

Supporting Information

Multifunctional Ionic Liquid-Assisted Interfacial Engineering towards ZnS Nanodots with

Ultrastable High-Rate Lithium Storage Performance

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Characterizations

TEM analysis of ZnS@SNG and ZnS-0 was performed on the JEM-2100 high resolution transmission electron microscope of JEOL. SEM analysis of ZnS@SNG and ZnS-0 was performed on the JSM-7500F cold field emission scanning electron microscope of JEOL.

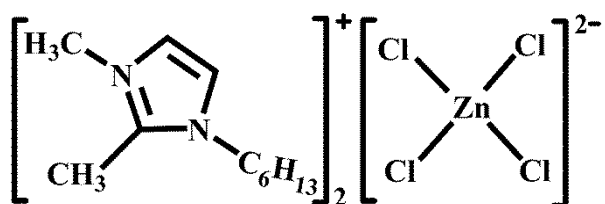


Fig. S1. The structure of [HMMIm]₂[ZnCl₄].

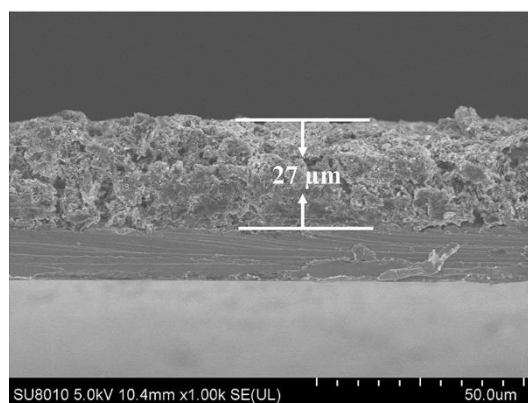


Fig. S2. SEM image of ZnS-NDs@SNG electrode.

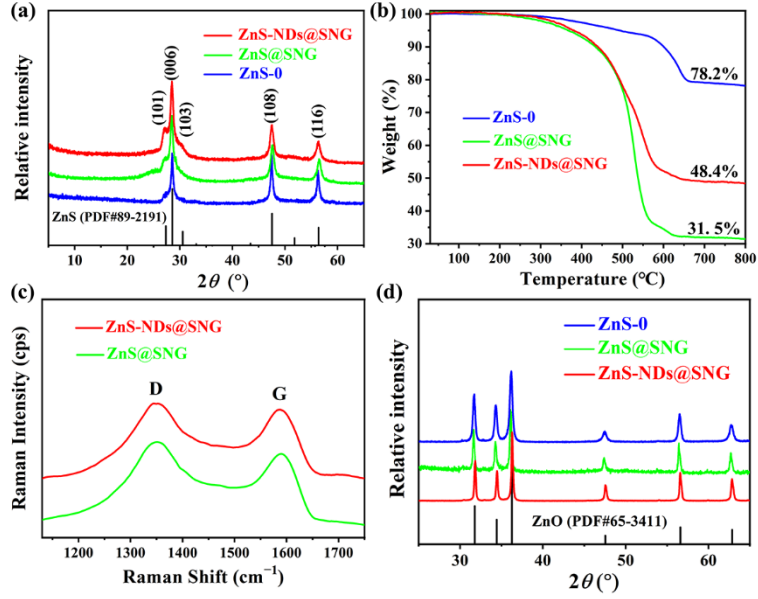


Fig. S3. (a) XRD patterns and (b) TG curves of ZnS-NDs@SNG, ZnS@SNG and ZnS-0; (c) Raman spectra of ZnS-NDs@SNG and ZnS@SNG nanocomposites; (d) XRD patterns of ZnS-0, ZnS@SNG and ZnS-NDs@SNG composites after heating at 800°C in the air.

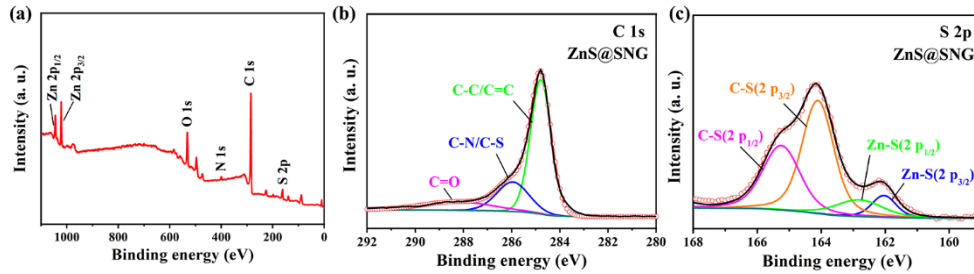


Fig. S4. XPS spectra of ZnS-NDs@SNG: (a) survey; XPS spectra of ZnS@SNG: (b) C 1s, (c) S 2p.

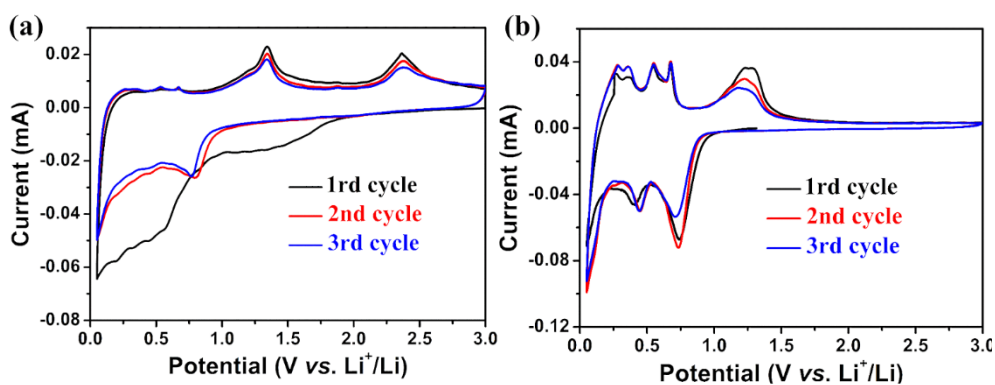


Fig. S5. CV curves of ZnS@SNG (a) and ZnS-0 (b) at a scan rate of 0.1 mV s^{-1} between 0.05 and 3.0 V.

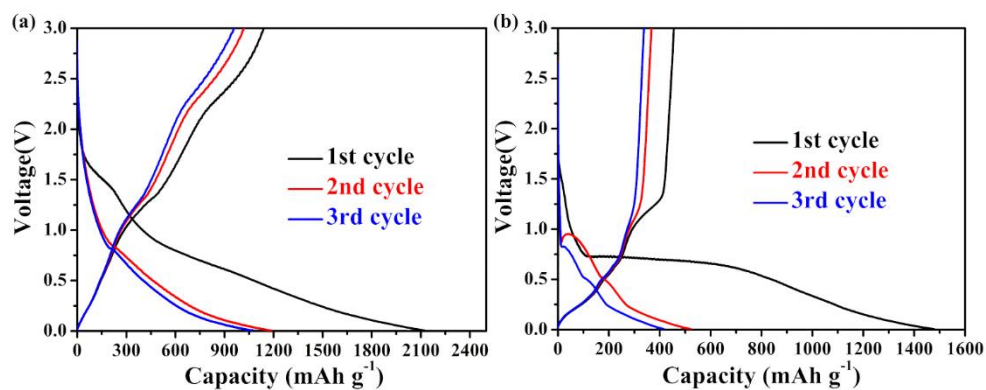


Fig. S6. Charge/discharge curves of ZnS@SNG (a) and ZnS-0 (b) at the current density of 60 mA g^{-1} .

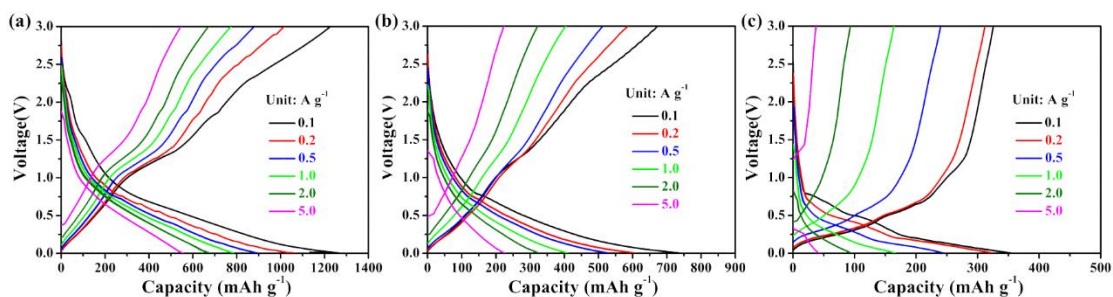


Fig. S7. Charge-discharge curves of ZnS-NDs@SNG (a), ZnS@SNG (b), and ZnS-0 (c) at various current densities.

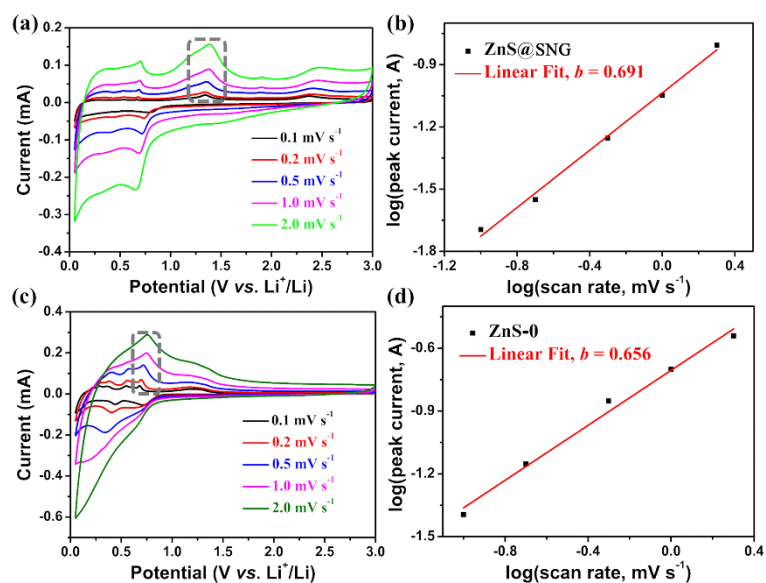


Fig. S8. Kinetic analysis of different anodes: CV curves of ZnS@SNG (a) and ZnS-0 (c) with scan rates from 0.1 to 2 mV s^{-1} . Log(v) versus Log(i) plots used to calculate the b values at the anodic peak for ZnS@SNG (b) and ZnS-0 (d).

Table S1. Details of the samples.

electrode	C (wt %)	S (wt %)	N (wt %)
ZnS-NDs@SNG	35.58	19.27	1.58
ZnS@SNG	49.90	15.47	1.13
ZnS-0	2.44	30.76	0.31

Table S2. Comparison of different ZnS-based materials for LIBs.

electrode	current density (mA g^{-1})	cycle number	discharge capacity (mAh g^{-1})	Ref.
ZnS-NDs@SNG	10000	5000	648.1	The work
ZnS/C	0.1 C	50	948.9	[1]
CC-ZnS/CNT	100	200	730	[2]
	2000	4000	333	
ZnS/NC	200	100	757	[3]
	2000	1000	~500	

ZnS-CNTs	5000	1200	451.3	[4]
ZnS-CFC	100	300	658	[5]
ZnS@(2D)Gra	100	300	444	[6]
ZnS/C	100	100	483	[7]
ZnS/C-800	300	80	624	[8]
ZnS-C/G	1000	200	452	[9]
ZnS/C	1000	1200	659	[10]
ZnS@SNG	1000	500	480.9	[11]
ZnS/CC	1C	100	487	[12]
ZnS/NC	200	150	521.8	[13]
ZnS/PCNFs	100	150	718	[14]
ZnS-QDs@mNC	840	300	506	[15]
GLC@ZnS	1000	200	890	[16]
ZnS@HPC	1000	200	~400	[17]
nano-ZnS-C	500	600	506	[18]
Nanotube Structured ZnS	0.2C	100	450	[19]
ZnS/C	500	300	~750	[20]
ZnS NR@HCP-600	600	300	694	[21]

References

- [1] A. Wang, X. Chen, G. Yu, Y. Wang, Carbon-coated ZnS composites for lithium-ion battery anode materials, *Ionics* 27 (2021) 541-550.
- [2] T. Hou, B. Liu, X. Sun, A. Fan, Z. Xu, S. Cai, C. Zheng, G. Yu, A. Tricoli, Covalent coupling-stabilized transition-metal sulfide/carbon nanotube composites for lithium/sodium-ion batteries, *ACS Nano* (2021) 10.1021/acsnano.0c10121.
- [3] P. Wang, A. Yuan, Z. Wang, X. Shen, H. Chen, H. Zhou, Self-templated formation of hierarchically yolk-shell-structured ZnS/NC dodecahedra with superior lithium storage properties, *Nanoscale* 13 (2021) 1988-1996.
- [4] W. L. Zhang, Z. Y. Huang, H. H. Zhou, S. L. Li, C. Q. Wang, H. X. Li, Z. H. Yan, F. Wang, Y. F. Kuang, Facile synthesis of ZnS nanoparticles decorated on defective CNTs with excellent

- performances for lithium-ion batteries anode material, *J. Alloys Compd.* 816 (2020) 152633.
- [5] V. M. Rangaraj, A. A. Edathil, P. Kadirvelayutham, F. Banat, Chicken feathers as an intrinsic source to develop ZnS/carbon composite for Li-ion battery anode material, *Mater. Chem. Phys.* 248 (2020) 122953.
- [6] J. Yoon, I. T. Kim, J. Bae, J. Hur, High-performance ZnS@graphite composites prepared through scalable high-energy ball milling as novel anodes in lithium-ion batteries, *J. Ind. Eng. Chem.* 76 (2019) 258-267.
- [7] Q. Yang, L. Xu, S. Luo, M. Chen, X. Wang, L. Ma, One-pot hydrothermal synthesis of ZnS/C microsphere as an electrode for reversible lithium-storage, *Mater. Lett.* 254 (2019) 386-389.
- [8] H. D. Wu, G. Li, Y. Li, Z. X. Geng, T. Q. Ren, T. F. Cai, Z. X. Yang, Synthesis of ZnS/C composites by metal-organic framework as high-performance lithium-ion batteries, *Cryst. Res. Technol.* 54 (2019) 1800281.
- [9] G. Y. Tian, Z. J. Zhao, A. Sarapulova, C. Das, L. H. Zhu, S. Y. Liu, A. Missiul, E. Welter, J. Maibach, S. Dsoke, Understanding the Li-ion storage mechanism in a carbon composited zinc sulfide electrode, *J. Mater. Chem. A* 7 (2019) 15640-15653.
- [10] Y. Q. Teng, H. Liu, D. D. Liu, H. He, Y. C. Chen, Pitaya-like carbon-coated ZnS/carbon nanospheres with inner three-dimensional nanostructure as high-performance anode for lithium-ion battery, *J. Colloid Interface Sci.* 554 (2019) 220-228.
- [11] H. B. Reb, Z. Y. Wen, G. Z. Wu, S. Chen, S. W. Joo, J. R. Huang, Preparation of zinc sulfide@reduced graphene oxide nanocomposites with enhanced energy storage performance, *J. Phys. Chem. Solids* 134 (2019) 43-51.
- [12] L. Y. Huang, Y. G. Zhang, C. Q. Shang, X. Wang, G. F. Zhou, J. Z. Ou, Y. C. Wang, ZnS nanotubes/carbon cloth as a reversible and high-capacity anode material for lithium-ion batteries, *ChemElectroChem* 6 (2019) 461-466.
- [13] H. Ding, H. C. Huang, X. K. Zhang, L. Xie, J. Q. Fan, T. Jiang, D. Shi, N. Ma, F. C. Tsai, Zinc sulfide decorated on nitrogen-doped carbon derived from metal-organic framework composites for highly reversible lithium-ion battery anode, *ChemElectroChem* 6 (2019) 5617-5626.
- [14] L. Y. Wang, J. G. Ju, N. P. Deng, G. Wang, B. W. Cheng, W. M. Kang, ZnS nanoparticles anchored on porous carbon nanofibers as anode materials for lithium ion batteries,

Electrochem. Commun. 96 (2018) 1-5.

- [15] D. L. Fang, S. M. Chen, X. Wang, Y. Bando, D. Golberg, S. J. Zhang, ZnS quantum dots@multilayered carbon: geological-plate-movement-inspired design for high-energy Li-ion batteries, *J. Mater. Chem. A* 6 (2018) 8358-8365.
- [16] H. W. Du, X. C. Gui, R. L. Yang, H. Zhang, Z. Q. Lin, B. H. Lang, W. J. Chen, H. Zhu, J. Chen, ZnS nanoparticles coated with graphene-like nano-cell as anode materials for high rate capability lithium-ion batteries, *J. Mater. Sci.* 53 (2018) 14619-14628.
- [17] H. Z. Chen, B. Zhang, Y. Cao, X. Wang, Y. Y. Yao, W. J. Yu, J. C. Zheng, J. F. Zhang, H. Tong, ZnS nanoparticles embedded in porous honeycomb-like carbon nanosheets as high performance anode material for lithium ion batteries, *Ceram. Int.* 44 (2018) 13706-13711.
- [18] X. F. Du, H. L. Zhao, Z. J. Zhang, Y. Lu, C. H. Gao, Z. L. Li, Y. Q. Teng, L. N. Zhao, K. Swierczek, Core-shell structured ZnS-C nanoparticles with enhanced electrochemical properties for high-performance lithium-ion battery anodes, *Electrochim. Acta* 225 (2017) 129-136.
- [19] W. Zhang, J. F. Zhang, Y. Zhao, T. Z. Tan, T. Yang, High Electrochemical performance of nanotube structured zns as anode material for lithium-ion batteries, *Materials* 11 (2018) 1537.
- [20] X. Du, H. Zhao, Y. Lu, Z. Zhang, A. Kulka, K. Swierczek, Synthesis of core-shell-like ZnS/C nanocomposite as improved anode material for lithium ion batteries, *Electrochim. Acta* 228 (2017) 100-106.
- [21] Z. L. Chen, R. B. Wu, H. Wang, Y. K. Jiang, L. Jin, Y. H. Guo, Y. Song, F. Fang, D. L. Sun, Construction of hybrid hollow architectures by in-situ rooting ultrafine ZnS nanorods within porous carbon polyhedra for enhanced lithium storage properties, *Chem. Eng. J.* 326 (2017) 680-690.