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.
**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) $\alpha$-Ni(OH)$_2$/C and (b) NiAl-LDH/C electrodes.
**Fig. S7** Comparative XRD results for (a) the $\alpha$-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 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.

<table>
<thead>
<tr>
<th>System</th>
<th>Anode</th>
<th>Cathode</th>
<th>Voltage (V)</th>
<th>Capacity (mAh g(^{-1}))</th>
<th>Energy density (Wh kg(^{-1}))</th>
<th>Ref.</th>
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<tbody>
<tr>
<td>Li-ion</td>
<td>Graphite</td>
<td>LiCoO(_2)</td>
<td>3.9</td>
<td>120</td>
<td>468</td>
<td>2</td>
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<tr>
<td>NiMH</td>
<td>Metal hydride</td>
<td>Ni(OH)(_2)</td>
<td>1.28</td>
<td>160</td>
<td>205</td>
<td>3</td>
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<tr>
<td>Aqueous Na-ion</td>
<td>Na(_{0.44})MnO(_2)</td>
<td>Carbon</td>
<td>1.1</td>
<td>45</td>
<td>49.5</td>
<td>4</td>
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<td>TiO(_2)/Ni(OH)(_2)</td>
<td>TiO(_2)</td>
<td>Ni(OH)(_2)</td>
<td>1.74</td>
<td>68.7</td>
<td>119.5</td>
<td>5</td>
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<tr>
<td>Fe/Ni</td>
<td>FeO(_x)</td>
<td>β-Ni(OH)(_2)</td>
<td>1.04</td>
<td>115</td>
<td>120</td>
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<tr>
<td>Zn/Co</td>
<td>Zn</td>
<td>Co(_3)O(_4)</td>
<td>1.78</td>
<td>135</td>
<td>288</td>
<td>7</td>
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<tr>
<td>Zn/Mn</td>
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<td>MnO(_2)</td>
<td>1.44</td>
<td>285</td>
<td>410</td>
<td>8</td>
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<td>Na/Ni</td>
<td>Na</td>
<td>NiCoAl-LDH/C</td>
<td>3.1</td>
<td>350</td>
<td><strong>1085</strong></td>
<td>This work</td>
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Supplementary references


