

## ***Supporting Information for***

### **Surface defect strategy of NiCo-layered double hydroxide decorated MXene layers for durable solid-state supercapacitors**

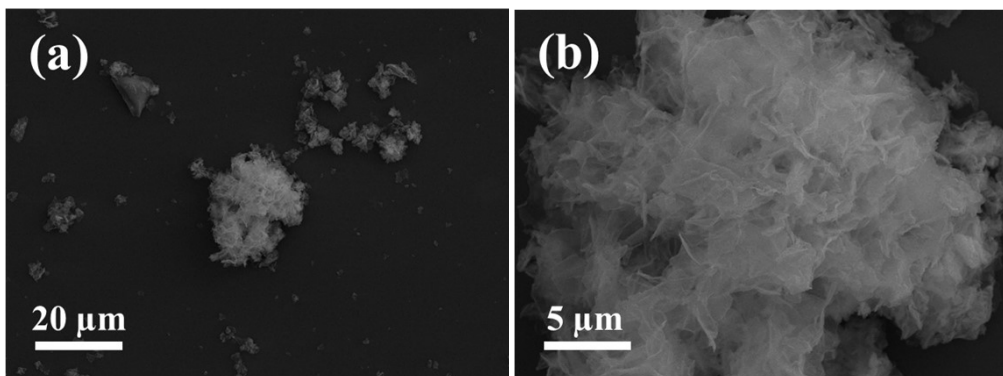
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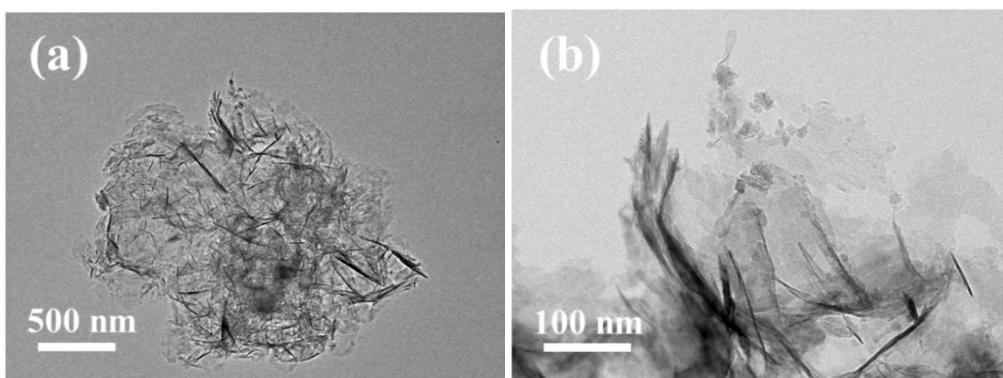
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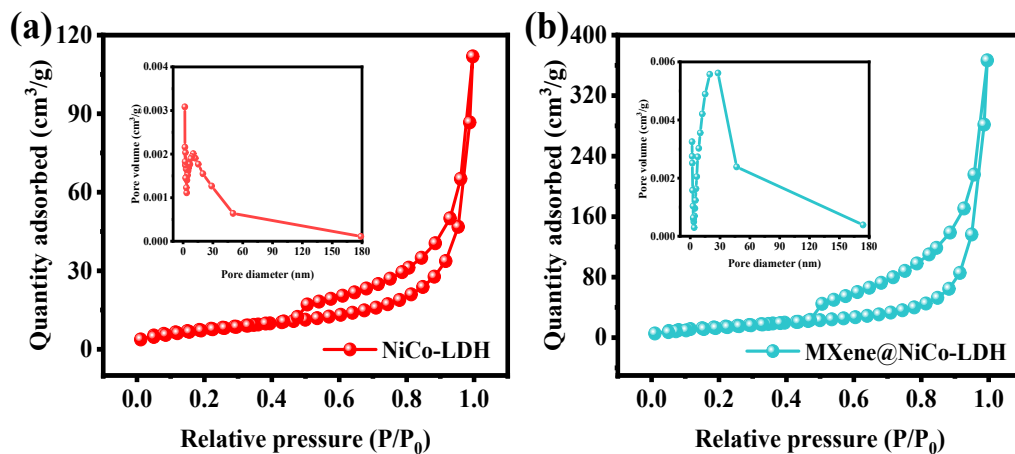
\* Corresponding author: qyouyang@hrbeu.edu.cn



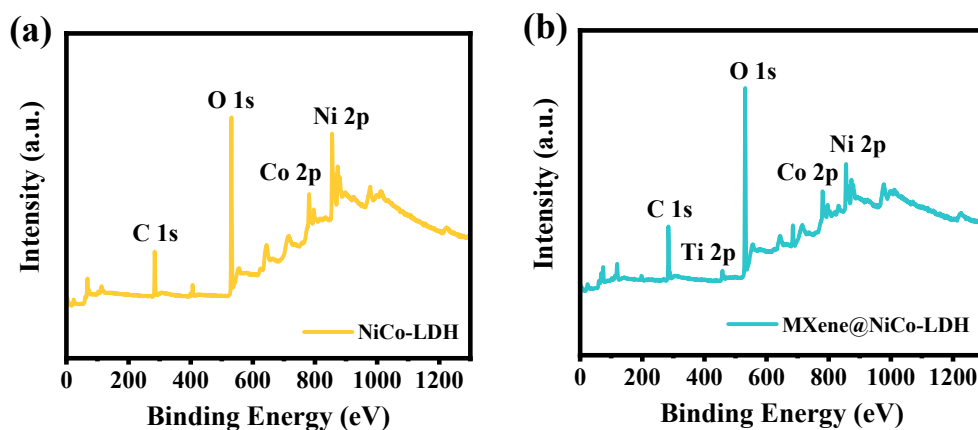
**Fig. S1** SEM images of (a,b) NiCo-LDH at different magnification.



**Fig. S2** TEM images of (a,b) NiCo-LDH at different magnification.



**Fig. S3** The BET isotherm and pore size distribution profile of (a) NiCo-LDH and (b) MXene@NiCo-LDH.



**Fig. S4** The XPS spectra of (a) NiCo-LDH and (b) MXene@NiCo-LDH samples: Survey spectrum.

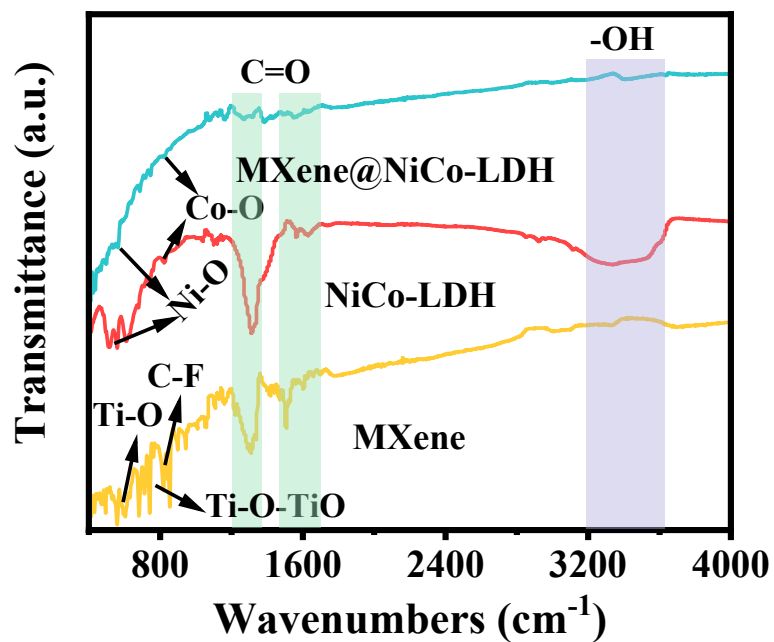


Fig. S5 FT-IR spectra of MXene, NiCo-LDH and MXene@NiCo-LDH.

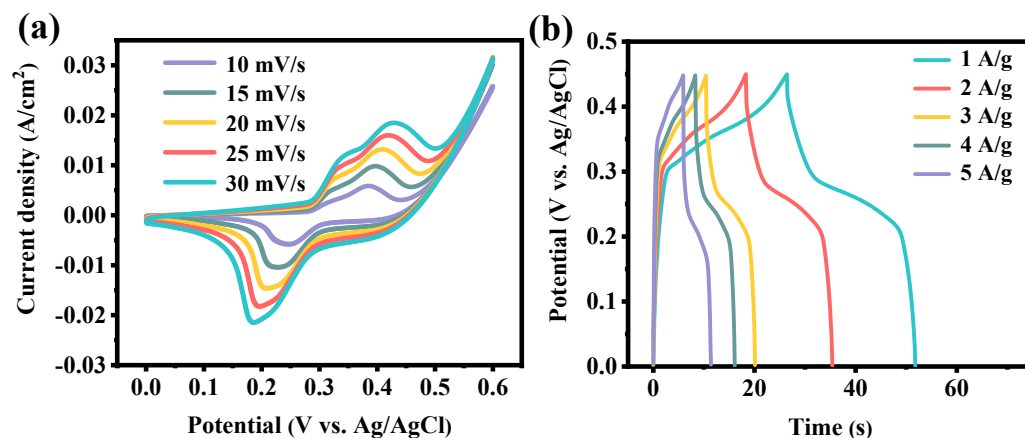
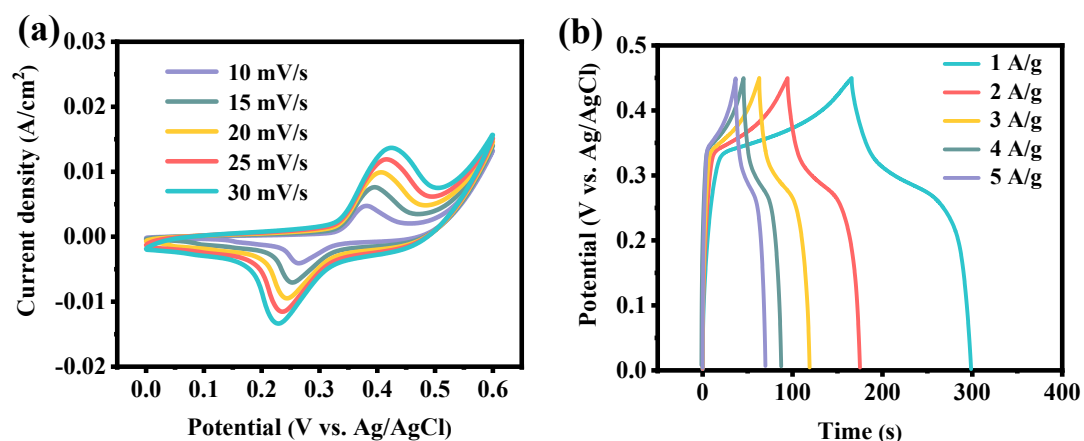
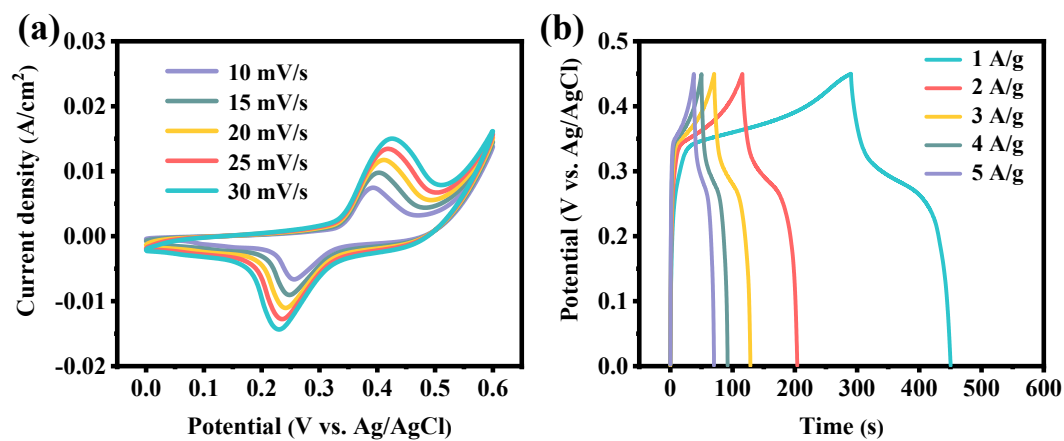


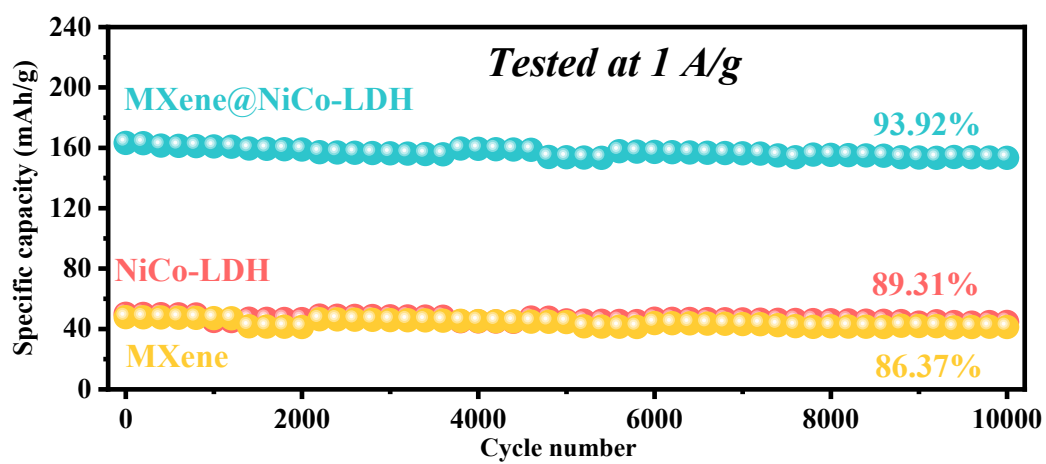
Fig. S6 (a) CV curves of NF electrode at different scan rates; (b) GCD curves of NF electrode at different specific currents.



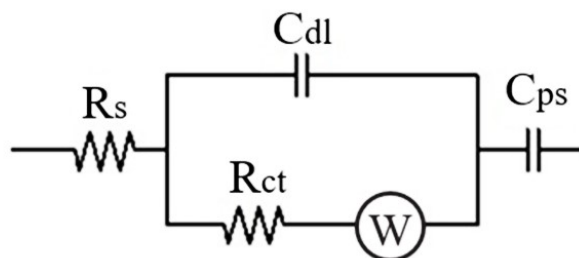
**Fig. S7** (a) CV curves of MXene electrode at different scan rates; (b) GCD curves of MXene electrode at different specific currents.



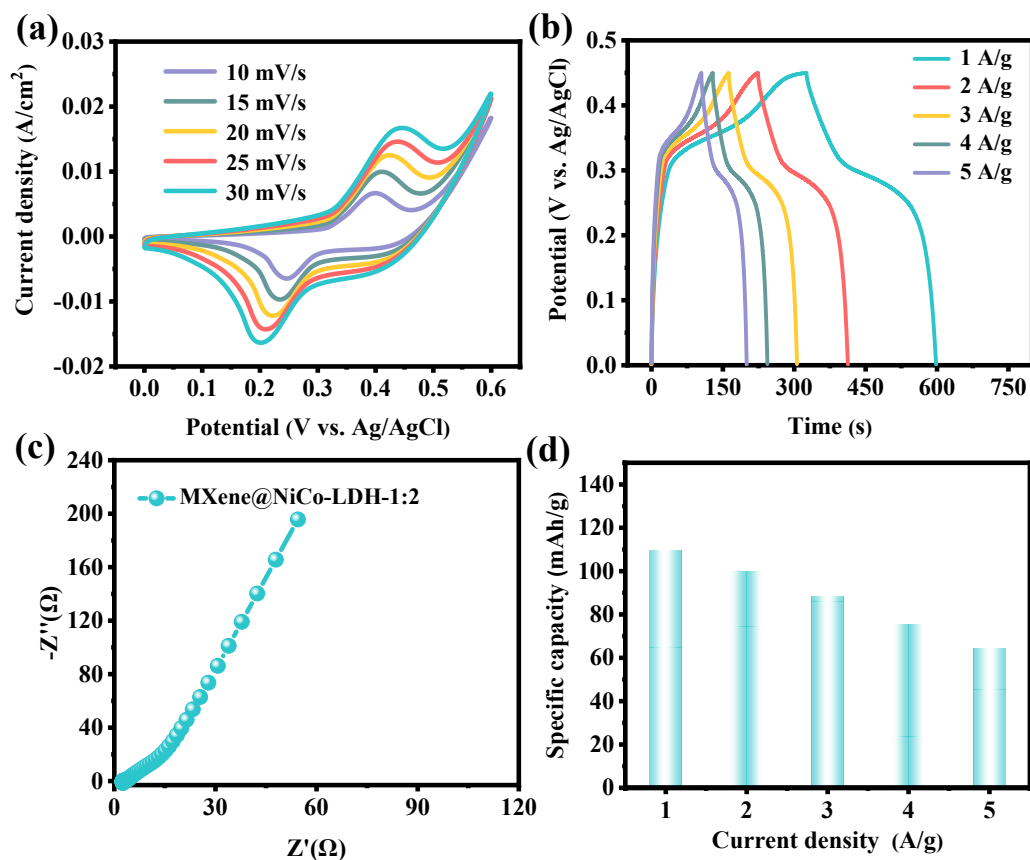
**Fig. S8** (a) CV curves of NiCo-LDH electrode at different scan rates; (b) GCD curves of NiCo-LDH electrode at different specific currents.



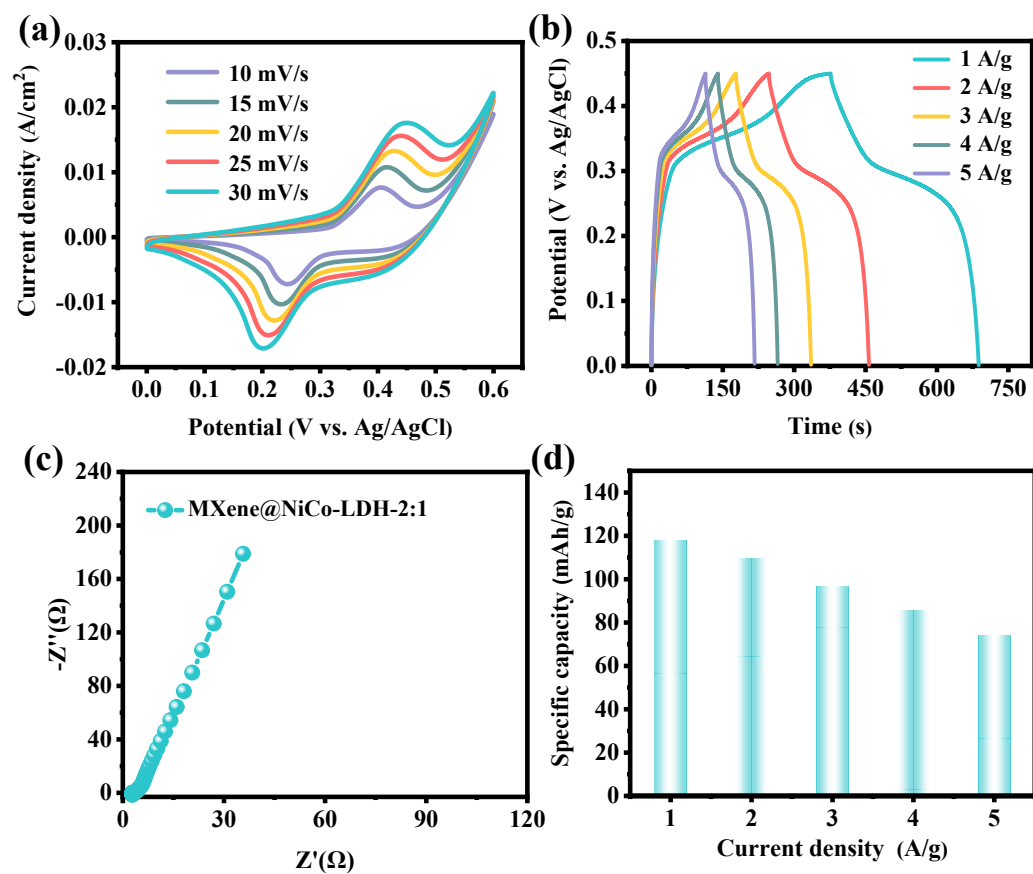
**Fig. S9** Cycling stability for 10000 cycles of as-prepared electrode.



**Fig. S10** Circuit used to fit EIS data.

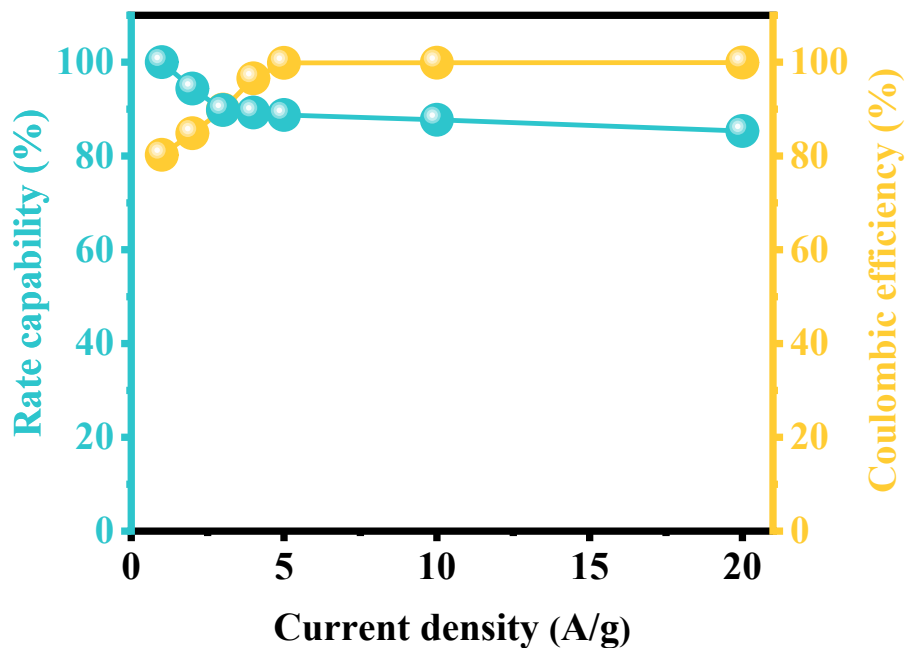


**Fig. S11** (a) CV curves of MXene@NiCo-LDH-1:2 electrode at different scan rates; (b) GCD curves of MXene@NiCo-LDH-1:2 electrode at different specific currents; (c) EIS curves; (d) Specific capacitance of MXene@NiCo-LDH-1:2 electrode.

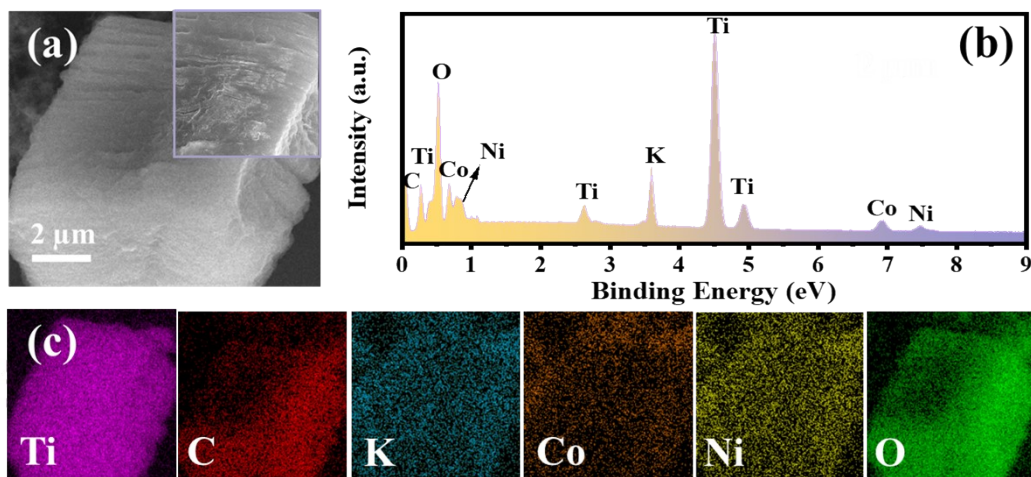


**Fig. S12** (a) CV curves of MXene@NiCo-LDH-2:1 electrode at different scan rates; (b) GCD curves of MXene@NiCo-LDH-2:1 electrode at different specific currents; (c) EIS curves; (d) Specific capacitance of MXene@NiCo-LDH-2:1 electrode.

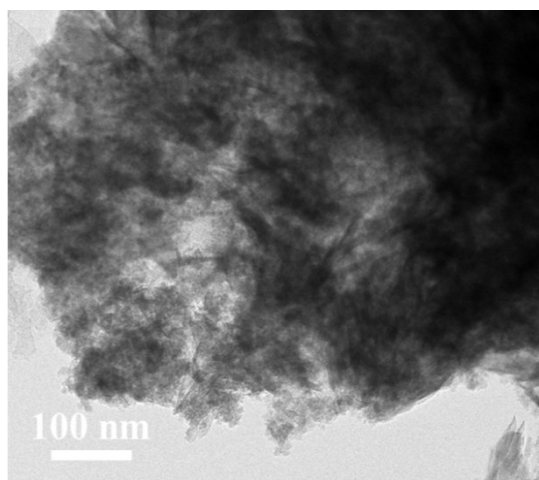




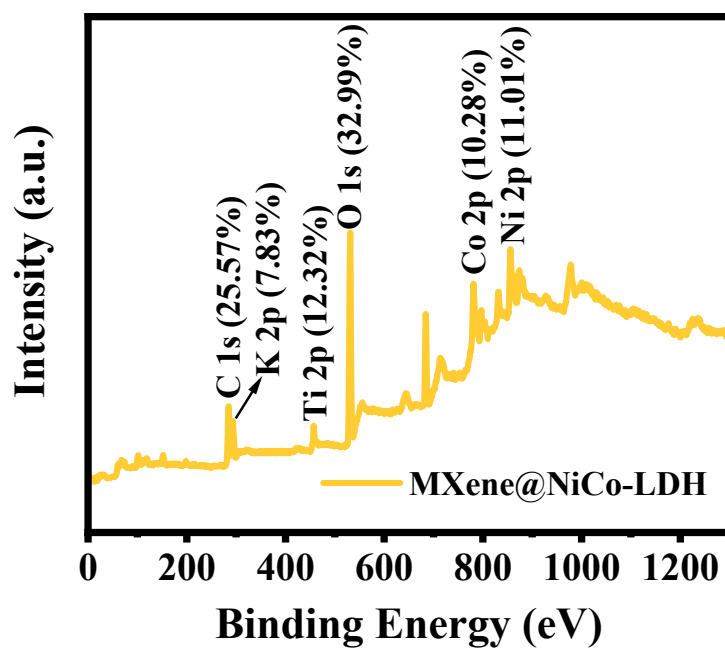
**Fig. S13** Rate capability and coulombic efficiency of the MXene@NiCo-LDH.



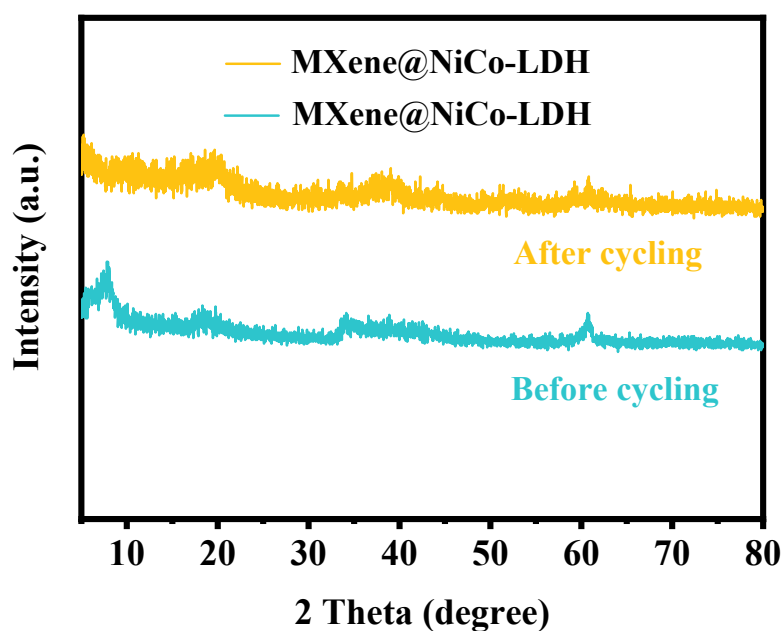
**Fig. S14** Post-electrochemical characterization of MXene@NiCo-LDH after stability test (a) SEM image; (b) SEM-EDX spectrum; (c) Elemental mapping area and elemental mapping spectra of different elements.



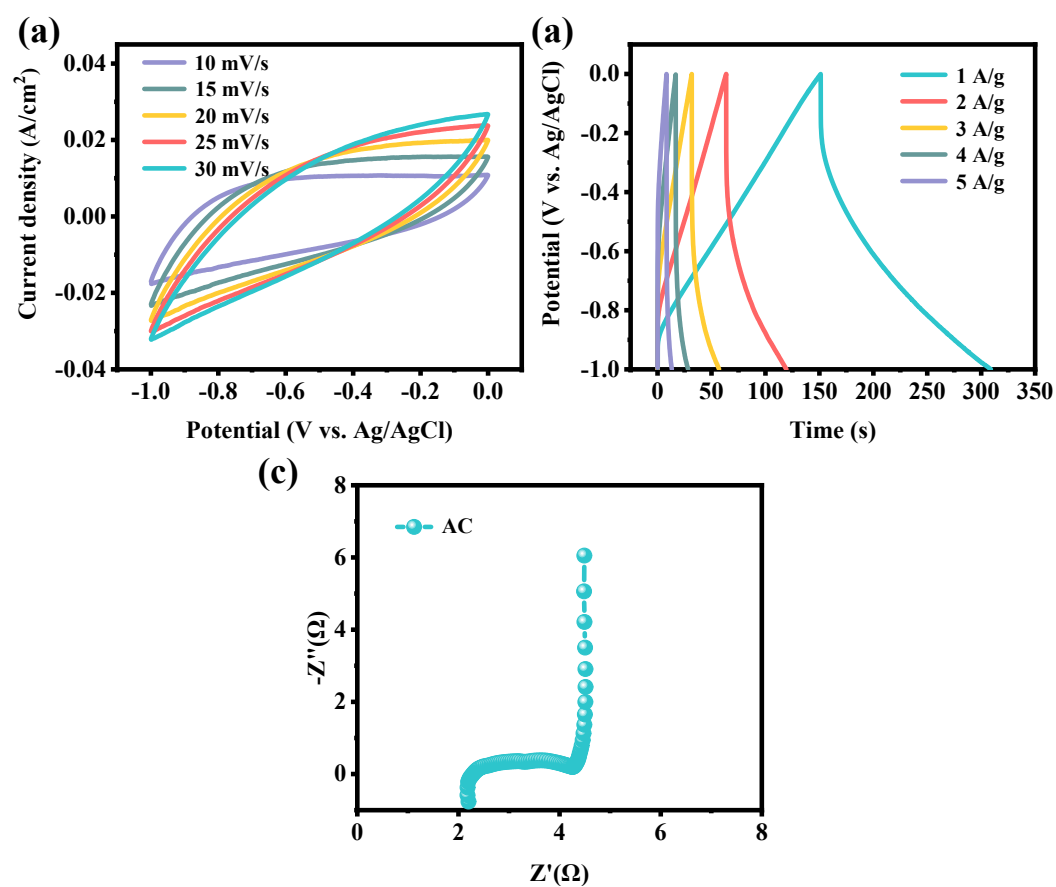
**Fig. S15** TEM images of MXene@NiCo-LDH after electrochemical test.



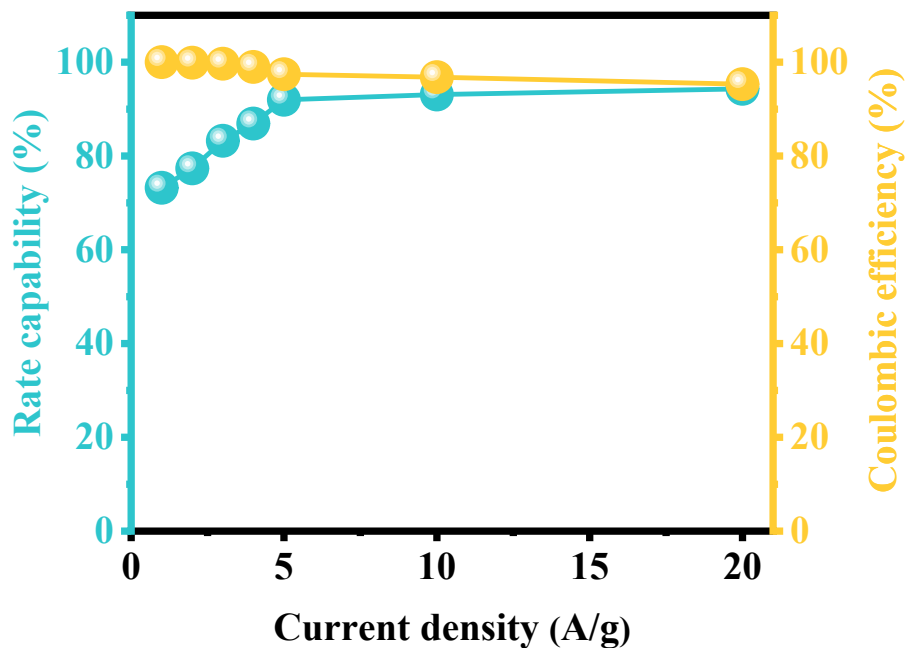
**Fig. S16** XPS survey spectra of MXene@NiCo-LDH after the electrochemical test.



**Fig. S17** XRD patterns of MXene@NiCo-LDH before and after electrochemical stability test.



**Fig. S18** (a) CV curves of AC; (b) GCD curves of AC; (c) EIS curve of AC.



**Fig. S19** Rate capability and coulombic efficiency of the MXene@NiCo-LDH//AC.

**Table S1** Calculation of adsorption energy of hydroxyl group on MXene@NiCo-LDH.

DFT calculation				
Materials	Slab	Slab+OH <sup>-</sup>	OH <sup>-</sup>	Adsorption energy
NiCo-LDH	-1847.63	-1856.09	-6.12	-2.34
MXene@NiCo-LDH	-3292.58	-3302.17	-6.12	-3.47

**Table S2** The electrochemical properties of the MXene based nanocomposite supercapacitors.

Electrode materials	Electrolyte	Potential window [V]	Performance	Cyclic stability	Ref.
<b>MXene@NiCo-LDH</b>	<b>2 M KOH</b>	<b>0 to 0.45</b>	<b>1306 F·g<sup>-1</sup> at 1 A·g<sup>-1</sup> (163.25 mAh·g<sup>-1</sup> at 1 A·g<sup>-1</sup>)</b>	<b>93.92 % after 10000 cycles</b>	<b>This work</b>
MoS <sub>2</sub> /MXene	3 M KOH	-0.8 to 0	583 F·g <sup>-1</sup> at 1 A·g <sup>-1</sup>	96.5 % after 5000 cycles	[1]
CoF/MXene	1 M KOH	0 to 0.4	1268.7 F·g <sup>-1</sup> at 1 A·g <sup>-1</sup>	96 % after 5000 cycles	[2]
PPy/MXene@CF	1 M H <sub>2</sub> SO <sub>4</sub>	0 to 0.5	506.6 F·g <sup>-1</sup> at 1 A·g <sup>-1</sup>	83.3 % after 2000 cycles	[3]
MnO <sub>2</sub> @MXene/CNT	1 M Na <sub>2</sub> SO <sub>4</sub>	0 to 1.0	181.8 F·g <sup>-1</sup> at 1 A·g <sup>-1</sup>	91 % after 5000 cycles	[4]
MXene@MnO <sub>2</sub>	1 M Na <sub>2</sub> SO <sub>4</sub>	0 to 1.0	348.5 F·g <sup>-1</sup> at 0.5 A·g <sup>-1</sup>	91.9 % after 5000 cycles	[5]
MXene@CNT-KOH	3 M H <sub>2</sub> SO <sub>4</sub>	-0.6 to 0.2	404.4 F·g <sup>-1</sup> at 1 A·g <sup>-1</sup>	99 % after 20000 cycles	[6]
MXene@CNTs	1 M H <sub>2</sub> SO <sub>4</sub>	0.2 to -0.8	300 F·g <sup>-1</sup> at 1 A·g <sup>-1</sup>	92 % after 10000 cycles	[7]
CoSe <sub>2</sub> /Ni <sub>3</sub> Se <sub>4</sub> /MXene	3 M KOH	0 to 0.5	283 mAh·g <sup>-1</sup> at 1 A·g <sup>-1</sup>	80 % after 5000 cycles	[8]
NiCo <sub>2</sub> S <sub>4</sub> @Ti <sub>3</sub> C <sub>2</sub> T <sub>x</sub>	3 M KOH	0 to 0.55	1076 F·g <sup>-1</sup> at 1 A·g <sup>-1</sup>	80 % after 5000 cycles	[9]
NiCo <sub>2</sub> S <sub>4</sub> /Ti <sub>3</sub> C <sub>2</sub> T <sub>x</sub>	0.5 M K <sub>2</sub> SO <sub>4</sub>	-0.2 to 0.5	167.28 F·g <sup>-1</sup> at 4 A·g <sup>-1</sup>	81 % after 3000 cycles	[10]
Ti <sub>3</sub> C <sub>2</sub> T <sub>x</sub> @NiO-RGO	1 M KOH	0 to 1	966 F·g <sup>-1</sup> at 1 A·g <sup>-1</sup>	94.5 % after 10000 cycles	[11]

**Table S3** The electrochemical properties of the NiCo-LDH based nanocomposite supercapacitors.

Electrode materials	Electrolyte	Potential window [V]	Performance	Cyclic stability	Ref.
MXene@NiCo-LDH	2 M KOH	0 to 0.45	1306 F·g <sup>-1</sup> at 1 A·g <sup>-1</sup> (163.25 mAh·g <sup>-1</sup> at 1 A·g <sup>-1</sup> )	93.92 % after 10000 cycles	This work
Ni <sub>2</sub> Co <sub>1</sub> -LDHs/BC50	6 M KOH (34–35°)	-0.1 to 0.30	2390 F·g <sup>-1</sup> at 1 A·g <sup>-1</sup>	75.9 % after 4000 cycles	[12]
Ni <sub>x</sub> Co <sub>1-x</sub> LDH/AC	6 M KOH	0 to 0.45	947 F·g <sup>-1</sup> at 1 A·g <sup>-1</sup>	83.5 % after 5000 cycles	[13]
NiAl/LDH	6 M KOH	0 to 0.55	884.5 F·g <sup>-1</sup> at 1 A·g <sup>-1</sup>	90 % after 7000 cycles	[14]
Ni@Co-Fe/LDH	6 M KOH	-0.8 to 0	1289 F·g <sup>-1</sup> at 1 A·g <sup>-1</sup>	76.66 % after 5000 cycles	[15]
Co(OH) <sub>2</sub> @NiCo/LDH	2 M KOH	0 to 0.5	996 F·g <sup>-1</sup> at 1 A·g <sup>-1</sup>	75.46 % after 5000 cycles	[16]
Ni <sub>2</sub> CoMn <sub>1</sub> /LDH	3 M KOH	0 to 0.5	1634.4 F·g <sup>-1</sup> at 0.5 A·g <sup>-1</sup>	73.1 % after 2000 cycles	[17]
NiMn-LDH	3 M KOH	0 to 0.45	1010.4 F·g <sup>-1</sup> at 0.2 A·g <sup>-1</sup>	70 % after 5000 cycles	[18]
Co-Al LDH/graphene	6 M KOH	0 to 0.55	712 F·g <sup>-1</sup> at 1 A·g <sup>-1</sup>	81 % after 2000 cycles	[19]
NiCo-LDH/NCF	2 M KOH	0 to 0.5	756 C·g <sup>-1</sup> at 0.5 A·g <sup>-1</sup>	54.7% from 0.5 to 20 A·g <sup>-1</sup>	[20]
NiCoMn LDH/rGO	2 M KOH	0 to 0.5	912 F·g <sup>-1</sup> at 1 A·g <sup>-1</sup>	63.3 % after 2500 cycles	[21]

**Table S4** Comparison of the electrochemical performances of MXene@NiCo-LDH composites with similar electrode materials.

Electrode materials	Electrolyte	Potential window [V]	Performance	Cyclic stability	Ref.
MXene@NiCo-LDH	2 M KOH	0 to 0.45	1306 F·g <sup>-1</sup> at 1 A·g <sup>-1</sup> (163.25 mAh·g <sup>-1</sup> at 1 A·g <sup>-1</sup> )	93.92 % after 10000 cycles	This work
QD-Ti <sub>3</sub> C <sub>2</sub> Cl <sub>2</sub> @ NiAlLDHs	1 M KOH	0 to 0.6	2010.8 F·g <sup>-1</sup> at 1 A·g <sup>-1</sup>	94.1 % after 10000 cycles	[22]
MXene/NiCo-LDHs	1 M (NH <sub>4</sub> ) <sub>2</sub> SO <sub>4</sub>	0 to 0.6	1207 F·g <sup>-1</sup> at 0.5 A·g <sup>-1</sup>	93 % after 5000 cycles	[23]
V <sub>2</sub> CT <sub>x</sub> /NiV-LDH	6 M KOH	0 to 0.5	1658.19 F·g <sup>-1</sup> at 1 A·g <sup>-1</sup>	80.95 % after 10000 cycles	[24]
Fe <sub>1</sub> Ni <sub>3</sub> -LDH/Ti <sub>3</sub> C <sub>2</sub> T <sub>x</sub> - MXene	6 M KOH	0 to 0.45	1015 F·g <sup>-1</sup> at 1 A·g <sup>-1</sup>	88 % after 10000 cycles	[25]
NiCo <sub>2</sub> - LDHs@MXene/rGO	2 M KOH	0 to 0.45	332.2 mAh·g <sup>-1</sup> at 1 A·g <sup>-1</sup>	87.5 % after 5000 cycles	[26]
Ni <sub>2</sub> CoLDHs@AL-Ti <sub>3</sub> C <sub>2</sub> MXene	6 M KOH	0 to 0.5	227 mAh·g <sup>-1</sup> at 1 A·g <sup>-1</sup>	90 % after 10000 cycles	[27]
Ti <sub>3</sub> C <sub>2</sub> /Ni-Co-Al-LDH	1 M KOH	0 to 0.6	748.2 F·g <sup>-1</sup> at 1 A·g <sup>-1</sup>	748.2 to 569.8 F·g <sup>-1</sup> from 1 to 20 A·g <sup>-1</sup>	[28]
LDH-MXene-LDH	6 M KOH	0 to 0.5	179 mAh·g <sup>-1</sup> at 1 A·g <sup>-1</sup>	79.1 % after 5000 cycles	[29]
NiMn-LDH/MXene	6 M KOH	0 to 0.45	1575 F·g <sup>-1</sup> at 0.5 A·g <sup>-1</sup>	90.3 % after 10000 cycles	[30]
NiMn-LDH/MXene	1 M LiPF <sub>6</sub>	0 to 0.4	133 mAh·g <sup>-1</sup> at 1 A·g <sup>-1</sup>	88.6 % after 3000 cycles	[31]

**Table S5** Nyquist impedance parameters of the different positrodes (MXene, NiCo-LDH, MXene@NiCo-LDH).

Sample	$R_s$ [ $\Omega$ ]	$R_{ct}$ [ $\Omega$ ]	$R_w$ [ $\Omega$ ]
MXene	3.87	2.31	0.48
NiCo-LDH	3.61	1.33	0.54
MXene@NiCo-LDH	2.10	1.10	0.12

**Table S6** Nyquist impedance parameters of MXene@NiCo-LDH//AC before and after stability.

Sample	$R_s$ [ $\Omega$ ]	$R_{ct}$ [ $\Omega$ ]	$R_w$ [ $\Omega$ ]
MXene@NiCo-LDH//AC			
Before stability	4.14	1.39	0.43
After stability	4.51	1.72	0.67



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