Electronic Supplementary Material (ESI) for Nanoscale.

Electronic Supplementary Information

Hierarchical bimetallic hydroxides/chalcogenides core-sheath microarrays for free-standing ultrahigh rate supercapacitors

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1. Materials characterizations

Morphologies of composites were observed at field-emission scanning electron microscopy (FESEM, JEOL JEM7800F) and low-resolution transmission electron microscope (LR-TEM, FEI Tecnai-12 at 120 kV). High-resolution transmission electron microscopy (HR-TEM) images were recorded on a FEI Tecnai F30 at an accelerating voltage of 80 kV. X-ray diffraction (XRD) patterns were obtained from Bruker D8 Advance Diffractometer by using the Cu Ka radiation (0.154 nm) at 40 kV and 30 mA. Raman spectroscopy (RM 2000 microscopic confocal Raman spectrometer) were performed by employing a 514 nm laser beam. X-ray photoelectron spectroscopy (XPS) analysis were carried out on a Thermo ESCALAB 250 X-ray photoelectron spectrometer with Al Kα radiation (1486.7 eV).

2. Electrochemical measurements

The three-electrode electrochemical performance was investigated in a 3 M KOH aqueous solution and the two-electrode HSCs were measured by using PVA/KOH gel as solid electrolyte. All the electrochemical performance were taken at the CHI760E electrochemical workstation (Shanghai, China). The areal capacitances of electrodes were measured by galvanostatic charge/discharge method according to the following equation:

$$C_A = \frac{I \times \Delta t}{S \times \Delta V} \qquad (1)$$

where C_A (F cm^{-2}) is the areal capacitance, I (A) is the constant discharging current, Δt is the discharging time, S (cm^2) is the surface area and ΔV (V) is the potential window.

The volumetric capacitance of the $Ni(OH)_2$-$Co(OH)_2$/NiSe-Ni_3S_2/NF//AC/NF HSCs were calculated from galvanostatic charge/discharge method according to the following equation:

$$C_V = \frac{I \times \Delta t}{V \times \Delta V} \qquad (2)$$

Where C_V (F cm^{-3}) is the volumetric capacitance, I (A) is constant discharging current, V (cm^3) is the volume of the whole device (The area and thickness of the $Ni(OH)_2$-$Co(OH)_2$/NiSe-Ni_3S_2/NF//AC/NF HSCs device is about 1.0 cm^2 and 0.12 cm. Hence, the whole volume of the HSCs device is ~ 0.12 cm^3), Δt (s) is the discharging time, ΔV (V) is the voltage window.

The volumetric energy density (E) and power density (P) of devices were calculated according to Equation (3)&(4):

$$E = \frac{1}{2 \times 3600} \times C_V \times \Delta V^2 \qquad (3)$$

$$P = \frac{E}{\Delta t} \qquad (4)$$

where E (Wh cm^{-3}) is the energy density, C_V is the volumetric capacitance obtained from Equation (2) and ΔV (V) is the voltage window of the two-electrode device. P (W cm^{-3}) is the power density. Δt (s) is the discharging time.

3. Morphology observation

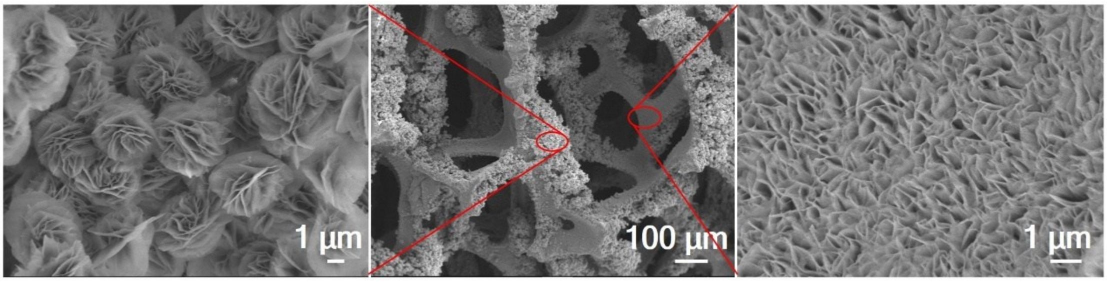

Fig. S1. FE-SEM images of $Ni(OH)_2$-$Co(OH)_2$/NF.

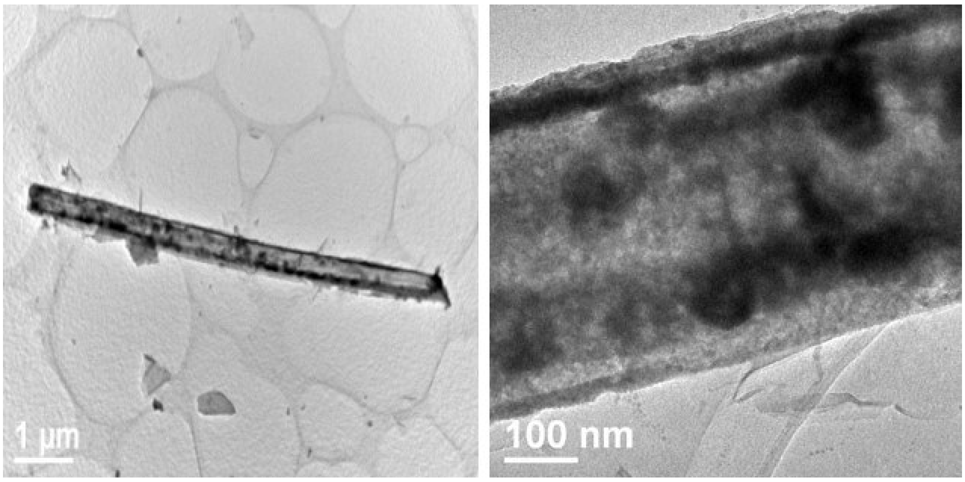

Fig. S2. TEM images of NiSe-Ni_3S_2/NF.

4.XRD pattern

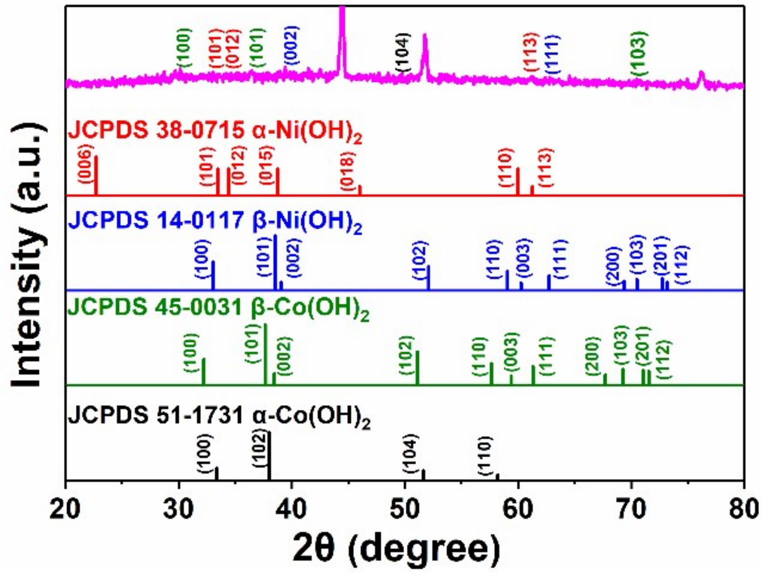

Fig. S3. XRD patterns of single $Ni(OH)_2$-$Co(OH)_2$/NF.

5. XPS spectra

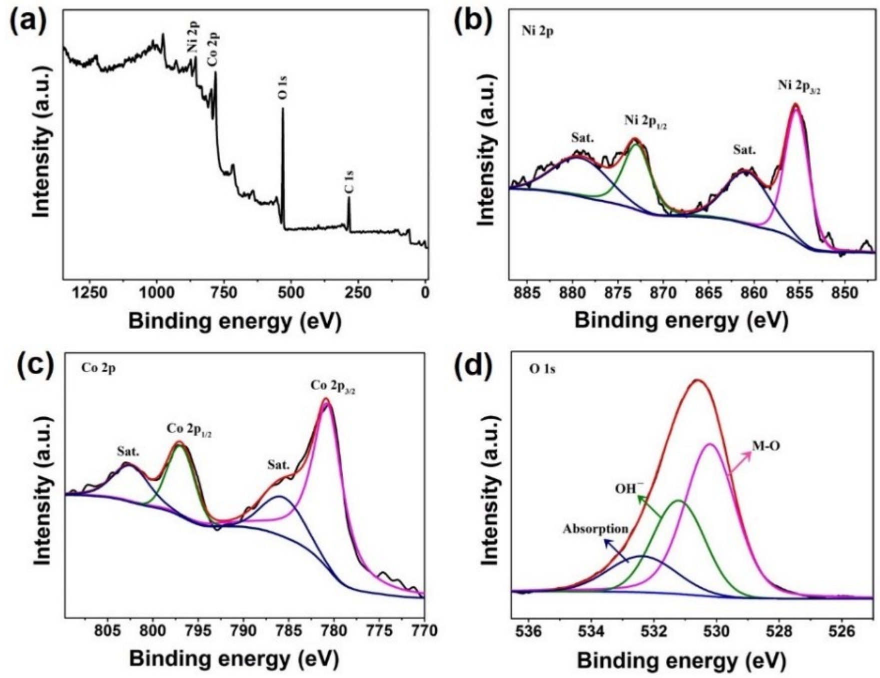

Fig. S4. (a) XPS full survey $Ni(OH)_2$-$Co(OH)_2$/NF and (c-d) corresponding spectra of Ni 2p, Co 2p and O 1s regions.

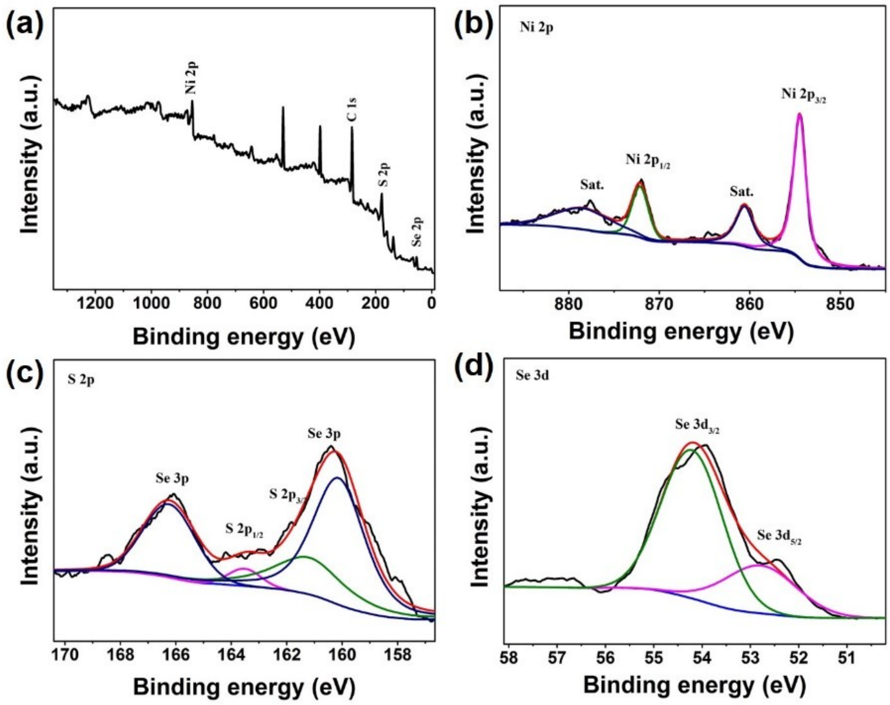

Fig. S5. (a) XPS full survey NiSe-$N_{i3}S_2$/NF and (c-d) corresponding spectra of Ni 2p, S 2p and Se 3d regions.

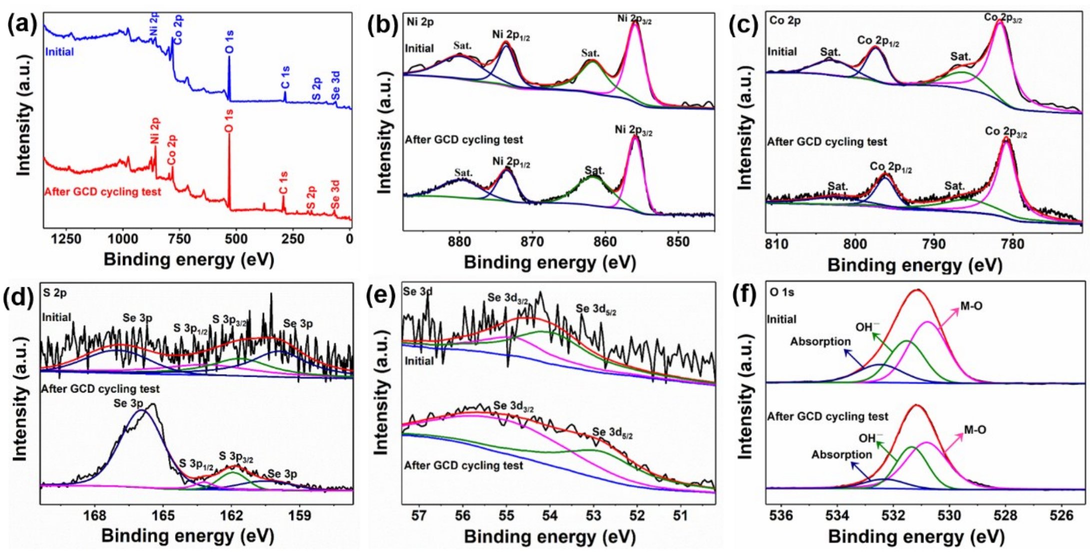

Fig. S6. (a) XPS full survey $Ni(OH)_2$-$Co(OH)_2$/NiSe-Ni_3S_2/NF before (above) and after 2500 galvanostatic charge-diacharge (GCD) cycles (below), and (c-d) corresponding spectra of Ni 2p, Co 2p, S 2p, Se 3d, and O 1s regions.

6.BET results

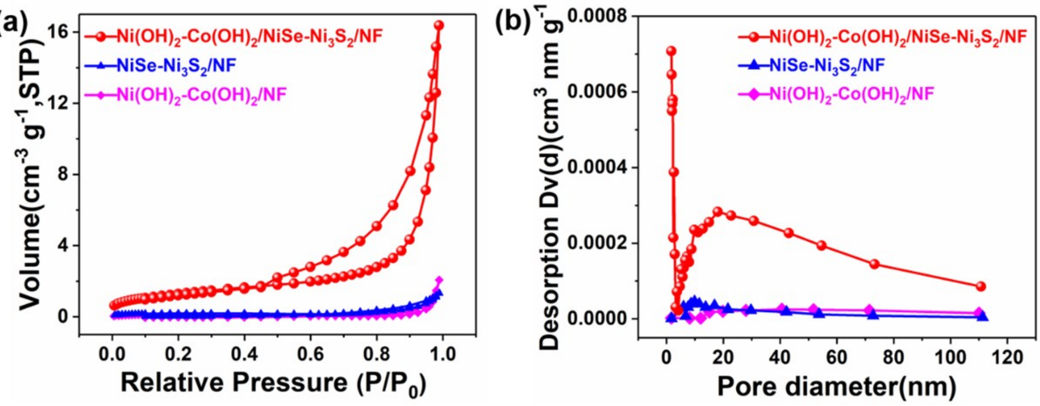

Fig. S7. N_2 adsorption-desorption isotherms and pore size distributions (inset) of $Ni(OH)_2$-$Co(OH)_2$/NF, NiSe-Ni_3S_2/NF and $Ni(OH)_2$-$Co(OH)_2$/NiSe-$N_{i3}S_2$/NF.

7. Electrochemical studies

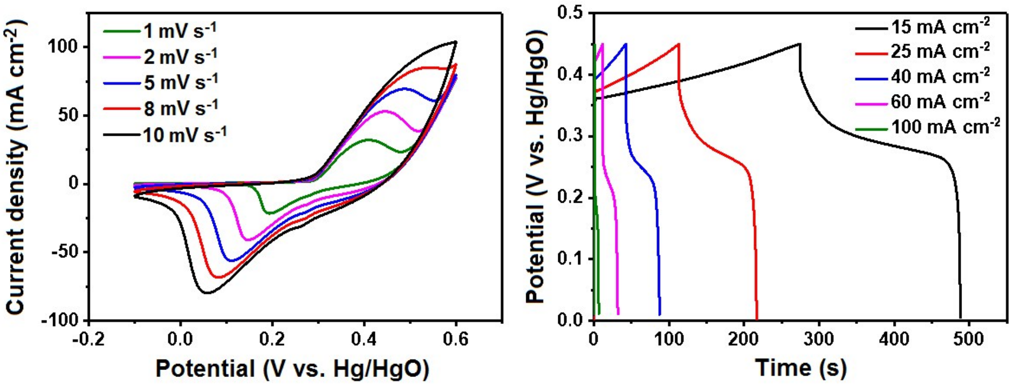

Fig. S8. CV and GCD curves of NiSe-Ni_3S_2/NF.

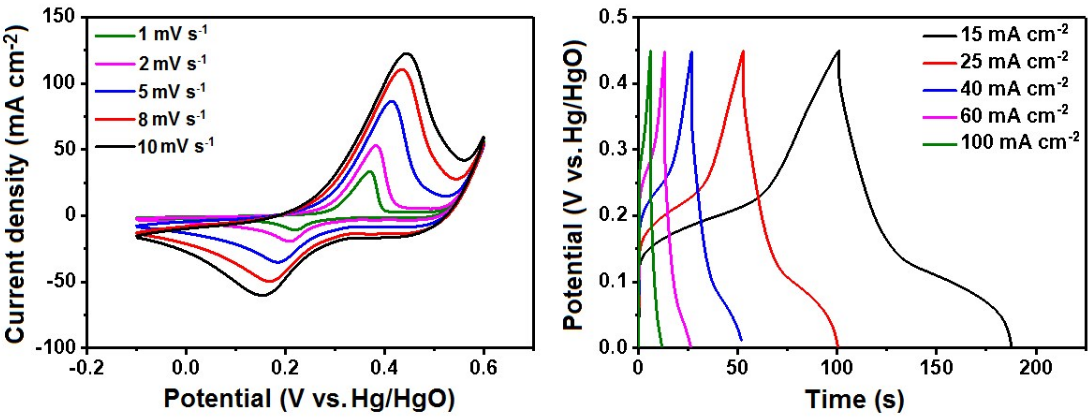

Fig. S9. CV and GCD curves of $Ni(OH)_2$-$Co(OH)_2$/NF.

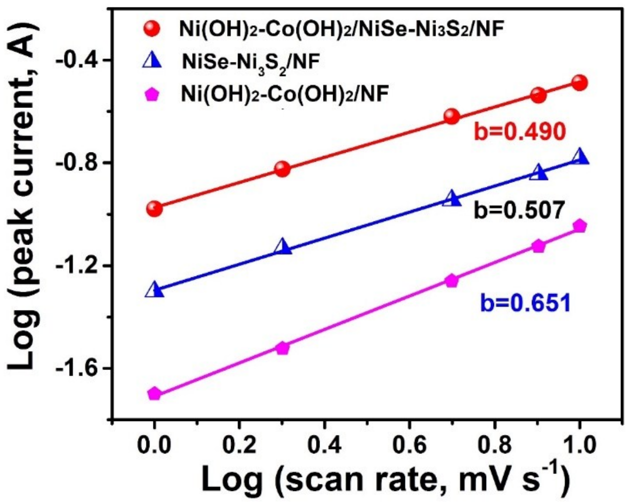

Fig. S10. Logarithmic peak current against logarithmic scan rate of 1-10 mV s-1.

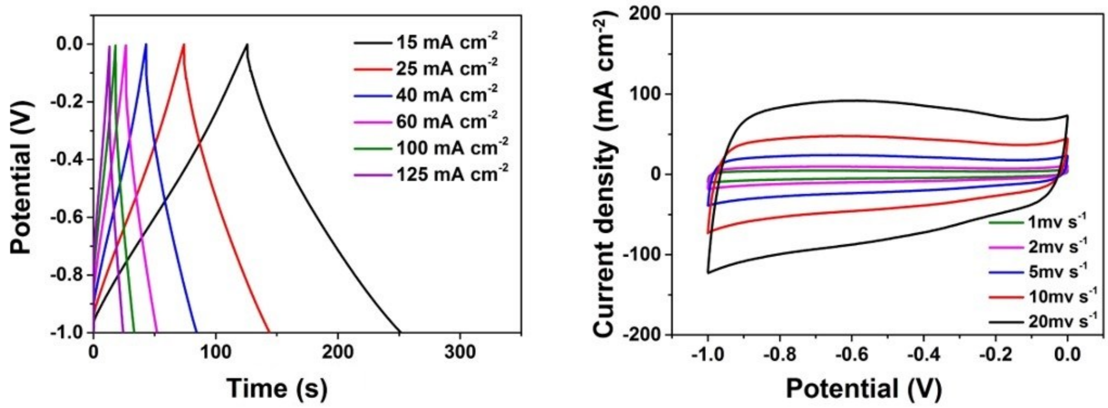

Fig. S11. CV and GCD curves of AC electrode.

Table S1. Comparison of electrochemical performance of representative free-standing electrodes on NF

Material	Specific capacitance	Current density	Reference
$NiCo_2S_4$@$Ni(OH)_2$@PPy	9.1125 F cm^{-2}	5 mA cm^{-2}	[1]
NiO nanosheets	2.01 F cm^{-2}	8 mA cm^{-2}	[2]
Ni–Tp/PANI	10.327 F cm^{-2}	20 mA cm^{-2}	[3]
Ni@NiO Nanowires	3.99 F cm^{-2}	8 mA cm^{-2}	[4]
$Ni(OH)_2$-Cu	8.66 F cm^{-2}	1 mA cm^{-2}	[5]
Ni_3S_2	4.52 F cm^{-2}	1.25 mA cm^{-2}	[6]
$Co(OH)_2$/HNNF	3.17 F cm^{-2}	5 mA cm^{-2}.	[7]
$NiCo_2O_4$@rGO	3.6 F cm^{-2}	5 mA cm^{-2}	[8]
2D-CMO	2.018 F cm^{-2}	10 mA cm^{-2}	[9]
$NiCo_2S_4$@PPy	9.781 F cm^{-2}	5 mA cm^{-2}	[10]

NF/S-Co_3O_4@$NiCo_2S_4$	5.4 F cm^{-2}	15 mA cm^{-2}	[11]
$Ni(OH)_2$-$Co(OH)_2$/NiSe-Ni_3S_2	19.01 F cm^{-2}	15 mA cm^{-2}	This work

References

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