

Supporting Information for

High-Quality Epitaxial N Doped Graphene on SiC with Tunable Interfacial Interactions via Electron/Ion Bridges for Stable Lithium-Ion Storage

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Supplementary Figures and Tables

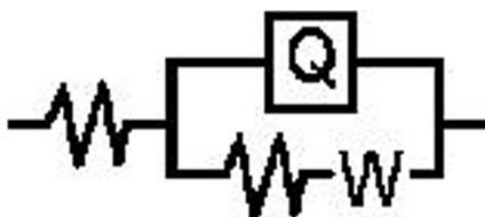


Fig. S1 Equivalent circuit model of the EIS spectrum

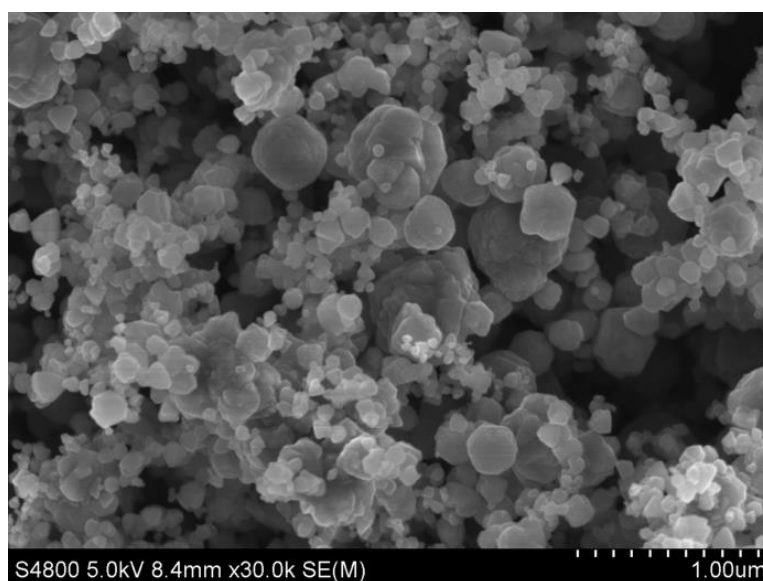


Fig. S2 Morphology change of NG@SiC particles after 1000 cycles

at 10.0 A g^{-1} , the SEM image of the NG@SiC particles does not show particle pulverization or crack can be observed after high current density reaction, revealing the outstanding structural integrity after long-term lithiation and delithiation reactions.

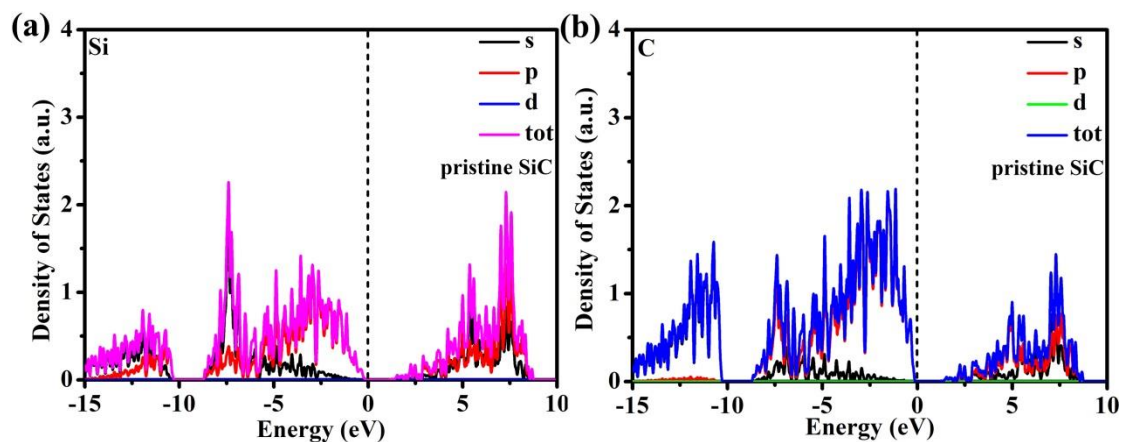


Fig. S3 Partial density of states (PDOS) of the Si and C in pristine SiC. The calculated TDOS result shows that the pristine SiC is direct band gap semiconductor with discrete electronic state, and the calculated PDOS of Si and C in pristine Si also confirm this result.

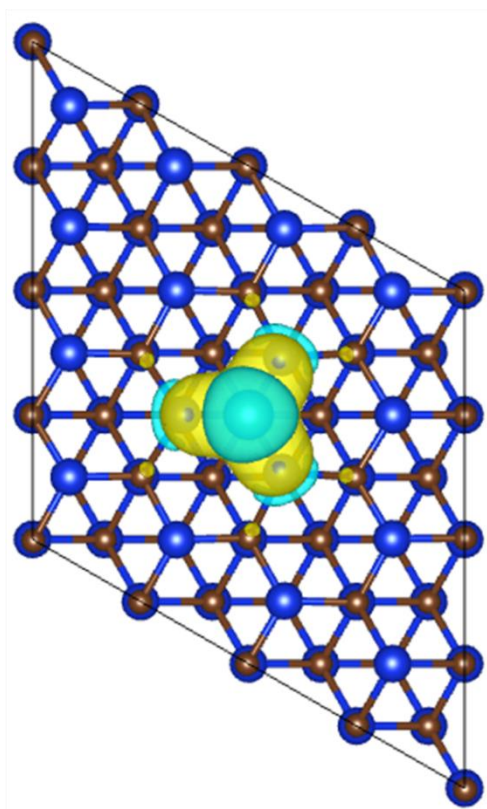


Fig. S4 Charge density distribution of pristine SiC after lithium-ion adsorption

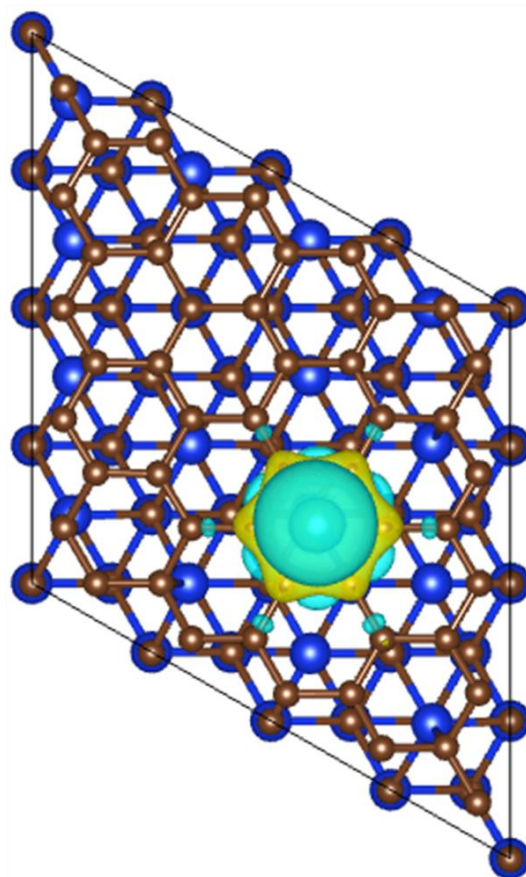


Fig. S5 Charge density distribution of NG@ SiC after lithium-ion adsorption. Charge density distribution analysis after lithium-ion adsorption demonstrates the charge transfer from lithium ion to NG@SiC, and the charge accumulation intensity is larger than that of pristine SiC.

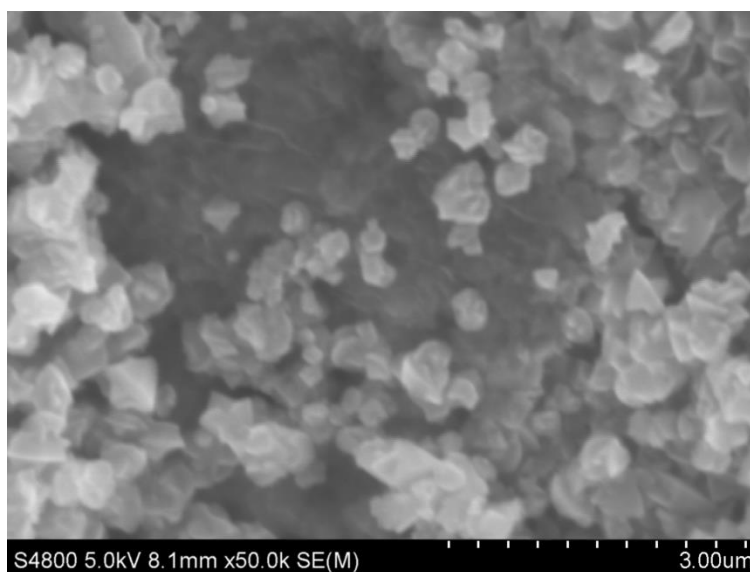


Fig. S6 SEM image of the commercialized LiFePO₄/C cathode. As shown in Figure S6, it can be seen that the commercial LiFePO₄/C is composed of irregular particles with the particle size of about 1.0 μm.

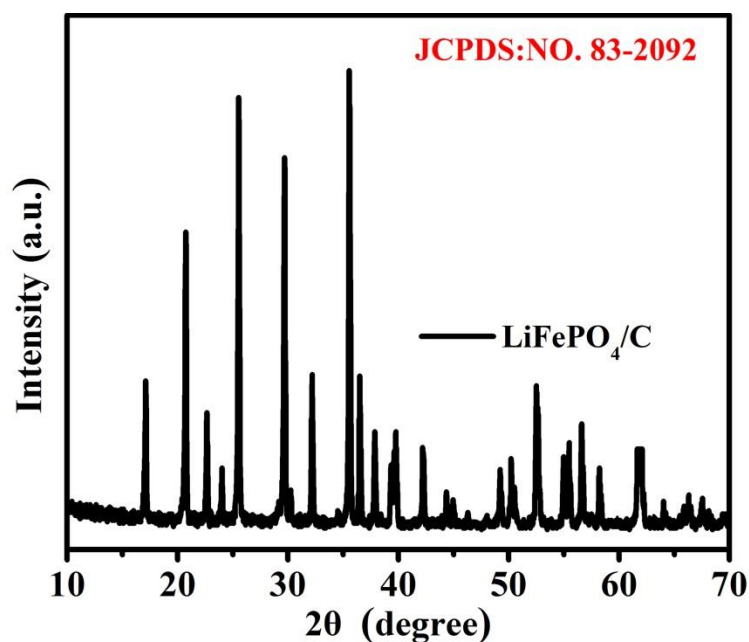


Fig. S7 XRD pattern of the commercialized LiFePO_4/C cathode. The strong diffraction peaks show the good crystallinity of the commercial LiFePO_4/C particles (ICDD PDF no. 83-2092)

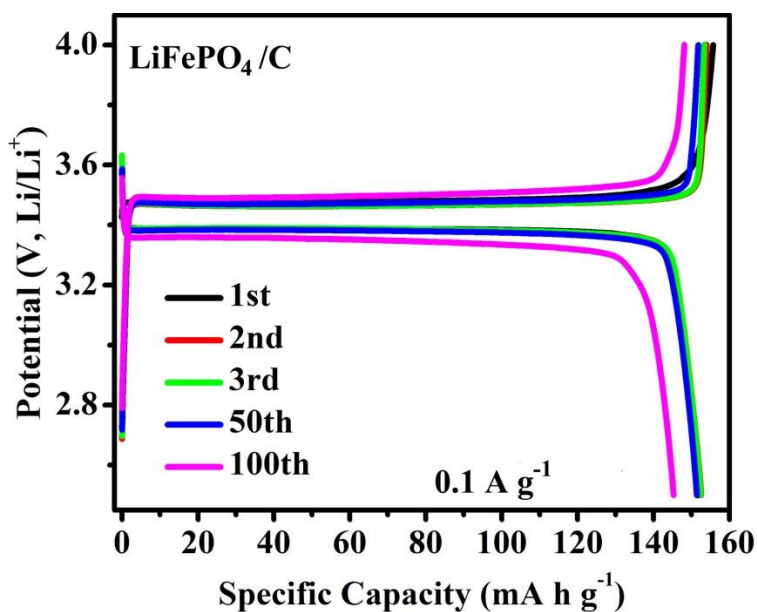


Fig. S8 Charge and discharge curves of the commercialized LiFePO_4/C cathode at 0.1 A g^{-1} of the initial three cycles.

The initial discharge and charge capacities of the commercialized LiFePO_4/C cathode are 152.4 and $151.8 \text{ mA h g}^{-1}$, respectively. The distinct charge-discharge platform indicates stable output voltage at about 3.4 V during the electrochemical reaction.

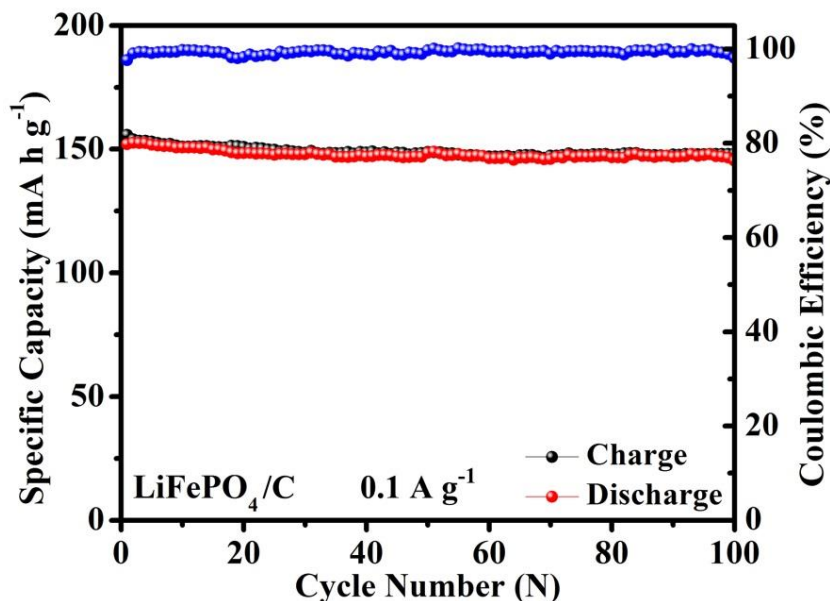


Fig. S9 Cycling stability at 0.1 A g^{-1} of the commercialized LiFePO_4/C cathode in the Li-ion half cells for 100 cycles. After 100 cycles, the reversible capacity is about $148.6 \text{ mA h g}^{-1}$ after 100 cycles, and the Coulombic efficiency can stabilize at $\sim 100\%$, showing good structural stability of the commercial LiFePO_4/C particles.

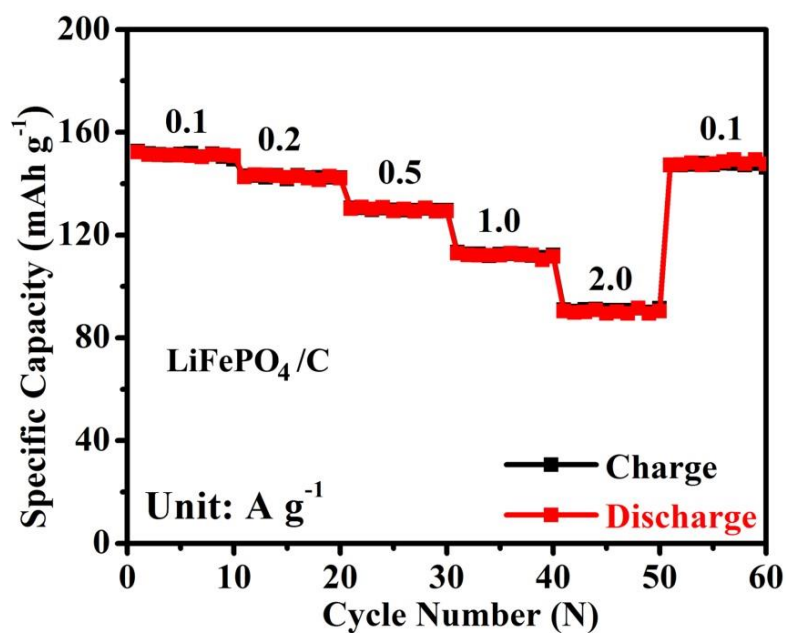


Fig. S10 Rate performance of the commercialized LiFePO_4/C at various current densities. The commercialized LiFePO_4/C cathode can deliver the reversible capacities of 151.8 , 141.3 , 129.7 , 112.9 , and 89.4 mA h g^{-1} at 0.1 , 0.2 , 0.5 , 1.0 , 2.0 and A g^{-1} , respectively, revealing the good structural tolerance and lithium-ion storage reversibility of the commercialized LiFePO_4/C cathode.

Table S1 Comparisons of the synthetic method, morphology, cycle number, current density, and capacity between NG@SiC anode and other previously reported Si-based LIBs anodes

Material	Method	Morphology	Current density (A g ⁻¹)	Cycle number	Capacity (mAh g ⁻¹)
NG@SiC(this work)	pyrolysis reaction	particles	0.110.0	2001000	1197.5447.8
SiC@HGSs ^{ref.1}	surface graphitization	nanoshells	0.63.0	6001000	1345742
SiO ₂ ^{ref.2}	mechanical milling	particles	0.5	200	800
SiO _x /C ^{ref.3}	CVD	particles	0.5	500	972
SiC-Sb-C ^{ref.4}	mechanical milling	microspheres	2.0	120	440
SiC ^{ref.5}	ICP-CVD	thin films	0.3 C	100	376
Si/SiO _x ^{ref.6}	direct heating	thin films	0.1	100	1186
SiO _x /C ^{ref.7}	sand milling	particles	0.325	500	645
Si-O-C ^{ref.8}	thermolysis	nanocomposite	1.6	970	200
SiOC/Sn ^{ref.9}	pyrolysis	nanocomposite	0.074	20	562
SiC/C ^{ref.10}	pyrolysis	nanofibers	0.1	250	254.5
SiN _{0.92} ^{ref.11}	pulsed laser deposition	thick films	0.02 C	100	700
SiN _x @Si ^{ref.12}	vacuum CVD	nanocomposite	0.5	200	1400
C@SiO _x ^{ref.13}	graphitization	nanospheres	5.0	500	350
NC@SiO _x ^{ref.14}	directly calcining	nanosheets	5.0	1000	427.6
C@SiO _x ^{ref.15}	mixed and heated	hollow spheres	0.51.0	300300	823682
Si/SiO ₂ @C ^{ref.16}	ball milling	nanoclusters	0.51.0	200200	534.3512.7
SiO _x /G ^{ref.17}	calcination	nanocomposite	1.0	1000	780
SiO ₂ /TiO ₂ /C ^{ref.18}	annealing	nanocomposite	2.0	400	410
ZnO-Si@C ^{ref.19}	electrospinning	nanofibers	0.81.8	10001000	1050920

Supplementary References

- [S1] H. Li, H. Yu, X. Zhang, G. Guo, J. Hu et al., Bowl-like 3C-SiC nanoshells encapsulated in hollow graphitic carbon spheres for high-rate lithium-ion batteries. *Chem. Mater.* **28**, 1179 (2016). <https://doi.org/10.1021/acs.chemmater.5b04750>
- [S2] W.-S. Chang, C.-M. Park, J.-H. Kim, Y.-U. Kim, G. Jeong et al., Quartz (SiO₂): a new energy storage anode material for Li-ion batteries. *Energy Environ. Sci.* **5**, 6895 (2012). <https://doi.org/10.1039/c2ee00003b>
- [S3] Z. Liu, Y. Zhao, R. He, W. Luo, J. Meng et al., Yolk@Shell SiO_x/C microspheres with semi-graphitic carbon coating on the exterior and interior surfaces for durable lithium storage. *Energy Storage Mater.* **19**, 299 (2019). <https://doi.org/10.1016/j.ensm.2018.10.011>
- [S4] Z. Chen, Y. Cao, J. Qian, X. Ai, H. Yang, Antimony-coated SiC nanoparticles as

- stable and high-capacity anode materials for Li-ion batteries. *J. Phys. Chem. C* **114**, 15196 (2010). <https://doi.org/10.1021/jp104099r>
- [S5] X.D. Huang, F. Zhang, X.F. Gan, Q.A. Huang, J.Z. Yang et al., Electrochemical characteristics of amorphous silicon carbide film as a lithium-ion battery anode. *RSC Adv.* **8**, 5189 (2018). <https://doi.org/10.1039/C7RA12463E>
- [S6] W. Yang, H. Liu, Z. Ren, N. Jian, M. Gao et al., A Novel multielement, multiphase, and B-containing SiO_x composite as a stable anode material for Li-Ion batteries. *Adv. Mater. Interfaces* **6**, 1801631 (2019). <https://doi.org/10.1002/admi.201801631>
- [S7] Q. Xu, J.-K. Sun, Y.-X. Yin, Y.-G. Guo, Facile synthesis of blocky SiO_x/C with graphite-like structure for high-performance lithium-ion battery anodes. *Adv. Funct. Mater.* **28**, 1705235 (2018). <https://doi.org/10.1002/adfm.201705235>
- [S8] L. David, R. Bhandavat, U. Barrera, G. Singh, Silicon oxycarbide glass-graphene composite paper electrode for long-cycle lithium-ion batteries. *Nat. Commun.* **7**, 10998 (2016). <https://doi.org/10.1038/ncomms10998>
- [S9] J. Kaspar, C. Terzioglu, E. Ionescu, M. Graczyk-Zajac, S. Hapis, H.-J. Kleebe, R. Riedel, Stable SiOC/Sn nanocomposite anodes for lithium-ion batteries with outstanding cycling stability. *Adv. Funct. Mater.* **24**, 4097 (2014). <https://doi.org/10.1002/adfm.201303828>
- [S10] X. Sun, C. Shao, F. Zhang, Y. Li, Q.-H. Wu et al., SiC nanofibers as long-life lithium-ion battery anode materials. *Front. Chem.* **6**, 166 (2018). <https://doi.org/10.3389/fchem.2018.00166>
- [S11] N. Suzuki, R.B. Cervera, T. Ohnishi, K. Takada, Silicon nitride thin film electrode for lithium-ion batteries. *J. Power Sources* **231**, 186 (2013). <https://doi.org/10.1016/j.jpowsour.2012.12.097>
- [S12] R.C. de Guzman, J. Yang, M. Ming-Cheng Cheng, S.O. Salley, K.Y.S. Ng, High capacity silicon nitride-based composite anodes for lithium ion batteries. *J. Mater. Chem. A* **2**, 14577 (2014). <https://doi.org/10.1039/C4TA02596B>
- [S13] G. Zhu, F. Zhang, X. Li, W. Luo, L. Li et al., Engineering the distribution of carbon in silicon oxide nanospheres at the atomic level for highly stable anodes. *Angew. Chem. Int. Ed.* **58**, 6669 (2019). <https://doi.org/10.1002/anie.201902083>
- [S14] X. Guo, Y.-Z. Zhang, F. Zhang, Q. Li, D.H. Anjum et al., A novel strategy for the synthesis of highly stable ternary SiO_x composites for Li-ion-battery anodes. *J. Mater. Chem. A* **7**, 15969 (2019). <https://doi.org/10.1039/C9TA04062E>
- [S15] T. Xu, Q. Wang, J. Zhang, X. Xie, B. Xia, Green Synthesis of Dual Carbon Conductive Network-Encapsulated Hollow SiO_x Spheres for Superior Lithium-Ion Batteries. *ACS Appl. Mater. Interfaces* **11**, 19959 (2019). <https://doi.org/10.1021/acsami.9b03070>
- [S16] M. Cui, L. Wang, X. Guo, E. Wang, Y. Yang et al., Designing of hierarchical mesoporous/macroporous silicon-based composite anode material for low-cost high-performance lithium-ion batteries. *J. Mater. Chem. A* **7**, 3874 (2019).

<https://doi.org/10.1039/C8TA11684A>

- [S17] Q. Xu, J.-K. Sun, Z.-L. Yu, Y.-X. Yin, S. Xin et al., SiO_x Encapsulated in graphene bubble film: an ultrastable li-ion battery anode. *Adv. Mater.* **30**, 1707430 (2018). <https://doi.org/10.1002/adma.201707430>
- [S18] L. Zhang, X. Gu, C. Yan, S. Zhang, L. Li et al., Reactive B/Ti nanomultilayers with superior performance in plasma generation. *ACS Appl. Mater. Interfaces* **10**, 44463 (2018). <https://doi.org/10.1021/acsami.8b08120>
- [S19] J. Li, Z. Li, W. Huang, L. Chen, F. Lv et al., A facile strategy to construct silver-modified, ZnO-incorporated and carbon-coated silicon/porous-carbon nanofibers with enhanced lithium storage. *Small* **15**, 1900436 (2019). <https://doi.org/10.1002/sml.201900436>