Supplementary Information

Multifunctional sandwich-structured double-carbon-layer modified SnS nanotubes with high capacity and stability for Liion batteries

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Figure S1. Pore size distribution (a) and N_2 adsorption-desorption isotherms (b) of the as-prepared N-DCSNs.



Figure S2. TG analysis of N-DCSNs sample at temperatures ranging from 200 to 800

°C in air.



Figure S3. XPS spectra of N-DCSNs for all elements.



Figure S4. SEM images of the N-DCSNs (a, d), the CN/SnS composites (b, e) and the

bare SnS (c, f).



Figure S5. Cyclic voltammetry curves of CN/SnS (a) and bare SnS (b) between 0.01 and 3.0 V with a scan rate of 0.5 mV s⁻¹.



Figure S6. Cyclic voltammetry curves of N-DCSNs (a), CN/SnS (b) and bare SnS (c) between 0.01 and 3.0 V with scan rate of 0.2, 0.4, 0.6, 0.8, and 1.0 mV s⁻¹, respectively.



Figure S7. Discharge-charge voltage profiles of CN/SnS (a) and bare SnS (b) at a current density of 0.2 A g^{-1} , respectively.

Samular	Specific capacity	Cycles	Current	Def
Samples	$(mAh g^{-1})$		density (A g ⁻¹)	KeI.
SnS/N-G	1120	130	0.1	1
SnS@C HSs	532	100	0.1	2
SnS/N-C particles	535	300	1.0	3
SnS/C nanofibers	648	500	0.2	4
3D porous SnS/C	607	200	1.0	5
SnS nanoparticles	410	50	0.1	6
SnS nanoflowers	600	30	0.05	7
SnS/C-CP	696	200	0.5	8
SnS NS/RGO	560	100	0.1	9
SnS-ZnS@C	302	500	0.5	10
3D SnS flowers	360	50	0.8	11
SnS-PNA	900	50	0.3	12
C@SnS/SnO ₂ @CNFs	917	200	0.2	13
N-DCSNs	911.5	270	0.2	This moult
	511.3	1000	1.0	I IIIS WOFK

 Table S1. Electrochemical performance of SnS-based anode materials for LIBs.



Figure S8. Electrochemical performance of SnS-based anode materials for LIBs.



Figure S9. (a) Nyquist plots of N-DCSNs electrodes measured in the frequency region of 10^{5} - 10^{-2} Hz after 10 and 50 cycles. (b) The real part of the complex impedance versus $\omega^{-1/2}$ at open circuit voltage for N-DCSNs electrodes after 10 and 50 cycles.



Figure S10. Equivalent circuit model for the simulation of the Nyquist plots.

 R_s is the electrolyte resistance, R_f is surface film resistance, R_{ct} is charge transfer resistance, W_o is the Warburg impedance related to the diffusion of Li-ion into electrodes, *CPE1* and *CPE2* represent the constant phase elements.



Figure S11. SEM images of N-DCSNs electrodes after 60 cycles (a, d) and 270 cycles (b, e) at 0.2 A g^{-1} . (e, f) EDS images with the corresponding element distribution images of N-DCSNs.

Samples	$R_{s}(\Omega)$	$R_{ m f}(\Omega)$	$R_{ct}(\Omega)$
N-DCSNs	1.928	48.98	36.68
CN/SnS	2.905	51.2	57.19
SnS	2.104	72.06	61.77

 Table S2. Impedance parameters of N-DCSNs, CN/SnS and SnS electrodes before

 cycling obtained by the equivalent circuit model.

 Slope
 N-DCSNs
 CN/SnS
 SnS

 Before cycle
 105.57
 116.08
 75.99

 After 50 cycles
 187.90
 372.11
 313.28

Table S3. The linear relevant fitting result of the N-DCSNs, CN/SnS and bare SnS electrodes, respectively.

Depiction S1. Electric conductivity & Li-ion diffusion coefficient at open circuit state:¹⁴⁻¹⁷

$$D = R^{2}T^{2}/2A^{2}n^{4}F^{4}C^{2}\sigma^{2}\cdots(1) \quad Z_{Re} = R_{e} + R_{ct} + \sigma\omega^{-1/2\cdots(2)}$$

where *D* is the diffusion coefficient (cm² s⁻¹), *R* is the gas constant (8.31 J mol⁻¹ K⁻¹), *T* is the absolute temperature (298 K), *A* is the surface area of the anode (0.36 cm²), *n* is the number of electrons transferred in the half-reaction for the redox couple, *F* is the Faraday constant (96485 C mol⁻¹), *C* is the is the molar concentration of Li-ion in N-DCSNs, R_e is the resistance between the electrolyte and electrode, R_{cl} is the charge transfer resistance, ω is frequency, and σ is the Warburg factor which corresponds to the slope of the curve shown in **Figure 5c, d**. **Depiction S2.** The chemical diffusion coefficients of the electrodes in LIBs are determined by formulas 3 and 4:¹⁸

$$D = \frac{4}{\pi} \left(\frac{I_0 V_M}{SF z_i} \right)^2 \left[\frac{dE}{d\delta} / \frac{dE}{d\sqrt{\tau}} \right]^2 \left(\tau \ll \frac{L^2}{D} \right) (3) \quad D = \frac{4}{\pi \tau} \left(\frac{m_B V_M}{SM_B} \right)^2 \left(\frac{\Delta E_s}{\Delta E_\tau} \right)^2 \left(\tau \ll \frac{L^2}{D} \right) (4)$$

The parameters required for D value can be obtained from the known conditions according to formula 4, τ is the excitation current time (s), S is the electrode area (cm²), ΔEs is the steady-state voltage change (V), $\Delta E\tau$ is the transient voltage change (V), V_M is the molar volume of electrode material (cm³ mol⁻¹), m_B is the mass of the electrode material (g), and M_B is the molar mass of the electrode material (g mol⁻¹).

References:

1. Yao, L.; Nie, M.; Zhu, C.; Cai, R.; Xia, W.; Sun, L.; Xu, F., Revealing a conversion-alloying reaction mechanism behind high capacity and rate capability of SnS/N-doped graphene anode by in situ TEM. *Electrochim. Acta.* **2019**, *297*, 46-54.

2. Guo, W.; Ding, K.; Mei, S.; Li, X.; Feng, X.; Guo, S.; Fu, J.; Zhang, X.; Gao, B.; Huo, K.; Chu, P. K., Hollow spheres consisting of SnS nanosheets conformally coated with S-doped carbon for advanced lithium/sodium-ion battery anodes. *ChemElectroChem* **2020**, *7* (4), 914-921.

3. Jin, A.; Kang, N.; Um, J. H.; Ko, I. H.; Kim, M. S.; Kim, K.; Kim, S. H.; Yu, S. H.; Sung, Y. E., Sn(salen)-derived SnS nanoparticles embedded in N-doped carbon for high performance lithium-ion battery anodes. *Chem. Commun.* **2020**, *56* (58), 8095-8098.

4. Xia, J.; Liu, L.; Jamil, S.; Xie, J.; Yan, H.; Yuan, Y.; Zhang, Y.; Nie, S.; Pan, J.; Wang, X.; Cao, G., Free-standing SnS/C nanofiber anodes for ultralong cycle-life lithium-ion batteries and sodium-ion batteries. *Energy Stor. Mater.* **2019**, *17*, 1-11.

5. Zhu, C.; Kopold, P.; Li, W.; van Aken, P. A.; Maier, J.; Yu, Y., A general strategy to fabricate carbon-coated 3D porous interconnected metal sulfides: case study of SnS/C nanocomposite for high-performance lithium and sodium ion batteries. *Adv. Sci.* **2015**, *2* (12), 1500200.

 Manukumar, K. N.; Nagaraju, G.; Kishore, B.; Madhu, C.; Munichandraiah, N., Ionic liquid-assisted hydrothermal synthesis of SnS nanoparticles: Electrode materials for lithium batteries, photoluminescence and photocatalytic activities. *J. Energy Chem.* 2018, 27 (3), 806-812.

7. Vaughn, D. D.; Hentz, O. D.; Chen, S.; Wang, D.; Schaak, R. E., Formation of SnS nanoflowers for lithium ion batteries. *Chem. Commun.* **2012**, *48* (45), 5608-5610.

8. Zheng, J.; Luo, Y.; Xie, D.; Xiong, X.; Lin, Z.; Wang, G.; Yang, C.; Liu, M., Onepot synthesis of SnS/C nanocomposites on carbon paper as a high-performance freestanding anode for lithium ion batteries. *J. Alloys Compd.* **2019**, *779*, 67-73.

9. Li, S.; Zheng, J.; Zuo, S.; Wu, Z.; Yan, P.; Pan, F., 2D hybrid anode based on SnS

nanosheet bonded with graphene to enhance electrochemical performance for lithiumion batteries. *RSC Adv.* **2015**, *5* (58), 46941-46946.

10. Zhang, Y.; Wang, P.; Yin, Y.; Zhang, X.; Fan, L.; Zhang, N.; Sun, K., Heterostructured SnS-ZnS@C hollow nanoboxes embedded in graphene for high performance lithium and sodium ion batteries. *Chem. Eng. J.* **2019**, *356*, 1042-1051.

11. Cho, E.; Song, K.; Park, M. H.; Nam, K. W.; Kang, Y. M., SnS 3D flowers with superb kinetic properties for anodic use in next-generation sodium rechargeable batteries. *Small* **2016**, *12* (18), 2510-2517.

Choi, J.; Kim, N. R.; Lim, K.; Ku, K.; Yoon, H. J.; Kang, J. G.; Kang, K.; Braun,
 P. V.; Jin, H. J.; Yun, Y. S., Tin Sulfide-Based Nanohybrid for High-Performance
 Anode of Sodium-Ion Batteries. *Small* 2017, *13* (30), 1700767.

13. Lian, Q.; Zhou, G.; Zeng, X.; Wu, C.; Wei, Y.; Cui, C.; Wei, W.; Chen, L.; Li, C., Carbon coated SnS/SnO₂ heterostructures wrapping on CNFs as an improvedperformance anode for Li-ion batteries: lithiation-induced structural optimization upon cycling. *ACS Appl. Mater. Interfaces* **2016**, *8* (44), 30256-30263.

14. Yin, H.; Shen, W.; Qu, H.-Q.; Li, C.; Zhu, M.-Q., Boosted charge transfer and Naion diffusion in cooling-fins-like Sb₂Te-Te nano-heterostructure for long cycle life and high rate capability anode. *Nano Energy* **2020**, *70*, 104468.

15. Jae-Min; Jeong, Ultrathin sandwich-like $MoS_2@N$ -doped carbon nanosheets for anodes of lithium ion batteries. *Nanoscale* **2015**, *1* (7), 324-329.

16. Ai, W.; Huang, Z.; Wu, L.; Du, Z.; Zou, C.; He, Z.; Shahbazian-Yassar, R.; Huang,
W.; Yu, T., High-rate, long cycle-life Li-ion battery anodes enabled by ultrasmall tinbased nanoparticles encapsulation. *Energy Stor. Mater.* 2018, *14*, 169-178.

17. Yin, H.; Yu, X.-X.; Zhao, H.; Li, C.; Zhu, M.-Q., Towards high-performance cathode materials for lithium-ion batteries: Al₂O₃-coated LiNi_{0.8}Co_{0.15}Zn_{0.05}O₂. *J. Solid. State. Electr.* **2018**, *22* (8), 2395-2403.

18. Zhu, S.; Yin, H.; Wang, Y.; Hui, K. S.; Wu, X.-L.; Mai, W.; Hong, X.; Chen, F.; Hui, K. N., Heteroatomic interface engineering of MOF-derived metal-embedded P and N-codoped Zn node porous polyhedral carbon with enhanced sodium-ion storage. *ACS Appl. Energy Mater.* **2020**, *3* (9), 8892-8902.