Supporting Information

Binder-free Ni$_3$S$_2$@PANI Core-Shell Nanosheets as Durable and High-Energy Cathode for Aqueous Rechargeable Nickel-Zinc battery

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Calculations:

The loading mass $m$ (mg cm$^{-2}$) of the Ni$_3$S$_2$ on nickel foam was evaluated by a method that a piece of Ni$_3$S$_2$ sample (cut into 0.5 cm $\varnothing$ 2 cm) was calcined in the H$_2$/Ar (or H$_2$/N$_2$) atmosphere (10 : 90 by volume) at 500 °C for 4 h, then weighed the loss mass and calculated the weigh of Ni$_3$S$_2$ from the following equation 1-3:

$$m = \frac{\Delta m \times M_{Ni_3S_2}}{2M_S}$$  \hspace{1cm}  (1)

Where $\Delta m$ is the weight loss after calcining obtained by electronic scales (BT25S, 0.01 mg), $M_{Ni_3S_2}$ and $M_S$ are the molecular weight of Ni$_3$S$_2$ and S, respectively.

The specific capacity $C$ (mA h g$^{-1}$) of the battery was calculated from the
discharge curve with the equations as followed:

\[ C = \frac{\int_{0}^{\Delta t} I \times dt}{m} \quad (2) \]

Where \( I \) (mA cm\(^{-2}\)) is the applied discharging current density, \( \Delta t \) (h) is the discharging time and \( m \) (mg cm\(^{-2}\)) is the mass loading of the electrodes (3.5 mg cm\(^{-2}\)).

The mass energy density \( E \) and mass power density \( P \) of the battery were obtained from the following equations:

\[ E = C \times \Delta V \quad (3) \]

\[ P = \frac{C \times \Delta V}{1000 \times \Delta t} \quad (4) \]

Where \( E \) (Wh kg\(^{-1}\)) is the energy density, \( C \) is the mass capacity obtained from Equation (1), (2) and \( \Delta V \) (V) is the discharging voltage. \( P \) (kW kg\(^{-1}\)) is the specific power density and \( \Delta t \) (h) is the discharging time.

\[ \text{Figure S1.} \ XRD \text{ patterns of pristine Ni}_{3}\text{S}_{2}, \text{ Ni}_{3}\text{S}_{2}@PANI-4, \text{ Ni}_{3}\text{S}_{2}@PANI-6 \text{ and } \text{Ni}_{3}\text{S}_{2}@PANI-8 \text{ electrodes.} \]
Figure S2. SEM images of the (a) Ni$_3$S$_2$@PANI-4 and (b) Ni$_3$S$_2$@PANI-8 electrodes (lower insets).

Figure S3. (a) CV curves at scaning rate of 10 mV s$^{-1}$, (b) GCD curves at 14.2 A g$^{-1}$ of the Ni$_3$S$_2$, Ni$_3$S$_2$@PANI-4, Ni$_3$S$_2$@PANI-6 and Ni$_3$S$_2$@PANI-8 electrodes.

Figure S4. SEM images of Ni$_3$S$_2$@PANI-6 electrode after 10000 cycles.
Figure S5. (a) CV curves at a scan rate of 10 mV s\(^{-1}\), (b) GCD curves at 14.2 A g\(^{-1}\) of NF@PANI and Ni\(_3\)S\(_2\)@PANI-6 electrodes.

Figure S6. The rate performance of Ni\(_3\)S\(_2\)@PANI//Zn battery.
Figure S7. GCD curves at 5.1 A g$^{-1}$ of Ni$_3$S$_2$@PANI//Zn battery in repeated experiments.

Table S1. The average value of capacities from three experiments (Figure S7).

<table>
<thead>
<tr>
<th>No.</th>
<th>Original</th>
<th>Repeat 1</th>
<th>Repeat 2</th>
<th>Average</th>
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<tr>
<td>Capacity (mAh g$^{-1}$)</td>
<td>241.8</td>
<td>232.5</td>
<td>254.1</td>
<td>242.8 ± 8.8</td>
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Figure S8. EIS spectra of Ni$_3$S$_2$@PANI//Zn battery before and after 5000 cycles.
Figure S9. CV curves at various scan rates of (a) Ni$_3$S$_2$@PANI//Zn, (b) Ni$_3$S$_2$/Zn, and log (i) versus log (v) plots at specific peak currents.

References