Supplementary Information

Rechargeable Na/Ni battery based on the $Ni(OH)_2$/NiOOH redox couple with high energy density and good cycling performance

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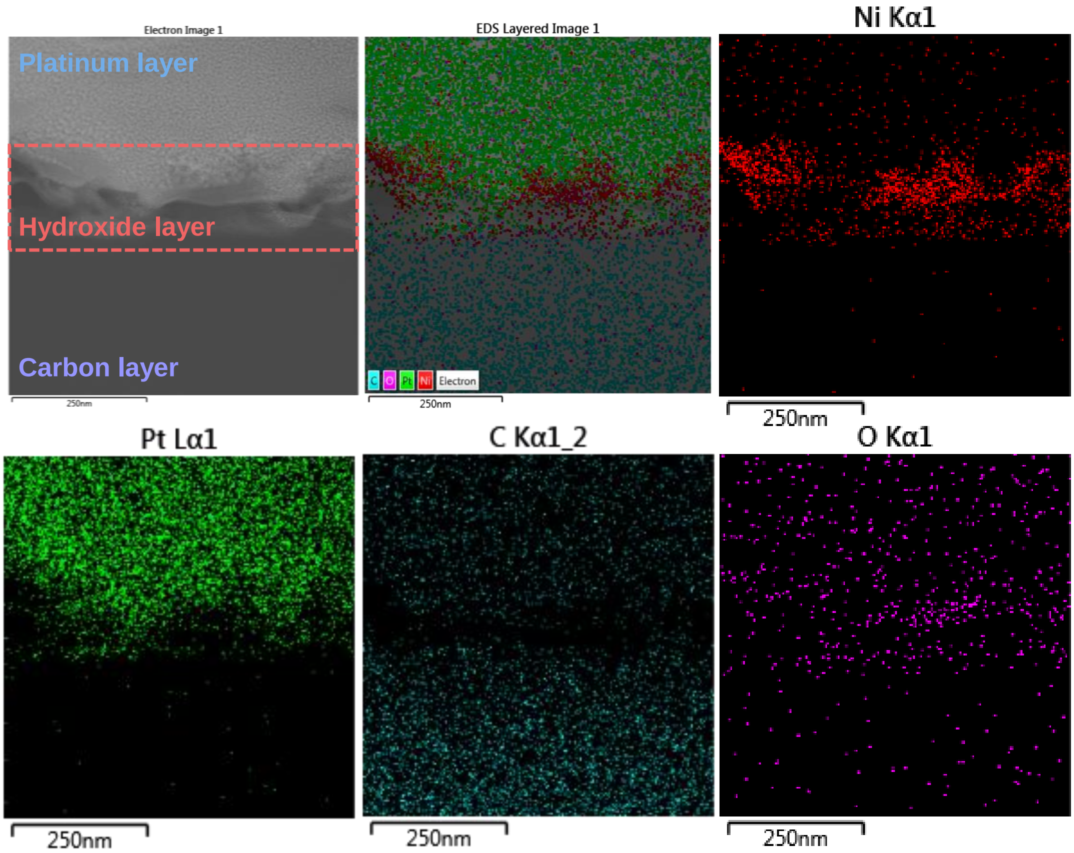


Fig. S1 STEM-EDS cross-sectional mapping images of the hydroxide layer on the carbon microfiber electrode.

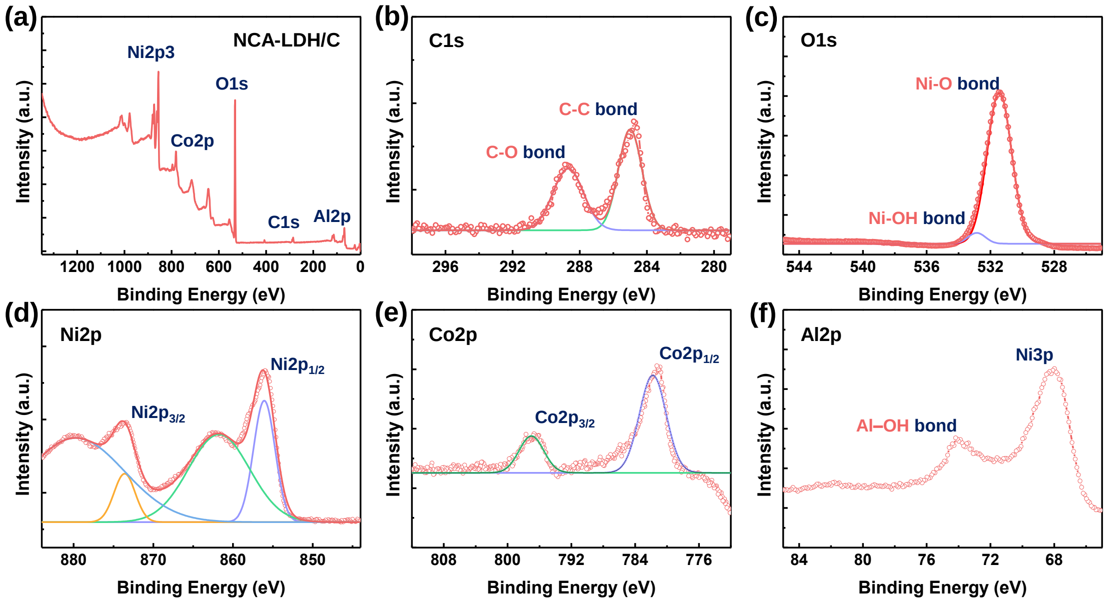


Fig. S2 Summary of XPS spectra recorded to analyze the chemical bonding in the NCA-LDH/C electrode. (a) Summary XPS spectra, (b) C 1s spectrum, (c) O 1s spectrum, (d) Ni 2p spectrum, (e) Co 2p spectrum, and Al 2p spectrum of NCA-LDH/C electrode.

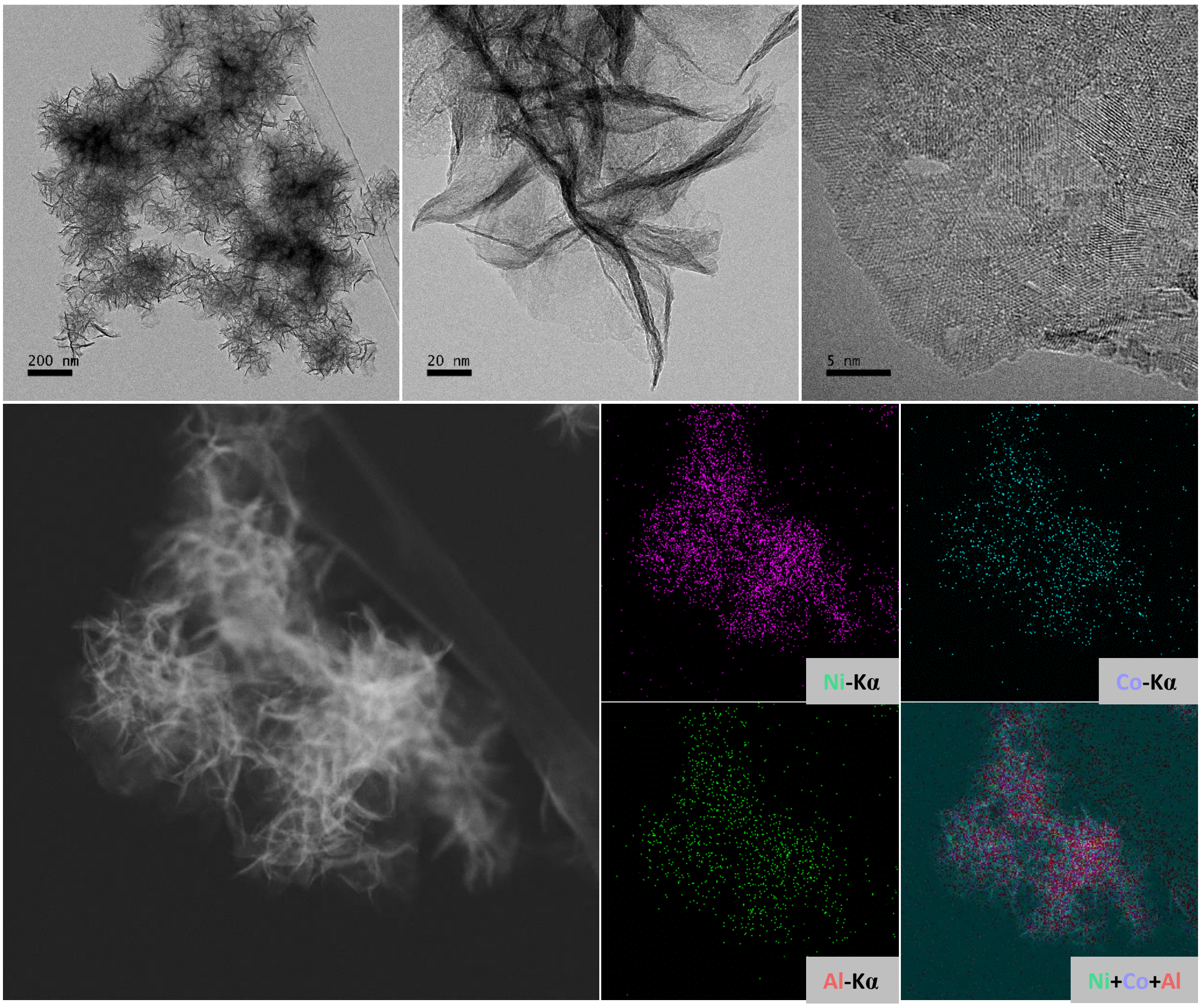
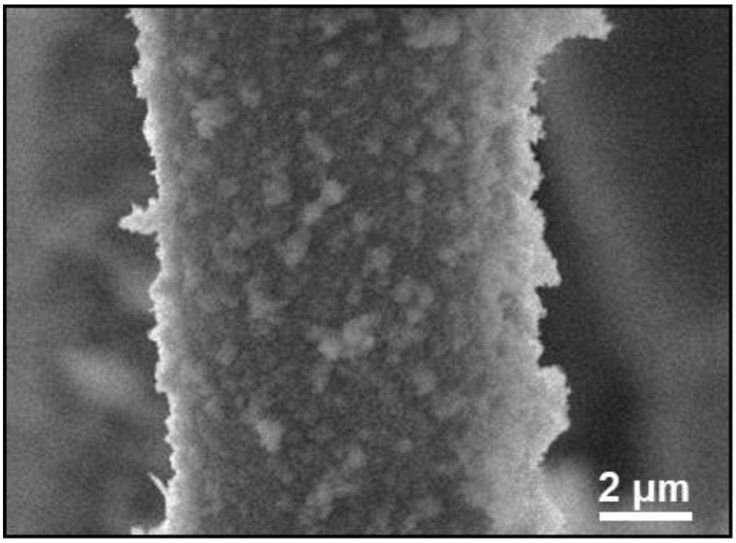
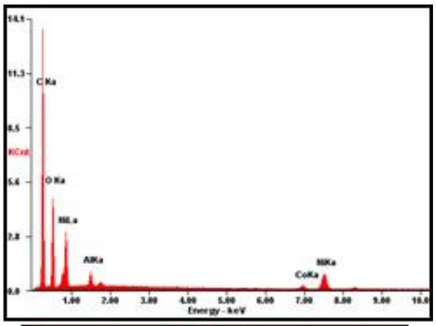


Fig. S3 TEM images and STEM-EDS mapping images of NCA-LDH sheets.





Element	Wt%	At%
CK	48.94	69.04
OK	20.23	21.42
AlK	1.91	1.20
CoK	4.16	1.20
NiK	24.76	7.15

Fig. S4 SEM image and SEM-EDS spectra of NCA/C electrode.

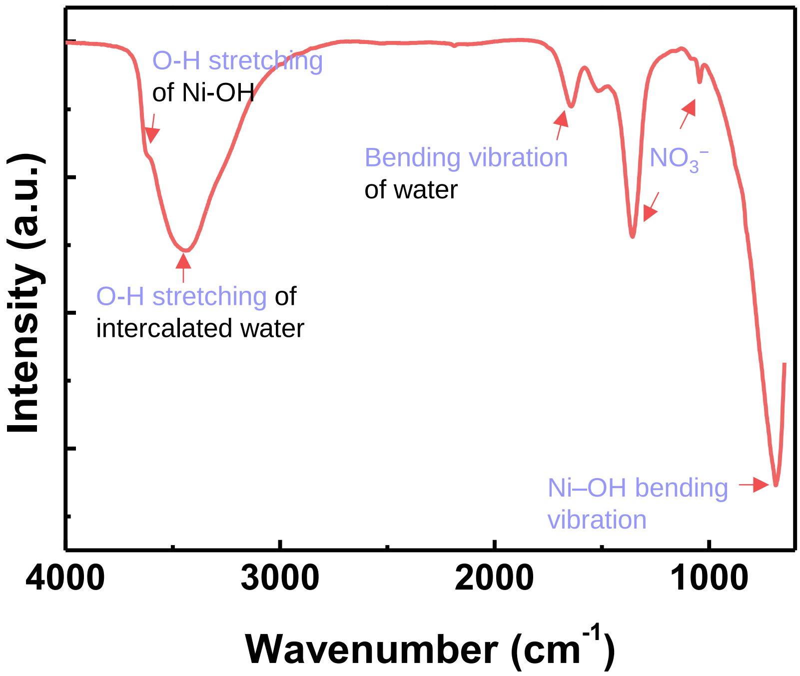


Fig. S5 FT-IR spectrum of NCA-LDH sheets.

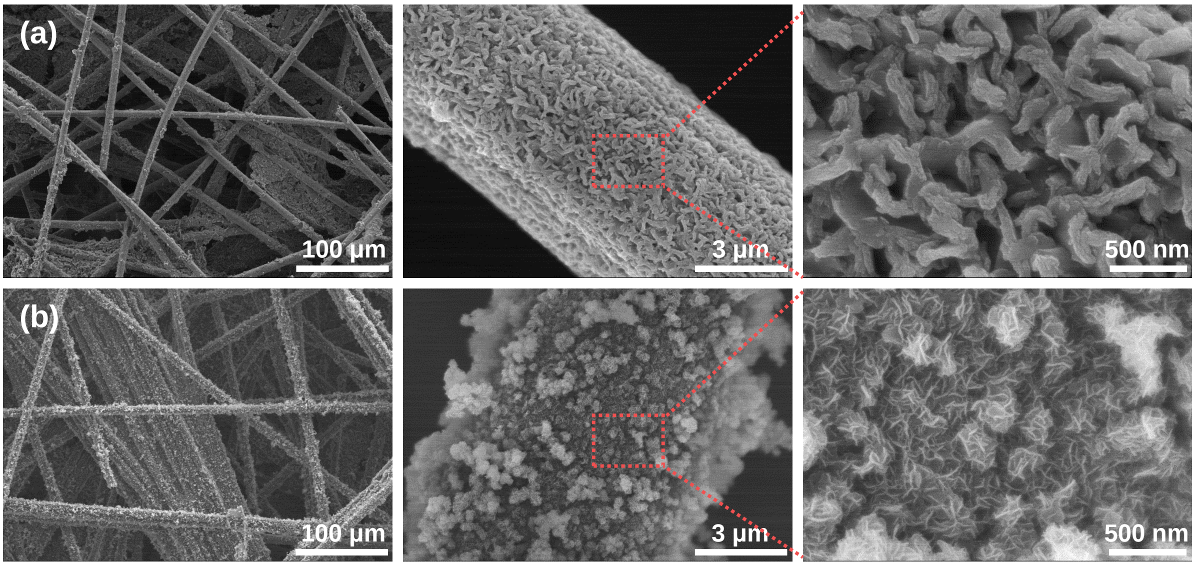


Fig. S6 SEM images of (a) α-$Ni(OH)_2$/C and (b) NiAl-LDH/C electrodes.

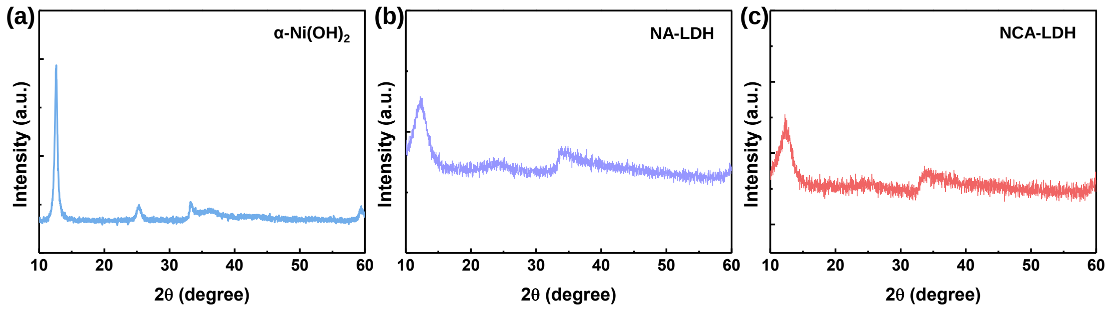


Fig. S7 Comparative XRD results for (a) the α-$Ni(OH)_2$/C, (b) NiAl-LDH/C, and (c) NCA-LDH/C electrodes.

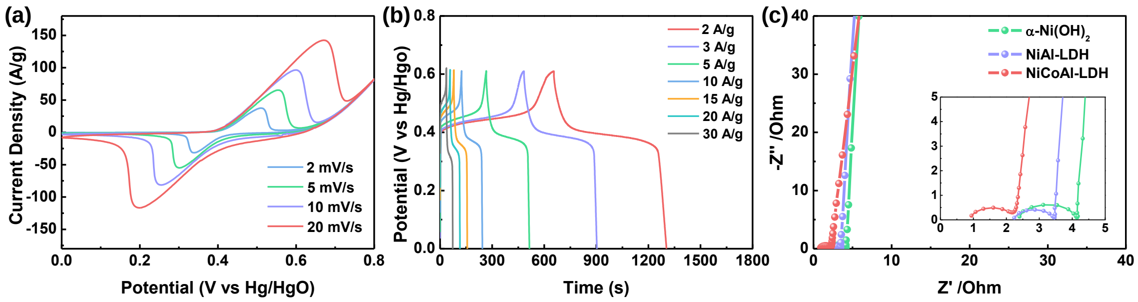


Fig. S8 Electrochemical results for NCA-LDH/C. (a) CV curves at different current rates. (b) Galvanostatic charge–discharge curves. (c) EIS analysis of NCA-LDH/C electrode.

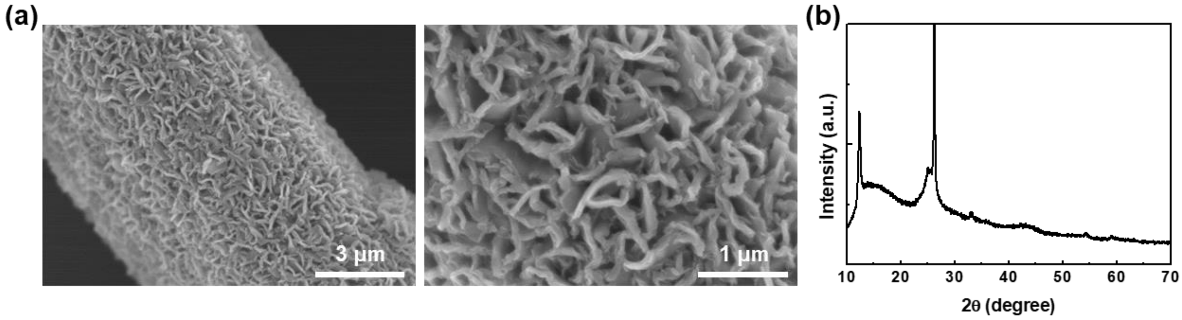


Fig. S9 (a) SEM images and (b) XRD spectra of NiCo-LDH/C electrode.

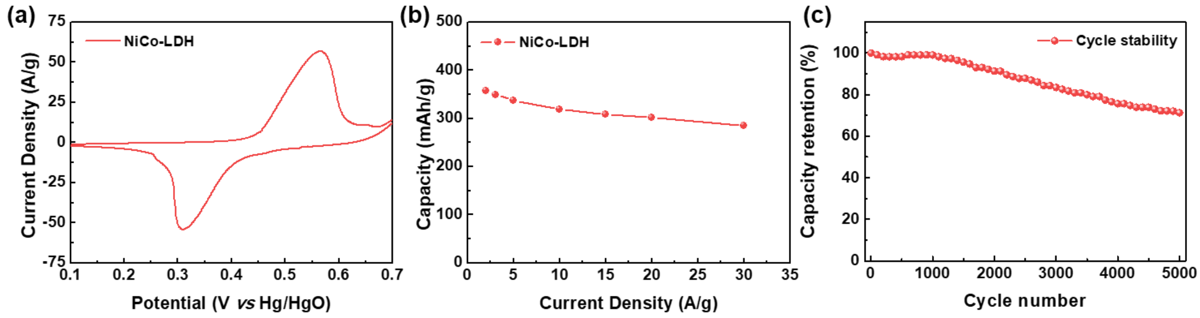


Fig. S10 (a) CV curve, (b) specific capacity as the current density and (c) cycle stability of NiCo-LDH/C electrode.

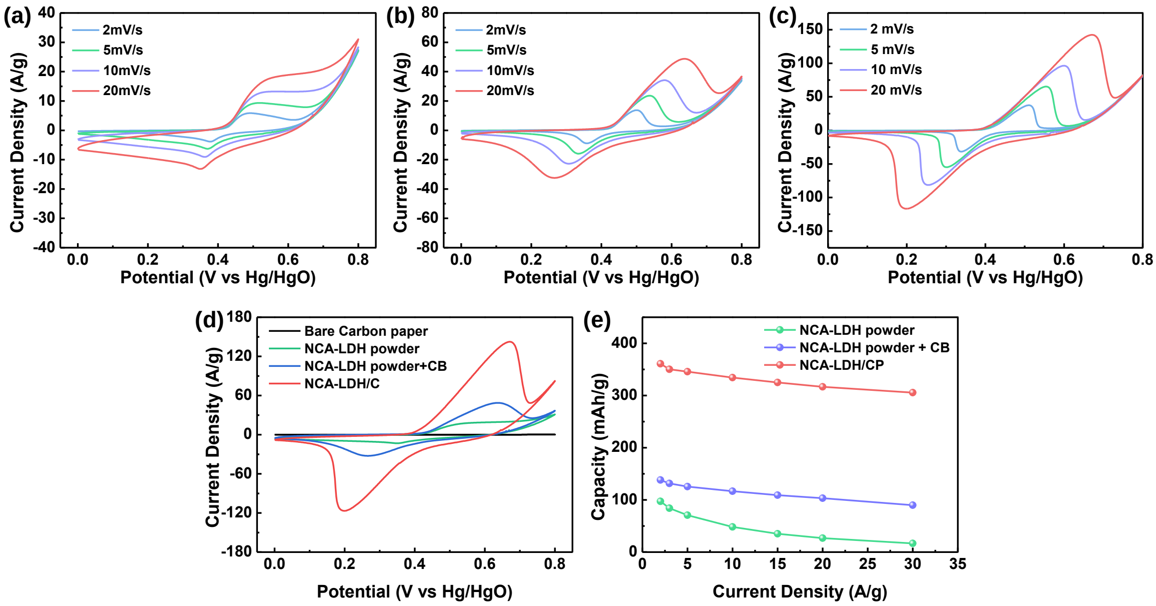


Fig. S11 Electrochemical results for NCA powder, NCA powder + carbon black (CB), and the NCA-LDH/C electrode. CV curves of (a) NCA powder, (b) NCA powder + CB, and (c) the NCA-LDH/C electrode. (d) Summary of CV curves and (e) Comparative rate capability of NCA powder, NCA powder + carbon black (CB), and the NCA-LDH/C electrode.

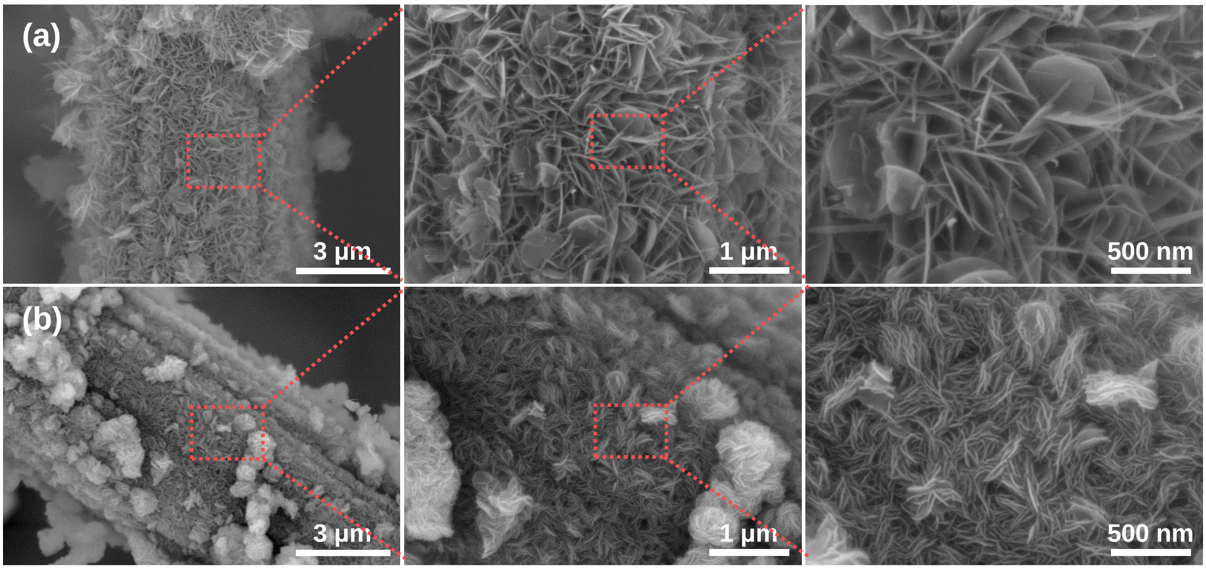


Fig. S12 SEM images of NCA-LDH/C electrode with different solvent mixtures. (a) DI water: DMF = 9:1. (b) DI water: DMF = 5:5.

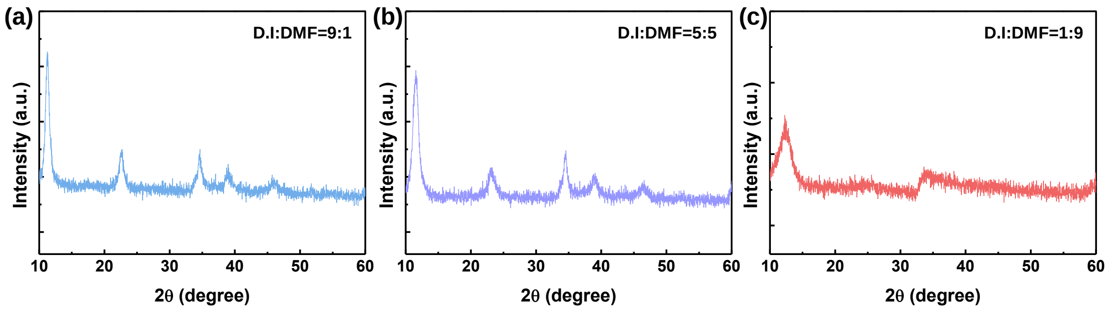


Fig. S13 Comparative XRD results with different solvent mixtures. (a) DI water: DMF = 9:1. (b) DI water: DMF = 5:5. (c) DI water: DMF = 1:9.

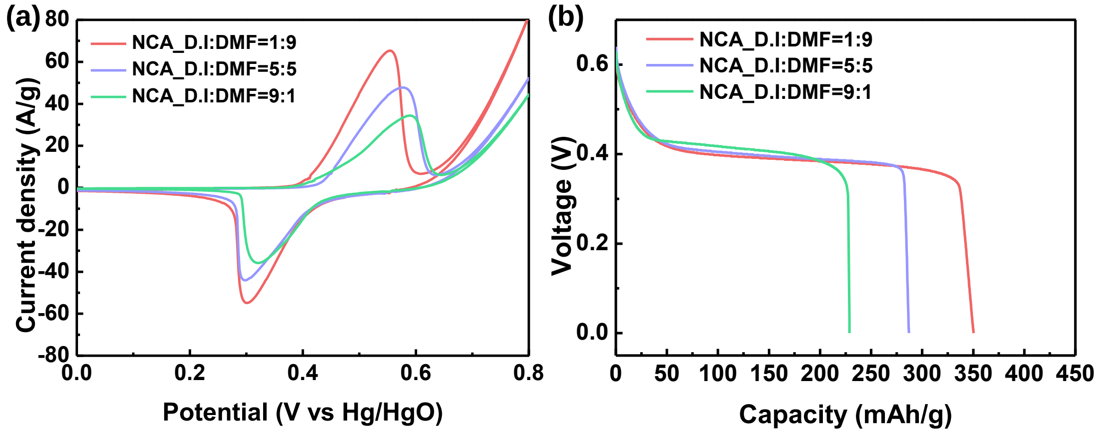


Fig. S14 Electrochemical results for NCA-LDH/C with different solvent mixtures. (a) Summary of CV curves and (b) Galvanostatic discharge curves with with different solvent mixtures

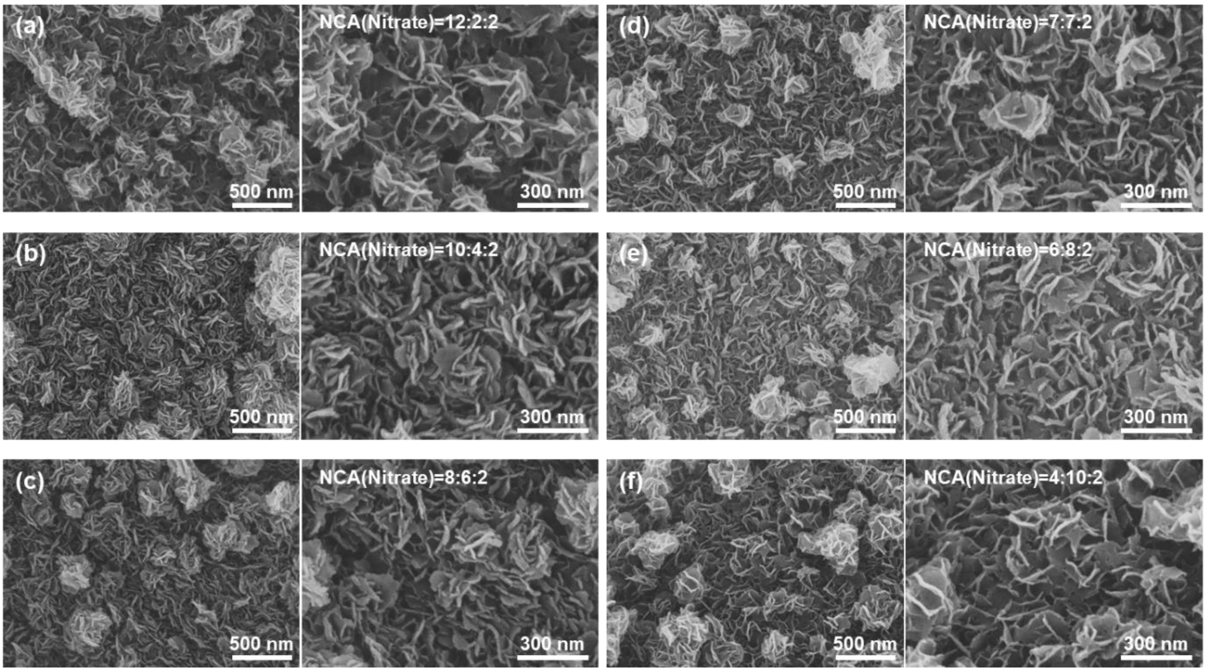


Fig. S15 SEM images of NCA-LDH/C electrode as the different ratio of Ni, Co, Al atom.

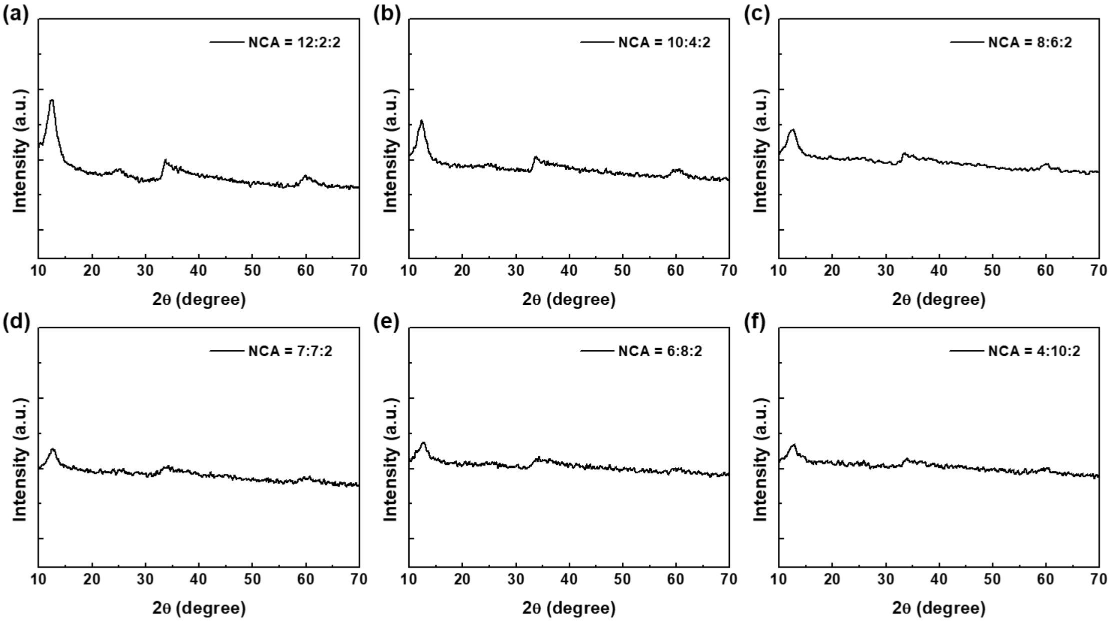


Fig. S16 XRD spectra of NCA-LDH electrode as the different ratio of Ni, Co, Al atom.

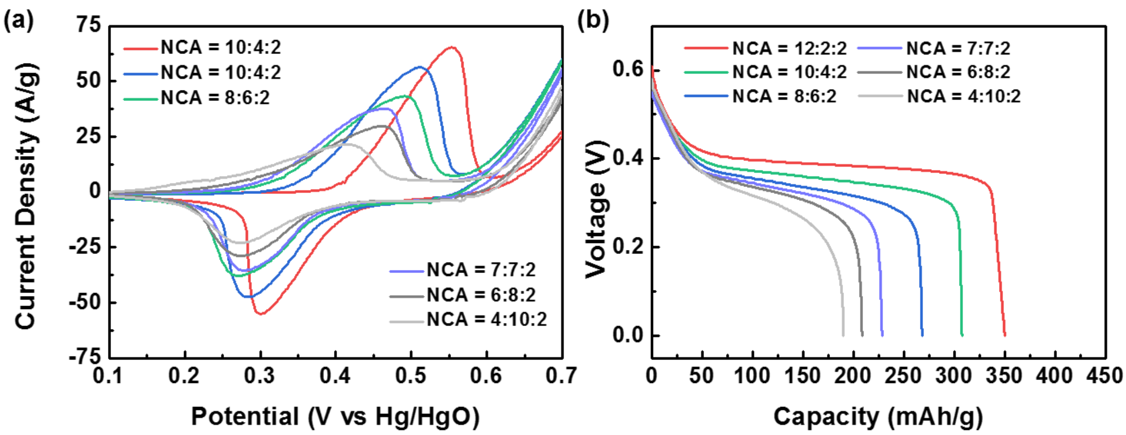


Fig. S17 (a) CV curve and (b) discharge curve of NCA-LDH/C electrode as the different ratio of Ni, Co, Al atom.

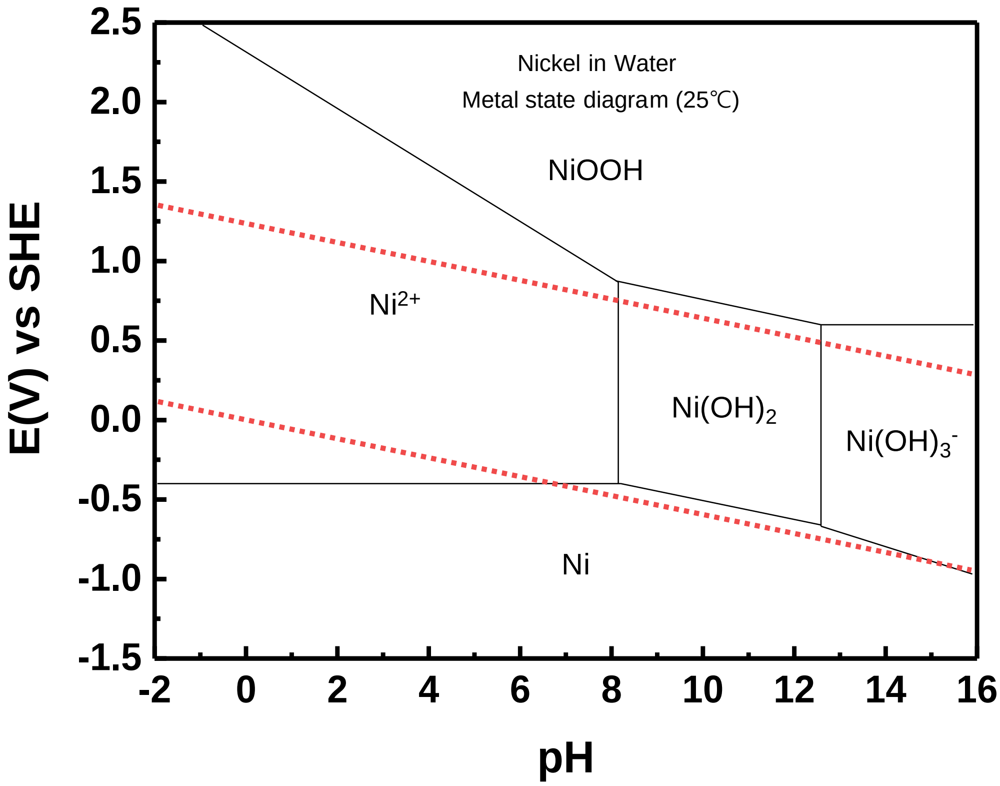


Fig. S18 Pourbaix diagram of $Ni(OH)_2$, which represents the relationship between the potential and pH.[1] The reduction potential of $Ni(OH)_2$ is dependent on the pH value (which corresponds to the concentration of NaOH solution), leading to the variable operating voltage of the Na/Ni battery as the concentration of NaOH.

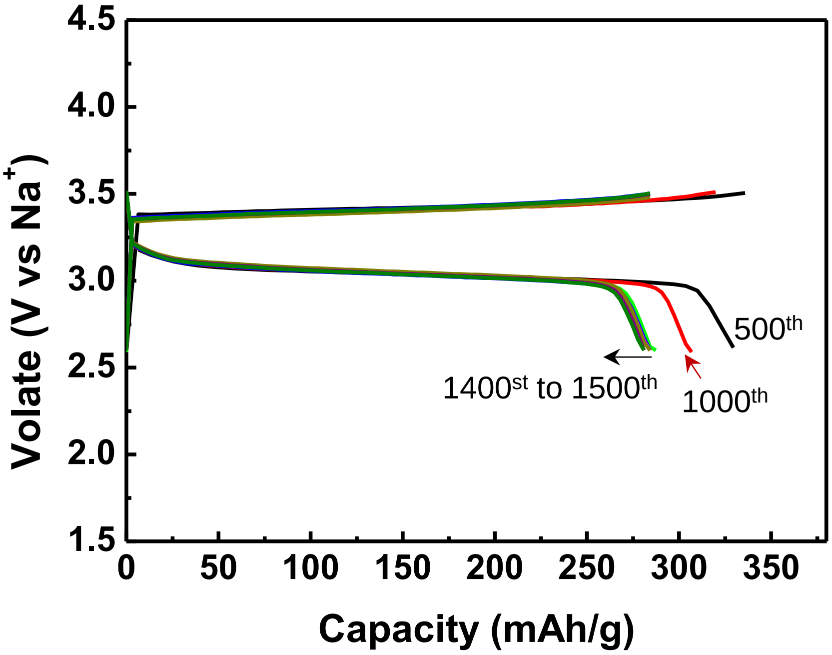


Fig. S19 Typical charge-discharge curves of Na-Ni battery during 1500 cycles.

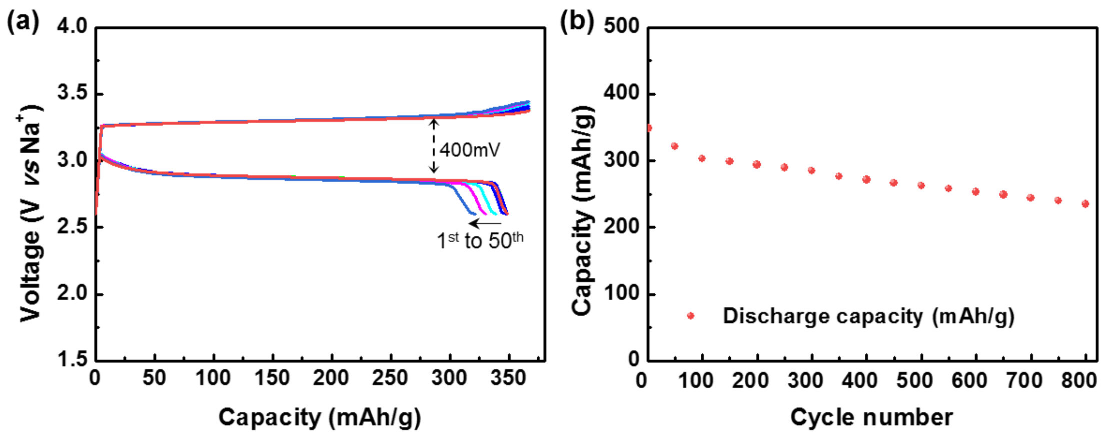


Fig. S20 (a) Charge-discharge profile and (b) cycle stability of Na-Ni battery in 5M NaOH.

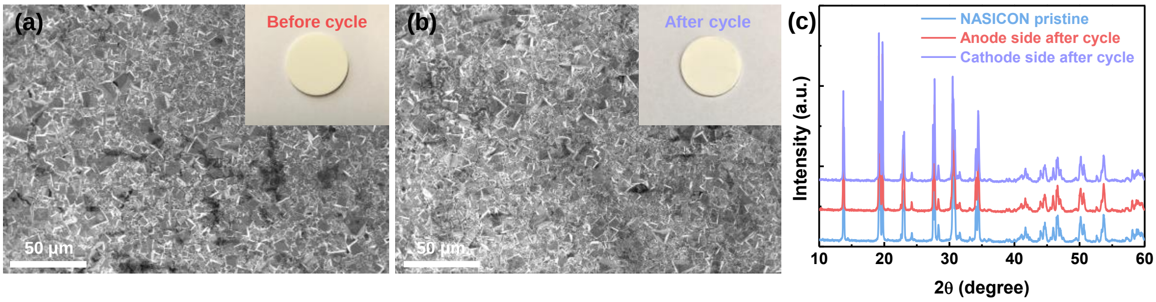


Fig. S21 Characteristics of NASICON after charge–discharge cycling. (a) SEM images of NASICON (a) before and (b) after cycle test of Na/Ni battery. The inset shows the photo images of NASICON. (c) XRD pattern of NASICON before and after cycle test.

Table S1. Comparative results for rechargeable battery systems.

	Anode	*Cathode*	*Voltage (V)*	*Capacity (mAh g^{-1})*	*Energy density (Wh kg^{-1})*	*Ref.*
Li-ion	Graphite	$LiCoO_2$	3.9	120	468	2
NiMH	Metal hydride	$Ni(OH)_2$	1.28	160	205	3
Aqueous Na-ion	$Na_{0.44}MnO_2$	Carbon	1.1	45	49.5	4
$TiO_2/Ni(OH)_2$	TiO_2	$Ni(OH)_2$	1.74	68.7	119.5	5
Fe/Ni	FeOx	β-$Ni(OH)_2$	1.04	115	120	6
Zn/Co	Zn	Co_3O_4	1.78	135	288	7
Zn/Mn	Zn	MnO_2	1.44	285	410	8
Na/Ni	Na	NiCoAl-LDH/C	3.1	350	**1085**	**This work**

Supplementary references

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