Supporting Information

Rational construction of core-shell Ni$_3$S$_2$@Ni(OH)$_2$ nanostructure as the battery-like electrode for supercapacitors

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**Fig. S1** SEM images of Ni$_3$S$_2$@Ni(OH)$_2$ samples with different immersion time: (a) 5 min, (b) 10 min, (c) 15 min, (d) 20 min.
Fig. S2 CV and GCD curves of Ni$_3$S$_2$@Ni(OH)$_2$ samples with different immersion time: (a, b) 5min, (c, d) 10 min and (e, f) 20min. (g) The GCD curves at a current density of 2 mA cm$^{-2}$ for Ni$_3$S$_2$@Ni(OH)$_2$ samples with different immersion time. (h) The areal capacitance of Ni$_3$S$_2$@Ni(OH)$_2$ samples with different immersion time at 2 mA cm$^{-2}$.

For comparison, Ni$_3$S$_2$@Ni(OH)$_2$ samples with different immersion time (5, 10, 15, 20 min) also prepared. Firstly, the electrochemical performances of Ni$_3$S$_2$@Ni(OH)$_2$ samples with different immersion time (5, 10, 15, 20 min) were compared. Fig. S1 shows the SEM images of Ni$_3$S$_2$@Ni(OH)$_2$ samples with different immersion time. As the immersion time increases, the surface of Ni$_3$S$_2$ nanorods is gradually covered by Ni(OH)$_2$ nanosheets. Undoubtedly, the size of Ni(OH)$_2$ nanosheets as shell can be adjusted by controlling the immersion time. Fig. S2g shows the GCD curves of Ni$_3$S$_2$@Ni(OH)$_2$ samples with different immersion time. Obviously, Ni$_3$S$_2$@Ni(OH)$_2$-15min electrode showed the longest discharge time at 2 mA cm$^{-2}$, indicating the highest specific capacitance. According to the above results, the following performance tests and discussion were focused on the Ni$_3$S$_2$@Ni(OH)$_2$ obtained with 15 min immersion time.
Fig. S3 The corresponding current density \( i^{1/2} \)–\( v^{1/2} \) (scan rate\(^{1/2} \)) plots of \( \text{Ni}_3\text{S}_2@\text{Ni(OH)}_2 \).

Fig. S4 CV and GCD curves of (a, b) \( \text{Ni}_3\text{S}_2 \) and (c, d) \( \text{Ni(OH)}_2 \).
Fig. S5 SEM images of Ni$_3$S$_2$@Ni(OH)$_2$ after 10000 cycles.

Fig. S6 The relationship between \( Z' \) and \( \omega^{-1/2} \) in the low-frequency region of Ni$_3$S$_2$@Ni(OH)$_2$ and Ni$_3$S$_2$.

According to previous researches [1-4], the OH$^-$ ion diffusion coefficient (\( D_{OH^-} \), cm$^2$ s$^{-1}$) was calculated by the following equation:

\[
D_{OH^-} = \frac{(RT)^2}{2(A n^2 F^2 C_{OH^-} \sigma)^2}
\]

where \( R \), \( T \), \( A \), \( n \), \( F \), \( C_{OH^-} \) and \( \sigma \) represent the gas constant, absolute temperature, electrode area, number of electrons transfer, Faraday constant, the OH$^-$ concentration and Warburg factor. And \( \sigma \) could be calculated by the slope of line in the low frequency region, according to the following equation [4,5]:

\[
Z' = R_e + R_{ct} + \sigma
\]

Based on above equation, the \( D_{OH^-} \) has been calculated and the results are shown in Fig. S6. As a result, the calculated \( D_{OH^-} \) of Ni$_3$S$_2$@Ni(OH)$_2$ is larger than that of pristine Ni$_3$S$_2$, suggesting the faster ion transport.
In order to investigate influence of crystallinity on the electrochemical performances, the obtained Ni$_3$S$_2$@Ni(OH)$_2$ was annealed at Ar atmosphere in 150 °C for 2h (labeled as Ni$_3$S$_2$@Ni(OH)$_2$-Ar). As shown in Fig. S7a-b, SEM images suggested that the morphology and structure of Ni$_3$S$_2$@Ni(OH)$_2$-Ar show no obvious changes. As shown in Fig. S7c, XRD result shows that the peaks of Ni(OH)$_2$ become obvious and sharp after annealing process, suggesting the improvement of crystallinity of the Ni(OH)$_2$.

Fig. S7d-S7f show the electrochemical performance of Ni$_3$S$_2$@Ni(OH)$_2$-Ar. As a results, Ni$_3$S$_2$@Ni(OH)$_2$-Ar shows the lower capacitance than that of pristine Ni$_3$S$_2$@Ni(OH)$_2$, only 2.37 F cm$^{-2}$ at 2 mA cm$^{-2}$. According to previous results [6-8], electrodes with low crystallinity could be able to achieve better electrochemical performances than that of crystalline counterparts, due to more structure disorder and defects.
Fig. S8 (a and b) CV and GCD curves of AC at various scan rate and (b) corresponding specific capacitance.

Fig. S9 (a) The specific capacitance vs. current density of Ni$_3$S$_2$@Ni(OH)$_2$//AC, (b) Cycling performance of Ni$_3$S$_2$@Ni(OH)$_2$//AC, which was determined by GCD tests at the current density of 10 A g$^{-1}$ for 5000 cycles.
Table S1  The specific capacitance of various electrodes in the three-electrode system in references

<table>
<thead>
<tr>
<th>Electrode materials</th>
<th>Electrolyte</th>
<th>Specific capacitance</th>
<th>Potential Windows (V)</th>
<th>Reference</th>
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<tbody>
<tr>
<td>Surface-enriched Ni-Co-S/Graphene</td>
<td>6 M KOH</td>
<td>1436 F·g⁻¹</td>
<td>-0.05-0.55</td>
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<tr>
<td>Ni₃S₂@β-NiS</td>
<td>6 M KOH</td>
<td>1158 F·g⁻¹</td>
<td>-0.2-0.5</td>
<td>10</td>
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<td>Nickel sulfides/MoS₂-CNT</td>
<td>3 M KOH</td>
<td>757 F·g⁻¹</td>
<td>0-0.55</td>
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<tr>
<td>NiS₂ hollow sphere</td>
<td>2 M KOH</td>
<td>1643 F·g⁻¹</td>
<td>0-0.7</td>
<td>12</td>
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<tr>
<td>Ni₃S₂@CdS</td>
<td>3 M KOH</td>
<td>3.15 F·cm⁻²</td>
<td>0-0.5</td>
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<td>NiS nanosheets array</td>
<td>1 M NaOH</td>
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<td>0-0.8</td>
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<td>NiS/carbon aerogel</td>
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<td>1606 F·g⁻¹</td>
<td>0-0.5</td>
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<td>graphene-wrapped nickel sulfide nanoprisms</td>
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<td>1495 F·g⁻¹</td>
<td>0-0.5</td>
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<td>NiMoO₄@Ni-Co-S</td>
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<td>2.27 F·cm⁻²</td>
<td>0-0.65</td>
<td>17</td>
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<td>Co₃O₄@CdS</td>
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<td>Hierarchical carbon@Ni₃S₂@MoS2</td>
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<td>0-0.4</td>
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<td>Ni₃S₂@Ni(OH)₂</td>
<td>6M KOH</td>
<td>1775 F·g⁻¹</td>
<td>0-0.5 (3.55 F·cm⁻²)</td>
<td>This work</td>
</tr>
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Reference: