

Supporting Information

Silicon/graphite composite anode with constrained swelling and stable solid electrolyte interphase enabled by spent graphite

Qi Xu,^a Qianwen Wang,^a Dequan Chen,^a Yanjun Zhong,^a Zhenguo Wu,^{*a} Yang Song,^a Gongke Wang,^b Yuxia Liu,^c Benhe Zhong^a and Xiaodong Guo^a

^a*School of Chemical Engineering, Sichuan University, Chengdu 610065, P. R. China*

^b*School of Materials Science and Engineering, Henan Normal University, XinXiang 453007, P. R. China*

^c*School of Chemistry and Chemical Engineering, Qufu Normal University, Qufu 273165, P. R. China.*

**Corresponding author:*

E-mail address: zhenguowu@scu.edu.cn

Table S1 Crystal plane spacings of (002) peaks in AG and SG.

Samples	$2\theta_{(002)}$ (°)	$d_{(002)}$ (nm)
AG	26.288	0.3387
SG	26.235	0.3394

Table S2 Surface element distributions of AG and SG.

Samples	C (%)	O (%)	Li (%)	F (%)	P (%)
AG	98.05	1.95	/	/	/
SG	94.72	3.33	1.49	0.33	0.14

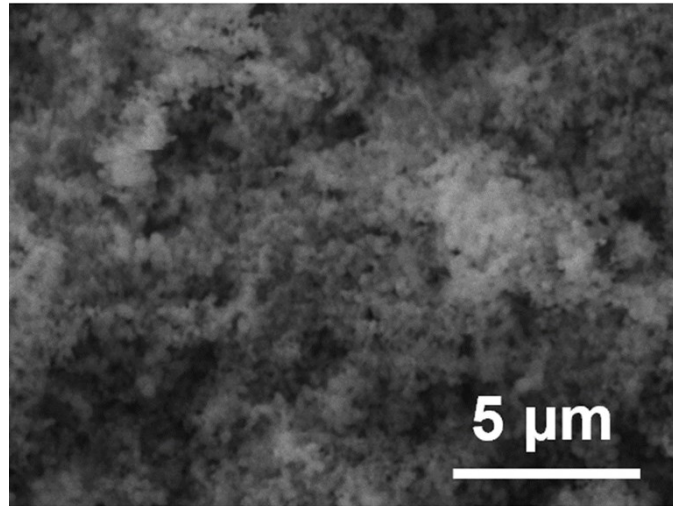


Fig. S1 SEM image of Si nanoparticles.

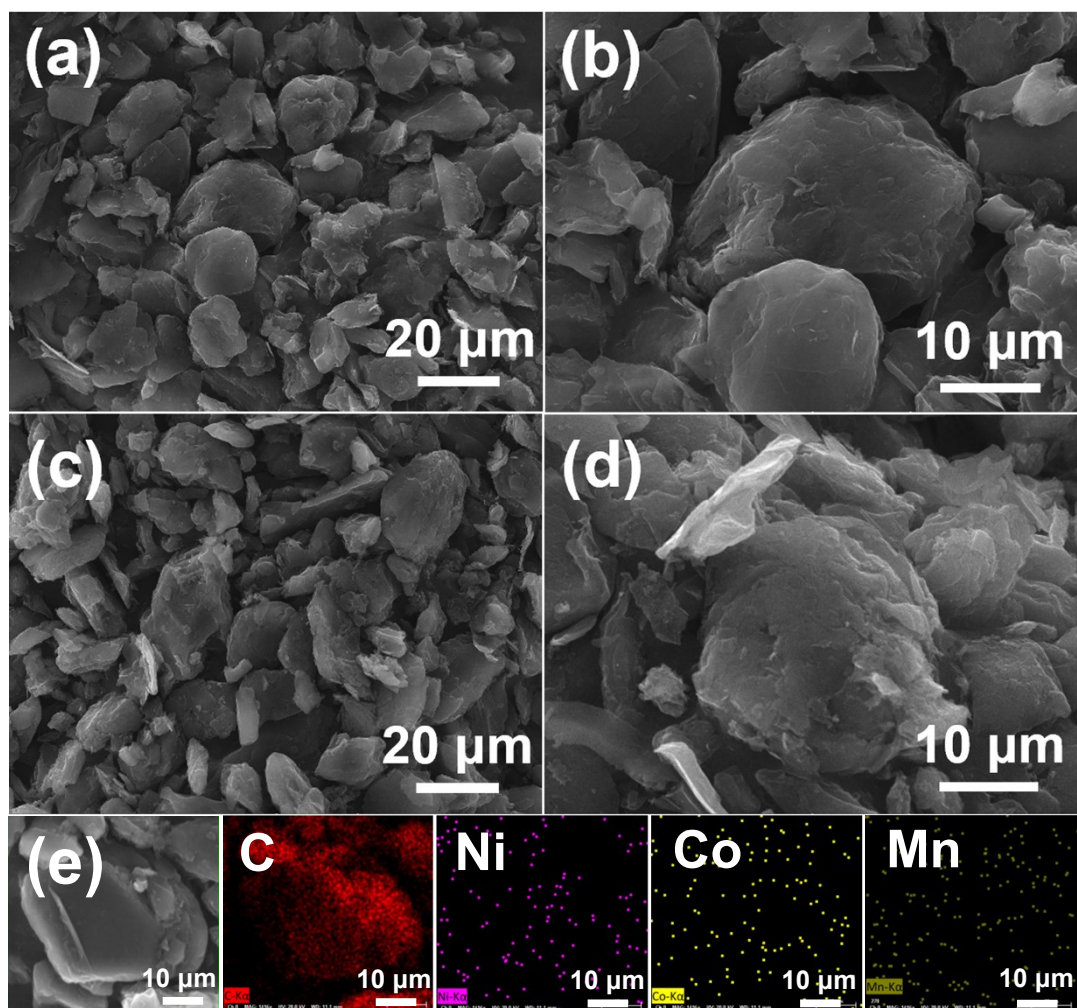


Fig. S2 SEM images of (a-b) AG and (c-d) SG, (e) corresponding EDS mapping images of SG.

Table S3 ICP-OES results of SG.

Elements	Li	Ni	Co	Mn	Al	Cu	Fe
Content (ppm)	266.29	215.05	75.33	365.88	137.20	139.21	120.79

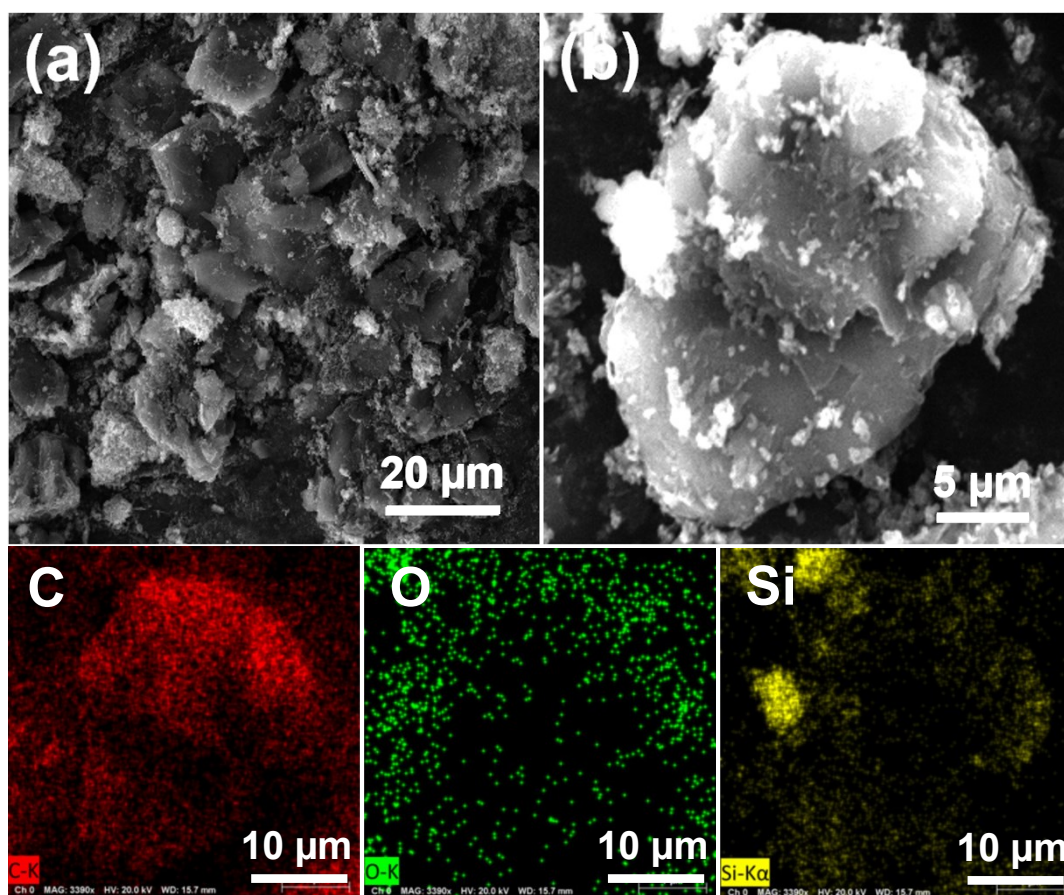


Fig. S3 (a) SEM image and (b) corresponding EDS mapping images of Si/AG composite.

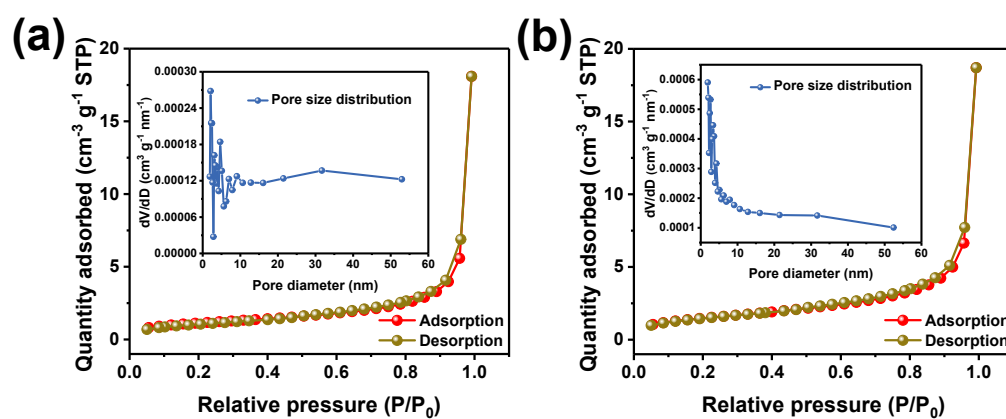


Fig. S4 Nitrogen adsorption-desorption isotherms and corresponding pore size distributions of (a) Si/AG and (b) Si/SG composites.

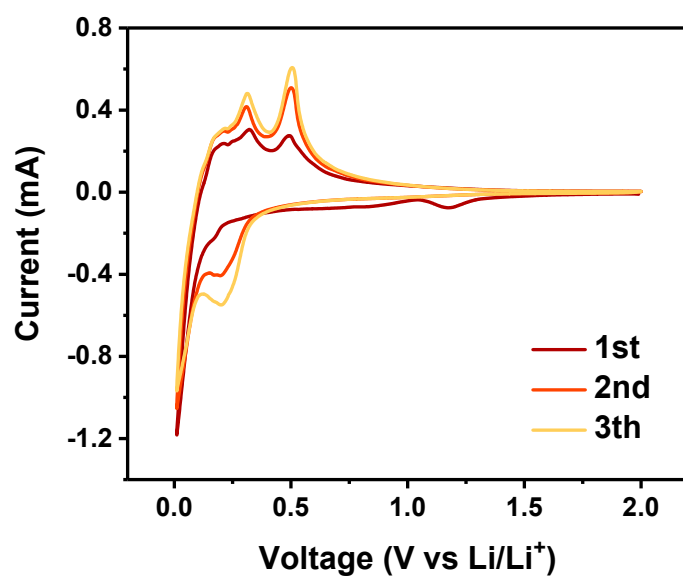


Fig. S5 CV curves of Si/AG composite at a scan rate of 0.1 mV s^{-1} for the first three cycles.

Table S4 Comparison of electrochemical performance with the other Si/graphite anodes in recent reported literatures.

Anodes	Synthesis strategy	Si content (wt%)	Mass loading (g cm ⁻²)	ICE (%)	Cycling stability (mAh g ⁻¹)	Rate capacity (mAh g ⁻¹)
Si _{FS} /G@C ₁	Ball milling	25	/	76.3	730 after 100 cycles at 0.1 A g ⁻¹	~680, 1 A g ⁻¹
SiGC ²	Ball milling + spray drying	12.8	1.1	80.5	610 after 300 cycles at 0.5 A g ⁻¹	458, 2 A g ⁻¹
CSG ³	CVD	16.8	1.0-1.05	80.5	530 after 100 cycles at 0.074 A g ⁻¹	260, 1.86 A g ⁻¹
Si/C-AG ⁴	Ball milling	21.5	1	64	334 after 500 cycles at 1 A g ⁻¹	350, 2 A g ⁻¹
c-Gr + 15% Si ⁵	Calcination + etching	15.4	0.9-1.1	74.2	485 after 400 cycles at 1 A g ⁻¹	770, 2 A g ⁻¹
Nano-Si/G/C-2 ⁶	Ball milling	15.4	0.968	83	368 after 500 cycles at 1 A g ⁻¹	200, 4 A g ⁻¹
GSiWh ⁷	Ball milling	33.3	1.4	74	595 after 200 cycles at 2 A g ⁻¹	500, 0.5 A g ⁻¹
Si/G@C ⁸	Ball milling	24.5	0.95	83.7	~630 after 180 cycles at 0.2 A g ⁻¹	697, 1 A g ⁻¹
MSC-2 ⁹	Magnesiothermic reduction	21	2	81.3	648 after 105 cycles at 0.1 A g ⁻¹	218, 1.5 A g ⁻¹
Si/G/C ¹⁰	Electrospray + ball milling	21.8	/	56.3	400 after 200 cycles at 0.5 A g ⁻¹	538, 2 A g ⁻¹
This work	Ball milling	32.6	1.0-1.3	76.4	562 after 400 cycles at 1 A g ⁻¹	646, 3 A g ⁻¹

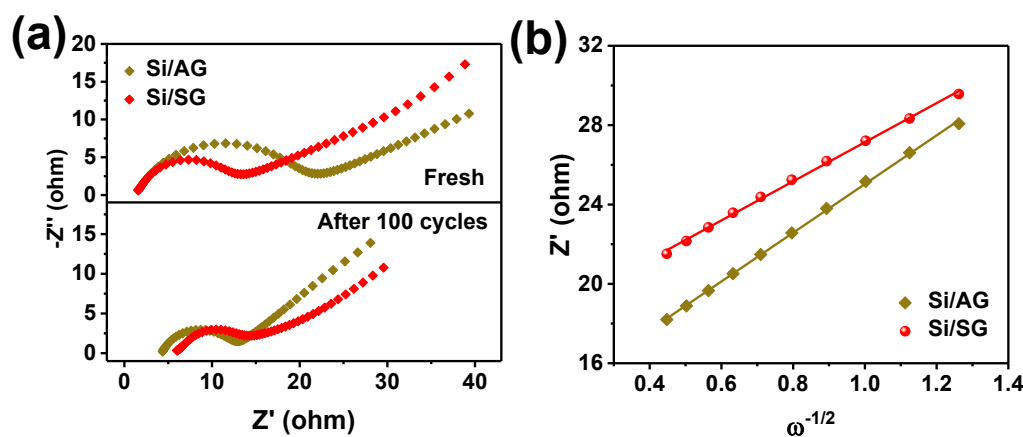


Fig. S6 (a) Nyquist plots of Si/AG and Si/SG composite electrodes before and after 100 cycles and (b) corresponding linear fits (relationship between Z' and $\omega^{-1/2}$) in the low-

frequency region after 100 cycles.

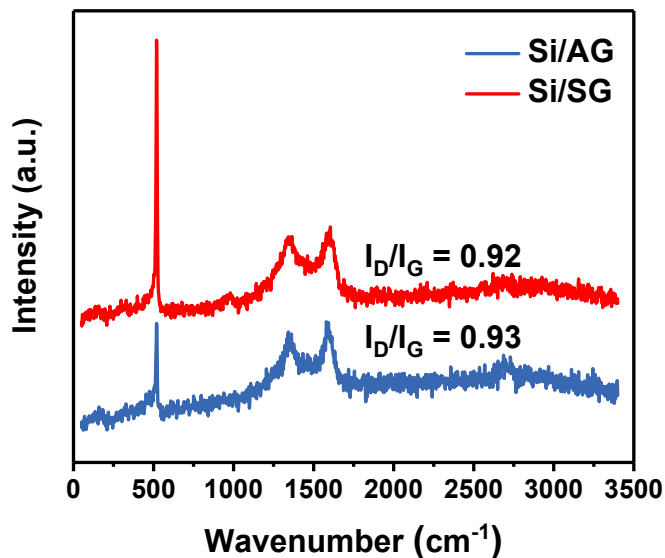


Fig. S7 Raman spectra of cycled Si/AG and Si/SG composite electrodes.

Table S5 Surface element distributions of Si/AG and Si/SG electrodes after 3 cycles.

Samples	C (%)	O (%)	Li (%)	F (%)	Si (%)
Si/AG	47.53	29.61	19.1	2.85	0.89
Si/SG	49.14	30.12	17.87	2.04	0.83

References

- 1 L. Geng, D. Yang, S. Gao, Z. Zhang, F. Sun, Y. Pan, S. Li, X. Li, P.-F. Cao and H. Yang, *Adv. Mater. Interfaces*, 2020, **7**, 1901726.
- 2 D. Sui, Y. Xie, W. Zhao, H. Zhang, Y. Zhou, X. Qin, Y. Ma, Y. Yang and Y. Chen, *J. Power Sources*, 2018, **384**, 328-333.
- 3 B. Liu, P. Huang, Q. Zhang, Q. Huang and Z. Xie, *J Mater Sci*, 2020, **55**, 12165-12176.
- 4 W. Yang, H. Ying, S. Zhang, R. Guo, J. Wang and W.-Q. Han, *Electrochim. Acta*, 2020, **337**, 135687.
- 5 M. Chen, W. Cao, L. Wang, X. Ma and K. Han, *ACS Appl. Energy Mater.*, 2021, **4**,

775-783.

- 6 A. Sun, H. Zhong, X. Zhou, J. Tang, M. Jia, F. Cheng, Q. Wang and J. Yang, *Appl. Surf. Sci.*, 2019, **470**, 454-461.
- 7 M. H. Parekh, V. P. Parikh, P. J. Kim, S. Misra, Z. Qi, H. Wang and V. G. Pol, *Carbon*, 2019, **148**, 36-43.
- 8 T. Mu, Z. Zhang, Q. Li, S. Lou, P. Zuo, C. Du and G. Yin, *J. Colloid Interface Sci.*, 2019, **555**, 783-790.
- 9 P. Fan, T. Mu, S. Lou, X. Cheng, Y. Gao, C. Du, P. Zuo, Y. Ma and G. Yin, *Electrochim. Acta*, 2019, **306**, 590-598.
- 10 W. Liu, Y. Zhong, S. Yang, S. Zhang, X. Yu, H. Wang, Q. Li, J. Li, X. Cai and Y. Fang, *Sustain. Energy Fuels*, 2018, **2**, 679-687.