Supporting Information for

A novel stalactiform structural CoNi-rGO for supercapacitors with enhanced
electrochemical performance

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S1 Experiment Characterization

The X-ray powder diffraction (XRD, MiniFlex600) was conducted on a D/teX Ultra2
diffractometer. Field emission scanning electron microscopy (FESEM) images were obtained on
a Hitachi S-4800 field emission scanning electron microscope with an operating voltage of 5 kV.
Transmission electron microscopy (TEM) and HAADF-STEM-EDX mapping images were
obtained on a Hitachi HT7700 transmission electron microscope, employing an accelerating
voltage of 120 kV. X-ray photoelectron spectroscopy (XPS) of the product was carried out using
a Thermo ESCALAB 250 instrument, employing monochromatic Al Kα (hv = 1486.6 eV) at a
power of 150 W. Nitrogen adsorption-desorption isotherms were measured at the liquid nitrogen
temperature (77 K), using a Micromeritics ASAP 2460 analyzer. Surface areas were calculated
by the Brunauer-Emmett-Teller (BET) method. Operando Raman measurements were recorded
using a Raman spectrometer (RENISHAW in Via system, U.K.) with a 532 nm He/Ne laser as the excitation source.

S2 Electrochemical Measurements

The electrochemical performance measurements were carried out by CHI 660E electrochemical workstation (Chenhua Instruments, China). In a three-electrode system, the samples were adopted directly as the working electrodes (1 × 1 cm²), Hg/HgO and Pt wires as reference and counter electrodes, respectively. Cyclic voltammetry (CV, 0 - 0.7 V), electrochemical impedance spectroscopy (EIS, at open circuit potential with an amplitude of 5 mV, 10⁵ - 0.01 Hz) and galvanostatic charge/discharge (GCD, 0 - 0.5 V) measurements were examined in 2 M KOH aqueous electrolytes. At different current densities, the specific gravimetric capacitance (Cₛ, or F g⁻¹) of the hybrid battery was calculated by equation (S1): [1]

\[ C_s = \frac{i \times \Delta T}{m \times \Delta V} \]  
(S1)

where \( m \) (g) is the mass of the active material, \( i \) (A) is the constant discharging current, \( \Delta T \) (s) is the duration of the discharge process and \( \Delta V \) (V) is the voltage window.

An asymmetric supercapacitor (ASC) was prepared using S-CoNi-rGO as the positive electrode, the activated carbon (AC) as the negative electrode, and glass fiber paper as the separator. The electrochemical test was carried out in 2 M KOH electrolyte. The AC electrode was made by mixing active materials, acetylene black and polyvinylidene fluoride with a mass ratio of 7:2:1 on a nickel foam and then drying at 60 °C for 24 h.

The charge balance of the supercapacitor follows \( q^+ = q^- \), where \( q^+ \) and \( q^- \) represent the charges stored by the positive and negative electrodes respectively. The optimized mass ratio
between the positive and negative electrode materials for S-CoNi-rGO//AC can be calculated based on the charge balance relationship \((q^+ = q^-)\) in the following equation: [2,3]

\[
\frac{m^+}{m^-} = \frac{C^- \Delta V^-}{C^+ \Delta V^+}
\]

(S2)

where \(C^{+/−} (\text{F g}^{-1})\) are the specific capacitances of positive and negative electrodes, \(\Delta V^{+/−} (\text{V})\) are the potential windows of positive and negative electrodes, and \(m^{+/−} (\text{g})\) stands for the loading masses of materials on the positive and negative electrodes.

The energy density \((E, \text{Wh kg}^{-1})\) and power density \((P, \text{kW kg}^{-1})\) were obtained from GCD curves according to the following formula: [4]

\[
E = \frac{C_s \Delta V^2}{7.2}
\]

(S3)

\[
P = \frac{3600E}{\Delta t}
\]

(S4)

where \(C_s (\text{F g}^{-1})\) represents the specific capacitance, \(\Delta V (\text{V})\) stands for the potential window, \(\Delta t (\text{s})\) stands for the time of discharging, respectively.
S3 Supplementary Tables and Figures

Fig. S1. XPS spectra of CoNi-rGO, S-CoNi-rGO for C 1s.

Fig. S2. Raman spectra of S-CoNi-rGO.
Fig. S3. N\textsubscript{2} adsorption/desorption isotherms and pore-size (inset) distributions of (a) CoNi-1, (b) CoNi-F, (c) CoNi-rGO, (d) S-CoNi-rGO, respectively.
Fig. S4. (a-c) CV curves at different scan rates, (d-f) GCD curves at different current densities, (g-i) the cycling performance at various current densities of CoNi-1, CoNi-F, CoNi-rGO.
Fig. S5. The cycling stability for CoNi-1, CoNi-F, CoNi-rGO and S-CoNi-rGO at current density of 10 A g$^{-1}$.

Fig. S6. The SEM images of S-CoNi-rGO after 10000 recycles tests.
Fig. S7. The SEM images of S-CoNi-rGO // AC ASC device after 5000 recycles tests.

Fig. S8. (a-h) LED-driven tests during 120 min.
Table S1. The specific surface area and pore volume of CoNi-1, CoNi-F, CoNi-rGO, S-CoNi-rGO.

<table>
<thead>
<tr>
<th>Materials</th>
<th>CoNi-1</th>
<th>CoNi-F</th>
<th>CoNi-rGO</th>
<th>S-CoNi-rGO</th>
</tr>
</thead>
<tbody>
<tr>
<td>Specific surface area (m$^2$ g$^{-1}$)</td>
<td>51.94</td>
<td>26.88</td>
<td>69.59</td>
<td>32.52</td>
</tr>
<tr>
<td>Pore volume (cm$^3$ g$^{-1}$)</td>
<td>0.119</td>
<td>0.110</td>
<td>0.175</td>
<td>0.132</td>
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</table>

Table S2. Comparison of S-CoNi-rGO//AC device electrochemical performances with those reported literatures.

<table>
<thead>
<tr>
<th>Hybrid battery</th>
<th>Electrolyte</th>
<th>Energy density W h kg$^{-1}$</th>
<th>Power density kW kg$^{-1}$</th>
<th>Ref.</th>
</tr>
</thead>
<tbody>
<tr>
<td>Co$_3$O$_4$@Ni(OH)$_2$ //AC</td>
<td>PVA:KOH</td>
<td>40.0</td>
<td>3.45</td>
<td>[5]</td>
</tr>
<tr>
<td>NiCo-MOF//AC</td>
<td>PVA:KOH</td>
<td>45.3</td>
<td>0.85</td>
<td>[4]</td>
</tr>
<tr>
<td>NiCo$_2$O$_4$@Ni(OH)$_2$//AC</td>
<td>2M KOH</td>
<td>98.5</td>
<td>0.85</td>
<td>[6]</td>
</tr>
<tr>
<td>CuCo HNSs/NF//AC</td>
<td>1M KOH</td>
<td>144.4</td>
<td>40</td>
<td>[7]</td>
</tr>
<tr>
<td>CF@CuO@CoNi LDH//RGO</td>
<td>2M KOH</td>
<td>92.5</td>
<td>0.4</td>
<td>[8]</td>
</tr>
<tr>
<td>Ni(OH)$_2$/CNT//AC</td>
<td>3M KOH</td>
<td>32.5</td>
<td>1.8</td>
<td>[9]</td>
</tr>
<tr>
<td>NiCo$_2$S$_4$@Ni(OH)$_2$//AC</td>
<td>2M KOH</td>
<td>53.3</td>
<td>0.29</td>
<td>[10]</td>
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<tr>
<td>Co(OH)$_2$/fCNT</td>
<td></td>
<td>fCNT</td>
<td>1M KOH</td>
<td>17</td>
</tr>
<tr>
<td>NMCOH-2.4//NDG</td>
<td>3 M KOH</td>
<td>92</td>
<td>1.7</td>
<td>[12]</td>
</tr>
<tr>
<td>S-CoNi-rGO //AC</td>
<td>2 M KOH</td>
<td>114.03</td>
<td>0.85</td>
<td>This work</td>
</tr>
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</table>

Table S3. S-CoNi-rGO//AC device electrochemical performances at different current densities.

<table>
<thead>
<tr>
<th>Hybrid battery</th>
<th>The current density A g$^{-1}$</th>
<th>The capacitance F g$^{-1}$</th>
<th>Energy density Wh kg$^{-1}$</th>
<th>Power density kW kg$^{-1}$</th>
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<tbody>
<tr>
<td>S-CoNi-rGO//AC</td>
<td>1</td>
<td>284.1</td>
<td>114.03</td>
<td>0.854</td>
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<td>S-CoNi-rGO//AC</td>
<td>2</td>
<td>276.9</td>
<td>111.14</td>
<td>1.724</td>
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<tr>
<td>S-CoNi-rGO//AC</td>
<td>3</td>
<td>267.9</td>
<td>107.53</td>
<td>2.579</td>
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<tr>
<td>S-CoNi-rGO//AC</td>
<td>4</td>
<td>259.2</td>
<td>104.04</td>
<td>3.419</td>
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<tr>
<td>S-CoNi-rGO//AC</td>
<td>5</td>
<td>253.5</td>
<td>101.75</td>
<td>4.249</td>
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<tr>
<td>S-CoNi-rGO//AC</td>
<td>8</td>
<td>246.0</td>
<td>96.57</td>
<td>6.803</td>
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<tr>
<td>S-CoNi-rGO//AC</td>
<td>10</td>
<td>234.7</td>
<td>94.21</td>
<td>8.501</td>
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Supplementary References


