Support Information

Three-dimensional Ni_{0.85}Se@ZnSe nanostrucures on carbon cloth for

flexible asymmetric battery-type supercapacitor

Shuling Liu*, Rui Wang, Qiuting Wang, Xian Cui, Zeyu Liu

College of Chemistry & Chemical Engineering, Shaanxi University of Science & Technology,

Xi'an, Shaanxi 710021, PR China

Contents:

- 1. Electrochemical Measurements
- 2. Material characterization
- 3. Supplementary figures S1-S9
- 4. Supplementary tables S1-S5
- 5. Supplementary References

1. Electrochemical performance test

After washing, the electrode materials obtained by electrodeposition were put into a vacuum drying oven and dried for 12 h at 60 °C. The electrolyte solution required for all electrochemical tests was 4 M KOH. The reference electrode, counter electrode and working electrode of the test system were Mercury oxide (Hg/HgO), platinum sheet electrode and electrodeposited active material electrode respectively. The voltage window range of cyclic voltammetry curve (CV) is set as 0-0.6V, and the voltage window range of constant current charge-discharge curve (GCD) is set as 0-0.5V (mono is: 0-0.45), the Energy Quest curve (EIS) was tested at an open circuit potential in the frequency range of 0.01Hz-100khz, and electrochemical performance was tested at the electrochemical workstation (CHI660E, Shanghai Chenhua). In the two-electrode test system, Ni0.85Se@ZnSe was used as the positive electrode and activated carbon (AC) as the negative electrode.

The discharge time (Δt) of the GCD curve is proportional to the electrochemical capacity (C_m, F g⁻¹) of the electrode material. Eq (S1) as follows:

$$C_m = \frac{I \times \Delta t}{m \times \Delta v} \tag{S1}$$

where I is the discharge current density (A g^{-1}); m represents the mass of active materials grown on CC for three-electrode configurations (g); Δt is the discharge time (s); and ΔV is the potential window (V).

The energy density (E, Wh kg^{-1}) and power density (P, W kg^{-1}) of the ASCs are were investigated by eqs (S2) and (S3), respectively, as follows:

$$E = \frac{C_m \times \Delta V^2}{3.6 \times 2}$$
(S2)
$$P = \frac{3600E}{\Delta t}$$
(S3)

2. Material characterization

The phase characterization of the sample Ni_{0.85}Se@ZnSe nanospheres was

performed by X-Ray powder diffractometer (XRD, Japanese, Rigaku D/Max-3c) Cu Ka (λ =1.5418 Å), the test voltage was 40 kV; The morphology, energy spectrum (EDS) and elemental analysis (mapping) of the sample were characterized by emission scanning electron microscope (FE-SEM, Hitachi S-4800, Japan); X-ray electron spectroscopy (XPS, UK AXIS SUPRA (Kratos)) was used. The composition and valence state of the sample elements were analyzed; the transmission electron microscope (TEM, FEI Tecnai G2F20, USA) characterized the microstructure of the sample.

3. Supplementary figures



Figure S1. The photographs of the Ni_{0.85}Se@ZnSe CC electrode under folded and twisted conditions



Figure S2. CV curves during electrodeposition process



Figure S3. (a, c) CV curves of the Ni_{0.85}Se, ZnSe at different scan rates, (b, d) GCD curves of the Ni_{0.85}Se, ZnSe at different electric current density.



Figure S4. (a,b, c) CV curves of the Ni_{0.85}Se@ZnSe-5, Ni_{0.85}Se@ZnSe-10,
Ni_{0.85}Se@ZnSe-20 at different scan rates, (d, e, f) GCD curves of the Ni_{0.85}Se@ZnSe-5, Ni_{0.85}Se@ZnSe-10, Ni_{0.85}Se@ZnSe-20 at different electric current density.



Figure S5. XRD patterns of the Ni_{0.85}Se@ZnSe-10 (blue line) and Ni_{0.85}Se@ZnSe-10 electrode after cycling tests (black line)



Figure S6. (a) SEM images of $Ni_{0.85}$ Se@ZnSe-10, (b) SEM images of $Ni_{0.85}$ Se@ZnSe-10 cycling at 10 A g⁻¹ for 5000 times



Figure S7. Quantitative capacitive analysis of energy storage behavior: (a) the CV curves of Ni_{0.85}Se@ZnSe-5, (b) the curves of log(i) and log(V), (c)the capacitive contribution for Ni_{0.85}Se@ZnSe-5 and diffusion contribution (blue) at a scan of 10 mV s⁻¹, (d) comparison of charge storage for Ni_{0.85}Se@ZnSe-5 electrodes at different scan rates.



Figure S8. Quantitative capacitive analysis of energy storage behavior: (a) the CV curves of Ni_{0.85}Se@ZnSe-20, (b) the curves of log(i) and log(V), (c)the capacitive contribution for Ni_{0.85}Se@ZnSe-20 and diffusion contribution (blue) at a scan of 10 mV s⁻¹, (d) comparison of charge storage for Ni_{0.85}Se@ZnSe-20 electrodes at different scan rates.



Figure S9. The capacity ratio of Ni_{0.85}Se@ZnSe-10//AC ACS

4. Supplementary tables S1-S4

samples	$R_{s}\left(\Omega ight)$	$R_{ct}\left(\Omega ight)$
ZnSe	1.387	1.98
Ni _{0.85} Se	1.174	1.46
Ni _{0.85} Se@ZnSe-5	0.69	1.38
Ni _{0.85} Se@ZnSe-10	0.47	1.14
Ni _{0.85} Se@ZnSe-20	0.64	1.26

Table S1. Fitted parameters of the Nyquist plots in Fig. 5f using equivalent circuits.

Table S2. The Ni_{0.85}Se@ZnSe-10 of elemental composition of Smart quant results by the EDS.

Element	Wt%	At%
SeL	58.14	52.83
NiK	24.06	29.45
ZnK	16.13	17.72

Table S3. The specific capacity (F g⁻¹) at different current densities in Figure 5e.

	1 A g ⁻¹	2 A g ⁻¹	3 A g ⁻¹	5 A g ⁻¹	10 A g ⁻¹
ZnSe	258	216.5	187	138.9	118.5
Ni _{0.85} Se	917	832.5	538.7	467	405
Ni _{0.85} Se@ZnSe-5	1228	1077	992	900	766
Ni _{0.85} Se@ZnSe-10	1927	1629,	1448,	1310	1058
Ni _{0.85} Se@ZnSe-20	1674	1448	1296	1085	912

Table S4. Comparison of the capacity performance of as-prepared $Ni_{0.85}Se@ZnSe$ and other metallic selenides electrode materials reported previously.

Electrode material	Special	Morphology	Current	Ref
	capacity		density	
Ni _{0.85} Se@ZnSe	nanostrucures	1927 F g ⁻¹	1A g ⁻¹	This work

Ni _{0.85} Se@MoSe ₂	nanostructures	$774 \ F \ g^{-1}$	$1 \mathrm{A} \mathrm{g}^{-1}$	Manuscript 34
NiFe ₂ Se ₄	particle-like	$372.2 \text{ mA h g}^{-1}$	$1 \mathrm{A} \mathrm{g}^{-1}$	Manuscript 29
	microstructure	(1339.92F g ⁻¹)		
NiCoSe ₂	nanostructure	2185 F g^{-1}	$1 \mathrm{A} \mathrm{g}^{-1}$	Manuscript 22
NiMoO ₄	heterostructured	1061 F g ⁻¹	$2 \ A \ g^{-1}$	Manuscript 36
Cu _{0.5} Co _{0.5} Se ₂	nanosheets	1695 F g ⁻¹	1 A g ⁻¹	1
Mn-Co-Se	urchin-like	1656 F g ⁻¹	$1 \mathrm{A} \mathrm{g}^{-1}$	2
CoNi ₂ Se ₄	flake-like	1505 F g-1	$1 \mathrm{A} \mathrm{g}^{-1}$	3
NiCoSe ₂	ellipsoid	1408 F g ⁻¹	1 A g ⁻¹	4
	structure			
Ni _{0.9} Co _{1.92} Se ₄	coral-like	510.55 C g ⁻¹	2 mA	6
		(1021.1 F g ⁻¹)	cm ⁻²	
(Ni,Co)Se ₂	cactus-like	972 F g ⁻¹	2 A g ⁻¹	7
	structure			
Co-Cd-Se	nanorods	192 mA h g ⁻¹	1 A g ⁻¹	5
		(1382 F g ⁻¹)		
CoFe ₂ Se ₄ -CoNiSe ₂	composite	183.4 mA h g ⁻¹	1 A g ⁻¹	8
		(1320.5 F g ⁻¹)		
NiFe ₂ Se ₄	particle-like	126.9 F g ⁻¹	1 A g ⁻¹	9
	microstructure			
Ni-Co-Se	hollow	584 C g ⁻¹	1 A g ⁻¹	10
	nanoparticles	(1168 F g ⁻¹)		
$Ni_{0.6}Co_{0.4}Se_2$	reed mat-like	1580 F g ⁻¹	1 A g ⁻¹	11
	sheets			
NiCo ₂ O ₄ @Ni _{0.85} Se	core-shell	1454 F g ⁻¹	1 A g ⁻¹	13
	architecture			

Supercapacitors	Specific energy	Specific power	Ref
	(Wh Kg ⁻¹)	W Kg ⁻¹	
Ni _{0.85} Se@ZnSe //AC ASC	74.67 Wh kg ⁻¹	800 W kg^{-1}	This work
NFSe-20//AC	$45.6 \text{ Wh } \text{kg}^{-1}$	$800 \mathrm{~W~kg^{-1}}$	manuscript 29
Ni _{0.85} Se@MoSe ₂ //GNS	$25.2 \text{ Wh } \text{kg}^{-1}$	$420 \mathrm{~W~kg^{-1}}$	Manuscript 34
ZNSe@NCS@NF//AC@NF	52.37Wh kg $^{-1}$	800 W kg $^{-1}$	Manuscript 24
NCSe-5//AC	41.8Wh kg ⁻¹	800W kg ⁻¹	Manuscript 22
NiMoO ₄ //APC	31.8Wh kg ⁻¹	786.5W kg ⁻¹	Manuscript 36
$Cu_{0.5}Co_{0.5}Se_2$ //Ti ₃ C ₂	84.17 Wh kg $^{-1}$	604 W kg $^{-1}$	1
Mn-Co-Se//AC	55.1 Wh kg $^{-1}$	880 W kg $^{-1}$	2
Ni _{0.9} Co _{1.92} Se ₄ //AC	26.29 Wh kg $^{-1}$	265 W kg $^{-1}$	6
(Ni,Co)Se ₂ /NiCo-LDH//PC	39 Wh kg $^{-1}$	1650 W kg $^{-1}$	7
Co-Cd-Se// /Fe alkaline	57.6Wh Kg $^{-1}$	10.9 kW kg $^{-1}$	5
CFS-CNS//CFS-CN	80.2 W h kg ⁻¹	1000 W kg-1	8
NiFe ₂ Se ₄ //AC	45.6 W h kg ⁻¹	$800 \mathrm{~W~kg^{-1}}$	9
Ni-Co-Se//RGO	41.8 W h kg ⁻¹	$750 \mathrm{~W~kg^{-1}}$	10
Ni _{0.6} Co _{0.4} Se ₂ //AC	44.1 W h kg ⁻¹	691.3 W kg^{-1}	11
(Ni,Cu)Se ₂ //AC	44.46 $Wh kg^{-1}$	797.9 W kg^{-1}	12
NiCo ₂ O ₄ @Ni _{0.85} Se //AC	29.3 Wh kg^{-1}	799 W kg^{-1}	13

Table S5 Comparison of the performance of $Ni_{0.85}Se@ZnSe$ //AC with those of reported HSC devices based on metallic selenides.

6. Supplementary References

1. Dakka, Y. A.; Balamurugan, J.; Balaji, R.; Kim, N. H.; Lee, J. H. J. C. E. J., Advanced $Cu_{0.5}Co_{0.5}Se2$ nanosheets and MXene electrodes for high-performance asymmetric supercapacitors. *385* : *123455*.

2. Miao, C.; Xu, P.; Zhao, J.; Zhu, K.; Cheng, K.; Ye, K.; Yan, J.; Cao, D.; Wang, G.; Zhang, X., Binder-Free Hierarchical Urchin-like Manganese–Cobalt Selenide with High Electrochemical Energy Storage Performance. *ACS Applied Energy Materials* **2019**, *2* (5),

3595-3604.

3. Rajesh, J. A.; Lee, Y.-H.; Yun, Y.-H.; Vinh Quy, V. H.; Kang, S.-H.; Kim, H.; Ahn, K.-S., Bifunctional NiCo₂Se4 and CoNi₂Se₄ nanostructures: Efficient electrodes for battery-type supercapacitors and electrocatalysts for the oxygen evolution reaction. *Journal of Industrial and Engineering Chemistry* **2019**, *79*, 370-382.

4. Sakthivel, M.; Sukanya, R.; Chen, S. M.; Pandi, K.; Ho, K. C. J. R. e., Synthesis and characterization of bimetallic nickel-cobalt chalcogenides (NiCoSe_2, NiCo_2S_4, and NiCo_2O_4) for non-enzymatic hydrogen peroxide sensor and energy storage: Electrochemical properties dependence on the metal-to-chalcogen composition. **2019**, *138* (AUG.), 139-151.

5. Zhai, Z.-B.; Huang, K.-J.; Wu, X., Superior mixed Co-Cd selenide nanorods for high performance alkaline battery-supercapacitor hybrid energy storage. *Nano Energy* **2018**, *47*, 89-95.

6. An, W.; Liu, L.; Gao, Y.; Liu, Y.; Liu, J., Ni_{0.9}Co_{1.92}Se₄ nanostructures: binder-free electrode of coral-like bimetallic selenide for supercapacitors. *RSC Advances* **2016**, *6* (79), 75251-75257.

7. Li, X.; Wu, H.; Guan, C.; Elshahawy, A. M.; Dong, Y.; Pennycook, S. J.; Wang, J., (Ni,Co)Se2 /NiCo-LDH Core/Shell Structural Electrode with the Cactus-Like (Ni,Co)Se2 Core for Asymmetric Supercapacitors. *Small* **2019**, *15* (3), e1803895.

8. Gopi, C.; Reddy, A. E.; Kim, H. J. J. J. o. M. C. A., Wearable superhigh energy density supercapacitors using a hierarchical ternary metal selenide composite of CoNiSe2 microspheres decorated with CoFe2Se4 nanorods. **2018**, 10.1039.C8TA01141A.

9. Ye, B.; Cao, X.; Zhao, Q.; Wang, J., Electrodeposited NiFe₂Se₄ on Nickel Foam as a Binder-Free Electrode for High-Performance Asymmetric Supercapacitors. *Industrial & Engineering Chemistry Research* **2020**, *59* (31), 14163-14171.

10. Chen, H.; Fan, M.; Li, C.; Tian, G.; Lv, C.; Chen, D.; Shu, K.; Jiang, J. J. J. o. P. S., One-pot synthesis of hollow NiSe–CoSe nanoparticles with improved performance for hybrid supercapacitors. **2016**, *329*, 314-322.

11. Xie, S.; Gou, J.; Liu, B.; Liu, C. J. J. o. C.; Science, I., Nickel-cobalt selenide as high-performance and long-life electrode material for supercapacitor. **2019**, 540: 306-314.

12. Bo, C.; Tian, Y.; Yang, Z.; Ruan, Y.; Jiang, J.; Wang, C. J. C., Construction of (Ni, Cu) Se2//Reduced Graphene Oxide for High Energy Density Asymmetric Supercapacitor. **2017**, *4* (11): 3004-3010.

13. Sui, Y.; Ye, A.; Qi, J.; Wei, F.; He, Y.; Meng, Q.; Ren, Y.; Sun, Z., Construction of NiCo₂O₄@Ni_{0.85}Se core-shell nanorod arrays on Ni foam as advanced materials for an asymmetric supercapacitor. *Journal of Alloys and Compounds* **2019**, *778*, 234-238.