Biomineralization-Inspired: Rapid Preparation of Silicon-Based Composite as High-Performance Lithium-Ion Battery Anode

Runsheng Gao ^{a,b}, Jie Tang ^{a,b,*}, Shuai Tang ^a, Kun Zhang ^a, Kiyoshi Ozawa ^a,

Lu-Chang Qin ^c

^a National Institute for Materials Science, 1-2-1 Sengen, Tsukuba, Ibaraki 305-0047, Japan

^b University of Tsukuba, 1-1-1 Tennodai, Tsukuba, Ibaraki 305-0006, Japan

^c Department of Physics and Astronomy, University of North Carolina at Chapel Hill, Chapel Hill, NC 27599-3255, USA

Corresponding author: Jie Tang, tang.jie@nims.go.jp



Figure S1. Optical images of (a) preparation procedure of gel-like composite and (b) Examples of editable shapes of gel-like composite.



Figure S2. HRTEM images of n-Si@C composite with a carbon-coated structure.



Figure S3. XRD patterns of GO, PAA, and n-Si@C composite before treatment.



Figure S4. Cyclic voltammetry profiles with cutoff voltage of 0.01-1.5 V vs Li/Li⁺ at scan rate of 0.1 mV s⁻¹.



Figure S5. Lithiation-delithiation profiles of Si/C electrode under various current densities.



Figure S6. Rate capability of pure Si electrode.



Figure S7. Nyquist plots of n-Si@C and Si/C electrode before and after activation.



Figure S8. Charge-discharge curves of (a) n-Si@C and (b) Si/C electrode at 2nd, 20th, 50th, 100th, and 150th cycle.



Figure S9. SEM images of n-Si@C composite electrode. Surface images of (a, b) before and (e, f) after 50 cycles. Cross-sectional images of (c) before and (g) after 50 cycles.



Figure S10. TEM images of Si nanoparticle before (a) and after (b) 50 cycles.

Elastrada	Voltage(V	Current density	Cycle	Capacity retention	Def
Electrode)	$(mA g^{-1})$	number	$(mAh g^{-1})$	Kel.
Spherical Si/C	0.01-3.0	200	400	1391	1
3D hierarchical Si/C	0.005-3.0	200	300	710	2
Si@CNT/C	0.01-2.0	200	300	2056	3
Mesocarbon					
microbeads	0.001-1.5	200	100	632	4
@Si@C					
Plasma nano-Si@C	0.01-1.5	100	200	966	5
Si/C composite	0.01-3.0	360	200	1545	6
3D network	0.01.2.0	200	100	1260	7
Si@SiOx/C	0.01-2.0	200	100	1362	,
Hierarchical					
Graphene-	0.005-1.5	372	300	445	8
Scaffolded Si/C					
Metallic	0.01.1.2	500	100	1205	9
frameworks Si	0.01-1.2	500	100	1203	-
MXene-Si-CNT	0.01-1.5	500	200	1000	10
Si@C@ZIF-67-	0.01 - 1.5	1000	300	852	11
800N	0.01-1.3	1000	500	832	
SiFeCNT	0.005-1.5	1000	300	1163	12
SiO _x /C	0.005-2.0	1000	500	454	13
Si/CNT	0.005-1.5	1000	400	715	14
		500	150	1606	This
n-Si@C	0.005-1.5	1000	300	1308	- This - work
		2000	600	867	

Table S1. Comparison of electrochemical performance of Si-based composites for LIBs.

References

- D. Wang, C. Zhou, B. Cao, Y. Xu, D. Zhang, A. Li, J. Zhou, Z. Ma, X. Chen and H. Song, Energy Storage Mater., 2020, 24, 312–318.
- X. Zuo, X. Wang, Y. Xia, S. Yin, Q. Ji, Z. Yang, M. Wang, X. Zheng, B. Qiu and Z. Liu, J. Power Sources, 2019, 412, 93–104.
- 3 H. Wang, J. Fu, C. Wang, J. Wang, A. Yang, C. Li, Q. Sun, Y. Cui and H. Li, *Energy Environ. Sci.*, 2020, 13, 848–858.
- 4 Y. Lin, Y. Chen, Y. Zhang, J. Jiang, Y. He, Y. Lei, N. Du and D. Yang, *Chem. Commun.*, 2018, **54**, 9466–9469.
- 5 H. Chen, X. Hou, F. Chen, S. Wang, B. Wu, Q. Ru, H. Qin and Y. Xia, *Carbon*, 2018, 130, 433–440.
- Z. Hu, L. Zhao, T. Jiang, J. Liu, A. Rashid, P. Sun, G. Wang, C. Yan and L. Zhang, *Adv. Funct. Mater.* 2019, 29, 1906548.
- 7 J. Lee, J. Moon, S. A. Han, J. Kim, V. Malgras, Y.-U. Heo, H. Kim, S.-M. Lee, H. K. Liu and S. X. Dou, *ACS Nano*, 2019, **13**, 9607–9619.
- S. Zhu, J. Zhou, Y. Guan, W. Cai, Y. Zhao, Y. Zhu, L. Zhu, Y. Zhu and Y. Qian, *Small*, 2018, 14, 1802457.
- A. Zhang, Z. Fang, Y. Tang, Y. Zhou, P. Wu and G. Yu, *Nano Lett.*, 2019, 19, 6292–6298.
- S. Liu, X. Zhang, P. Yan, R. Cheng, Y. Tang, M. Cui, B. Wang, L. Zhang, X. Wang and Y. Jiang, *ACS Nano*, 2019, **13**, 8854–8864.
- N. Liu, J. Liu, D. Jia, Y. Huang, J. Luo, X. Mamat, Y. Yu, Y. Dong and G. Hu, *Energy Storage Mater.*, 2019, 18, 165–173.
- C. Jo, A. S. Groombridge, J. De La Verpilliere, J. T. Lee, Y. Son, H.-L. Liang, A. M. Boies and M. De Volder, *ACS Nano*, 2019, 14, 698–707.
- R. Gao, J. Tang, X. Yu, K. Zhang, K. Ozawa and L.-C. Qin, *Electrochimica Acta*, 2020, 360, 136958.
- Z. Yi, N. Lin, Y. Zhao, W. Wang, Y. Qian, Y. Zhu and Y. Qian, *Energy Storage Mater.*, 2019, 17, 93–100.