Supporting Information

Precise design and in-situ synthesis of hollow Co_9S_8@CoNi-LDH heterostructure for high performance supercapacitors

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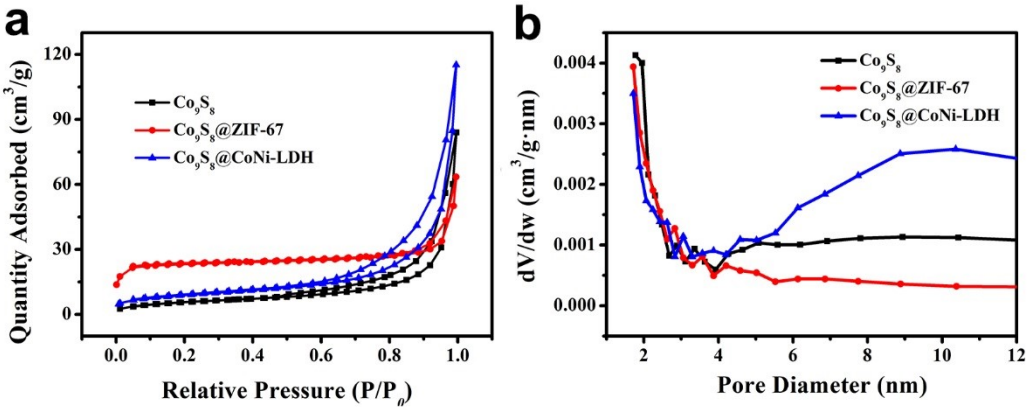

Fig. S1 (a) N_2 adsorption-desorption isotherms and (b) pore-size distribution of Co_9S_8, Co_9S_8@ZIF-67 and Co_9S_8@CoNi-LDH.

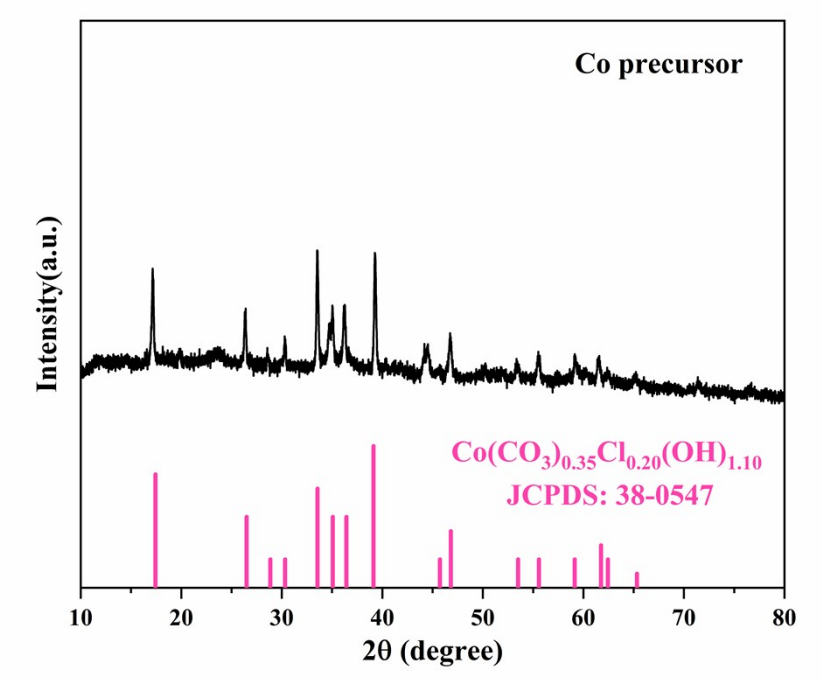

Fig. S2 XRD pattern of Co precursor.

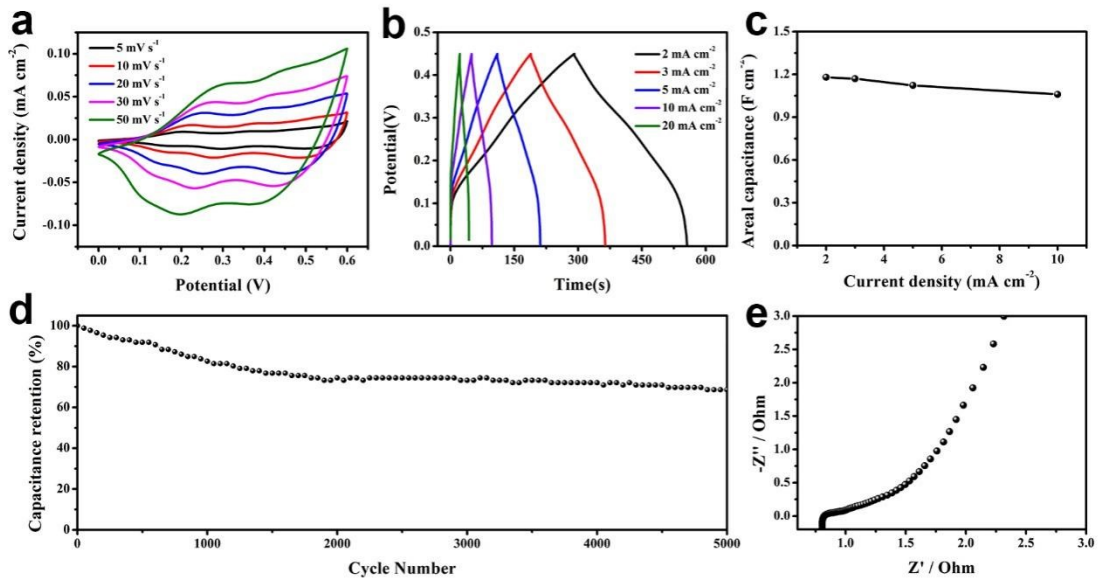

Fig. S3 The electrochemical properties of CoCH/NF. (a) CV curves at scan rates of 5-50 mV s^{-1}; (b) GCD curves at current density of 2-20 mA cm^{-2}; (c) The specific capacitance at current densities of 1-10 mA cm^{-2}; (d) The cycling capacity after 5000 cycles, (e) Nyquist plots of ESI.

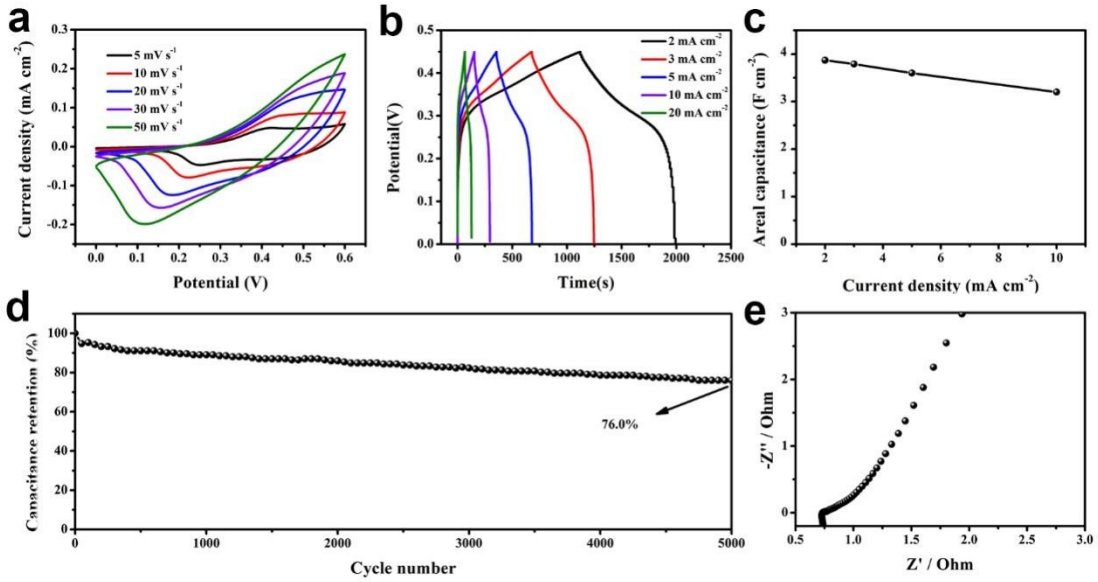

Fig. S4 The electrochemical properties of Co_9S_8/NF. (a) CV curves at scan rates of 5-50 mV s^{-1}; (b) GCD curves at current density of 2-20 mA cm^{-2}; (c) The specific capacitance at current densities of 1-10 mA cm^{-2}; (d) The cycling capacity after 5000 cycles, (e) Nyquist plots of ESI.

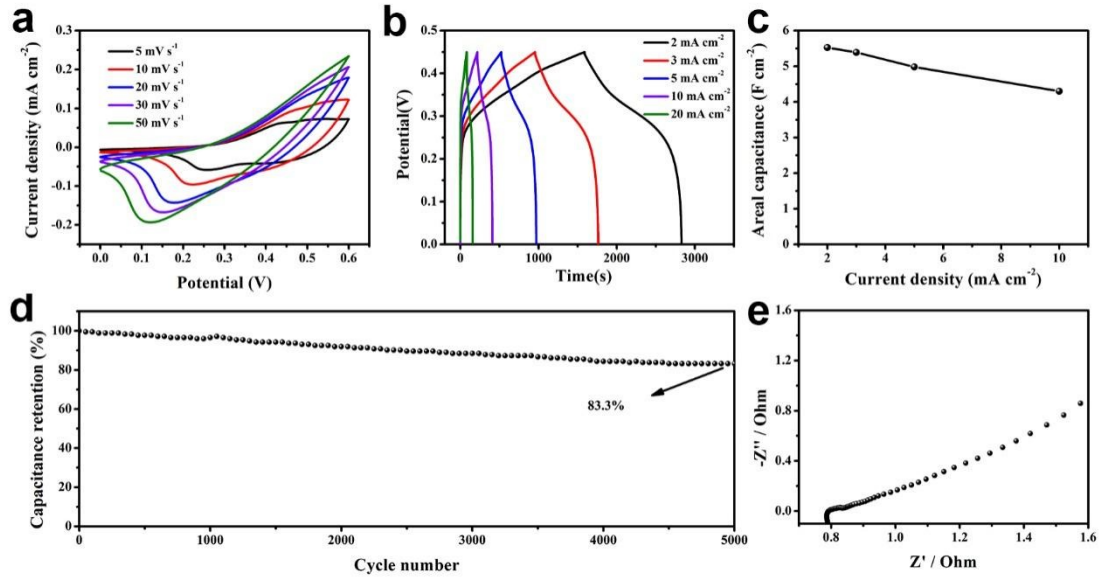

Fig. S5 The electrochemical properties of Co_9S_8@ZIF-67/NF. (a) CV curves at scan rates of 5-50 mV s^{-1}; (b) GCD curves at current density of 2-20 mA cm^{-2}; (c) The specific capacitance at current densities of 1-10 mA cm^{-2}; (d) The cycling capacity after 5000 cycles, (e) Nyquist plots of ESI.

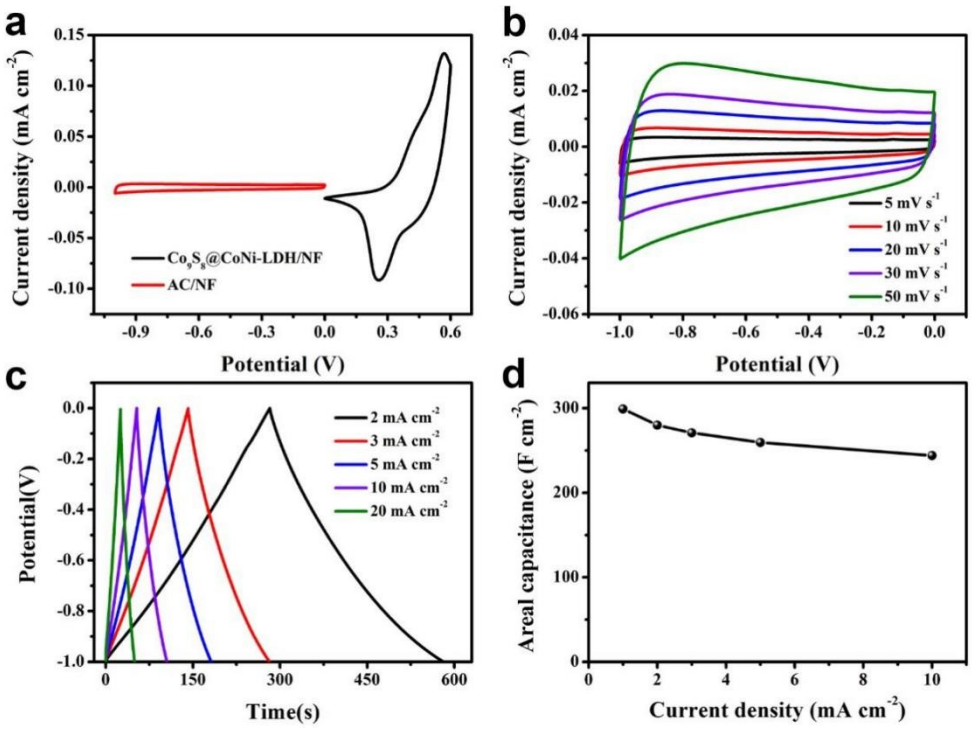

Fig. S6 (a) The CV curves of AC and Co_9S_8@CoNi-LDH/NF; (b) The CV curves of the AC at scan rates of 5-50 mV s^{-1}; (c) The GCD curves of AC at current density of 2-20 mA cm^{-2}; (d) The specific capacity of AC at current densities of 1-10mA cm^{-2}.

Table S1 The capacitance comparison of other CoNi-LDH based electrode materials

Electrode material	Specific capacitance	Cycle performance	Ref.
CNTs/Ni Co LDH	1628 F g^{-1}at1 A g^{-1}	78.073% after 5000 cycles	S1
H-NiCo LDH@ACC	1377mC/cm^2 at 1mA/cm^2	70% after 10000 cycles	S2
CoNi-LDH	986.3 F g^{-1}at1 A g^{-1}	92.3% after 10000 cycles	S3
CNMnL-m	1644 F g^{-1} at 1 A g^{-1}	94.2% after 5000 cycles	S4
Ni_xCo_{1-x} LDH/AC	947 F g^{-1} at 1 A g^{-1}	80% after 5000 cycles	S5
CoNi LDH@Co_3O_4@CC	1021.7 F g^{-1} at 10 A g^{-1}	Not mentioned	S6
Ni/Co-LDH	1652 F·g^{-1} at 1 A·g^{-1}	99% after 2000 cycles	S7
NiCoAl-LDH/NF	5691.25 mF cm^{-2} at 1 mA cm^{-2}	73.5% after 3000 cycles	S8
4M-P@NiCo LDH	7 F cm^{-2} at 50 mA cm^{-2}	72.5% after 5000 cycles	S9
(Ni,Co)Se_2/NiCo-LDH	1224 F g^{-1} at 2A·g^{-1}	89.5% after 3000 cycles	S10
Co_3O_4@Ni-Co LDH/NF	1069 C g^{-1} at 1 A·g^{-1}	67.9% after 5000 cycles	S11
Co_9S_8@CoNi-LDH	9.7 F cm^{-2} (2411 F g^{-1},1085 C g^{-1}) at 2 mA cm^{-2}	92% after 5000 cycles	This work

References

S1. M. Huang, Y. Wang, J. Chen, D. He, J. He and Y. Wang, Biomimetic design of Ni Co LDH composites linked by carbon nanotubes with plant conduction tissues characteristic for hybrid supercapacitors, Electrochim. Acta, 2021, **381**, 138289.

S2. X. Xuan, M. Qian, L. Han, L. Wan, Y. Li, T. Lu, L. Pan, Y. Niu and S. Gong, In-situ growth of hollow NiCo layered double hydroxide on carbon substrate for flexible supercapacitor, Electrochim. Acta, 2019, **321**, 134710.

S3. B. Huang, W. Wang, T. Pu, J. Li, J. Zhu, C. Zhao, L. Xie and L. Chen, Two-dimensional porous (Co, Ni)-based monometallic hydroxides and bimetallic layered double hydroxides thin sheets with honeycomb-like nanostructure as positive electrode for high-performance hybrid supercapacitors, J. Colloid Interface Sci., 2018, **532**, 630-640.

S4. W. Quan, Y. Xu, Y. Wang, S. Meng, D. Jiang and M. Chen, Hierarchically structured

Co_3O_4@glucose-modified LDH architectures for high-performance supercapacitors, Appl. Surf. Sci., 2019, **488**, 639-647.

S5. T. Chen, L. Luo, X. Wu, Y. Zhou, W. Yan, M. Fan and W. Zhao, Three dimensional hierarchical porous nickel cobalt layered double hydroxides (LDHs) and nitrogen doped activated biocarbon composites for high-performance asymmetric supercapacitor, J. Alloys Compd., 2021, **859**, 158318.

S6. D. Wang, L. Tian, D. Li, Y. Xu and Q. Wei, Rational design of Co–Ni layered double hydroxides electrodeposited on Co_3O_4 nanoneedles derived from 2D metal-organic frameworks for high-performance asymmetric supercapacitors, J. Electroanal. Chem., 2020, **873**, 114377.

S7. Z. Xiao, Y. Mei, S. Yuan, H. Mei, B. Xu, Y. Bao, L. Fan, W. Kang, F. Dai, R. Wang, L. Wang, S. Hu, D. Sun and H. C. Zhou, Controlled Hydrolysis of Metal-Organic Frameworks: Hierarchical Ni/Co-Layered Double Hydroxide Microspheres for High-Performance Supercapacitors, ACS Nano, 2019, **13**, 7024-7030.

S8. P. Li, Y. Jiao, S. Yao, L. Wang and G. Chen, Dual role of nickel foam in NiCoAl-LDH ensuring high-performance for asymmetric supercapacitors, New J. Chem., 2019, **43**, 3139-3145.

S9. G. Wang, Z. Jin and W. Zhang, A phosphatized NiCo LDH 1D dendritic electrode for high energy asymmetric supercapacitors, Dalton Trans., 2019, **48**, 14853-14863.

S10. X. Li, H. Wu, C. Guan, A. M. Elshahawy, Y. Dong, S. J. Pennycook and J. Wang, (Ni,Co)Se_2/NiCo-LDH Core/Shell Structural Electrode with the Cactus-Like (Ni,Co)Se_2 Core for Asymmetric Supercapacitors, Small, 2019, **15**, 1803895.

S11. Y. Zhang, Y. Wang, J. Zhu, X. Zhang and W. Cai, Regulating the core/shell electric structure of Co_3O_4@Ni-Co layered double hydroxide on Ni foam through electrodeposition for a quasi-solid-state supercapacitor, Nanotechnology, 2021, **32**, 345702.