Supplementary Information

Fabrication and Understanding of Cu₃Si-Si@Carbon@Graphene

Nanocomposites as High-performance Anode for Lithium-Ion

Batteries

Zhiming Zheng^{a,#}, Hong-Hui Wu^{b,#}, Huixin Chen^c, Yong Cheng^a, Qiaobao Zhang^{a,*}, Qingshui Xie^a, Laisen Wang^a, Kaili Zhang^d, Ming-Sheng Wang^a, Dong-Liang Peng^{a,*}, Xiao Cheng Zeng^{b,*}

^a Department of Materials Science and Engineering, Collaborative Innovation Center of Chemistry for Energy Materials, Xiamen University, Xiamen, Fujian 361005, China

^b Department of Chemistry, University of Nebraska-Lincoln, NE 68588 Lincoln, United States

^cXiamen Institute of Rare Earth Materials, Haixi institutes, Chinese Academy of Sciences, Xiamen 361024, China

^d Department of Mechanical and Biomedical Engineering, City University of Hong Kong, 83 Tat Chee Avenue, Hong Kong

*Corresponding authors: Qiaobao Zhang (<u>zhangqiaobao@xmu.edu.cn</u>), Dong-Liang Peng(<u>dlpeng@xmu.edu.cn</u>) and Xiao Cheng Zeng (<u>xzeng1@unl.edu</u>).

 $\underline{\#}$ These authors contributed equally to this work.

The agglomeration of SC nanoparticles is shown by the FESEM images in Fig. S1a and S1b, which are completely enfolded by graphene, forming a well-connected 3D conductive network. The TEM image in Fig. S1c further shows that the SC nanoparticles around 35-45 nm in diameter with regular and uniform structure encapsulated into some pieces of graphene sheets. A high-resolution TEM (HRTEM) image in the Fig. S1d presents the carbon shells of SC composites around 3-4 nm in thickness. The inset image in Fig. S1d depicts clear and continuous lattice-fringe with the neighboring fringes distance is about 0.314 nm, which corresponds to that of (111) lattice spacing of Si. The STEM image and the corresponding element mappings of SCG are displayed in Supplementary Fig. S1e-h, respectively. The clear distribution of C and Si elementals further confirms the encapsulation of SC composites in graphene sheets.



Fig. S1 (a, b) FESEM images of SCG composite. (c)TEM and (d) HRTEM image of SCG composite. (e -h) STEM image of SCG composite and corresponding element mappings of Si and C elements.



Fig. S2 Thermal analysis of SC and SCG composites.



Fig. S3 (a) Cycling performance of Cu₃Si-Si-2, Cu₃Si-SC-2 and Cu₃Si-SCG-2 composite at current density of 1 A g⁻¹. (b) Cycling performance of SCG together with SCG composite after heat treatment 2 h at 700, 800 and 900 °C at current density of 1 A g⁻¹



Fig. S4 Experimental (scatter diagram) and fitted (solid lines) Nyquist plots of SCG electrode before cycling, Cu₃Si-SCG -2 electrode before cycling, Cu₃Si-SCG-2 electrode after 500 cycles at 4 A g⁻¹.



Fig. S5. Cycling performance of the LCO cathode in half cell: at 1 C rate for 200 cycles.

Table S1 A summary of the performances of different inactive phase/Si-based anodes in the literature in comparison with the results in this work.

| Electrode material | Cycling stability | | | | |
|--|-------------------------------------|-------|----------------------|---------------|------|
| | Capacity after cycles | cycle | Current density | Capacity | Ref. |
| | [mA h g ⁻¹] | | [A g ⁻¹] | retention [%] | |
| Highly connected hollow Si-Cu | 1676 | 100 | 0.84 | 77.6 | 1 |
| alloy-nanotube | | | | | |
| Micro-sized FeCuSi ternary composites | 1054 | 50 | 0.5C | 90 | 2 |
| Nanoporous Si/Cu composites | 1306.2 | 150 | 0.2 | 55.6 | 3 |
| Si–Ti–Ni alloy | 900 (based on | 50 | 1C | _ | 4 |
| | Si–Ti–Ni alloy) | | | | |
| Antimony-doped tin oxide coated Si | 1320 | 50 | 0.5C | 88 | 5 |
| Si _{sere} | 1186.9 | 180 | 1.5 | _ | 6 |
| Cu ₃ Si@Si core-shell nanoparticles | 1560.6 | 50 | 1 | 89.2 | 7 |
| Si-Cu ₃ Si-Al ₂ O ₃ nanocomposite | 704 | 200 | 0.2 | 83.7 | 8 |
| Cu ₃ Si-SCG-2 | 522 (based on | 500 | 1 | 70.7 | This |
| | the total mass of Cu ₃ S | bi- | | | work |
| | SCG) | | | | |
| Cu ₃ Si-SCG-2 | 483 (based on | 500 | 4 | 80 | This |
| | the total mass of Cu ₃ S | 5i- | | | work |
| | SCG) | | | | |
| Cu ₃ Si-SCG-2 | 676 (based on | 500 | 4 | 80 | This |
| | the mass of Cu ₃ Si and | d | | | work |
| | Si) | | | | |
| Cu ₃ Si-SCG-2 | 1208 (based on | 500 | 4 | 80 | This |
| | the mass of Si) | | | | work |

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