

## Supporting Information

# Rational design of ultrathin 2D tin nickel selenide nanosheets for high-performance flexible supercapacitors

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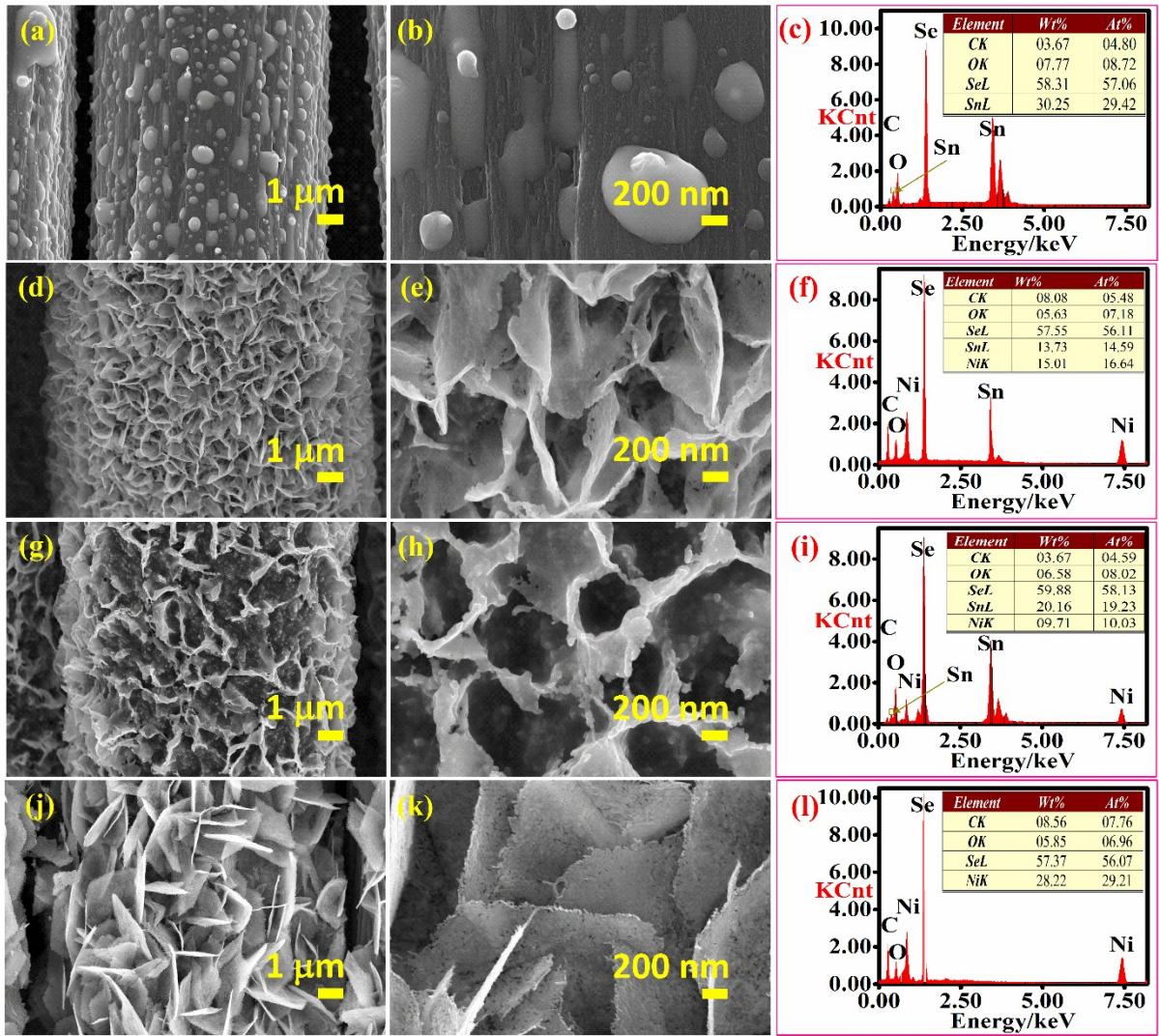
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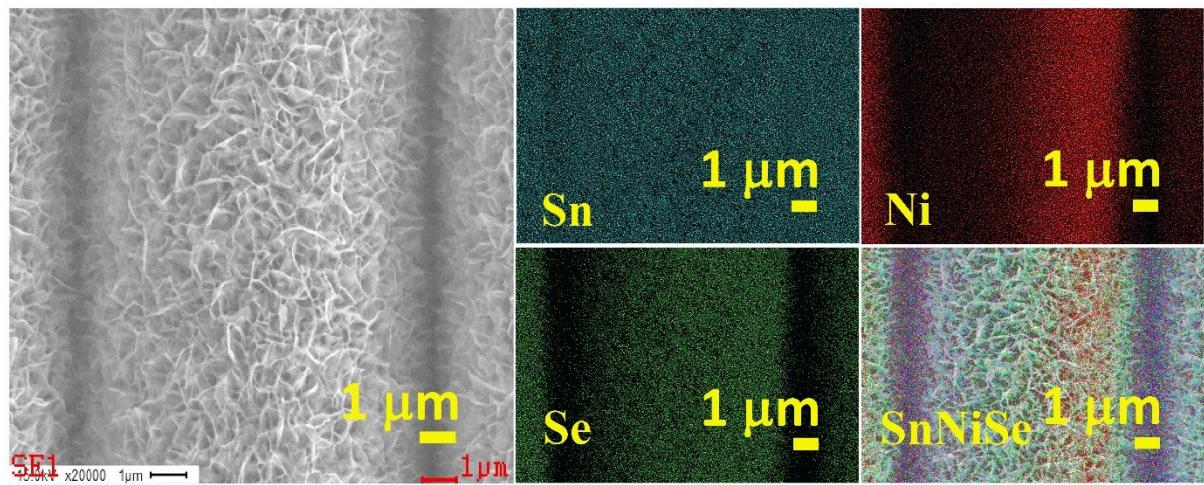
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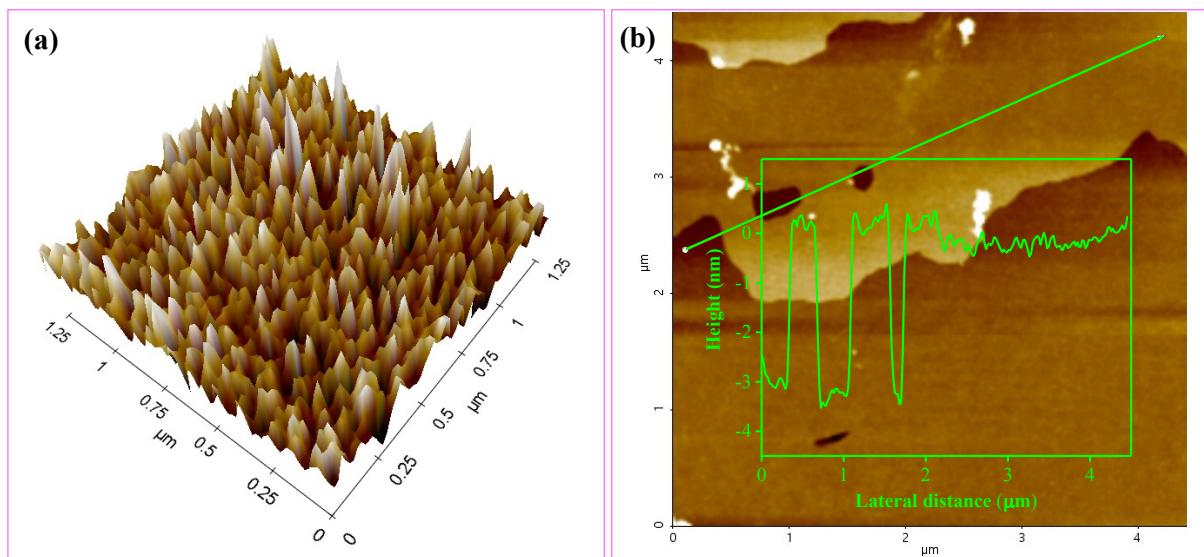
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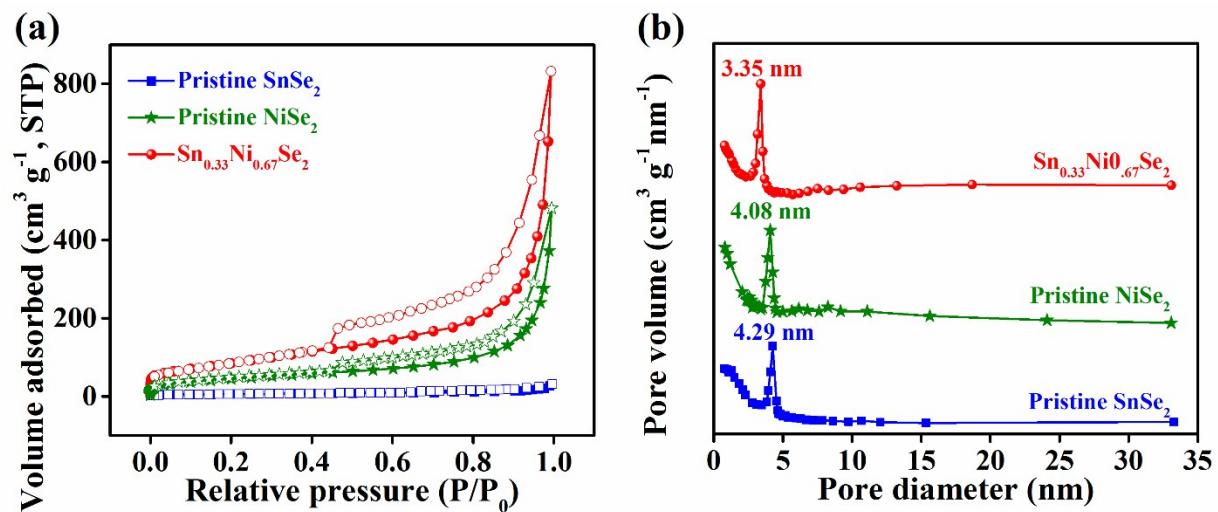
**Fig. S1** SEM and high-resolution SEM images of (a, b) SnSe<sub>2</sub>, (d, e) Sn<sub>0.5</sub>Ni<sub>0.5</sub>Se<sub>2</sub>, (g, h) Sn<sub>0.67</sub>Ni<sub>0.33</sub>Se<sub>2</sub>, and (j, k) NiSe<sub>2</sub> nanostructures. EDX spectra of (c) SnSe<sub>2</sub>, (f) Sn<sub>0.5</sub>Ni<sub>0.5</sub>Se<sub>2</sub>, (i) Sn<sub>0.67</sub>Ni<sub>0.33</sub>Se<sub>2</sub>, and (l) NiSe<sub>2</sub> nanostructures.



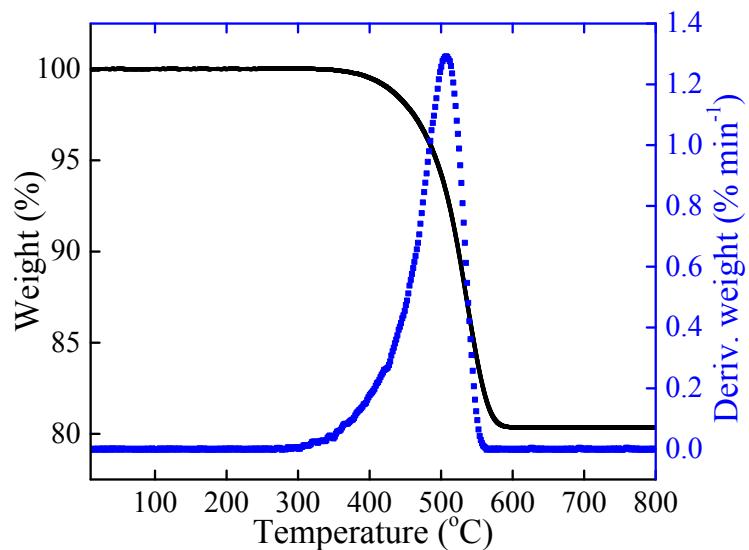
**Fig. S2** SEM-EDS color elemental mapping of  $\text{Sn}_{0.33}\text{Ni}_{0.67}\text{Se}_2$  NAs in terms of Sn-K, Ni-K, and Se-K.



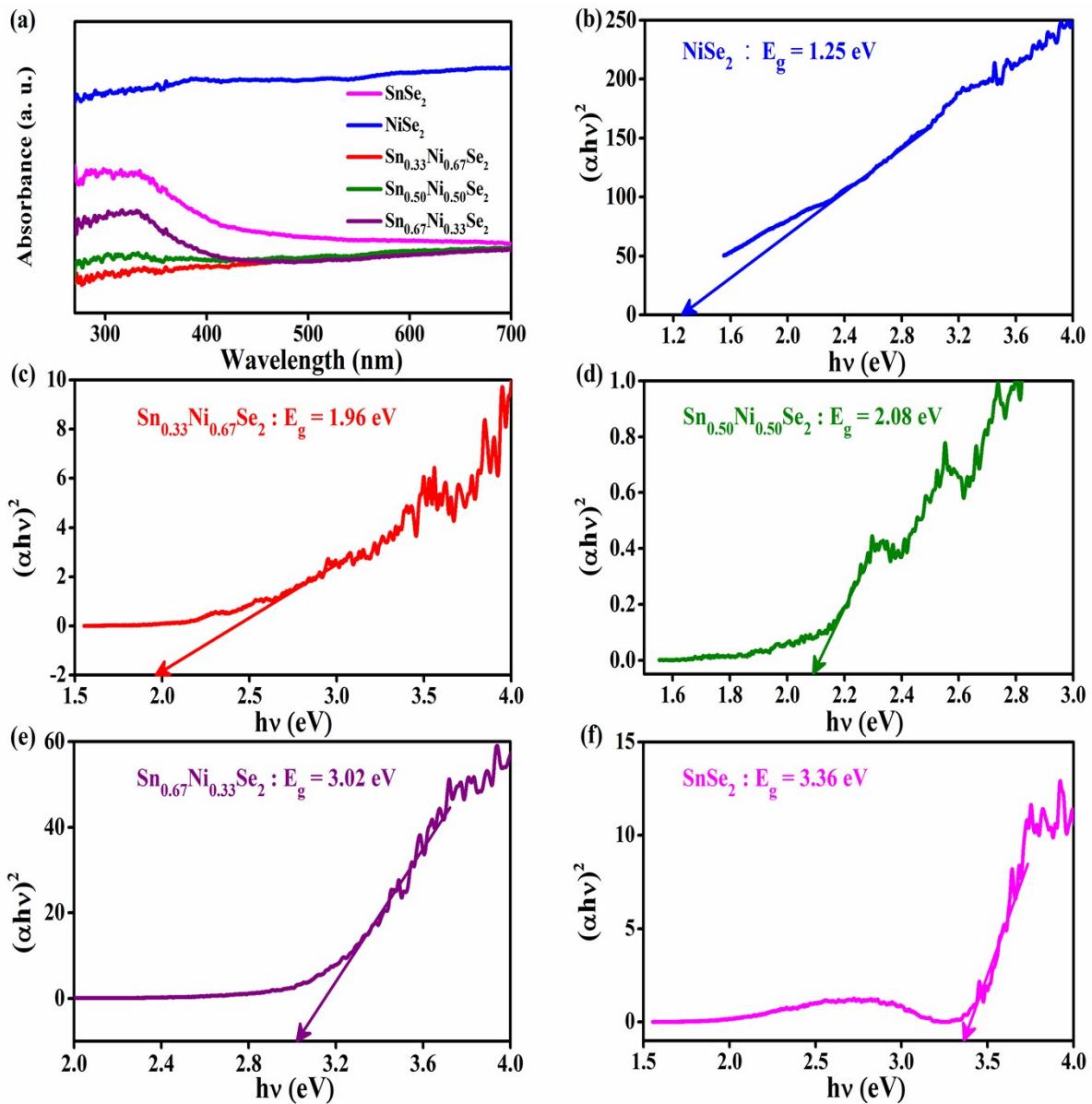
**Fig. S3** (a) 3D and (b) 2D AFM images and its height profile of  $\text{Sn}_{0.33}\text{Ni}_{0.67}\text{Se}_2$  representing the thickness of few nanometer and width  $> 4 \mu\text{m}$ .



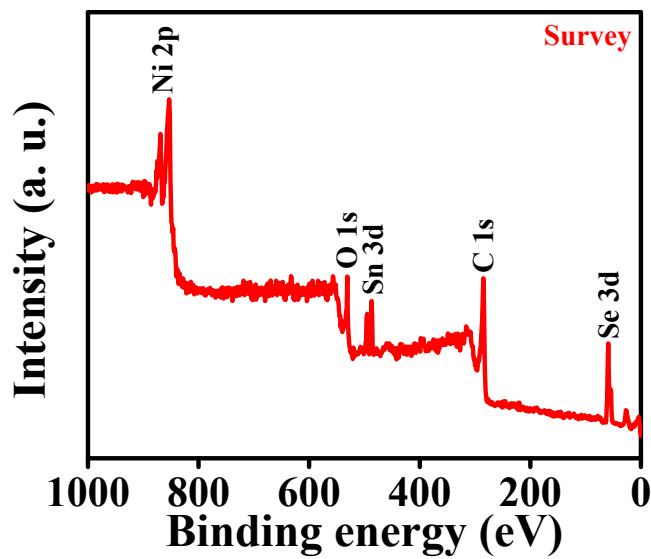
**Fig. S4** (a) N<sub>2</sub> adsorption-desorption isotherms and (b) their corresponding pore size distribution of pristine SnSe<sub>2</sub>, NiSe<sub>2</sub>, and Sn<sub>0.33</sub>Ni<sub>0.67</sub>Se<sub>2</sub> NAs.



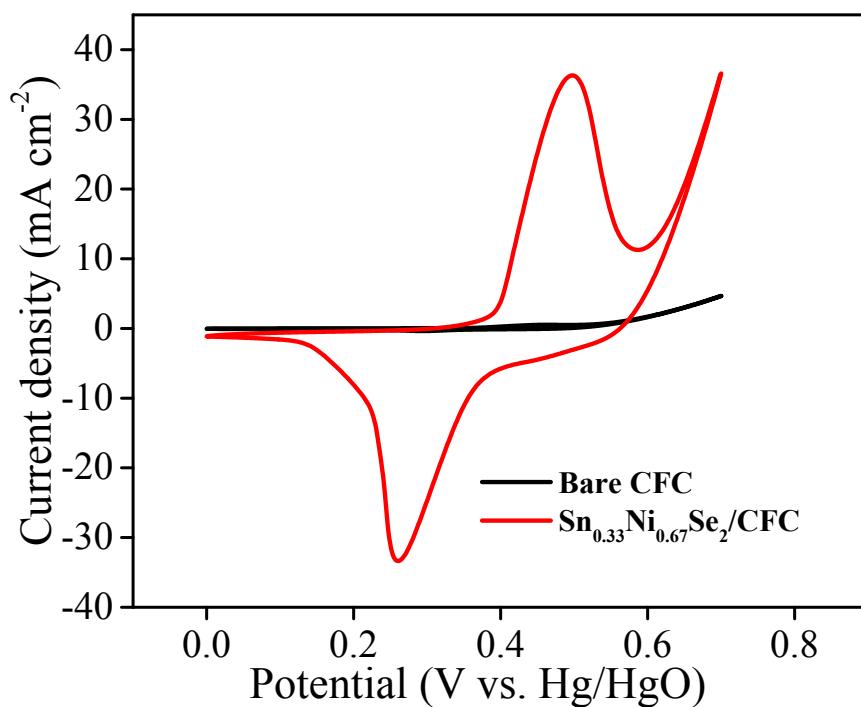
**Fig. S5** TGA curves of Sn<sub>0.33</sub>Ni<sub>0.67</sub>Se<sub>2</sub> NAs under air flow with a temperature ramp of ~5 °C min<sup>-1</sup>.



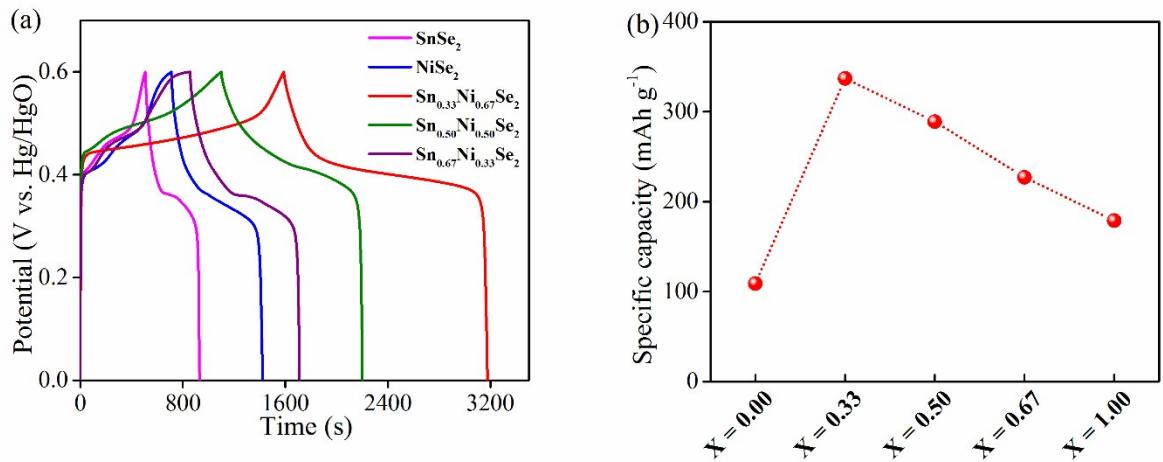
**Fig. S6** (a) UV–vis DRS spectra and (b-f) bandgap measurements of  $\text{Sn}_x\text{Ni}_{1-x}\text{Se}_2$  nanostructures.



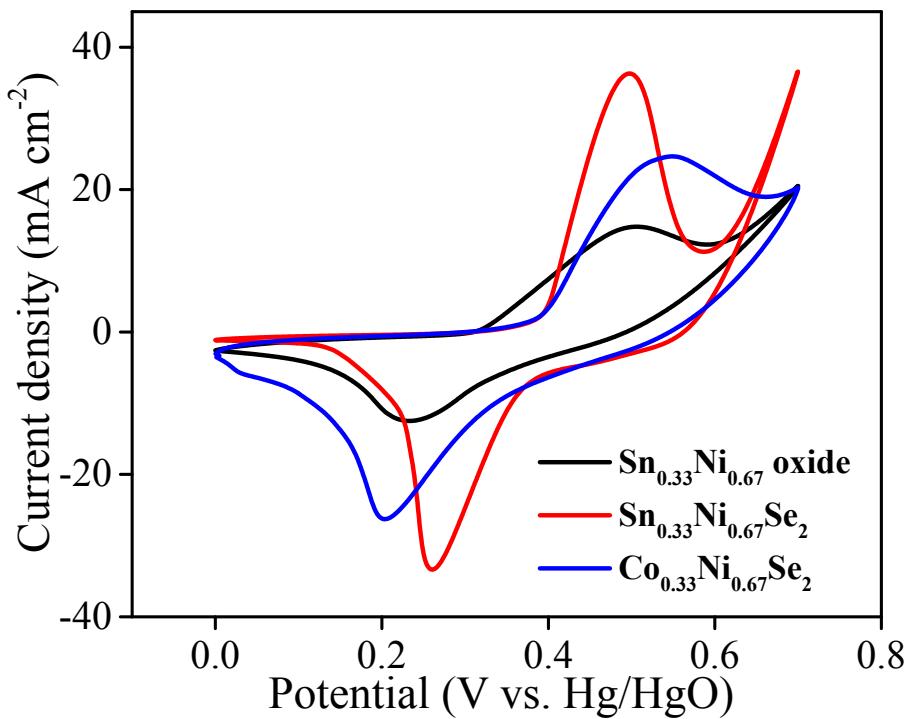
**Fig. S7** XPS survey spectrum of  $\text{Sn}_{0.33}\text{Ni}_{0.67}\text{Se}_2$  NAs.



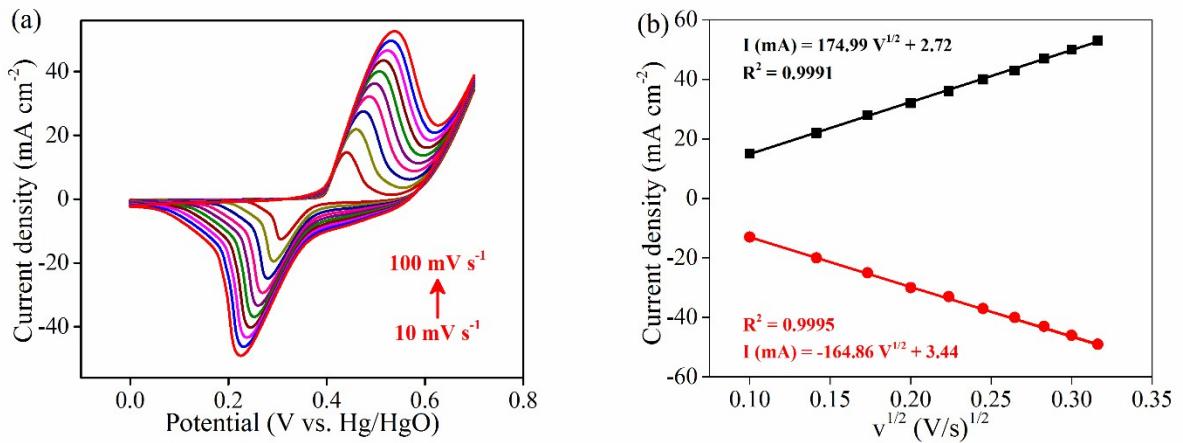
**Fig. S8** CV curves of bare CFC and  $\text{Sn}_{0.33}\text{Ni}_{0.67}\text{Se}_2$  NAs/CFC electrode at a scan rate of 50 mV s<sup>-1</sup>.



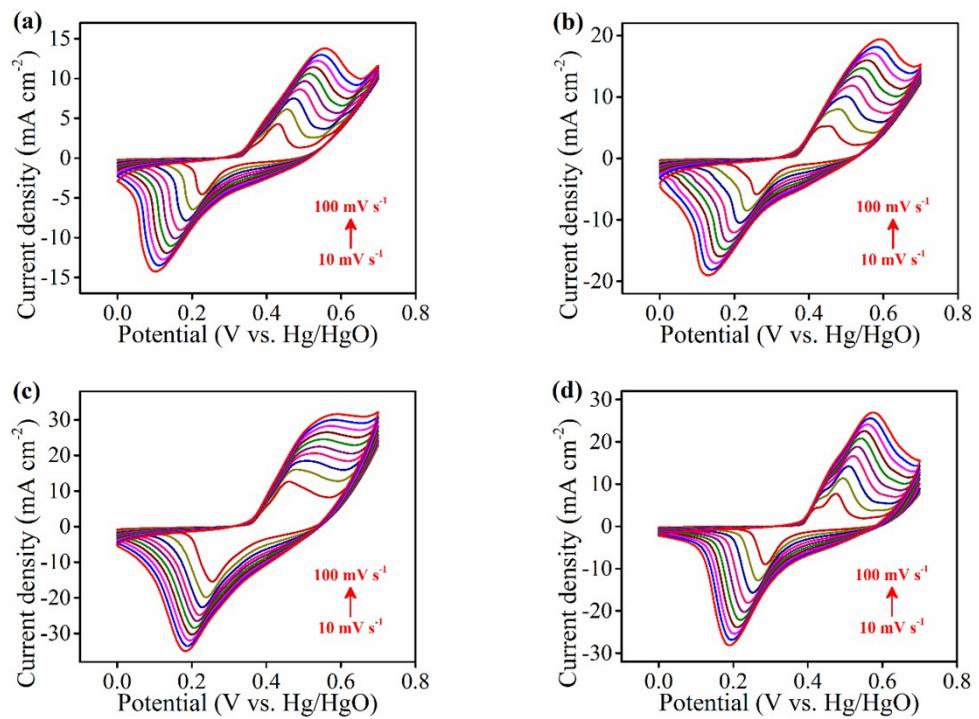
**Fig. S9** (a) GCD curves of  $\text{Sn}_x\text{Ni}_{1-x}\text{Se}_2$  electrodes with different  $x$  values using three-electrode system at a current density of  $1 \text{ mA cm}^{-2}$  and (b) specific capacity values calculated from GCD data at different  $x$  values.



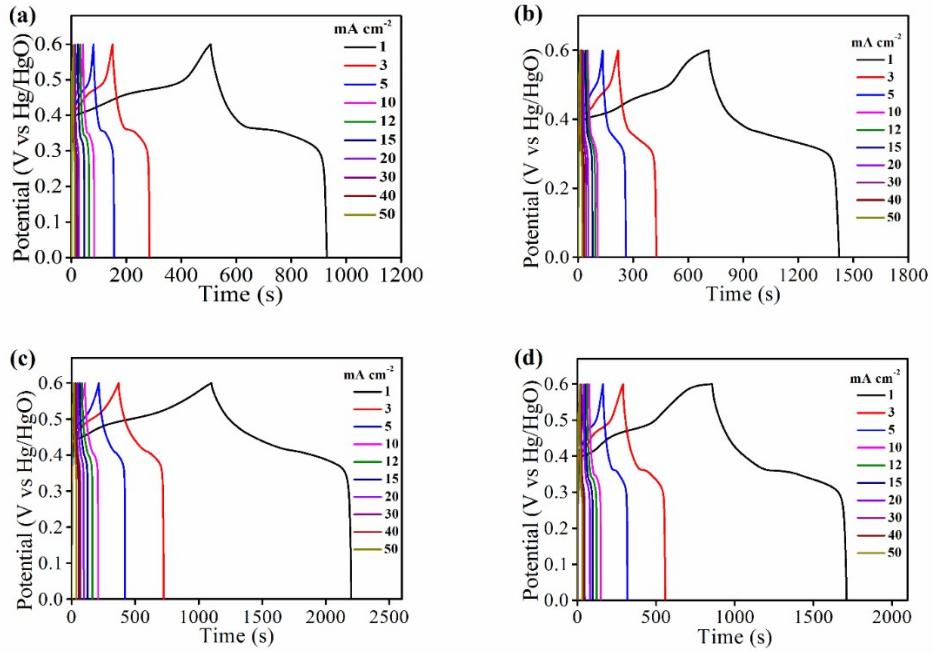
**Fig. S10** Comparisons between the  $\text{Sn}_{0.33}\text{Ni}_{0.67}\text{Se}_2$  electrode,  $\text{Co}_{0.33}\text{Ni}_{0.67}\text{Se}_2$  electrode, and  $\text{Sn}_{0.33}\text{Ni}_{0.67}$  oxide electrode perceived from heat treatment of their corresponding LDH precursors at  $350^\circ\text{C}$  at air atmosphere: CVs measured at a high scan rate of  $50 \text{ mV s}^{-1}$ .



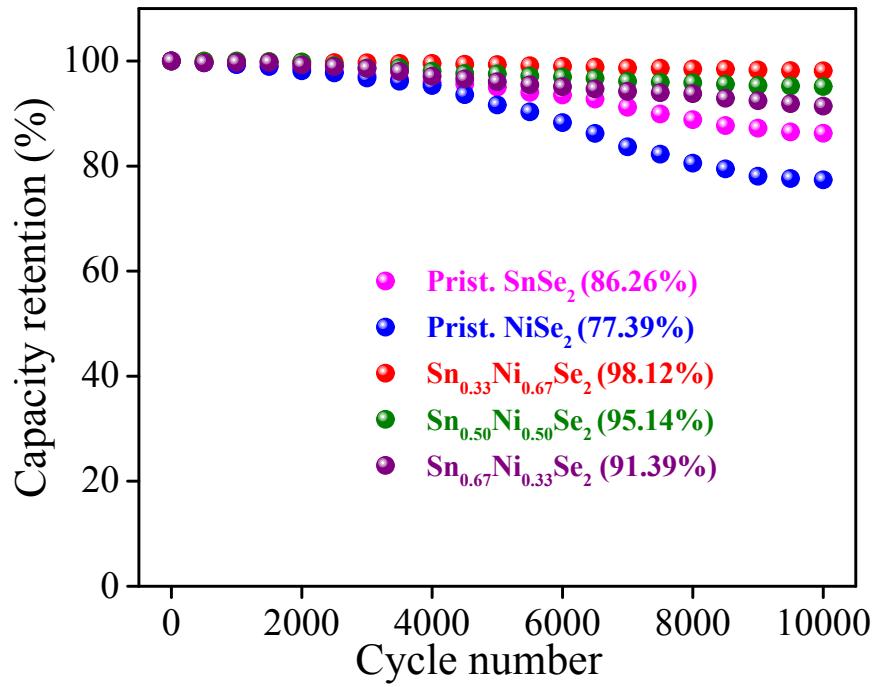
**Fig. S11** (a) CV curves of  $\text{Sn}_{0.33}\text{Ni}_{0.67}\text{Se}_2$  electrode with different scan rates from 10 to 100  $\text{mV s}^{-1}$  and (b) the relationship of redox peak current vs. square root of scan rate.



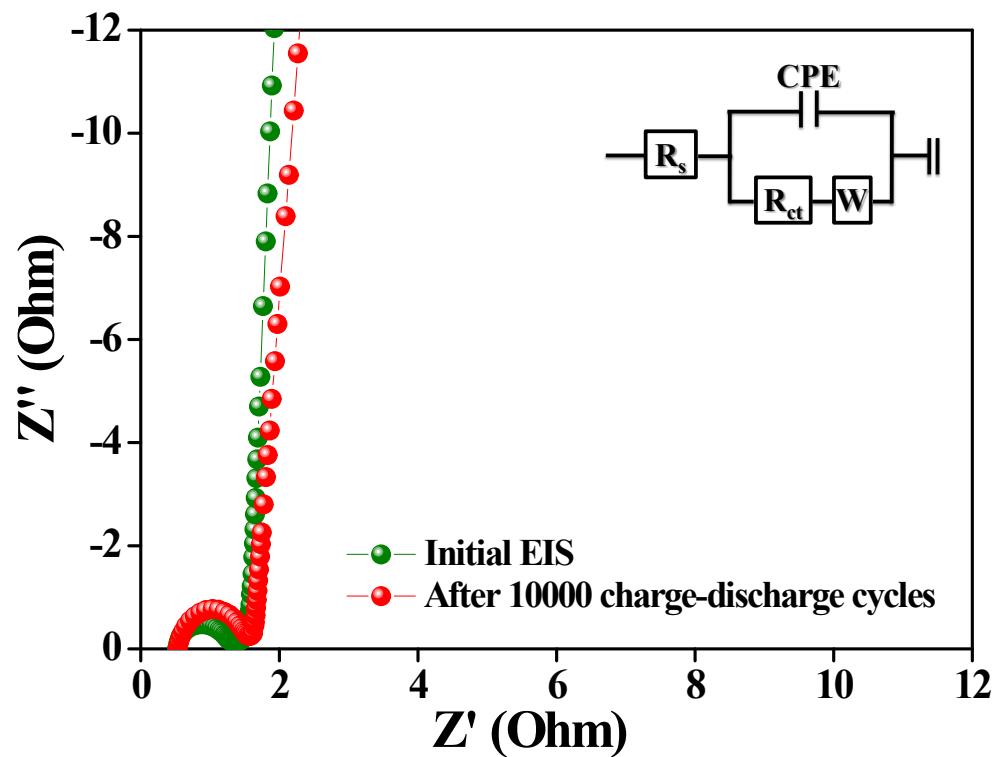
**Fig. S12** CV curves with different scan rates from 10 to 100  $\text{mV s}^{-1}$  of (a)  $\text{SnSe}_2$ , (b)  $\text{NiSe}_2$ , (c)  $\text{Sn}_{0.5}\text{Ni}_{0.5}\text{Se}_2$ , and (d)  $\text{Sn}_{0.67}\text{Ni}_{0.33}\text{Se}_2$  electrodes.



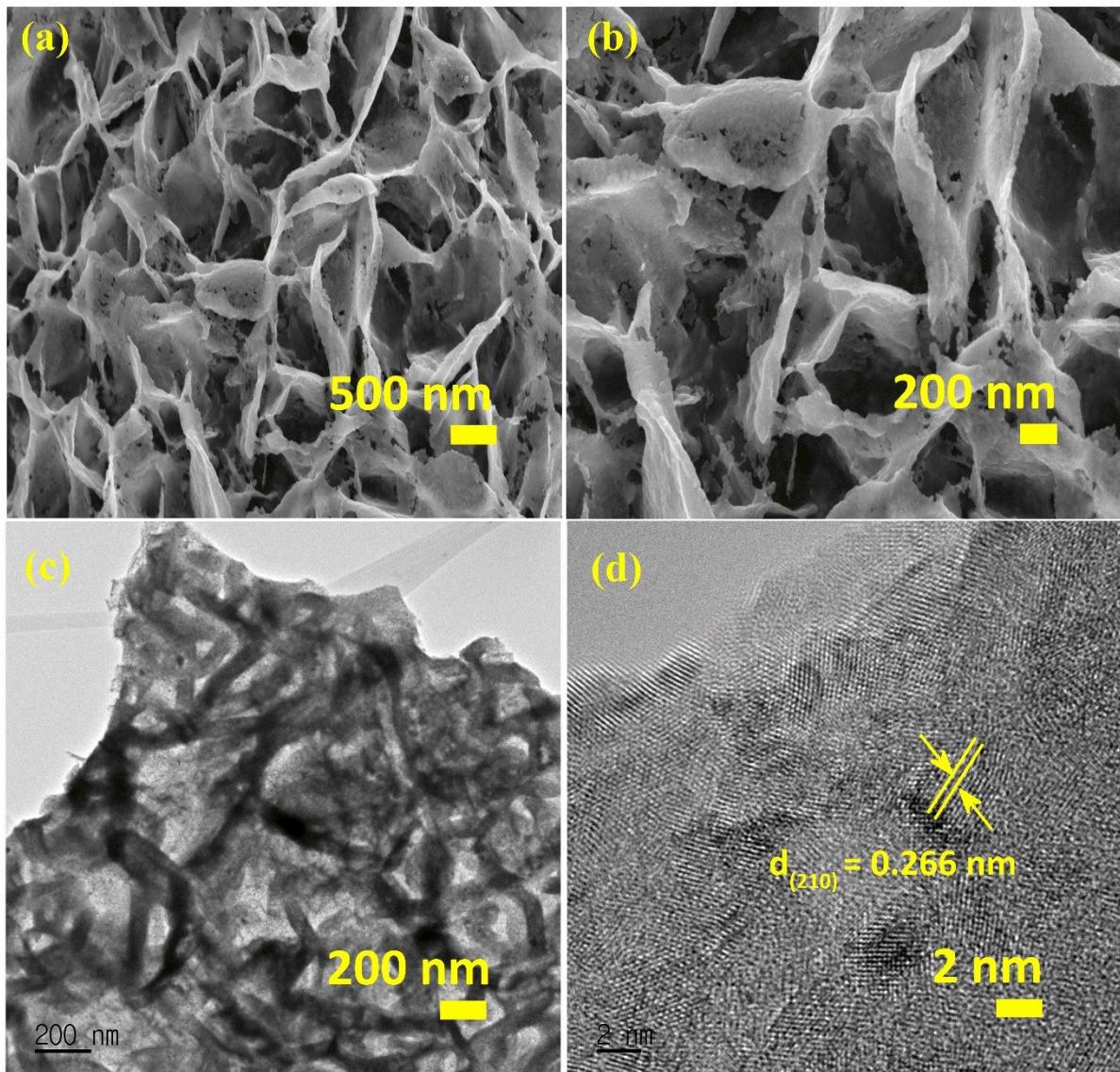
**Fig. S13** GCD curves with various 1 to 50 mA cm<sup>-2</sup> of (a) SnSe<sub>2</sub>, (b) NiSe<sub>2</sub>, (c) Sn<sub>0.5</sub>Ni<sub>0.5</sub>Se<sub>2</sub>, and (d) Sn<sub>0.67</sub>Ni<sub>0.33</sub>Se<sub>2</sub> electrodes.



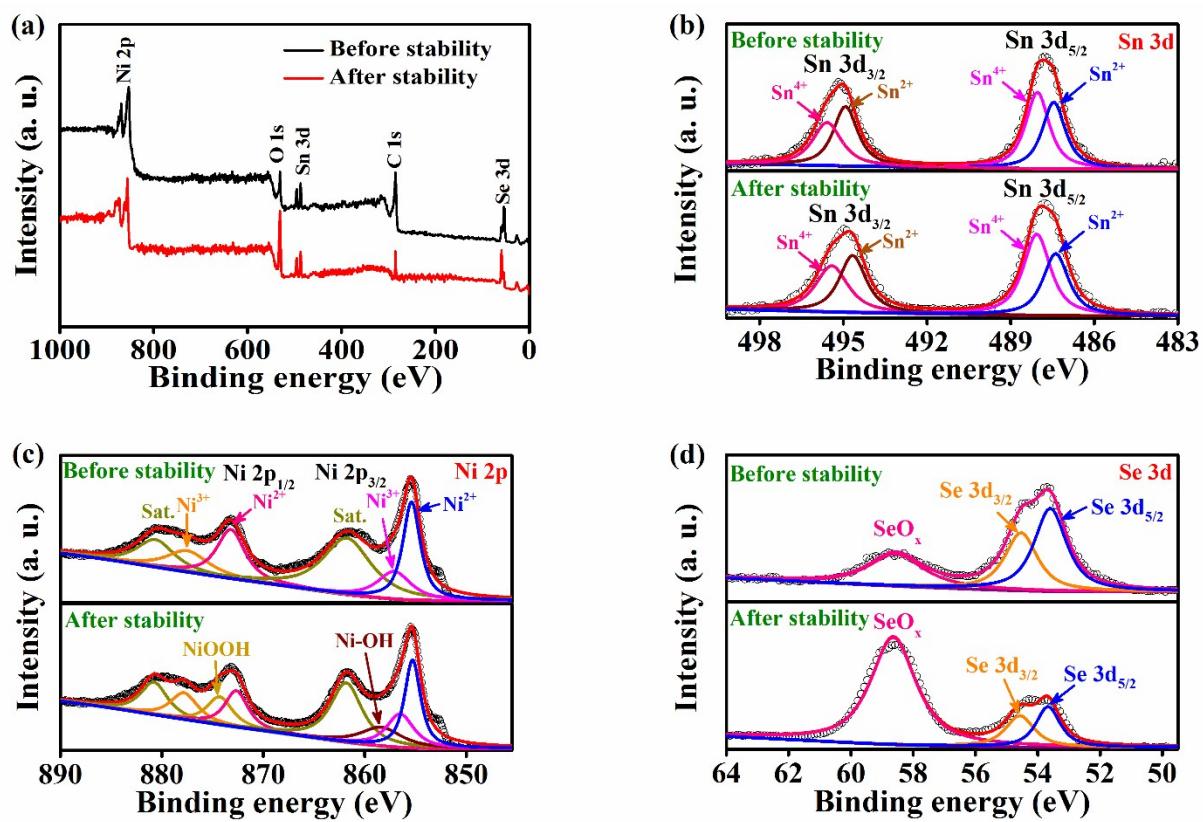
**Fig. S14** Cycling performance of SnSe<sub>2</sub>, NiSe<sub>2</sub>, Sn<sub>0.33</sub>Ni<sub>0.67</sub>Se<sub>2</sub>, Sn<sub>0.50</sub>Ni<sub>0.50</sub>Se<sub>2</sub>, and Sn<sub>0.67</sub>Ni<sub>0.33</sub>Se<sub>2</sub> electrodes at a higher current density of 30 mA cm<sup>-2</sup>.



**Fig. S15** EIS of optimal  $\text{Sn}_{0.33}\text{Ni}_{0.67}\text{Se}_2$  electrode using three-electrode system (initial and after successive 10000 charge–discharge cycles).



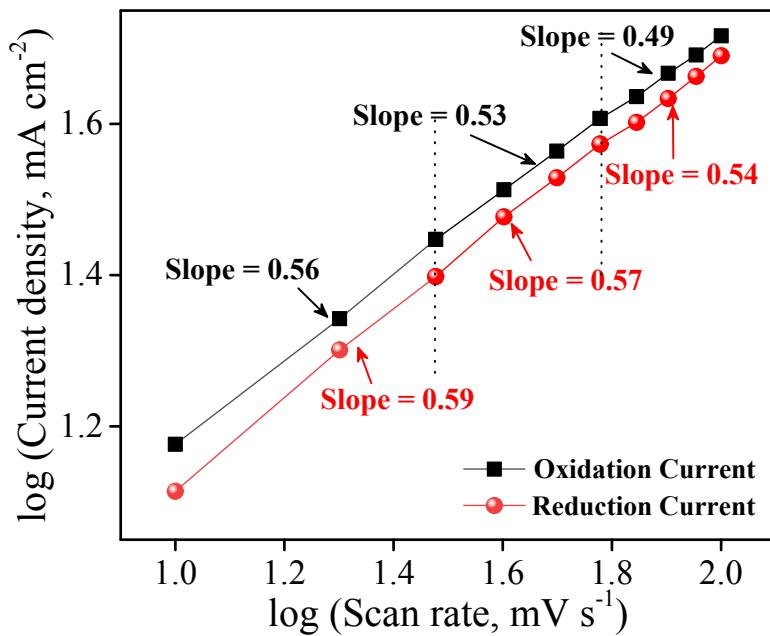
**Fig. S16** (a, b) SEM images with different magnifications and (c, d) TEM images with different magnifications of  $\text{Sn}_{0.33}\text{Ni}_{0.67}\text{Se}_2$  NAs (after successive 10000 charge–discharge cycles).



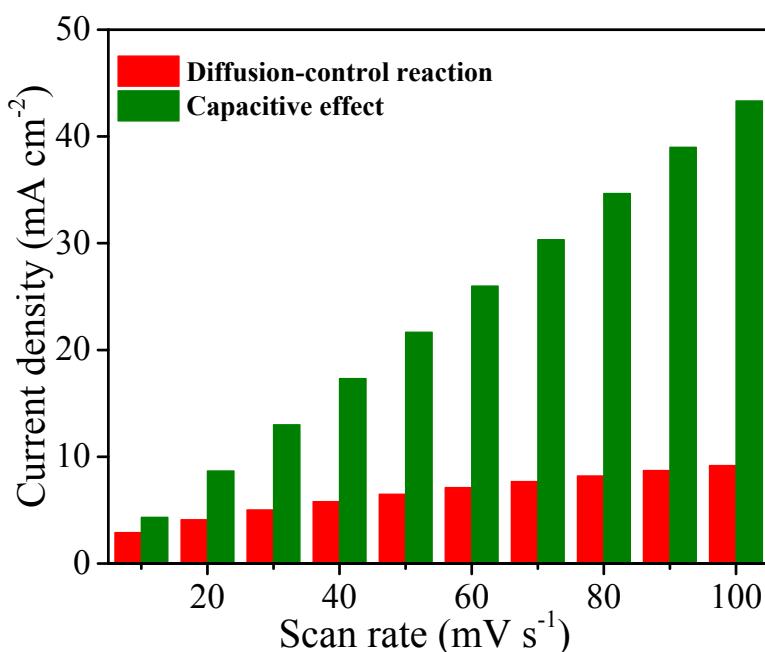
**Fig. S17** XPS before and after successive 10000 charge–discharge cycles.



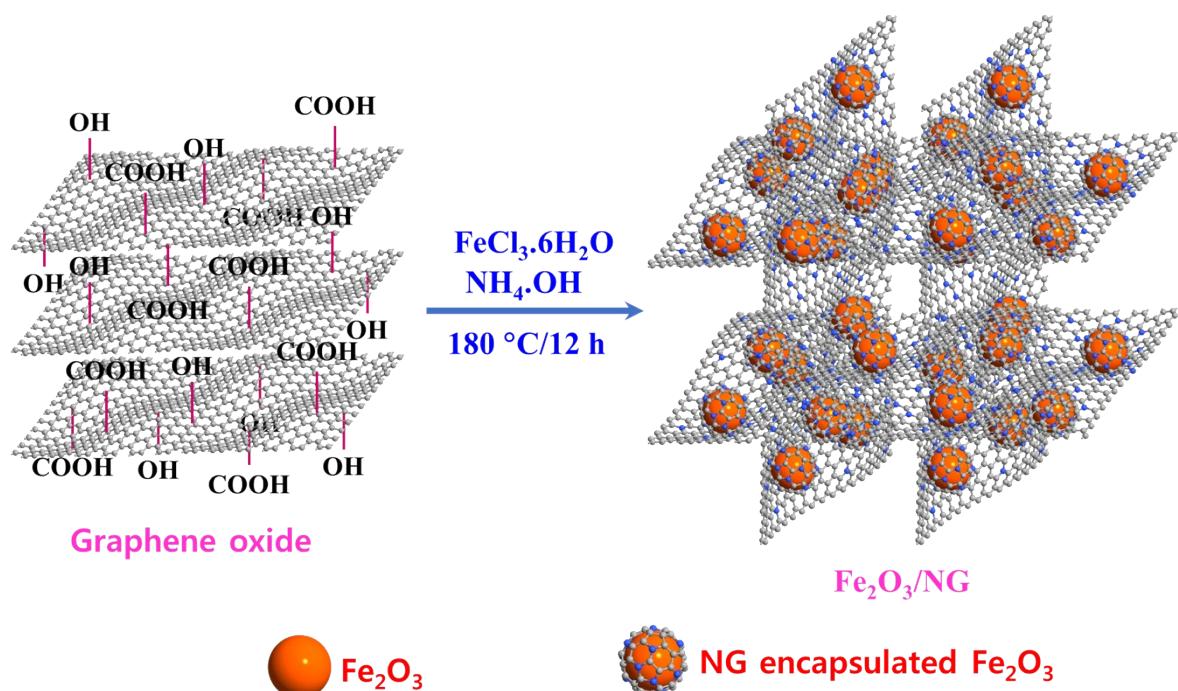
**Fig. S18** Schematic diagram of the virtues of the interconnected ultra-thin  $\text{Sn}_{0.33}\text{Ni}_{0.67}\text{Se}_2$  NAs.



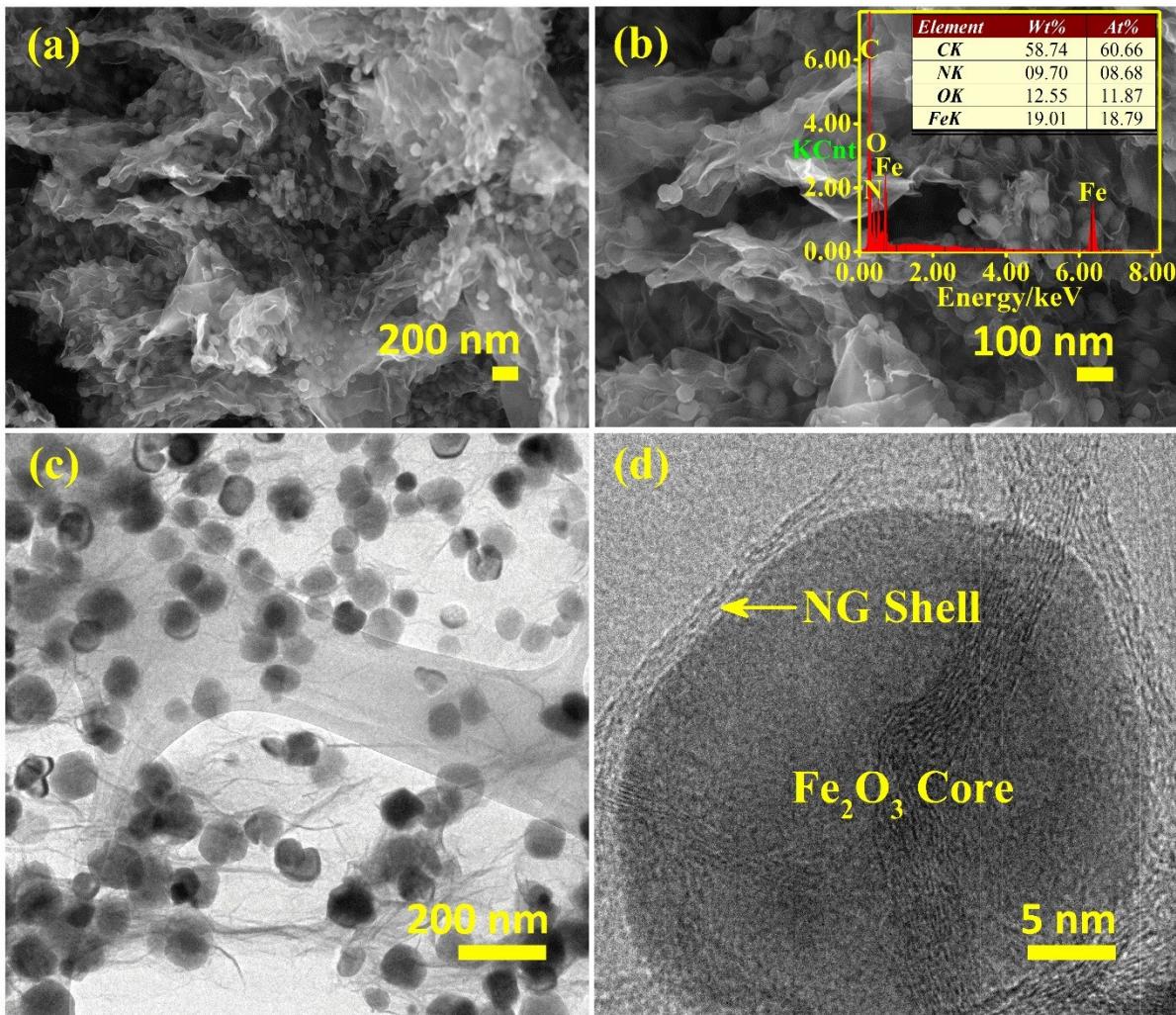
**Fig. S19** Calculated *b*-value of the oxidation and reduction peak currents at scan rate of 10 to 100 mV s<sup>-1</sup> of Sn<sub>0.33</sub>Ni<sub>0.67</sub>Se<sub>2</sub> electrode.



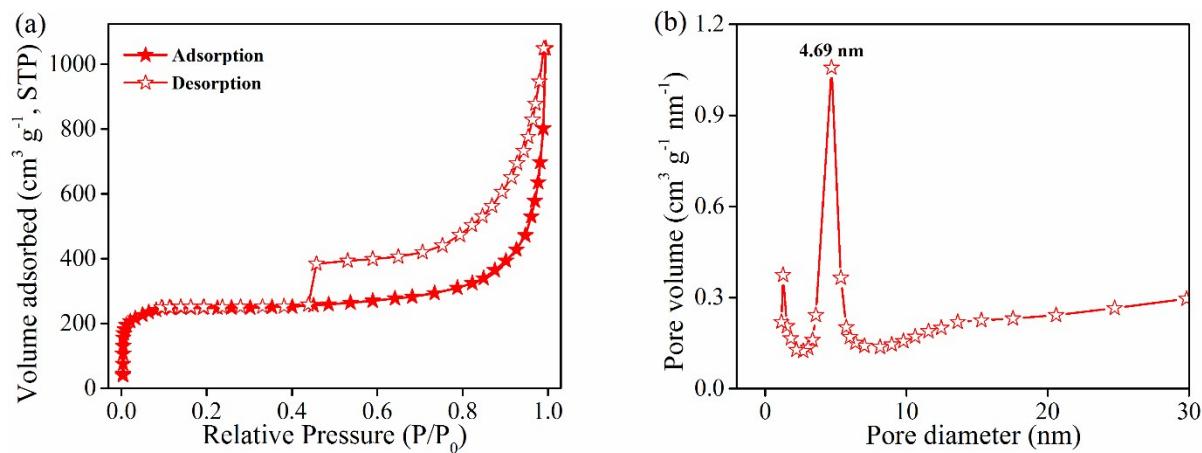
**Fig. S20** Calculated surface pseudocapacitive and bulk-insertion typed current density at various scan rates of Sn<sub>0.33</sub>Ni<sub>0.67</sub>Se<sub>2</sub> electrode.



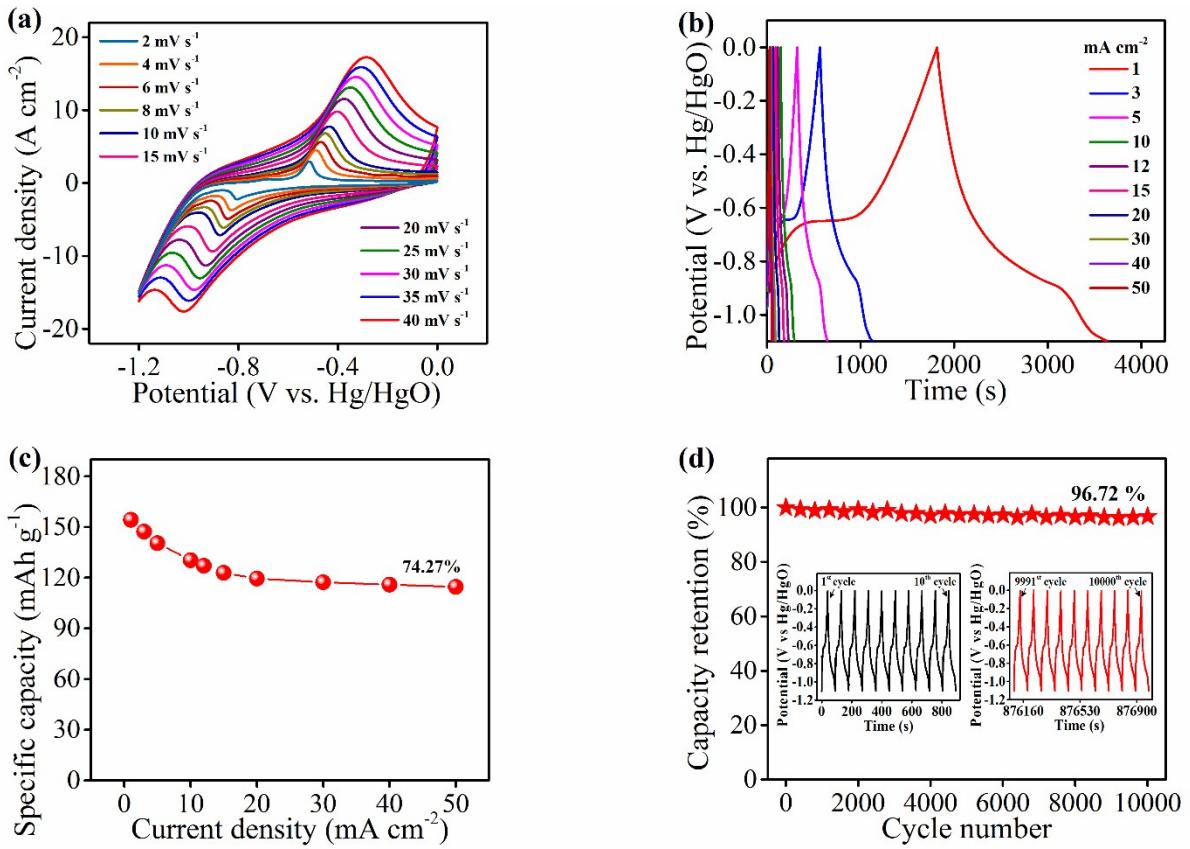
**Fig. S21** Schematic illustration for the synthesis of  $\text{Fe}_2\text{O}_3@\text{NG}$  hydrogel by in-situ hydrothermal method.



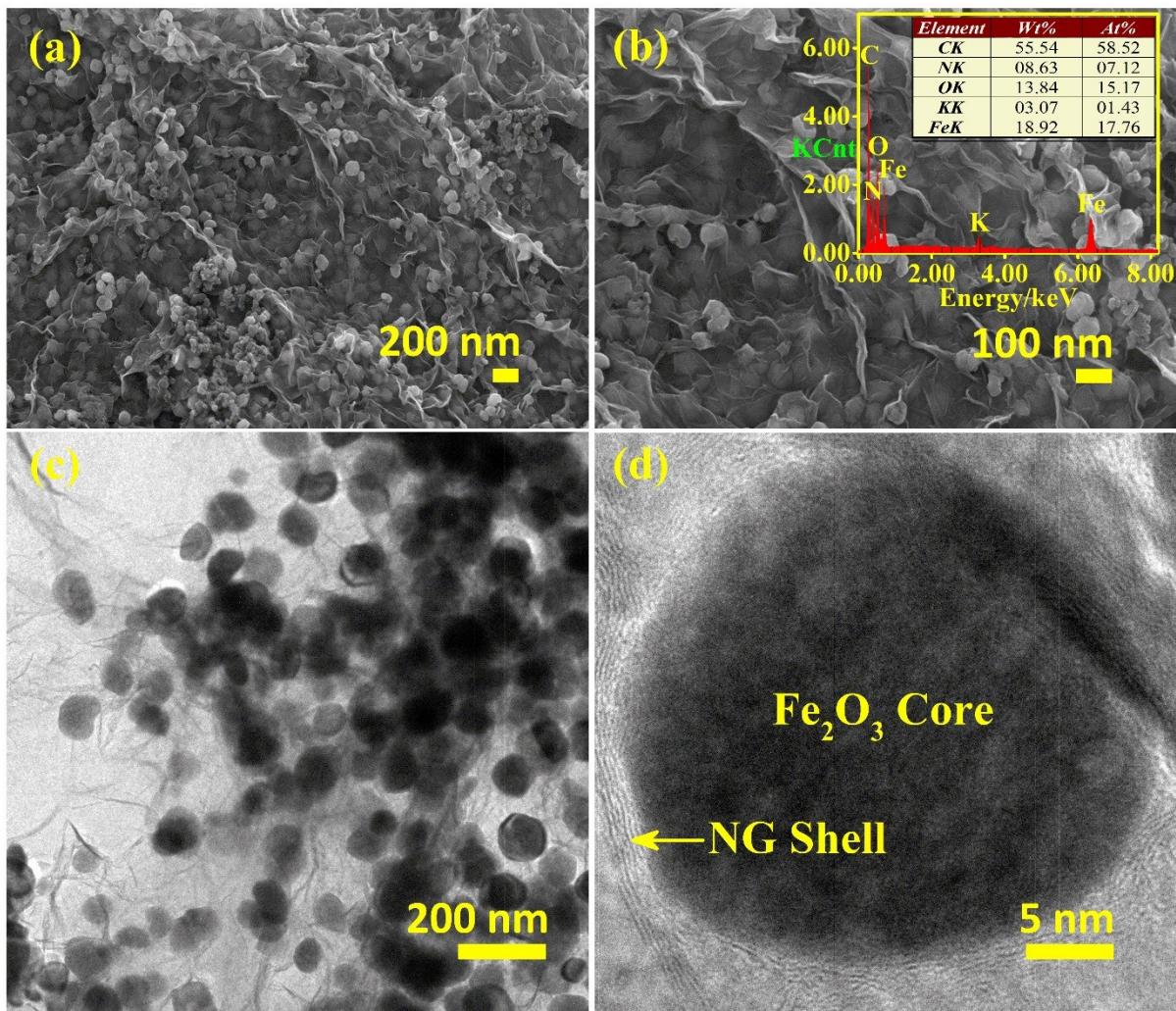
**Fig. S22** (a, b) SEM images with different magnifications (inset shows the corresponding EDX analysis), (c) TEM, and (d) HR-TEM images of the as-synthesized  $\text{Fe}_2\text{O}_3@\text{NG}$  hydrogel.



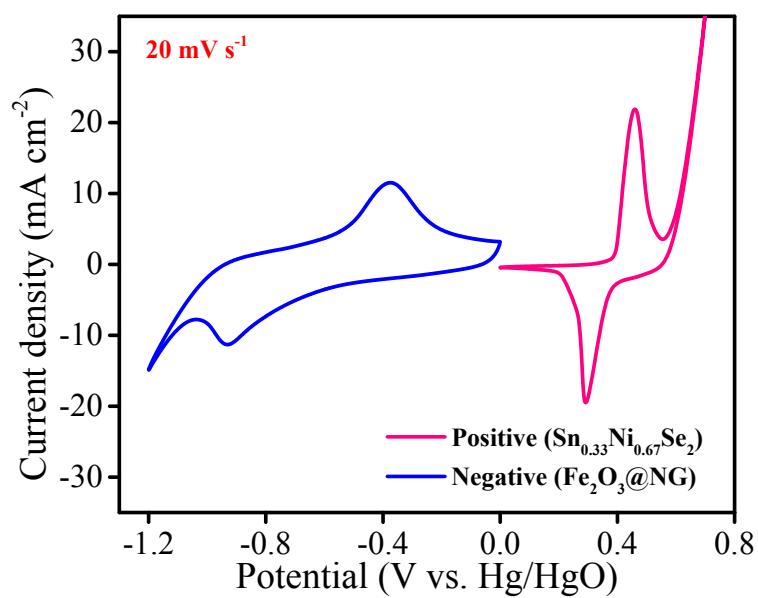
**Fig. S23** (a)  $\text{N}_2$  adsorption-desorption isotherms and (b) pore size distribution of the as-synthesized  $\text{Fe}_2\text{O}_3@\text{NG}$  hydrogel.



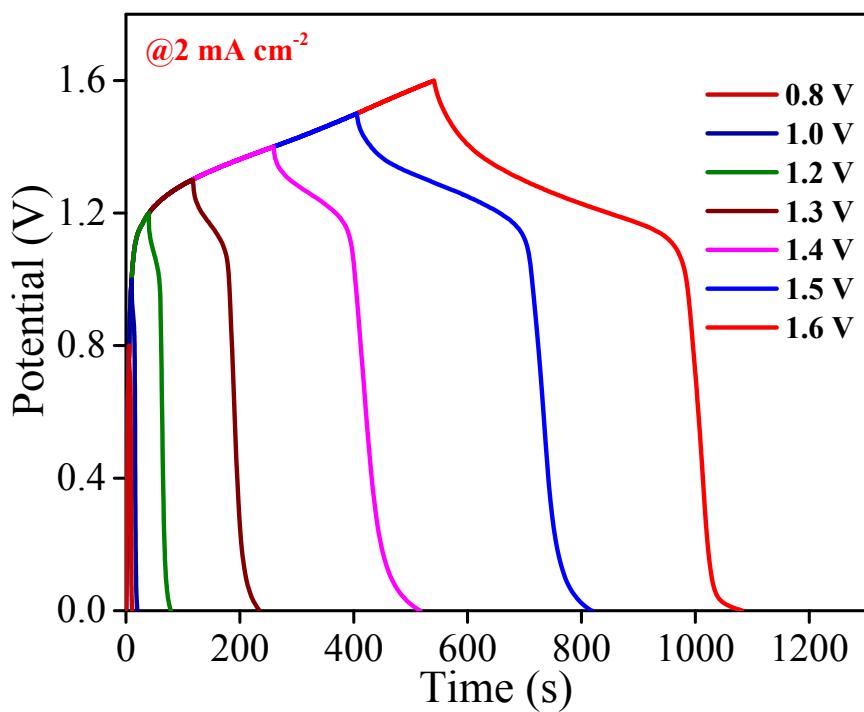
**Fig. S24** (a) CV curves of  $\text{Fe}_2\text{O}_3@\text{NG}$  hydrogel at different scan rates from 10 to 100  $\text{mV s}^{-1}$ , (b) GCD curves of  $\text{Fe}_2\text{O}_3@\text{NG}$  hydrogel at various current densities from 1 to 50  $\text{A g}^{-1}$ , (c) specific capacity vs. current density of  $\text{Fe}_2\text{O}_3@\text{NG}$  hydrogel, and (d) Cycling performance of  $\text{Fe}_2\text{O}_3@\text{NG}$  hydrogel electrode at a higher current density of 30  $\text{A g}^{-1}$  (inset shows the initial and last 10 charge–discharge cycles).



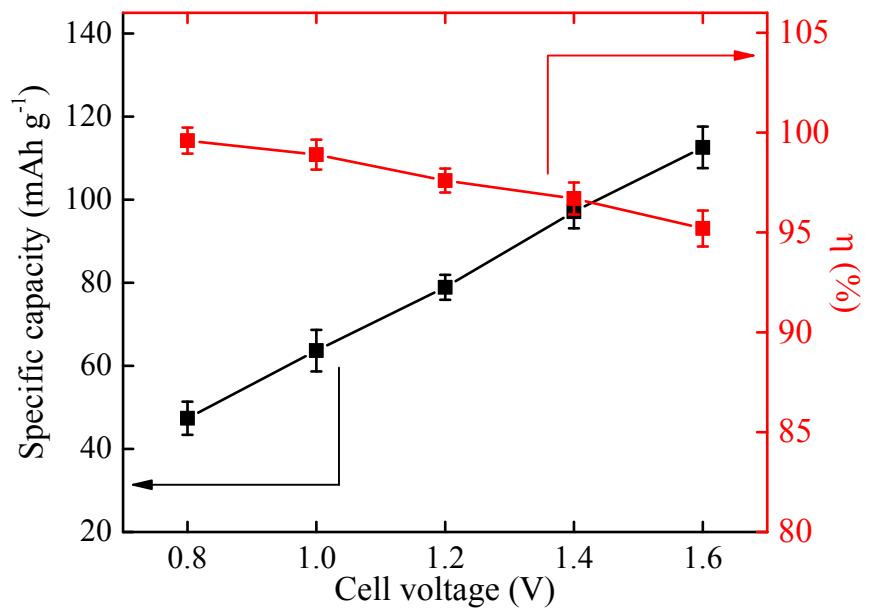
**Fig. S25** (a, b) SEM images with different magnifications (inset shows the corresponding EDX analysis), (c) TEM, and (d) HR-TEM images of the as-synthesized  $\text{Fe}_2\text{O}_3@\text{NG}$  hydrogel (after successive 10000 charge–discharge cycles).



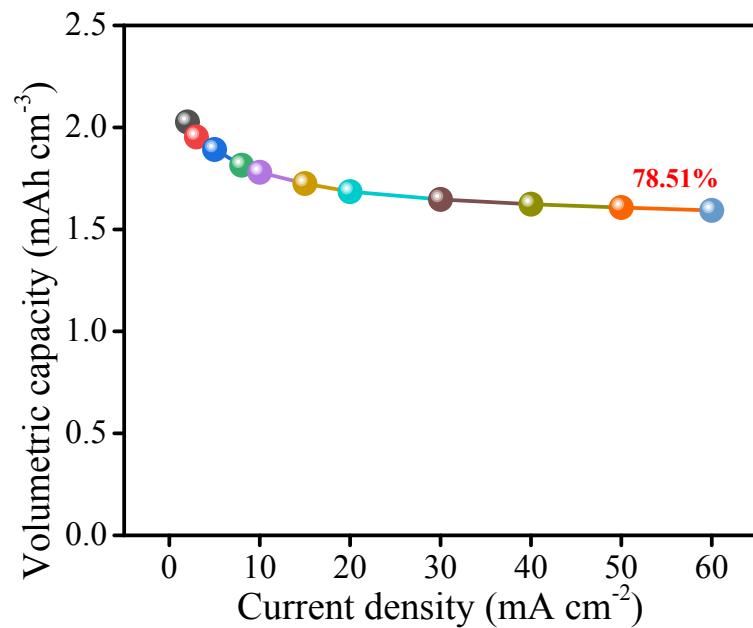
**Fig. S26** CV curves of the  $\text{Sn}_{0.33}\text{Ni}_{0.67}\text{Se}_2$  and  $\text{Fe}_2\text{O}_3@\text{NG}$  electrodes in three-electrode setup at a scan rate of  $20 \text{ mV s}^{-1}$ .



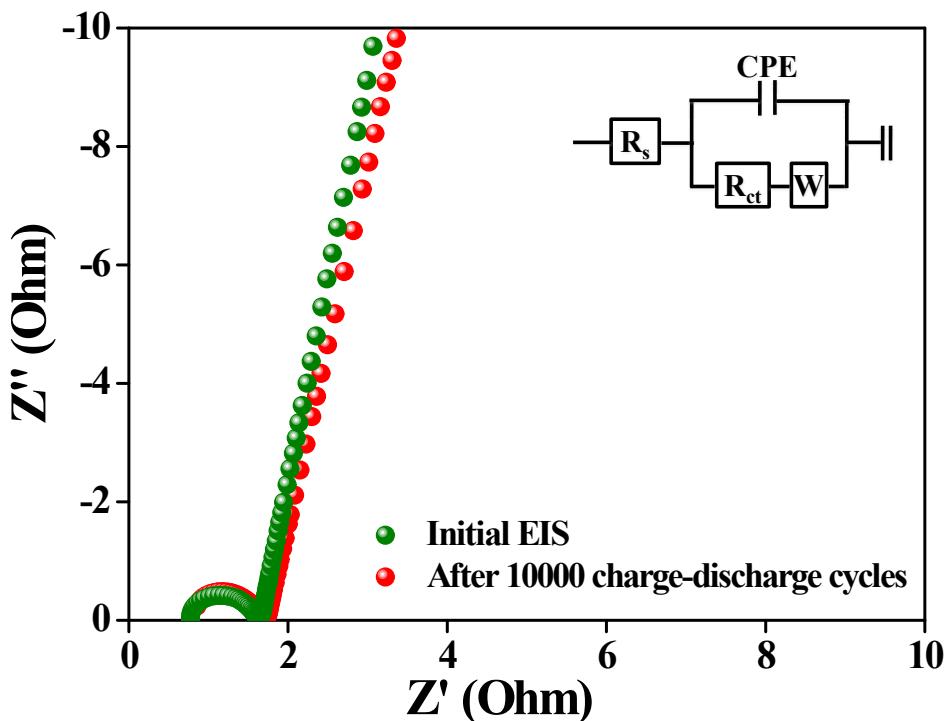
**Fig. S27** GCD curves (at a current density of  $2 \text{ mA cm}^{-2}$ ) of the flexible solid-state ASC collected at different potential windows from 0.8 to 1.6 V.



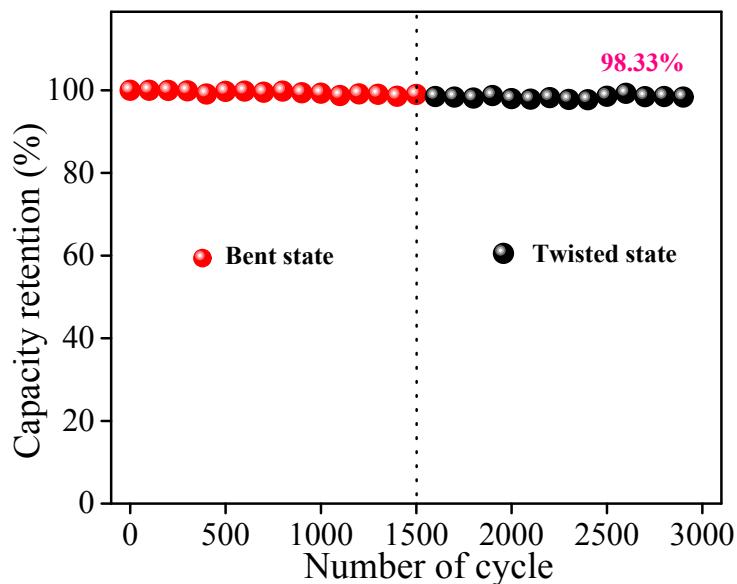
**Fig. S28** Coulombic efficiency and specific capacity of flexible solid-state ASC in gel PVA-KOH electrolyte *vs.* different cell voltage.



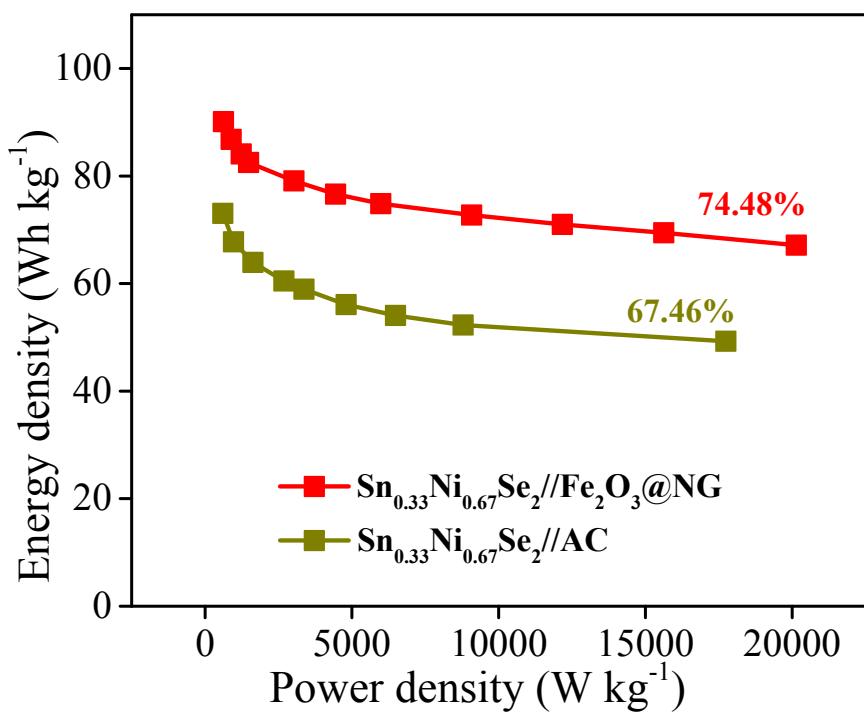
**Fig. S29** Volumetric capacity of flexible solid-state ASC at various current densities from 2 to 60 mA cm⁻².



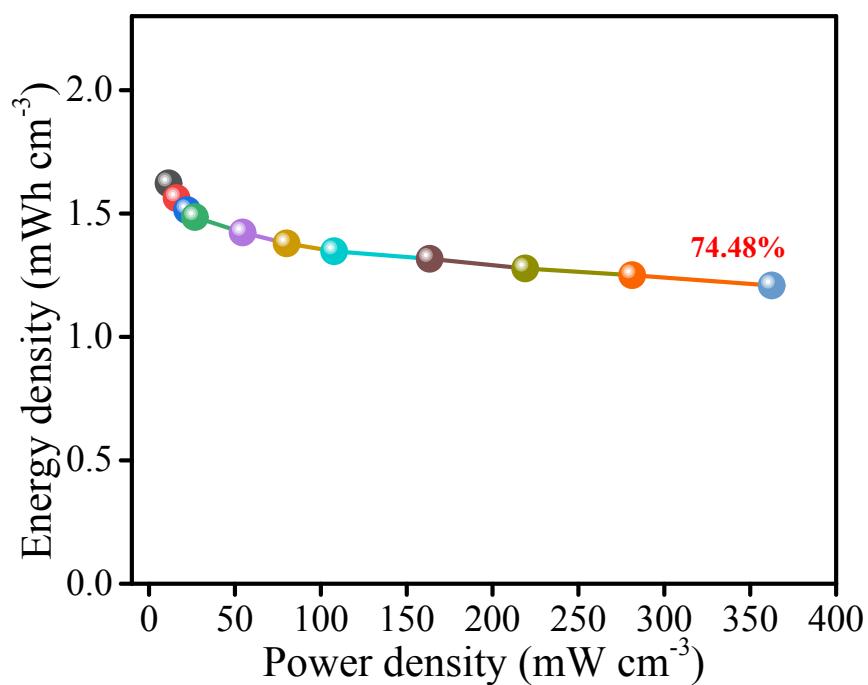
**Fig. S30** EIS of optimal flexible solid-state ASC device (initial and after successive 10000 charge–discharge cycles).



**Fig. S31** (a) CV curves of flexible solid-state ASC at normal, different twisting and bent state, and (b) Durability of the flexible solid-state ASC when tested under bending and twisting states. The flexible solid-state ASC retain ~98.33% of its initial capacity after successive 1500 cycles under the bent state, surveyed by another successive 1500 cycles under the twisted state.



**Fig. S32** Ragone plot (specific energy density vs. specific power density) of flexible solid-state ASC.



**Fig. S33** Volumetric energy density vs. volumetric power density of flexible solid-state ASC.

**Table S1.** Elemental composition of  $\text{Sn}_x\text{Ni}_{1-x}\text{Se}_2$  nanostructures with different  $x$  values estimated from ICP-OES.

Samples	Sn (at. %)	Ni (at. %)	Se (at. %)	O (at. %)	C (at. %)
$\text{SnSe}_2$	28.46		57.62	7.14	6.78
$\text{NiSe}_2$		28.29	58.67	6.82	6.22
$\text{Sn}_{0.33}\text{Ni}_{0.67}\text{Se}_2$	9.49	19.35	57.03	6.95	7.18
$\text{Sn}_{0.5}\text{Ni}_{0.5}\text{Se}_2$	14.77	15.13	56.32	6.72	7.06
$\text{Sn}_{0.67}\text{Ni}_{0.33}\text{Se}_2$	19.41	10.17	56.56	7.01	6.85

Sn, Ni, Se, O and C contents were detected by ICP-OES measurement.

**Table S2.** Flexible ASC device properties comparison with the reported literature.

Reported Devices	Electrolyte	Device window (V)	Energy density (W h kg <sup>-1</sup> )	Power density (W kg <sup>-1</sup> )	Stability (Cycles)	Reference
$\text{Sn}_{0.33}\text{Ni}_{0.67}\text{Se}_2/\text{Fe}_2\text{O}_3@ \text{NG}$	KOH-PVA	0-1.6 V	90.3	631	96.41% (10000 cycles)	This work
$\text{Ni}_3\text{Se}_2 \text{NSs}@\text{CF}/\text{AC}@\text{CC}$	1 M KOH + 0.02 m $\text{K}_3\text{Fe}(\text{CN})_6$	0-1.5 V	32.8	677	98% (5000 cycles)	1
AC//H-NiCoSe <sub>2</sub>	6 M KOH	0-1.5 V	25.5	3750	82.3% (5200 cycles)	2
$(\text{Ni}_{0.1}\text{Co}_{0.9})_9\text{Se}_8@\text{NF}/\text{rGO}@ \text{NF}$	PVA/KOH	0-1.6 V	17.0	3100	88.8% (3000 cycles)	3
Graphene//(NiCo) <sub>0.85</sub> Se	1 M KOH	0-1.8 V	12.5	112.5	85% (10 000 cycles)	4
CC/H-Ni@Al-Co-S//multilayer graphene/CNT	PVA-KOH	0-1.8 V	65.7	765.3	90.6% (10 000 cycles)	5
$\text{NiCo}_2\text{S}_4/\text{CFP-AC}$	2 M KOH	0-1.5 V	180	17.3	88.6% (5 000 cycles)	6
Ni-Co-S/G//PCNS	6 M KOH	0-1.6 V	43.3	800	85% (10 000 cycles)	7
$\text{NiCo}_2\text{S}_4/\text{G/CS}$	6 M KOH	0-1.6 V	42.3	476	78.6% (10 000 cycles)	8
$\text{NiCo}_2\text{S}_4/\text{NCF}/\text{OMC/NCF}$	6 M KOH	0-1.6 V	45.5	512	70.4% (10 000 cycles)	9
$\text{Ni}_3\text{Se}_2@\text{Ni}(\text{OH})_2/\text{AC}$	7 M KOH	0-1.6 V	50.9 Wh L <sup>-1</sup>	83.62 W L <sup>-1</sup>	83.6% (10 000 cycles)	10
$\text{Ni}_{0.67}\text{Co}_{0.33}\text{Se}/\text{RGO}$	6 M KOH	0-1.5 V	36.7	750	90.8% (2000 cycles)	11

NiSe-G//AC	6 M KOH	0-1.6 V	50.1	816	84.4% (3000 cycles)	12
Ni <sub>0.85</sub> Se nanoarray//AC	3 M KOH	0-1.6 V	65.02	103.33	93.7% (10 000 cycles)	13
NiCoP nanoplates//graphene	1 M KOH	0-1.5 V	32.9	1301	83% (5000 cycles)	14
CoMoO <sub>4</sub> //AC	2M KOH	0-1.8	21.1	300	87.42% (10 000 cycles)	15

## Notes and references

1. G. Nagaraju, S. M. Cha, S. C. Sekhar, J. S. Yu, *Adv. Energy Mater.*, 2017, **7**, 1601362.
2. L. Hou, Y. Shi, C. Wu, Y. Zhang, Y. Ma, X. Sun, J. Sun, X. Zhang, C. Yuan, *Adv. Funct. Mater.*, 2018, **28**, 1705921.
3. P. Yang, Z. Wu, Y. Jiang, Z. Pan, W. Tian, L. Jiang, L. Hu, *Adv. Energy Mater.*, 2018, **8**, 1801392.
4. C. Xia, Q. Jiang, C. Zhao, P. M. Beaujuge, H. N. Alshareef, *Nano Energy.*, 2016, **24**, 78.
5. J. Huang, J. Wei, Y. Xiao, Y. Xu, Y. Xiao, Y. Wang, L. Tan, K. Yuan, Y. Chen, *ACS Nano*, 2018, **12**, 3030.
6. X. Xiong, G. Waller, D. Ding, D. Chen, B. Rainwater, B. Zhao, Z. Wang, M. Liu, *Nano Energy*, 2015, **16**, 71.
7. J. Yang, C. Yu, X. Fan, S. Liang, S. Li, H. Huang, Z. Ling, C. Hao, J. Qiu, *Energy Environ. Sci.*, 2016, **9**, 1299.
8. L. Shen, L. Yu, H. B. Wu, X.-Y. Yu, X. Zhang, X. W. D. Lou, *Nat. Commun.*, 2015, **6**, 6694.
9. L. Shen, J. Wang, G. Xu, H. Li, H. Dou, X. Zhang, *Adv. Energy Mater.*, 2015, **5**, 1400977.
10. X. Shi, J. Key, S. Ji, V. Linkov, F. Liu, H. Wang, H. Gai, R. Wang, *Small*, 2018, 1802861.
11. H. Chen, S. Chen, M. Fan, C. Li, D. Chen, G. Tian, K. Shu, *J. Mater. Chem. A*, 2015, **3**, 23653.
12. B. Kirubasankar, V. Murugadoss, J. Lin, T. Ding, M. Dong, H. Liu, J. Zhang, T. Li, N. Wang, Z. Guo, *Nanoscale*, 2018, **10**, 20414.

13. L. Du, W. Du, H. Ren, N. Wang, Z. Yao, X. Shi, B. Zhang, J. Zai, X. Qian, *J. Mater. Chem. A*, 2017, **5**, 22527.
14. H. Liang, C. Xia, Q. Jiang, A. N. Gandi, U. Schwingenschlögl, H. N. Alshareef, *Nano Energy*, 2017, **35**, 331.
15. X. Yu, B. Lu, Z. Xu, *Adv. Mater.*, 2014, **26**, 1044.