing a 14.6% decrease compared to the Ni<sub>1</sub>Co<sub>2</sub>-LDH//MXene//AC device under identical conditions. Fig. S12(c) contrasts the CV curves of Ni<sub>1</sub>Co<sub>2</sub>-LDH//AC and Ni<sub>1</sub>Co<sub>2</sub>-LDH//MXene//AC at a scan rate of 50 mV s<sup>-1</sup>, revealing a significantly larger CV enclosed area for the Ni<sub>1</sub>Co<sub>2</sub>-LDH//MXene//AC, signifying enhanced electrochemical performance. This observation is corroborated by the GCD curves at a current density of 1 A g<sup>-1</sup> in Fig. S10(d). Additionally, Fig. S10(e) and Fig. S10(f) depict the proportions of capacitive contributions to total charge storage at 100 mV s<sup>-1</sup>. Compared to Ni<sub>1</sub>Co<sub>2</sub>-LDH//AC exhibits a lower pseudocapacitive contribution rate.



Fig. S13. Comparison of Nyquist plots of Ni $_1Co_2\text{-LDH}/MXene//AC$  and Ni $_1Co_2\text{-LDH}//AC$ 



Fig. S14. SEM images of (a) Ni $_1$ Co $_2$ -LDH/MXene and (b) Ni $_1$ Co $_2$ -LDH after cycling.

Note: The SEM images in Fig. S14 reveal that the LDH layers within the Ni<sub>1</sub>Co<sub>2</sub>-LDH/MXene structure exhibit significant thickening following charge-discharge cycling. This phenomenon is attributed to the repeated dissolution-deposition process occurring throughout the cycles, although the overall structural integrity remains intact and no collapse or major damage is observed (Fig. S14(a)). However, Fig. S14(b) shows clear signs of deformation in the LDH structure, with the layers beginning to aggregate, which contributes to the observed deterioration in electrochemical performance.

Table S2. A comprehensive comparison of the specific capacitance, potential window, and other performance metrics of the  $Ni_1Co_2$ -LDH/MXene//AC device with other relevant reported materials.

Electrode material	Electrolyte	Operating voltage (V)	Specific capacitance (F g <sup>-1</sup> )	Ref
P: Ni <sub>1</sub> Co <sub>2</sub> -LDH/MXene	6 М КОН	0~1.7	240.0 (1 A g <sup>-1</sup> )	This
N: AC				work
P: Ni <sub>1</sub> Co <sub>2</sub> -LDH/MXene	6 М КОН	0~1.7	221.7 (2 A g <sup>-1</sup> )	This
N: AC				work
P: Ni <sub>1</sub> Co <sub>2</sub> -LDH/MXene		0~1.7	261.9 (3 A g <sup>-1</sup> )	This
N: AC				work
P: Ni <sub>1</sub> Co <sub>2</sub> -LDH/MXene		0~1.7	204.9 (5 A g <sup>-1</sup> )	This
N: AC				work
P: Ni <sub>1</sub> Co <sub>2</sub> -LDH/MXene		0~17		This
N: AC		0 1.7	100.2 (10 A g <sup>-</sup> )	work
P: Ni <sub>1</sub> Co <sub>2</sub> -LDH/MXene		0~17	1797 (15 A g-1)	This
N: AC		0 1.7	178.7 (15 A g <sup>-</sup> )	work
P: NiCo-LDH@NCF		0~1.5	199.0 (1 A g <sup>-1</sup> )	4
N: AC				
P: CC@NiAl-		0~1.6	146.1 (1 A g <sup>-1</sup> )	
LDH@GO@NiCo-LDH	PVA/KOH			5
N: AC				
P: NiCo-LDH		0~16	177 Ο (1 Δ σ <sup>-1</sup> )	6
N: AC	_	0 1.0	177.0 (1 A g <sup>-</sup> )	0
P: CoMo-				
LDH@CuCo <sub>2</sub> S <sub>4</sub> NWAs/N		0~1.6	193.0 (1 A g <sup>-1</sup> )	7
F	o wincom			
N: AC				
P: CuCo <sub>2</sub> O <sub>4</sub>	ΡVΑ/ΚΟΗ	0~1.1	157 8 (1 35 A g <sup>−1</sup> )	8
N: CuCo <sub>2</sub> O <sub>4</sub>				
P: Ni-Al LDH-rGO	ΡVΑ/ΚΟΗ	0~1.6	214.0 (2 A g <sup>-1</sup> )	9
N: Fe <sub>2</sub> O <sub>3</sub> -rGO				
P: graphene/Ni–Al LDH	6 М КОН	-05~05	268 (0.43 A g <sup>-1</sup> )	10
N: graphene/Ni–Al LDH				
P: NiCo-LDH/rGO	2 М КОН	0~15	226 0 (1 A g <sup>-1</sup> )	11
N: AC	2			
P: Ni@ZnCo-LDH-2	3 М КОН	0~1.5	229.1 (0.5 A g <sup>-1</sup> )	12
N: Ni@ZnCo-LDH-2				
P: MC-LDH/rGO@NCS-6	2 М КОН	0~1.7	236.3 (1 A σ <sup>−1</sup> )	13
N: AC	2 10 1011	0 1.7	230.3 (1 A g -)	

Materials	Energy	Power	Ctability		
	density	density		Ref	
		(Wh kg <sup>-1</sup> )	(W kg <sup>-1</sup> )	(cycles)	
P:	Ni <sub>1</sub> Co <sub>2</sub> -LDH/MXene (1 A g <sup>-1</sup> )	02.0	764.8	١	This
N:	AC	82.9			work
P:	Ni <sub>1</sub> Co <sub>2</sub> -LDH/MXene (2 A g <sup>-1</sup> )	78.8	1576.8	١	This
N:	AC				work
P:	Ni <sub>1</sub> Co <sub>2</sub> -LDH/MXene (3 A g <sup>-1</sup> )	76.9	2364.9	١	This
N:	AC				work
P:	Ni <sub>1</sub> Co <sub>2</sub> -LDH/MXene (5 A g <sup>-1</sup> )	73.5	3950.3	١	This
N:	AC				work
P:	Ni <sub>1</sub> Co <sub>2</sub> -LDH/MXene (10 A g <sup>-1</sup> )	70.1	5872.2	١	This
N:	AC	70.1			work
P:	Ni <sub>1</sub> Co <sub>2</sub> -LDH/MXene (15 A g <sup>-1</sup> )	66.96	7770.0	89.5%	This
N:	AC		///0.0	(40000)	work
P:	NiCo-LDH@NCF	41 E	750.0	80.4%	л
N:	AC	41.5	750.0	(10000)	4
P:	CC@NiAl-LDH@GO@NiCo-LDH	E2 0	796.7	85.7%	5
N:	AC	52.0		(1500)	
P:	NiCo-LDH	62.0	800.0	73.3%	6
N:	AC	02.9		(3000)	
P:	CoMo-		983.9	02.2%	7
	LDH@CuCo <sub>2</sub> S <sub>4</sub> NWAs/NF	52.2		92.2%	
N:	AC			(10000)	
P:	CuCo <sub>2</sub> O <sub>4</sub>	26 5	763.8	88.0%	0
N:	CuCo <sub>2</sub> O <sub>4</sub>	26.5		(2000)	ð
P:	Ni-Al LDH-rGO	26.7	800.1	90.3%	9
N:	Fe <sub>2</sub> O <sub>3</sub> -rGO	50.7		(6000)	
P:	graphene/Ni–Al LDH	27.0	570.0	١	10
N:	graphene/Ni–Al LDH	57.0			
P:	NiCo-LDH/rGO	10.0	375.2	62.2%	11
N :	AC	49.9		(10000)	11
P:	Ni@ZnCo-LDH-2	40.2	15080.0	87.5%	12
N:	Ni@ZnCo-LDH-2	40.3		(6000)	12
P:	MC-LDH/rGO@NCS-6	EE 9	856.2	100%	13
N:	AC	٥.5٥		(6000)	

**Table S3.** Comparison of the energy density, power density and stability of  $Ni_1Co_2$ -LDH/MXene in this work with those of other LDHs

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