Electronic supplementary information

α -Ni(OH)₂/NiS_{1.97} heterojunction composites with excellent ion and

electron transport properties for advanced supercapacitors

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Electrochemical measurements.

The assemblage of supercapacitor devices.

The working electrode was prepared by mixing active materials (such as, NO, NO/NS-4, NO/NS-8 or NO/NS-10) (80 wt%) with acetylene black (10 wt%) and polyvinylidene fluoride (PVDF) (10 wt%) in mixed solution of absolute alcohol and isopropanol with a 1:1 volume ratio. The resulting slurry was then coated onto a piece of nickel foam (1 cm × 1 cm). The electrode was dried at 60 °C for 12 h in a vacuum oven. The counter electrode was prepared by using the similar methods, but changing the composition proportion of the slurry. The compositions of slurry for the counter electrode were 80 wt% active carbon (AC) and 20 wt% PVDF. The as-obtained working electrodes contained ~5 mg of active materials. The amount of AC in the counter electrode was ~15 mg. The working electrode and the AC electrode were put together and separated by a piece of non-woven separator and soaked in 2 M KOH aqueous solution. The energy storage performances of the as-assembled supercapacitor devices were evaluated by cyclic voltammetry (CV) and galvanostatic charge/discharge test (GCD).

Calculations.^{S1, S2}

(1) The specific capacitance for the working electrode at three-electrode system from CV curves as follow:

$$C_{electrode} = Q/(m\Delta V) \tag{1}$$

where $C_{electrode}$ is the specific capacitance of the working electrode in farads per gram, Q is quantity of electric charge during discharge process in coulomb, m indicates the mass of the active materials on both electrodes in grams and ΔV is the potential window after removing the voltage drop in volts. (2) The total capacitance for a supercapacitance device is calculated from GCD curves according to Equation (2):

 $C_{device} = I\Delta t_d / \Delta V \tag{2}$

where C_{device} is the total capacitance for a device in farads, *I* is the discharge current in amperes, Δt_d is the discharge time in second, and ΔV is the potential window after removing the voltage drop in volts. (3) Specific capacitance for a device is calculated based on the mass according to Equation (3):

$$C_{specific} = C_{device}/m$$

where $C_{specific}$ is the specific capacitance for a device in farads per gram, and m indicates the total mass of the active materials on both electrodes in grams.

(3)

(4)

(6)

(7)

(4) The specific capacitance of a single electrode was calculated by using Equation (4):

$$C_{electrode} = 4 \times C_{specific}$$

where $C_{electrode}$ is the specific capacitance of a single electrode in farads per gram.

(5) The energy density (E) and power density (P) of the device was obtained using Equations (5) and (6), respectively.

$E = 0.5 \times C_{specific} \times \Delta V^2$	(5)
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 $P = 3600 \times E/\Delta t_d$

where E is the energy density in watt hour per kilogram and P is the power density in watts per kilogram.

(6) Coulomb efficiency of electrode was calculated using Equation (7):

 $\eta = \Delta t_d / \Delta t_c$

where η is the Coulomb efficiency, and Δt_c is the charge time in second.



Figure S1. Morphology, structure, phase and elementary distribution characterization of peony-like NO/NS-2 material. (a-b) SEM images with different magnification. (c) XRD curves of NO and NO/NS-2 materials. (d) EDS mapping images of Ni, O, S, N and C.



Figure S2. Morphology, structure, phase and elementary distribution characterization of peony-like NO/NS-4 material. (a-b) SEM images with different magnification. (c) XRD curves of NO and NO/NS-4 materials. EDS mapping images of Ni, O, S, N and C at (d) low and (e) high magnification.



Figure S3. Morphology, structure, phase and elementary distribution characterization of peony-like NO/NS-6 material. (a-b) SEM images with different magnification. (c) XRD curves of NO and NO/NS-6 materials. (d) EDS mapping images of Ni, O, S, N and C.



Figure S4. CV curves of (a) peony-like NO microsphere, (b) NO/NS-4 heterojunction materials, (c) NO/NS-8 heterojunction materials, (d) NO/NS-10 nanocuboids at the scan rates of 10 mV s⁻¹ for 350 cycles in three-electrode system. (e) Cycle life curves of NO, NO/NS-4, NO/NS-8 and NO/NS-10 devices at 5 A g⁻¹ for 2500 cycles in three-electrode system. (f) Photo of the three-electrode system after cycle test.



Figure S5. The over-lapped CV curves of active carbon electrode and NO, NO/NS-4, NO/NS-8 and NO/NS-10 electrodes in three electrode system at a scan rate of 5 mV s⁻¹.

References

[S1] E. Frackowiak, V. Khomenko, K. Jurewicz, K. Lota and F. Béguin, *J. Power Sources*, 2006, **153**, 413.
[S2] N. Li, G. Z. Yang, Y. Sun, H. W. Song, H. Cui, G. W. Yang and C. X. Wang, *Nano Lett.*, 2015, **15**, 3195.