

Hierarchical electrodes of NiCo₂S₄ nanosheets anchored sulfur-doped Co₃O₄ nanoneedles with advanced performance for battery-supercapacitor hybrid devices

Yu Ouyang,^a Haitao Ye,^a Xifeng Xia,^a Xinyan Jiao,^a Guangmin Li,^a Sadaf Mutahir,^a
Liang Wang,^a Daniel Mandler,^b Wu Lei,^{*a} Qingli Hao^{*a}

^aKey Laboratory of Soft Chemistry and Functional Materials, Ministry of Education,
Nanjing University of Science and Technology, 210094, China

^bInstitute of Chemistry, The Hebrew University of Jerusalem, Jerusalem 9190401,
Israel

*E-mail: qinglihao@njust.edu.cn (Q. Hao); leiwuhao@njust.edu.cn (W. Lei).

*Fax: +86-25-84315190; Tel: +86-25-84315190

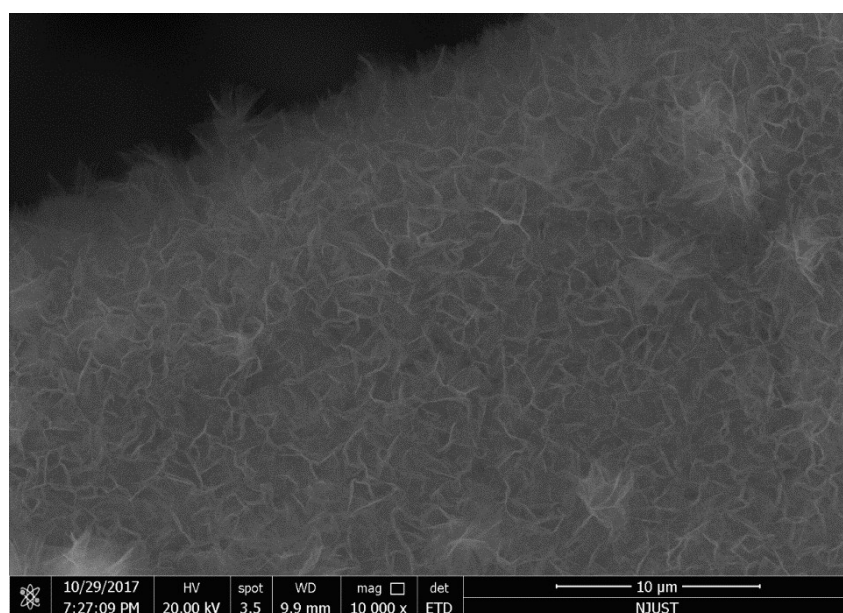


Fig. S1 SEM image of NF/NiCo₂S₄.

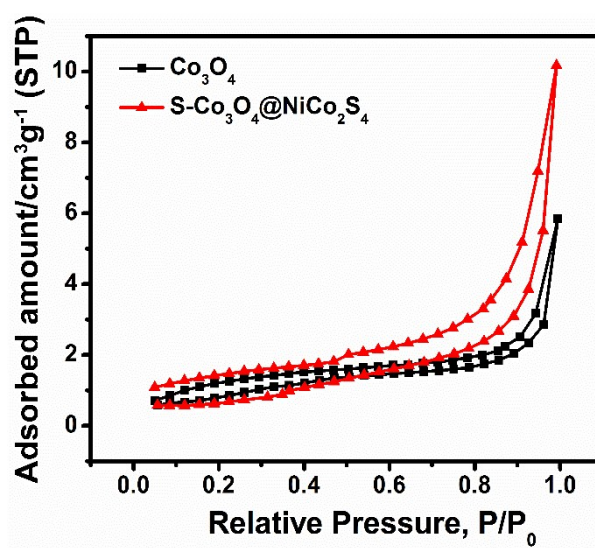


Fig. S2 Nitrogen adsorption/desorption of the NF/S-Co₃O₄ and NF/S-Co₃O₄@NiCo₂S₄.

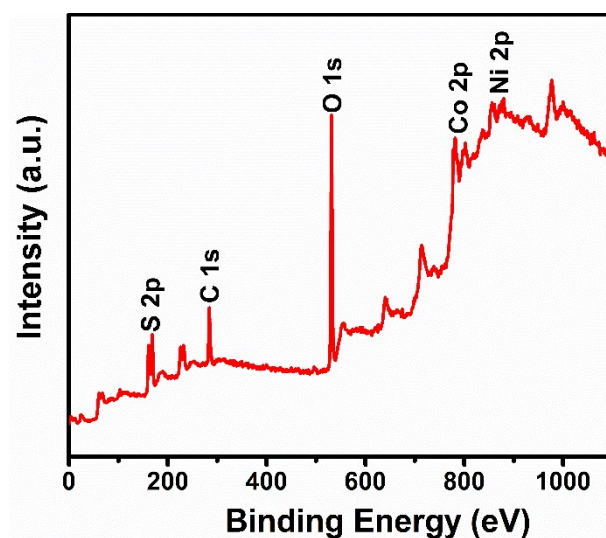


Fig. S3 XPS wide-scan spectra of S-Co₃O₄@NiCo₂S₄.

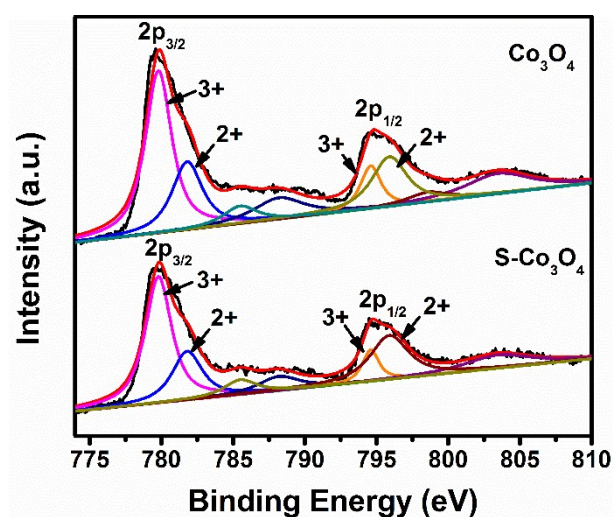


Fig. S4 High-resolution XPS spectra for Co of S-Co₃O₄.

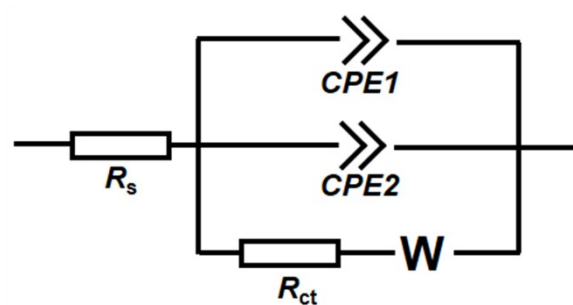


Figure S5. The equivalent electrical circuit in NF/S-Co₃O₄@NiCo₂S₄, NF/S-Co₃O₄, NF/Co₃O₄, and NF/NiCo₂S₄.

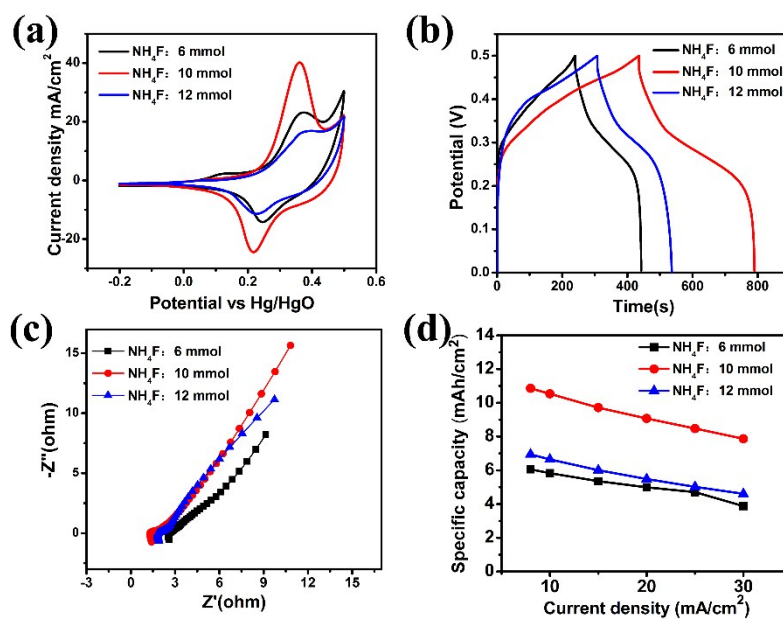


Fig. S6 Electrochemical performance of S-Co₃O₄@NiCo₂S₄ with different molar weight (6, 10, 12 mmol) of NH₄F. (a) EIS curves; (b) CV curves; (c) GCD curves; and (d) Specific capacity at different current densities from 8 to 30 mA/cm².

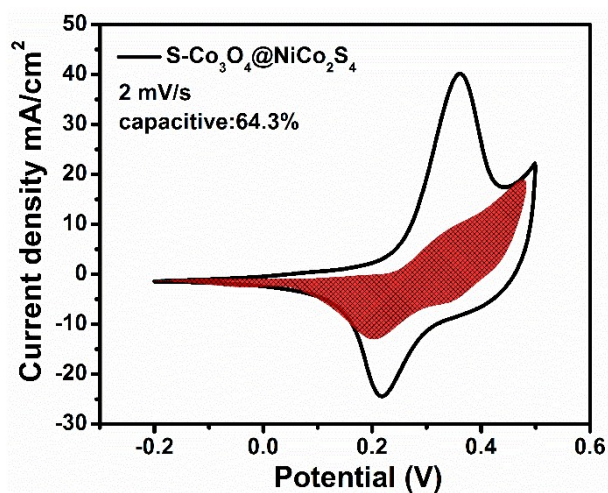


Fig. S7 Capacitive contribution of the NF/S-Co₃O₄@NiCo₂S₄.

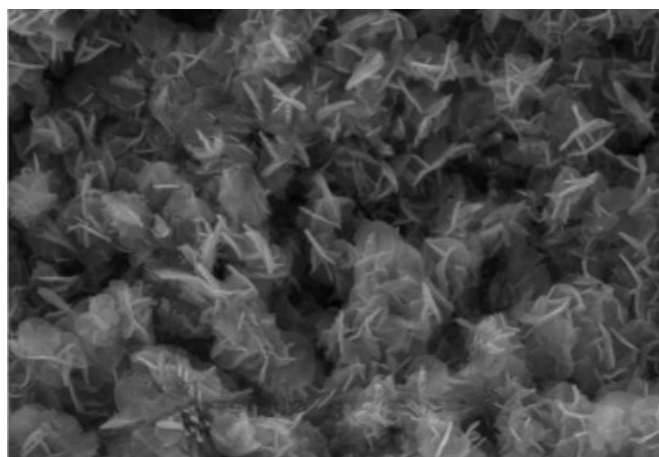


Fig. S8 SEM of NF/S-Co₃O₄@NiCo₂S₄ after 5,000 cycles.

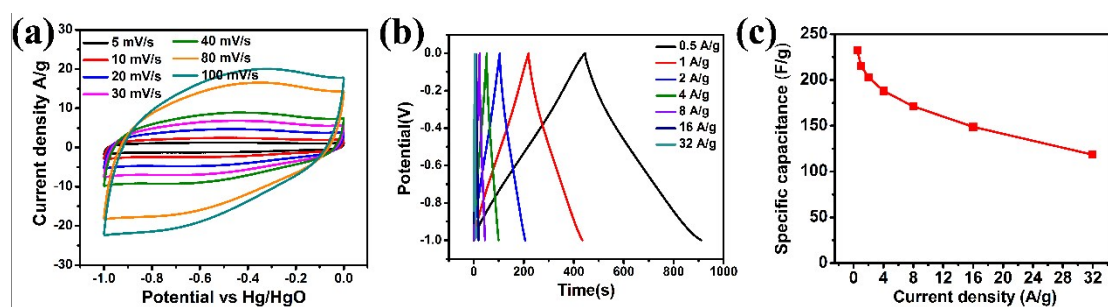


Fig. S9 (a) CV curves of AC at different scan rates. (b) GCD curves of AC at different current densities. (c) Specific capacity at different current densities from 0.5 to 30 A/g.

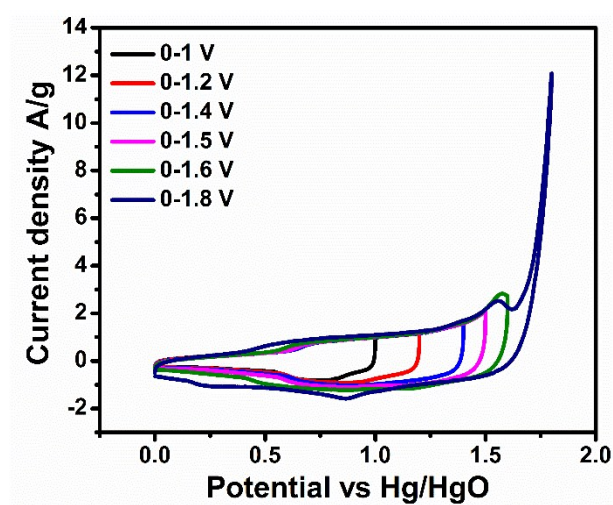


Fig. S10. CV curves of BSH device with different window voltages at 5 mV/s.

Table S1. Comparison of electrochemical performance between the NF/S-Co₃O₄@NiCo₂S₄ and the reported sulfides-based electrode materials.

Electrode material	Specific capacitance (capacity)	Cycling performance	Ref
hollow ellipsoid Ni–Mn sulfides	1636.8 F/g at 2 A/g	95.1% (4000 cycles)	[1]
NiCo ₂ S ₄ /NCF	1231 F/g at 2 A/g	90.4% (2000 cycles)	[2]
NiCo ₂ S ₄ @Ni–Mn LDH arrays/GS	1.48 F/cm ² at 5 mA/cm ²	88.3% (1000 cycles)	[3]
porous NiCo ₂ S ₄ aerogel	1268 F/g at 1 A/g	--	[4]
P-Doped NiCo ₂ S ₄ nanotubes	8.03 F/cm ² at 2 mA/cm ²	87.5% (5000 cycles)	[5]
NiCo ₂ O ₄ @NiCo ₂ S ₄ Nanocomposite	1872 F/g at 1 A/g	65% (4000 cycles)	[6]
Onion-like NiCo ₂ S ₄ particles	1016 F/g at 2 A/g	87% (10000 cycles)	[7]
FeCo ₂ S ₄ -NiCo ₂ S ₄ composite	1519 F/g at 5 mA/cm ²	--	[8]
NF/S-Co ₃ O ₄ @NiCo ₂ S ₄	1886 F/g at 2.5 A/g or (6 F/cm ² at 8 mA/cm ²)	97.3% (5000 cycles)	This work

Ref:

1. C. Cheng, D. Kong, C. Wei, W. Du, J. Zhao, Y. Feng, Q. Duan, *Dalton Transactions*, 2017, **46**, 5406-5413.
2. L. Shen, J. Wang, G. Xu, H. Li, H. Dou, X. Zhang, *Adv. Energy Mater*, 2015, **5**, 1400977.
3. H. Wan, J. Liu, Y. Ruan, L. Lv, L. Peng, X. Ji, J. Jiang, *Appl. Mater. Interfaces*, 2015, **7**, 15840-15847.
4. Q. Gao, X. Wang, Z. Shi, Z. Ye, W. Wang, N. Zhang, Z. Hong, M. Zhi, *Chem. Eng. J.*, 2018, **331**, 185-193.
5. J. Lin, Y. Wang, X. Zheng, H. Liang, H. Jia, J. Qi, J. Cao, J. Tu, W. Fei, J. Feng, *Dalton Transactions*, 2018, **47**, 8771-8778.
6. H. Rong, T. Chen, R. Shi, Y. Zhang, Z. Wang, *ACS Omega*, 2018, **3**, 5634-5642.
7. B. Guan, L. Yu, X. Wang, S. Song, X. Lou, *Adv. Mater.*, 2017, **29**, 1605051.
8. J. Zhu, S. Tang, J. Wu, X. Shi, B. Zhu, X. Meng, *Adv. Energy Mater.*, 2017, **7**, 1601234.