Electronic Supplementary Information (SI) for

Integration of ultrathin and modified NiCoAl-layered double hydroxide nanosheets with N-doped reduced graphene oxide for high-performance all-solid-state supercapacitors

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This information contains:

(1) Digital photographs for m-LDH/NRG NHs, pure m-LDH and NRG dispersions (Figure S1)

(2) The HAADF-STEM images for pure NiCoAl-LDH and m-LDH nanosheets as well as m-LDH/NRG NHs (**Figure S2**)

(3) The mapping analysis for N element in the m-LDH/NRG (**Figure S3**)

(4) Raman for NiCoAL-LDH, m-LDH and m-LDH/NRG at 200 to 1000 cm^{-1} (**Figure S4**)

(5) Raman for NRG and m-LDH/NRG at 1100 to 1900 cm^{-1} (**Figure S5**)

(6) The survey XPS spectrum for m-LDH/NRG NHs (**Figure S6**)

(7) Fine core-level XPS spectra of NiCoAl-LDH (**Figure S7**)

(8) Fine core-level XPS spectra of m-LDH (**Figure S8**)

(9) N_2 sorption isothermal plots for m-LDH/NRG NHs and NiCoAl-LDH nanosheets (**Figure S9**)

(10) CV plots for m-LDH/NRG NHs, NRG and pure Ni foam at a constant potential scan rate, and CV plots for m-LDH/NRG NHs, NiCoAl-LDH and m-LDH and at different potential scan rates (**Figure S10**)

(11) The detailed data for XPS spectra of m-LDH/NRG NHs, pure m-LDH and unmodified NiCoAl-LDH nanosheets (**Table S1**)

(12) ICP data for pure NiCoAl-LDH and m-LDH nanosheets (**Table S2**)

(13) The detailed integral areas of O1s peaks for NiCoAl-LDH and m-LDH nanosheets (**Table S3**)

(14) Comparing the electrochemical performance of m-LDH/NRG NHs with other reported electrode materials (**Table S4**)

(15) References

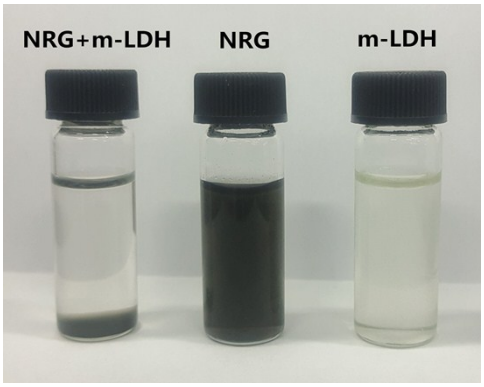

Fig. S1 Digital photographs for the dispersions of m-LDH/NRG NHs as well as pure m-LDH and NRG dispersions.

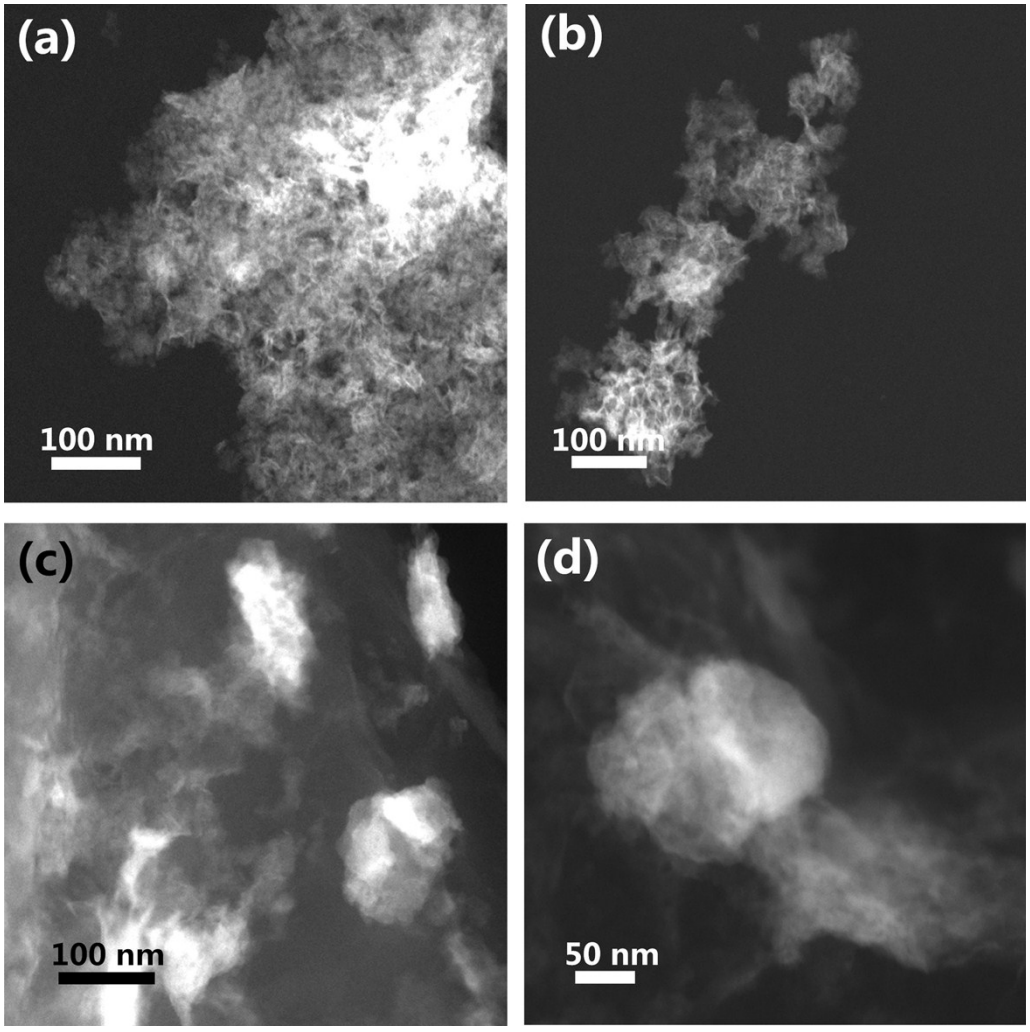

Fig. S2 (a-b) Low-magnification HAADF-STEM images of NiCoAl-LDH (a) and m-LDH (b) nanosheets. (c-d) Low and high-magnification HAADF-STEM images for m-LDH/NRG NHs.

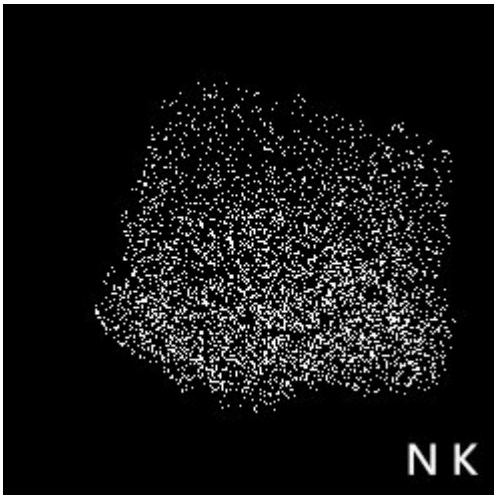

Fig. S3 The mapping analysis for N element in the m-LDH/NRG NHs.

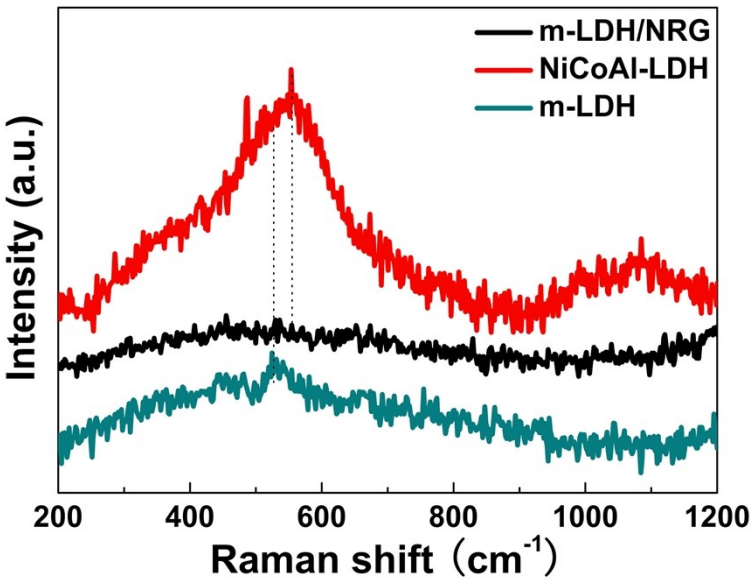

Fig. S4 Comparing the Raman spectra of m-LDH/NRG NHs, NiCoAl-LDH and m-LDH nanosheets at the low wavenumber region (ranging from 200 to 1100 cm^{-1}).

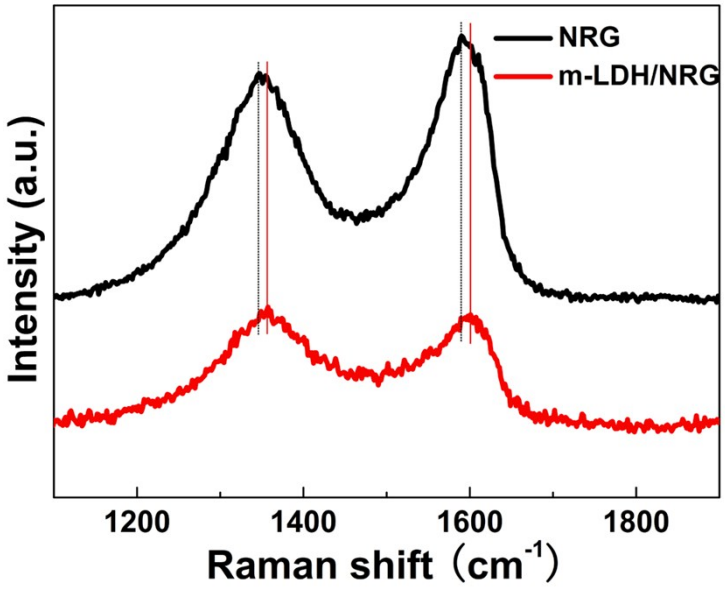

Fig. S5 Comparing the Raman spectra of m-LDH/RGN NHs and pure m-LDH nanosheets at the high wavenumber region (ranging from 1100 to 1900 cm^{-1}).

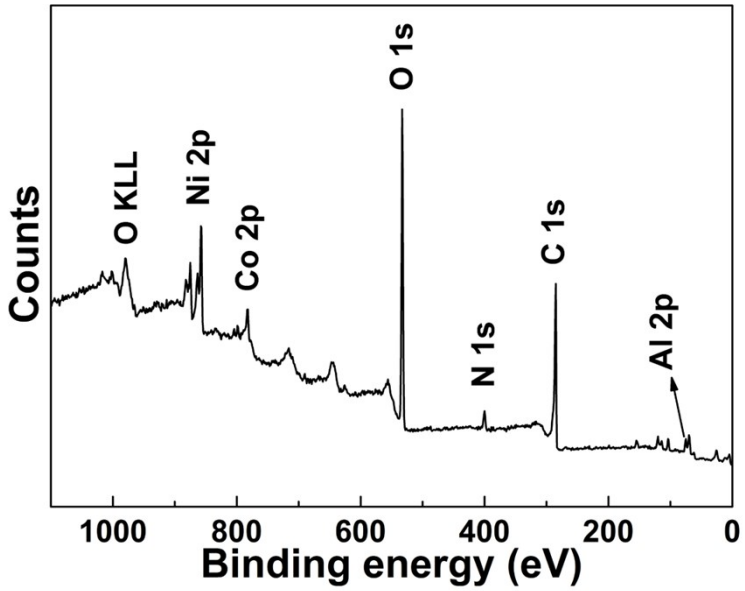

Fig. S6 The survey XPS spectrum for m-LDH/NRG NHs.

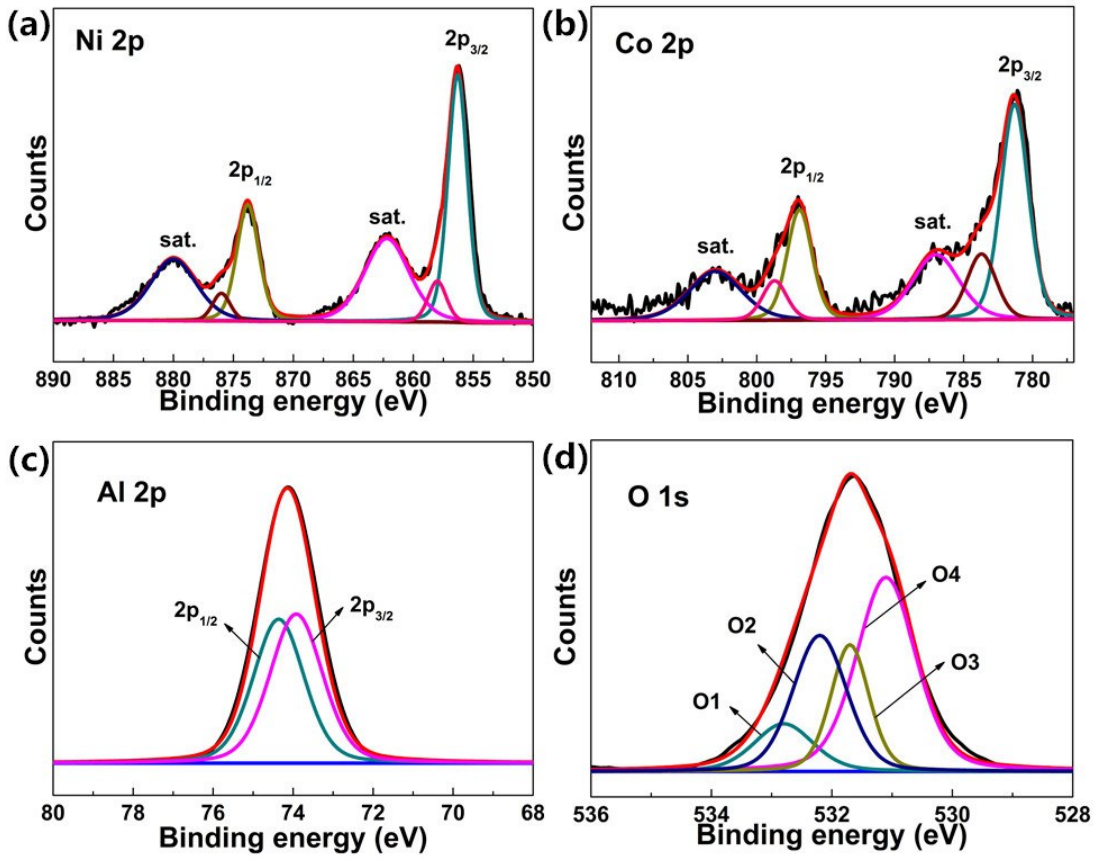

Fig. S7 Fine core-level XPS spectra for Ni 2p (a), Co 2p (b), Al 2p (c) and O 1s(d) peaks of the unmodified NiCoAl-LDH nanosheets.

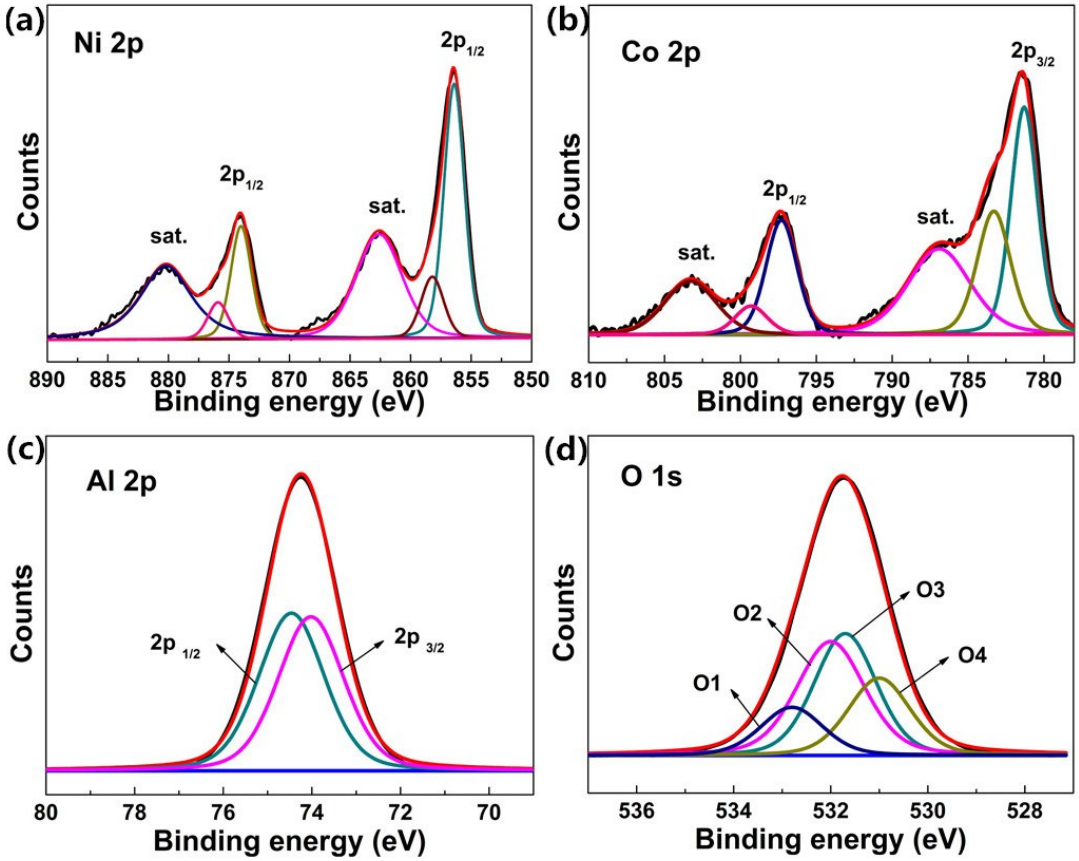

Fig. S8 Fine core-level XPS spectra for Ni 2p (a), Co 2p (b), Al 2p (c) and O 1s(d) peaks of pure m-LDH nanosheets.

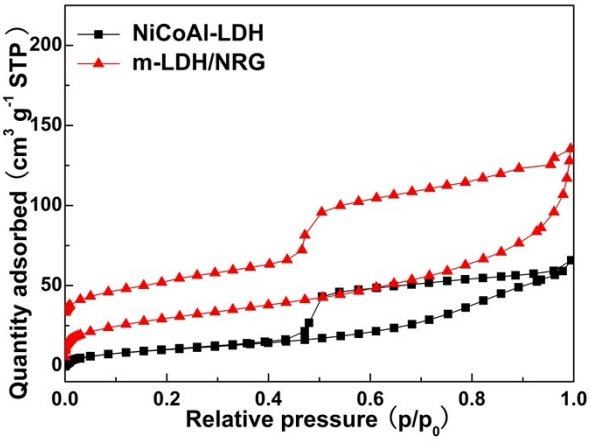

Fig. S9 N_2 adsorption-desorption isothermal plots for m-LDH/NRG NHs and the unmodified NiCoAl-LDH nanosheets.

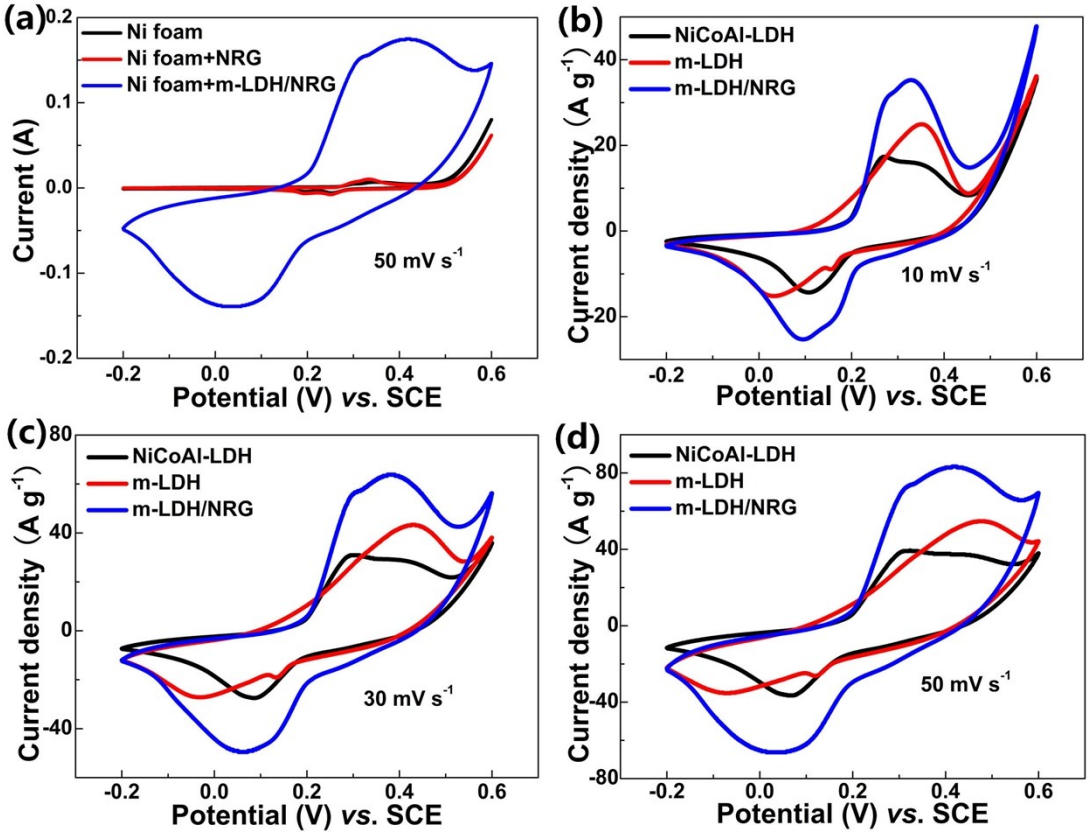

Fig. S10 (a) Comparing the CV curves of m-LDH/NRG NHs with that of pure NRG nanosheets and pure Ni foam at the potential scan rate of 50 mV s^{-1}. (b-d) Comparing the CV curves of m-LDH/NRG NHs with that of NiCoAl-LDH and m-LDH nanosheets at the potential scan rate of 10, 30 and 50 mV s^{-1}, respectively.

Table S1 The detailed data for fitting the Ni 2p, Co2p, Al 2p and O1s fine XPS spectra of m-LDH/NRG NHs, pure m-LDH and unmodified NiCoAl-LDH

	Spin-orbit	LDH	m-LDH	m-LDH/NRG
Ni	$2p_{1/2}$ +2	856.2eV	856.5eV	856.8eV
	$2p_{1/2}$ +3	858.1eV	858.2eV	858.3eV
	$2p_{3/2}$ +2	873.7eV	874.0eV	874.4eV
	$2p_{3/2}$ +3	875.6eV	875.9eV	876.5eV
Co	$2p_{1/2}$ +2	783.7eV	783.3eV	783.9eV
	$2p_{1/2}$ +3	781.2eV	781.3eV	781.8eV
	$2p_{3/2}$ +2	798.7eV	799.3eV	800.1eV
	$2p_{3/2}$ +3	796.9eV	797.3eV	797.8eV
Al	$2p_{1/2}$	73.9eV	74.0eV	74.7eV
	$2p_{3/2}$	74.3eV	74.4eV	75.2eV
O	O_1	532.8eV	532.8eV	533.6eV
	O_2	532.1eV	532.1eV	532.6eV
	O_3	531.7eV	531.7eV	532.2eV
	O_4	531.1eV	531.1eV	531.6eV

Table S2 ICP test results for the content of Co, Al and Ni in NiCoAl-LDH and m-LDH nanosheets

Sample	Element	Ion concentration (μg mL^{-1})	Mass percent
NiCoAl-LDH	Co	0.703	7.0%
	Al	0.415	4.2%
	Ni	2.061	20.6%
m-LDH	Co	0.592	5.9%
	Al	0.314	3.1%
	Ni	1.839	18.4%

[**Notes**]: For ICP tests, the NiCoAl-LDH and m-LDH nanosheets with the same mass were dissolved in 6 mL concentrated nitric acid, respectively. Then, these solution were added into 100 mL flask and diluted to 100 mL, respectively. The content of various metal elements were examined by ICP.

Table S3. The detailed integral areas of O1s peaks for NiCoAl-LDH and m-LDH nanosheets

peaks	NiCoAl-LDH (integration area)	m-LDH (integration area)
O1 H_2O	26204	45757
O2 O-H	65340	121574
O3 oxygen vacancies	44633	114272
O4 M-OH	107492	73501

[**Notes**]: By calculating from their integration areas, the content of O3 peak (O vancancies or defects) in m-LDH nanosheets (about 32.2%) is much higher than that of unmodified NiCoAl-LDH nanosheets (18.3%), implying that etching Al atoms can create more oxygen vacancies in m-LDH in relative to unmodified NiCoAl-LDH. This is consistent with previously reported works (*e.g.*, *Chem. Eur. J.* **2016**, *22*, 4000).

Table S4 Comparing the electrochemical performance of m-LDH/NRG NHs with other reported electrode materials

Electrode materials	Specific capacitance ($F\ g^{-1}$)	Rate performance	Capacitance retention (cycles)	Literatures
NiCoAl-LDH/ NRG	1877 (1 A g^{-1})	72% (10 A g^{-1})	74% (5000)	This work
NiAl-LDH/rGO	1630 (1 A g^{-1})	17% (10 A g^{-1})	95% (1500)	Ref S1
CoMn-LDH/ rGO	1635 (1 A g^{-1})	71% (10 A g^{-1})	100% (10000)	Ref S2
3D NiCoMn -LDH/rGO	912 (1 A g^{-1})	81% (10 A g^{-1})	64% (5000)	Ref S3
CNT@$NiCo_2O_4$	1038 (0.5 A g^{-1})	64% (10 A g^{-1})	100% (1000)	Ref S4
CoAl-LDH/rGO	825 (1 A g^{-1})	62% (8 A g^{-1})	89% (4000)	Ref S5
$Ni_{1-x}Co_xAl$- LDH/rGO	1902 (1 A g^{-1})	75% (10 A g^{-1})	62% (1500)	Ref S6
$Ni_xCo_{1-x}S_8$	1404 (2 A g^{-1})	38% (9A g^{-1})	75% (2000)	Ref S7
α-$(Ni/Co)(OH)_2$ /graphene	1809 (0.5 A g^{-1})	84% (40 A g^{-1})	83%（10000）	Ref S8
NiV-LDH	1581 (1 A g^{-1})	77% (1 A g^{-1})	40% (10000)	Ref S9

CoAl-LDH@ CoS	1205 (1 A g^{-1})	941 (10 A g^{-1})	88% (2000)	Ref S10
CoAl-LDH/ graphene	1296 (1 A g^{-1})	43% ((10 A g^{-1})	90% (1000)	Ref S11
HMCS/$Ni(OH)_2$	1632 (1 A g^{-1})	61% (10 A g^{-1})	81% (3000)	Ref S12
MoO_3/$Ni(OH)_2$	1622 (1 A g^{-1})	26% (10 A g^{-1})	80% (3000)	Ref S13
$Ni(OH)_2$/3D graphite foam	1500 (1 A g^{-1})	67% (10 A g^{-1})	65% (1000)	Ref S14

[Notes]: The abbreviation of "LDH" stands for the "layered double hydroxides", and the abbreviation of "NRG" represents "N-doped reduced graphene oxide". The "rGO" stands for "reduced graphene oxide".

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