

**One-Step Synthesis of Flower-Like Bi<sub>2</sub>O<sub>3</sub>/Bi<sub>2</sub>Se<sub>3</sub> Nanoarchitectures  
and NiCoSe<sub>2</sub>/Ni<sub>0.85</sub>Se Nanoparticles with Appealing Rate Capability  
for Constructing High-Energy and Long-Cycle-Life Asymmetric  
Aqueous Batteries**

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### Calculations:

(1) The specific capacity ( $Q$ ) and specific capacitance ( $C$ ) of the  $\text{Bi}_2\text{O}_3/\text{Bi}_2\text{Se}_3$  NFs or  $\text{NiCoSe}_2/\text{Ni}_{0.85}\text{Se}$  NPs on graphite substrate electrode calculated from GCD curves are obtained according to the following equation:

$$Q = \frac{I\Delta t}{\Delta V}; C = \frac{I\Delta t}{m\Delta V}$$

where  $I$  is the discharge current,  $\Delta t$  is the discharge time in GV test,  $m$  is the active material mass, and  $\Delta V$  is the voltage window.

(2) The specific capacitance of the  $\text{Bi}_2\text{O}_3/\text{Bi}_2\text{Se}_3$  NFs// $\text{NiCoSe}_2/\text{Ni}_{0.85}\text{Se}$  NPs asymmetric aqueous battery (AAB) device can be got in accordance with the following equation:

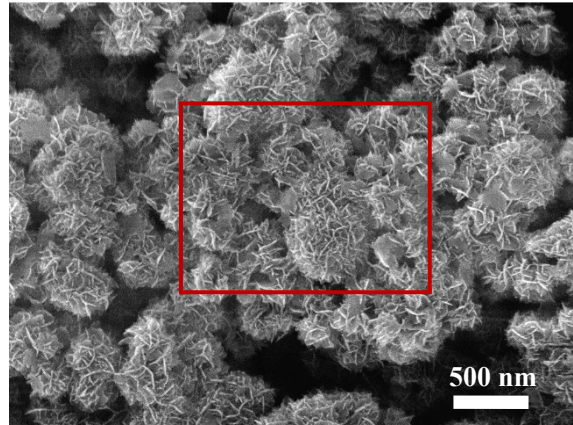
$$C_{\text{device}} = \frac{I\Delta t}{M\Delta V}$$

Herein,  $I$  is the discharge current,  $\Delta t$  is the discharge time in GCD test,  $M$  is the total mass of both positive and negative electrodes, and  $\Delta V$  is the voltage window of the device.

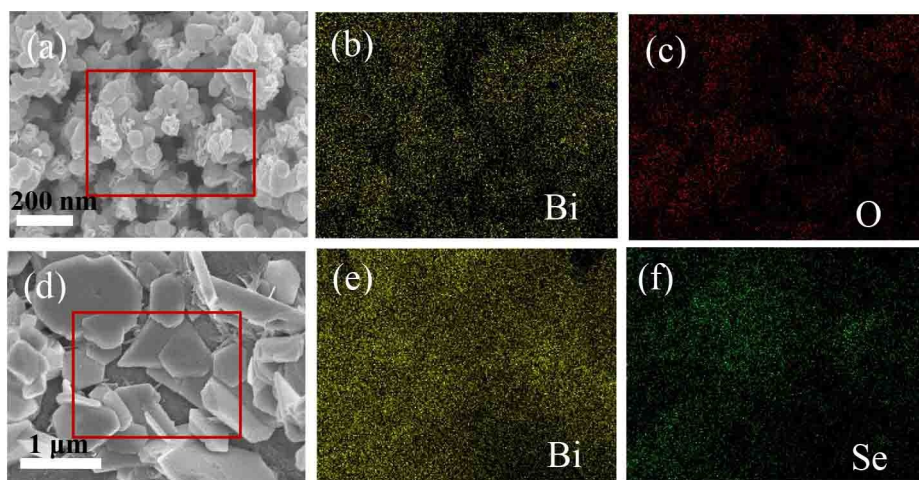
(3) Methods to calculate the energy and power density of the ASC device:

$$E = \frac{1}{2} C_{\text{device}} \Delta V^2; P = \frac{E}{t}$$

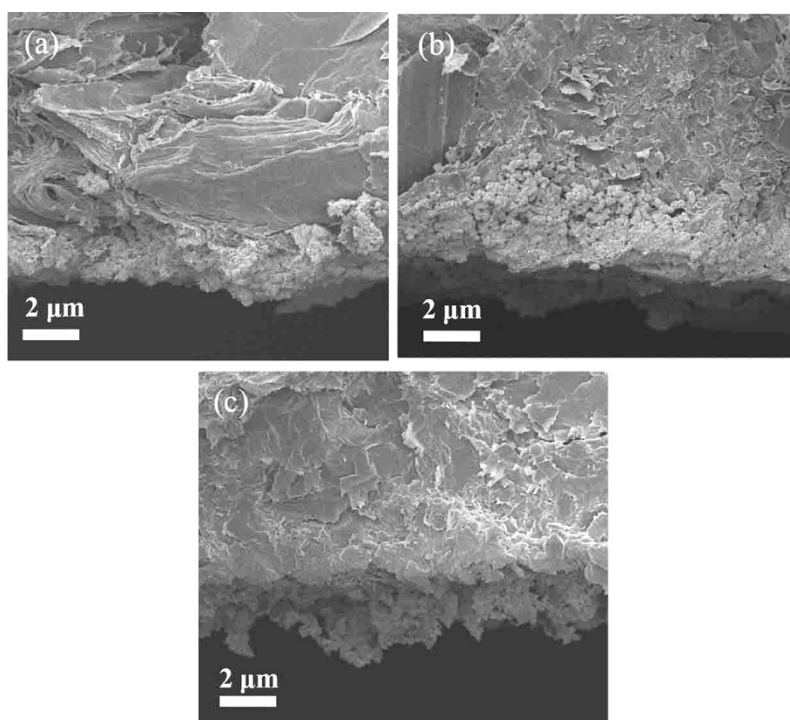
Here,  $C_{\text{device}}$  is the specific capacitance of the device,  $\Delta V$  is the potential window, and  $t$  is the discharge time.



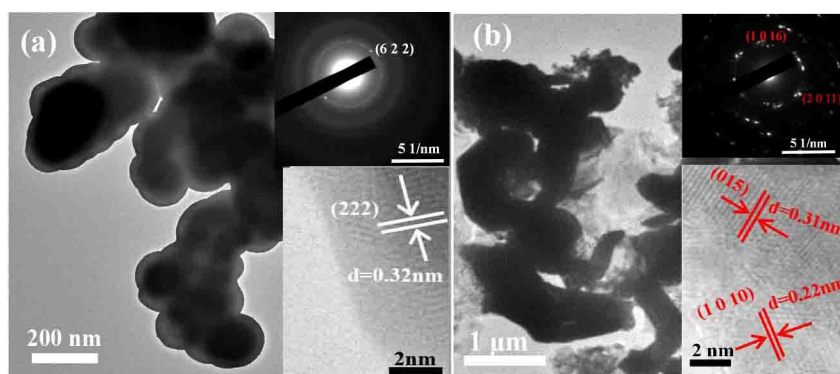
**Fig. S1.** SEM image of the obtained  $\text{Bi}_2\text{O}_3/\text{Bi}_2\text{Se}_3$  NFs.



**Fig. S2.** The elemental mapping of the  $\text{Bi}_2\text{O}_3$  (a-c) and  $\text{Bi}_2\text{Se}_3$  sample (d-f).



**Fig. S3.** Low-resolution cross-section SEM images of the  $\text{Bi}_2\text{O}_3$  (a),  $\text{Bi}_2\text{Se}_3$  (b) and  $\text{Bi}_2\text{O}_3/\text{Bi}_2\text{Se}_3$  (c).



**Fig. S4.** The typical TEM images of the Bi<sub>2</sub>O<sub>3</sub> (a) and Bi<sub>2</sub>Se<sub>3</sub> (b) with the insets showing the corresponding SAED patterns and HRTEM images.

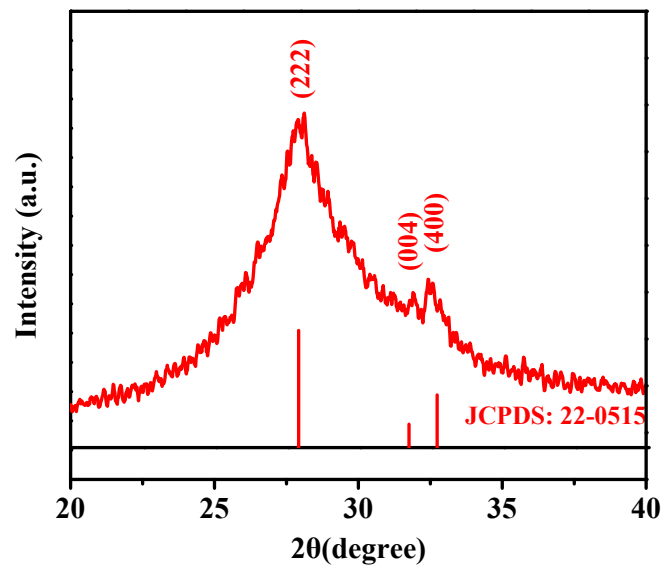


Fig. S5. Local-magnification XRD pattern of Bi<sub>2</sub>O<sub>3</sub> NSs.

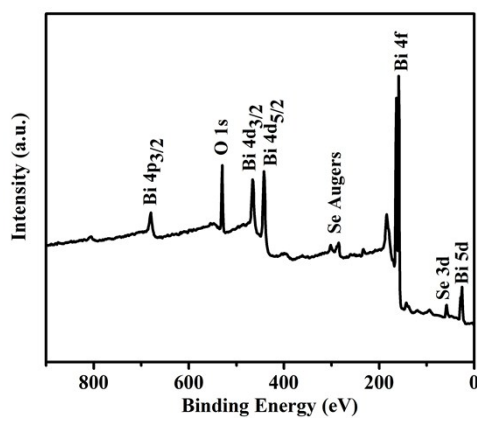


Fig. S6. XPS survey spectrum of Bi<sub>2</sub>O<sub>3</sub>/Bi<sub>2</sub>Se<sub>3</sub> NFs.

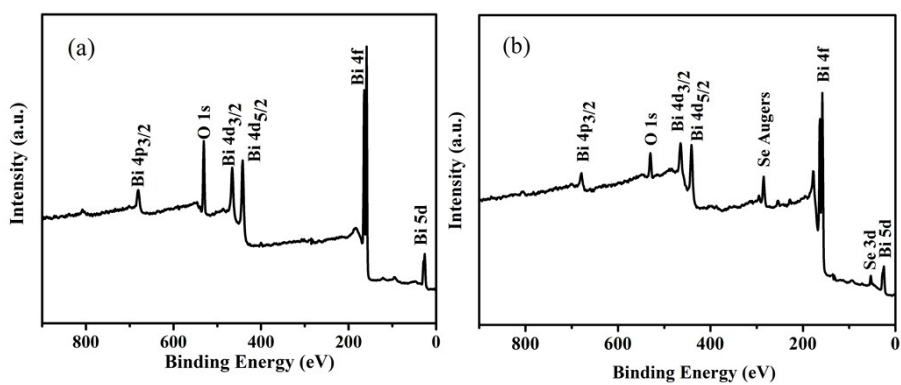


Fig. S7. XPS survey spectrum of the pristine  $\text{Bi}_2\text{O}_3$  (a) and  $\text{Bi}_2\text{Se}_3$  (b) products .

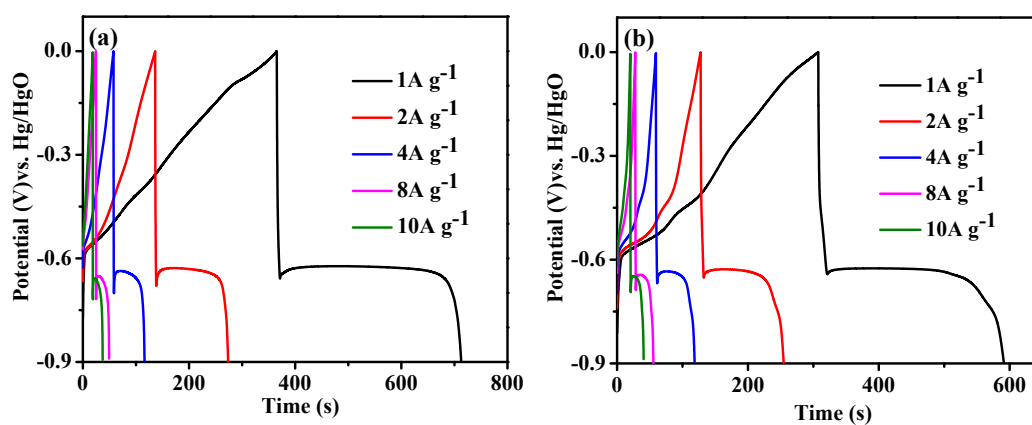
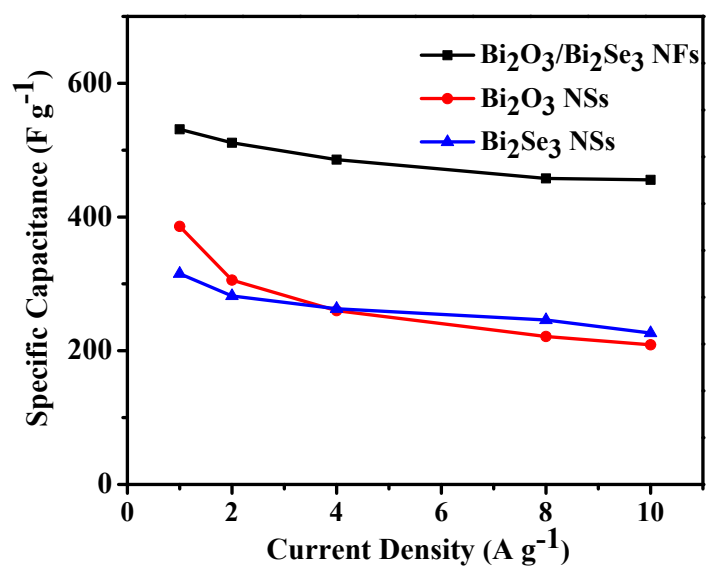


Fig. S8. GCD profiles at different current densities of the  $\text{Bi}_2\text{O}_3$  NSs (a) and  $\text{Bi}_2\text{Se}_3$  NSs (b) electrode

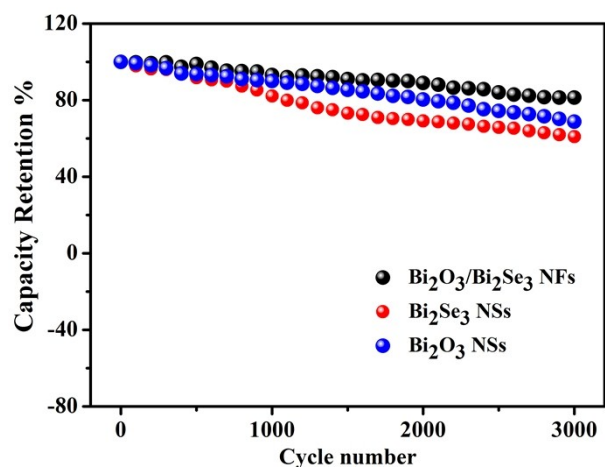


**Fig. S9.** specific capacitance values of the Bi<sub>2</sub>O<sub>3</sub> NSs, Bi<sub>2</sub>O<sub>3</sub>/Bi<sub>2</sub>Se<sub>3</sub> NFs and Bi<sub>2</sub>Se<sub>3</sub> NSs electrode versus different current densities



**Table S1.** Comparison of the electrochemical properties of the Bi<sub>2</sub>O<sub>3</sub>/Bi<sub>2</sub>Se<sub>3</sub> NFs with previously reported negative electrode materials.

Material	Fabrication method	current collector	Specific capacitance or capacity	Rate performance	Reference
Bi <sub>2</sub> O <sub>3</sub>	Hydrothermal	Ni foam	447 F g <sup>-1</sup> (2A g <sup>-1</sup> )	260 F g <sup>-1</sup> (10 A g <sup>-1</sup> )	S1
FeOOH	Electrodeposition	Polyamide Nanofiber Film	315 F g <sup>-1</sup> (0.5 A g <sup>-1</sup> )	194 F g <sup>-1</sup> (10 A g <sup>-1</sup> )	S2
Bi <sub>2</sub> S <sub>3</sub>	Hydrothermal and calcination	S-NCNF	466 F g <sup>-1</sup> (1 A g <sup>-1</sup> )	299 F g <sup>-1</sup> (8 A g <sup>-1</sup> )	S3
Bi <sub>2</sub> O <sub>3</sub>	Room-temperature wet chemical method.	Ni foam	155 mAh g <sup>-1</sup> (0.4 A g <sup>-1</sup> )	58 mAh g <sup>-1</sup> (1.8 A g <sup>-1</sup> )	S4
Fe <sub>2</sub> O <sub>3</sub>	Hydrothermal	Ni foam	158.9 mAh g <sup>-1</sup> (1 A g <sup>-1</sup> )	32.8 mAh g <sup>-1</sup> (10 A g <sup>-1</sup> )	S5
WO <sub>3</sub>	Calcination	GCE	508 F g <sup>-1</sup> (1 A g <sup>-1</sup> )	332.2 F g <sup>-1</sup> (20 A g <sup>-1</sup> )	S6
MoS <sub>2</sub>	Hydrothermal	RCF	225 F g <sup>-1</sup> (0.5A g <sup>-1</sup> )	106 F g <sup>-1</sup> (5 A g <sup>-1</sup> )	S7
ZnFe <sub>2</sub> O <sub>4</sub>	Calcination	Ni foam	58.7 mAh g <sup>-1</sup> (1 A g <sup>-1</sup> )	50.2 mAh g <sup>-1</sup> (1 A g <sup>-1</sup> )	S8
Fe <sub>3</sub> O <sub>4</sub>	Hydrothermal and calcination	Ni foam	379.8 F g <sup>-1</sup> (2 A g <sup>-1</sup> )	272.2 F g <sup>-1</sup> (10 A g <sup>-1</sup> )	S9
Fe <sub>2</sub> O <sub>3</sub>	Hydrothermal	Ni foam	269 mAh g <sup>-1</sup> (0.3A g <sup>-1</sup> )	67.3 mAh g <sup>-1</sup> (12.3 A g <sup>-1</sup> )	S10
<b>Bi<sub>2</sub>O<sub>3</sub>/Bi<sub>2</sub>Se<sub>3</sub></b>	<b>Solvothermal</b>	<b>Graphite substrate</b>	<b>132.7 mAh g<sup>-1</sup> (531F g<sup>-1</sup>) at 1 A g<sup>-1</sup></b>	<b>113.8 mAh g<sup>-1</sup> (455 F g<sup>-1</sup>) at 10 A g<sup>-1</sup></b>	<b>In this work</b>



**Fig. S10.** Cyclic stability of the Bi<sub>2</sub>O<sub>3</sub>/Bi<sub>2</sub>Se<sub>3</sub> NFs Bi<sub>2</sub>O<sub>3</sub> NSs and Bi<sub>2</sub>Se<sub>3</sub> NSs negative electrode materials over 3000 cycles at 8 A g<sup>-1</sup>.

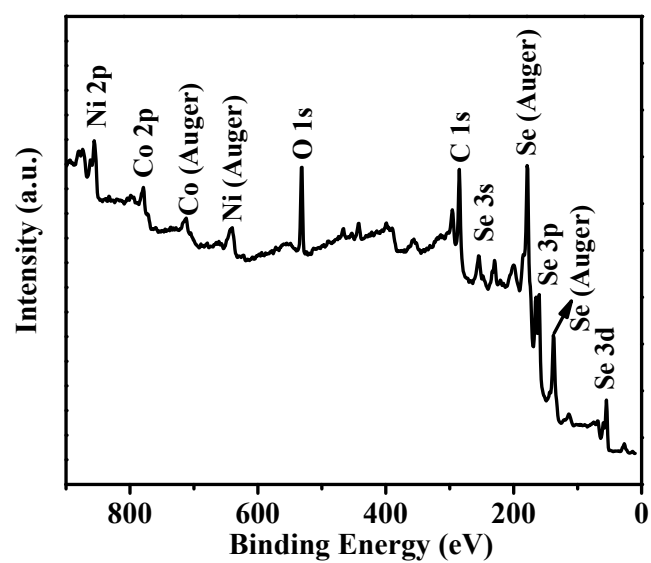


Fig. S11. XPS survey spectrum of the of the NiCoSe<sub>2</sub>/Ni<sub>0.85</sub>Se NPs.

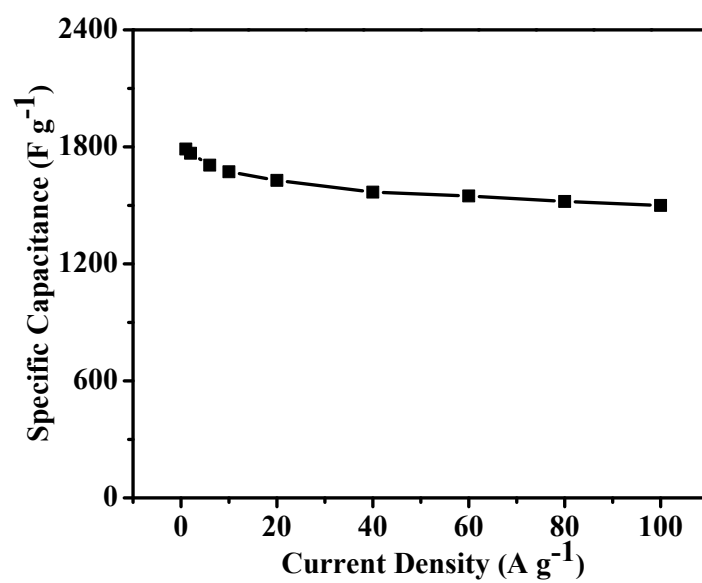
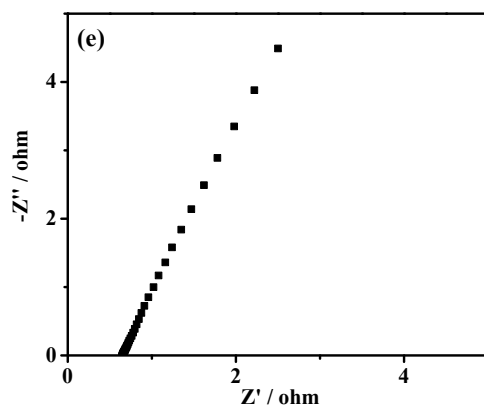


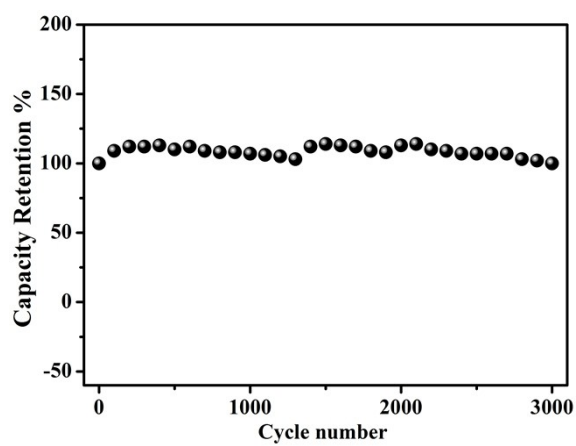
Fig. S12. Specific capacitance of the NiCoSe<sub>2</sub>/Ni<sub>0.85</sub>Se NPs as a function of current densities

**Table S2.** Electrochemical performances comparison of the CoNiSe<sub>2</sub>/Ni<sub>0.85</sub>Se NPs with other Ni-Co compound based positive electrodes fabricated by different methods.

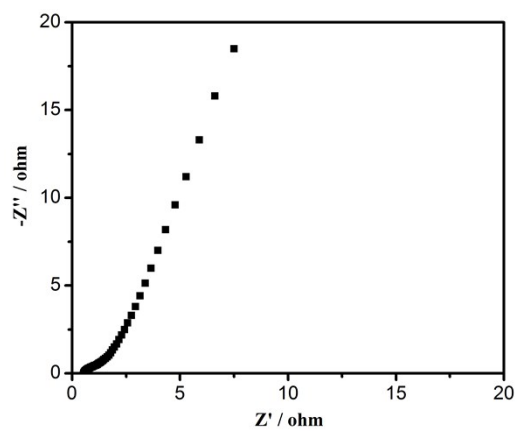
Material	Fabrication method	current collector	Specific capacitance or capacity	Rate performance	Reference
(Ni,Co)Se <sub>2</sub> /NiCo-LDH	Calcination and electrodeposition	Carbon substrate	170 mAh g <sup>-1</sup> (2 A g <sup>-1</sup> )	120.7 mAh g <sup>-1</sup> (20 A g <sup>-1</sup> )	S11
CoNi-MOF	Hydrothermal and calcination	Ni foam	1044 F g <sup>-1</sup> (2 A g <sup>-1</sup> )	569 F g <sup>-1</sup> (32A g <sup>-1</sup> )	S12
CoNi <sub>2</sub> S <sub>4</sub>	Chemical deposition	Ni foam	1530 F g <sup>-1</sup> (1 A g <sup>-1</sup> )	1346 F g <sup>-1</sup> (8 A g <sup>-1</sup> )	S13
NiCo <sub>2</sub> O <sub>4</sub> /CNT	Calcination	Ni foam	1596 F g <sup>-1</sup> (1 A g <sup>-1</sup> )	1406 F g <sup>-1</sup> (10 A g <sup>-1</sup> )	S14
Ni <sub>3</sub> S <sub>2</sub>	Hydrothermal	Ni foam	70 mAh g <sup>-1</sup> (2mA cm <sup>-2</sup> )	-----	S15
CoNiSe <sub>2</sub>	Solvothermal and calcination	Ni foam	750 F g <sup>-1</sup> (1 A g <sup>-1</sup> )	660 F g <sup>-1</sup> (20 A g <sup>-1</sup> )	S16
NiSe <sub>2</sub>	Plasma-exfoliation	Ni foam	466 F g <sup>-1</sup> (3 A g <sup>-1</sup> )	328 F g <sup>-1</sup> (20 A g <sup>-1</sup> )	S17
NiCo <sub>2</sub> O <sub>4</sub>	Hydrothermal and calcination	Carbon cloth	1055 F g <sup>-1</sup> (0.4 A g <sup>-1</sup> )	483 F g <sup>-1</sup> (10 A g <sup>-1</sup> )	S18
Co <sub>3</sub> O <sub>4</sub> /Ni-based MOFs	Hydrothermal	Carbon cloth	209 mAh g <sup>-1</sup> (2 A g <sup>-1</sup> )	~58 mAh g <sup>-1</sup> (10 A g <sup>-1</sup> )	S19
Co <sub>3</sub> O <sub>4</sub> /Co(OH) <sub>2</sub>	Hydrothermal	polyethylene naphthalate	184.9 mAh g <sup>-1</sup> (1 A g <sup>-1</sup> )	129.4 mAh g <sup>-1</sup> (16 A g <sup>-1</sup> )	S20
<b>NiCoSe<sub>2</sub>/Ni<sub>0.85</sub>Se</b>	<b>Electrodeposition</b>	<b>Graphite substrate</b>	<b>248.4 mAh g<sup>-1</sup> (1788 F g<sup>-1</sup>) at 1 A g<sup>-1</sup></b>	<b>208 mAh g<sup>-1</sup> (1500 F g<sup>-1</sup>) at 100A g<sup>-1</sup></b>	<b>In this work</b>



**Fig. S13.** Nyquist plot of the NiCoSe<sub>2</sub>/Ni<sub>0.85</sub>Se NPs hybrid electrode.



**Fig. S14.** Cyclic stability of the NiCoSe<sub>2</sub>/Ni<sub>0.85</sub>Se NPs hybrid positive electrode materials over 3000 cycles at 10 A g<sup>-1</sup>.



**Fig. S15.** The Nyquist plot of the device.

**Table. S3** Cycle performance comparison of the assembled AAB with other state-of-the-art devices with various positive and negative electrodes.

ASC devices	Cell voltage (V)	Cycle performance	Reference
Fe <sub>2</sub> O <sub>3</sub> -P//MnO <sub>2</sub>	1.6	88% retention after 9000 cycles	S21
MnO <sub>2</sub> //Fe <sub>2</sub> O <sub>3</sub>	2.2	83% retention after 3000 cycles	S22
Co <sub>3</sub> O <sub>4</sub> // $\gamma$ -Fe <sub>2</sub> O <sub>3</sub>	1.7	80.1% retention after 5000 cycles	S23
Ni-Co-S-W/NF//AC/NF	1.8	91.7% retention after 6000 cycles	S24
MnO <sub>2</sub> /CNT//CNT/PPy	1.5	80% retention after 5000 cycles	S25
Ni-Co-N/GP//GOP	1.5	95% retention after 5000 cycles	S26
NF@NiO//FeOOH HSCs	1.7	84.7% retention after 10000 cycles	S27
ZnNiCo-P//PPD-rGOs	1.6	89% retention after 8000 cycles	S28
NiO–CuO//PGH	1.6	90.4% retention after 5000 cycles	S29
CC/H-Ni@Al-Co-S//graphene/CNT	1.8	90.6% retention after 10000 cycles	S30
<b>Bi<sub>2</sub>O<sub>3</sub>/Bi<sub>2</sub>Se<sub>3</sub>//NiCoSe<sub>2</sub>/Ni<sub>0.85</sub>Se</b>	<b>1.6</b>	<b>91.5% retention after 10000 cycles</b>	<b>In this work</b>

## References

- S1 N. Shinde, Q. Xia, J. Yun, R. Mane and K. Kim, *Acs Appl. Mater. Interfaces*, 2018, **10**, 11037.
- S2 T. Gao, Z. Zhou, J. Yu, D. Cao, G. Wang, B. Ding and Y. Li, *ACS appl. mater. Interfaces*, 2018, **10**, 23834.
- S3 W. Zong, F. Lai, G. He, J. Feng, W. Wang, R. Lian, Y. Miao, G. Wang, I. Parkin and T. Liu, *Small*, 2018, **14**, 1801562.
- S4 N. Shinde, Q. Xia, J. Yun, P. Shinde, S. Shaikh, R. Sahoo, S. Mathur, R. Mane and K. Kim, *Electrochim. Acta*, 2019, **296**, 308.
- S5 X. Yun, J. Li, Z. Luo, J. Tang and Y. Zhu, *Electrochim. Acta*, 2019, **302**, 449.
- S6 D. Mandal, P. Routh and A. K. Nandi, *Small*, 2018, **14**, 1702881.
- S7 C. Zhao, Y. Zhou, Z. Ge, C. Zhao and X. Qian, *Carbon*, 2018, **127**, 699.
- S8 Yang, Z. Han, F. Zheng, J. Sun, Z. Qiao, X. Yang, L. Li, C. Li, X. Song and B. Cao, *Carbon*, 2018, **134**, 15.
- S9 F. Li, H. Chen, X. Liu, S. Zhu, J. Jia, C. Xu, F. Dong, Z. Wen and Y. Zhang, *J. Mater. Chem. A*, 2016, **4**, 2096.
- S10 J. Zhu, L. Li, Z. Xiong, Y. Hu and J. Jiang, *ACS Sustainable Chem. Eng.*, 2017, **5**, 269.
- S11 X. Li, H. Wu, C. Guan, A. Elshahawy, Y. Dong, S. Pennycook and J. Wang, *Small*, 2019, **15**, 1803895.
- S12 T. Deng, Y. Lu, W. Zhang, M. Sui, X. Shi, D. Wang and W. Zheng, *Adv. Energy Mater.*, 2018, **8**, 1702294.
- S13 X. Cao, J. He, H. Li, L. Kang, X. He, J. Sun, R. Jiang, H. Xu, Z. Lei and Z. Liu, *Small*, 2018, **14**, 1800998.



- S14 P. Wu, S. Cheng, M. Yao, L. Yang, Y. Zhu, P. Liu, O. Xing, J. Zhou, M. Wang and H. Luo, *Adv. Funct. Mater.*, 2017, **27**, 1702160.
- S15 K. Tao, Y. Gong, Q. Zhou and J. Lin, *Electrochim. Acta*, 2018, **286**, 65.
- S16 L. Hou, Y. Shi, C. Wu, Y. Zhang, Y. Ma, X. Sun, J. Sun, X. Zhang and C. Yuan, *Adv. Funct. Mater.*, 2018, **28**, 1705921.
- S17 A. Chang, C. Zhang, Y. Yu, Y. Yu and B. Zhang, *ACS appl. mater. Interfaces*, 2018, **10**, 41861.
- S18 C. Guan, X. Liu, W. Ren, X. Li, C. Cheng and J. Wang, *Adv. Energy Mater.*, 2017, **7**, 1602391.
- S19 L. Zhang, Y. Zhang, S. Huang, Y. Yuan, H. Li, Z. Jin, J. Wu, Q. Liao, L. Hu, J. Lu, S. Ruan and Y. Zeng, *Electrochim. Acta*, 2018, **281**, 189.
- S20 G. Lee and J. Jang, *J. Power Sources*, 2019, **423**, 115.
- S21 H. Liang, C. Xia, A. Emwas, D. Anjum, X. Miao and H. Alshareef, *Nano energy*, 2018, **49**, 155.
- S22 M. Zhang, Y. Li and Z. Shen, *J. Power Sources*, 2019, **414**, 479.
- S23 R. Wang, Y. Sui, S. Huang, Y. Pu and P. Cao, *Chem. Eng. J.*, 2018, **331**, 527.
- S24 W. He, Z. Liang, K. Ji, Q. Sun, T. Zhai and X. Xu, *Nano Res.*, 2018, **11**, 1415.
- S25 J. Yu, W. Lu, J. Smith, K. Booksh, L. Meng, Y. Huang, Q. Li, J. Byun, Y. Oh and Y. Yan, *Adv. Energy Mater.*, 2017, **7**, 1600976.
- S26 F. Liu, L. Zeng, Y. Chen, R. Zhang, R. Yang, J. Pang, L. Ding, H. Liu and W. Zhou, *Nano Energy*, 2019, **61**, 18.
- S27 Q. Chen, J. Li, C. Liao, G. Hu, Y. Fu, O. K. Asare, S. Shi, Z. Liu, L. Zhou and L.

Mai, *J. Mater. Chem. A*, 2018, **6**, 19488.

S28 J. Li, Z. Liu, Q. Zhang, Y. Cheng, B. Zhao, S. Dai, H.-H. Wu, K. Zhang, D. Ding and Y. Wu, *Nano Energy*, 2019, **57**, 22.

S29 Z. Fang, S. ur Rehman, M. Sun, Y. Yuan, S. Jin and H. Bi, *J. Mater. Chem. A*, 2018, **6**, 21131.

S30 J. Huang, J. Wei, Y. Xiao, Y. Xu, Y. Xiao, Y. Wang, L. Tan, K. Yuan and Y. Chen, *ACS nano*, 2018, **12**, 3030.