## **Supporting Information**

## Novel hierarchical NiS/N-doped carbon composite hollow spheres as an enhanced-performance electrode for hybrid supercapacitors

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## **Calculation equations**

For the three-electrode system, the specific capacitance,  $C_{s-CD}$  (F g<sup>-1</sup>), of the working electrode can be obtained from the GCD curves based on the following equations:

$$C_{s-CD} = \frac{I \times \Delta t}{m \times \Delta V} \tag{1}$$

Where *I* is the charge-discharge current (A),  $\Delta V$  is the width of potential window (V),  $\Delta t$  is the time of discharge (s), and *m* is the loading mass of active material (g).

For ASCs, charge balance is required to optimize the capacitive performance. Generally, the charge balance  $(q^+=q^-)$  is decided based on the capacitive performance of each electrode. The mass balancing is valuated from Equation 3 according to the specific capacitance  $(C_s)$  and potential range  $(\Delta V)$ .

$$q = m \times C_s \times \Delta V \tag{2}$$

$$\frac{m_+}{m_-} = \frac{C_{s-} \times \Delta V_-}{C_{s+} \times \Delta V_+} \tag{3}$$

Where  $m_+$  is the anode active-material mass and  $m_-$  is the cathode active-material mass.

Total capacitance, C (F g<sup>-1</sup>), energy density, E (Wh kg<sup>-1</sup>), and power density, P (W kg<sup>-1</sup>), of ASCs are determined by the following equations:

$$C = \frac{I \times \Delta t}{M \times V} \tag{4}$$

$$E = \frac{1}{2} \times \frac{1}{3.6} \times C \times V^2 \tag{5}$$

$$P = 3600 \times \frac{E}{\Delta t} \tag{6}$$

Where V is the operating voltage window (V),  $\Delta t$  is the time of discharge (s), and M is the total active material mass of these two electrodes (g).



Fig. S1. FESEM images of N-carbon/SiO<sub>2</sub> spheres (a and b) and NiSi/NHCS/SiO<sub>2</sub> (c and d).



Fig. S2. FESEM images of NiSi hollow spheres



Fig. S3. (a)  $N_2$  adsorption/desorption isotherm and (b) Pore size distribution of NiS HS and NiS/NHCS.



Fig. S4. XPS survey scan (a), C 1s (b) and N1s (c) high-resolution XPS spectrum for N-carbon/SiO<sub>2</sub>.



Fig. S5. XPS survey spectra for NiS/NHCS.



Fig. S6. XPS survey scan (a) and O1s (b) high-resolution XPS spectrum for NHCS.



**Fig. S7.** GCD and CV curves of NiS/NHCS (a and b) and NiS HS (c and d) measured at various current densities.



Fig. S8. Cycling stability tests of NiS/NHCS and NiS HS electrodes at 5 A g<sup>-1</sup>.



Fig. S9 (a) CV curves and (b) GCD curves of activated carbon electrode in a three-electrode system.



Fig. S10 (a) GCD curves (b) and CV curves of the NiS/NHCS||AC| device.



**Fig. S11** (a) CV curves of AC and NiS-HS from -1.0 to 0 V and 0 to 0.5 V at 10 mV s<sup>-1</sup> in a three-electrode system, respectively. (b) CV curves in various operation voltages at a scan rate of 10 mV s<sup>-1</sup>, (c) GCD curves and (d) CV curves of the NiS-HS||AC device.



Fig. S12. Cycling stability of NiS/NHCS||AC device at 3 A  $g^{-1}$ .



Fig. S13. TEM images of NiS/NHCS after 5000 cycles at 3 A g<sup>-1</sup>.