Supplementary material

SiO_x Anode Strengthened by Self-Catalytic Growth of Carbon Nanotubes

Hongjin Xue,^{a,b} Yong Cheng,^{a*} Qianqian Gu,^{a,b} Zhaomin Wang,^a Yabin Shen,^{a,b}

Dongming Yin,^{a,b} Limin Wang^{a,b} and Gang Huang^{a*}

^a State Key Laboratory of Rare Earth Resource Utilization, Changchun Institute of Applied Chemistry Chinese Academy of Sciences, Changchun, 130022, China ^b University of Science and Technology of China, Hefei, 230026, China

*Address correspondence to: <u>cyong@ciac.ac.cn; ghuang@ciac.ac.cn</u>.



Figure S1. XRD patterns of SiO, SiO_x, SiO_x@G, and SiO_x@CNTs.



Figure S2. (a,b) TEM images and (c) high-resolution TEM image of $SiO_x@G$. (d-g) TEM image and EDS elemental mapping images of C, Si, and O of $SiO_x@G$.



Figure S3. Raman spectra of $SiO_x@CNTs$ and $SiO_x@G$.



Figure S4. (a) XPS survey spectra of $SiO_x@CNTs$ and $SiO_x@G$. High resolution XPS spectra of (b) Si2p/Si2s, (c) C1s, and (d) O1s of $SiO_x@CNTs$ and $SiO_x@G$.



Figure S5. TGA profiles of SiO_x@CNTs, SiO_x@G and SiO_x.



Figure S6. N₂ adsorption/desorption isotherms and pore size distribution curves of (a,b) SiO_x@CNTs and (c,d) SiO_x@G.



Figure S7. CV curves of SiO_x@G at a scan rate of 0.1 mV s⁻¹.



Figure S8. Typical voltage vs. capacity profiles of $SiO_x@CNTs$ in the cycle test.



Figure S9. Cycle performance of $SiO_x@CNTs$ at a current density of 1.5 mA cm⁻².



Figure S10. Comparative cycle performance of the $SiO_x@CNTs$ and ball-milled $SiO_x/CNTs$ at a current density of 2 A g⁻¹.



Figure S11. Method of the HPPC test. (a) The battery was pulse discharged for 10 s at a rate of 3 C, which was subsequently relaxed at the open-circuit voltage (OCV) for 40 s, and then charged for 10 s with a regenerative pulse at 75% current (2.25 C rate) of the discharge pulse. The discharge procedure was repeated from 10 to 90% depth of discharge (DOD), which was followed by a 1 h rest period before applying the next sequence. (b) One sequence under 20% DOD.



Figure S12. Nyquist plots of $SiO_x@CNTs$ and SiO_x electrodes after the 1st cycle.



Figure S13. Ex-situ SEM images of SiO_x@G anode after 100 cycles.



Figure S14. Cross-sectional SEM images of (a) electrode before cycling, and (b) SiO_x and (c) $SiO_x@CNTs$ electrodes after 100 cycles.



Figure S15. Surface layer characterization of the SiO_x electrode after 100 cycles. (a) Full XPS spectrum and XPS expanded spectra of (b) Si2p, (c) C1s, and (d) O1s.



Figure S16. Surface layer characterization of the $SiO_x@CNTs$ electrode after 100 cycles. (a) Full XPS spectrum and XPS expanded spectra of (b) Si2p, (c) C1s, and (d) O1s.



Figure S17. Surface layer characterization of the $SiO_x@G$ electrode after 100 cycles. (a) Full XPS spectrum and XPS expanded spectra of (b) Si2p, (c) C1s, and (d) O1s.

| Materials | Current density (A g ⁻¹) | Cycle numbers | Capacity (mAh g ⁻¹) | Retention | Ref. |
|---|---|------------------|------------------------------------|-----------|------|
| SiO _x /G/C | 0.12 | 500 | 487 | 74.6% | [1] |
| SiO _x /C spheres | 1 | 400 | 493 | 80.9% | [2] |
| VG@SiO _x /NC | 2 | 500 | 642 | 84.2% | [3] |
| SiO/Sn | 0.2 | 100 | 850 | 60.0% | [4] |
| YS-SiO _x /C@C | 0.5 | 1000 | 557 | 76.4% | [5] |
| SiO _x -C | 0.1 | 100 | 674 | 83.5 | [6] |
| p-SiO _x @TiO ₂ @C | 0.7 | 500 | 588 | 61.1% | [7] |
| SiO _x @CNTs | 2 | 500 | 673 | 89.1 | This |
| composites | | | | | work |

Table S1. Comparative electrochemical performance of the as-prepared SiO_x @CNTs and those of the SiO_x -based composites reported previously.

The energy density of the full cell is calculated by the following equation:

Energy Density =
$$\frac{C_{cathode} \times C_{anode}}{C_{cathode} + C_{anode}} \times V_{nominal}$$

where $C_{cathode}$, C_{anode} , and $V_{nominal}$ are 173 mAh g⁻¹, 1012 mAh g⁻¹, and 3.4 V respectively.

For example, the energy density of the full cell at 0.3 C is:

Energy Density =
$$\frac{173 \text{ mAh } \text{g}^{-1} \times 1012 \text{ mAh } \text{g}^{-1}}{173 \text{ mAh } \text{g}^{-1} + 1012 \text{ mAh } \text{g}^{-1}} \times 3.4 \text{ V} = 502.3 \text{ Wh } \text{kg}^{-1}$$

References

- [1] G. Li, J.-Y. Li, F.-S. Yue, Q. Xu, T.-T. Zuo, Y.-X. Yin, Y.-G. Guo, Nano Energy 60 (2019) 485–492.
- [2] M. Han, J. Yu, J. Power Sources 414 (2019) 435–443.
- [3] M. Han, Y. Mu, F. Yuan, J. Liang, T. Jiang, X. Bai, J. Yu, J. Mater. Chem. A 8 (2020) 3822–3833.
- [4] R. Fu, Y. Wu, C. Fan, Z. Long, G. Shao, Z. Liu, Chemsuschem 12 (2019) 3377– 3382.
- [5] Y. Zhang, G. Hu, Q. Yu, Z. Liu, C. Yu, L. Wu, L. Zhou, L. Mai, Mater. Chem. Front. 4 (2020) 1656–1663.
- [6] W. Wu, J. Shi, Y. Liang, F. Liu, Y. Peng, H. Yang, Phys. Chem. Chem. Phys. 17 (2015) 13451–13456.
- [7] F. Dou, Y. Weng, G. Chen, L. Shi, H. Liu, D. Zhang, Chem. Eng. J. 387 (2020) 124106.