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Supporting Information

Building Sponge-Like Robust Architectures of CNT-Graphene-Si Composites with Enhanced Rate and Cycling Performance for Lithium-Ion Batteries

Xiaolei Wang,[a] Ge Li,[a] Fathy M Hassan,[a] Matthew Li,[a] Kun Feng,[a] Xingcheng Xiao,[b] and Zhongwei Chen[a]**

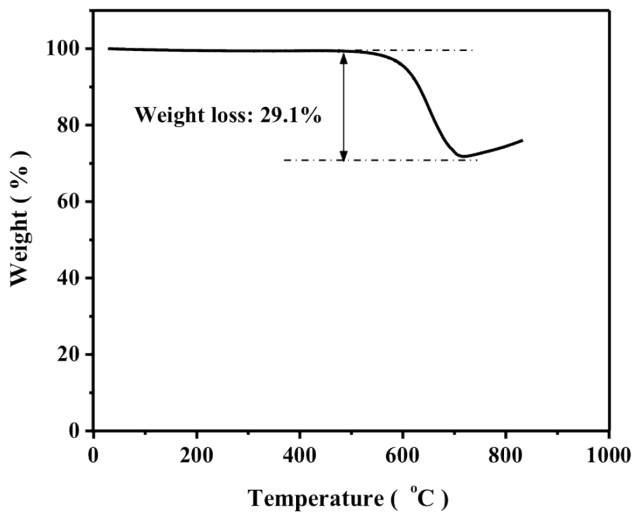

Figure S1. Thermogravmetric analysis of CNT-graphene-Si composite materials conducted in air from 25 to 850 °C. A significant mass loss is observed from ~500 to 700 °C due to the carbon content being burned away. A mass increase is observed at higher temperature, which can be ascribed to the partial oxidation of silicon forming silicon oxides. To further investigate the ratio of Si in the composite materials, the composite was calcinated at 850 °C for 24 hours in air. A 66% Si composition can be obtained based on the calculation of mass of the white powder (SiO_2) remained.

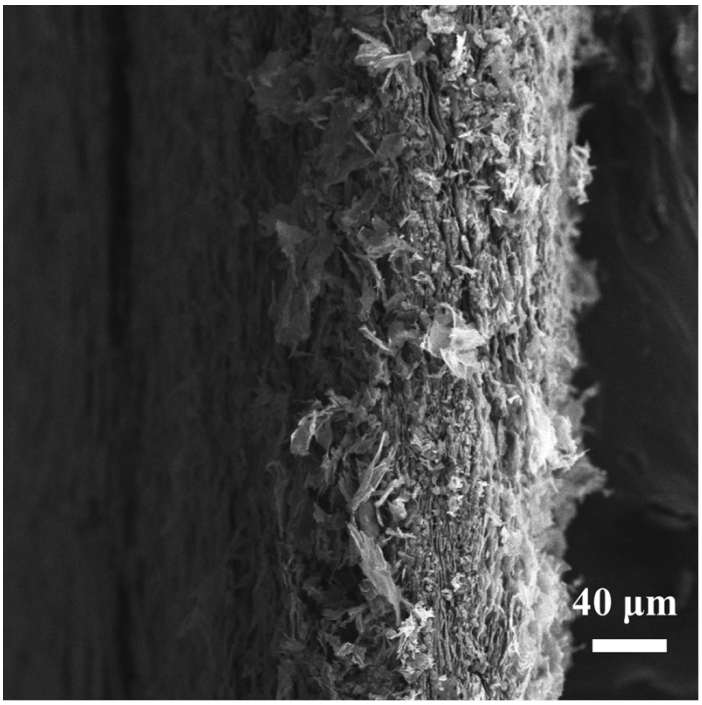

Figure S2. Representative SEM images of the cross-section of the CNT-graphene-Si composites after being pressed at 10 MP. The porous structure can still be observed.

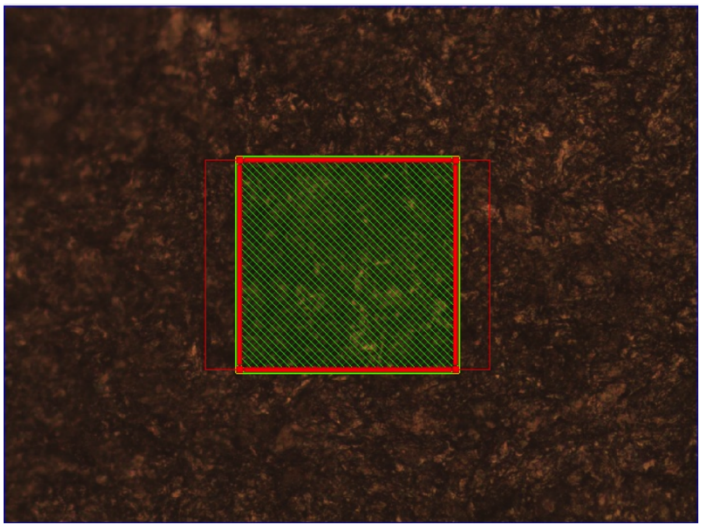

Figure S3. Optical image of an electrode from Raman spectroscopy with dispersive microscope. The marked area is 100×100 μm^2, which is used for Raman mapping of Si peak.

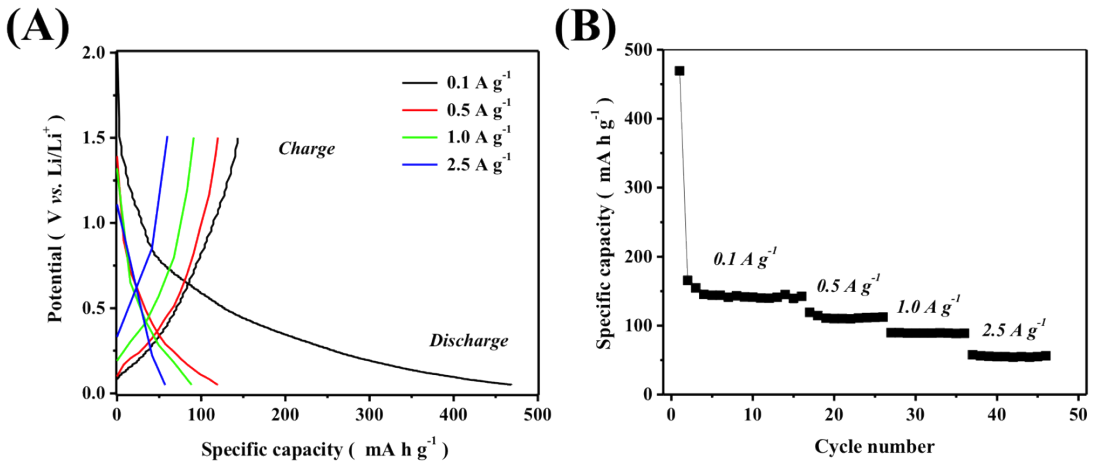

Figure S4. (A) Galvanostatic charge/discharge curves of a pure CNT-graphene electrode at various current densities from 0.1 to 2.5 A g^{-1} between 1.5 and 0.050 V. Such an electrode delivers a high initial discharge capacity of ~470 mAh g^{-1} with a low Coulombic efficiency. The large irreversible capacity could be attributed to the SEI formation on the large surface which is created by assembling CNTs and graphene sheets. This phenomenon is consistent with previous reports on carbon-based materials with high surface area as LIBs anode materials.[S1-S3] (B) Corresponding rate performance of the electrode.

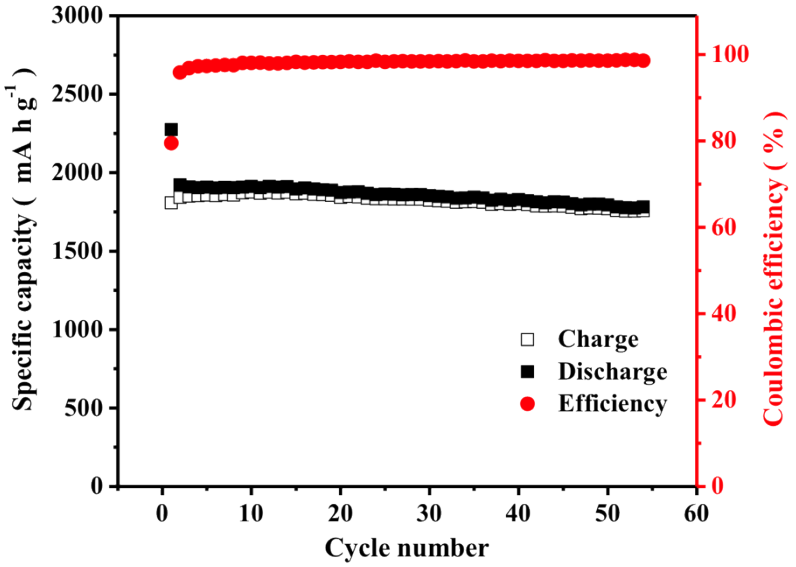

Figure S5. Cycling stability of the CNT-graphene-Si electrode at 0.1 A g^{-1} between 1.500 and 0.005 V *vs.* Li/Li^+. An initial discharge capacity of 2275 mA h g^{-1} can be delivered with a Coulombic efficiency of 79.5%. After 50 cycles, a capacity retention of 78.9% can be achieved, corresponding to ~1.58% capacity loss for each cycle.

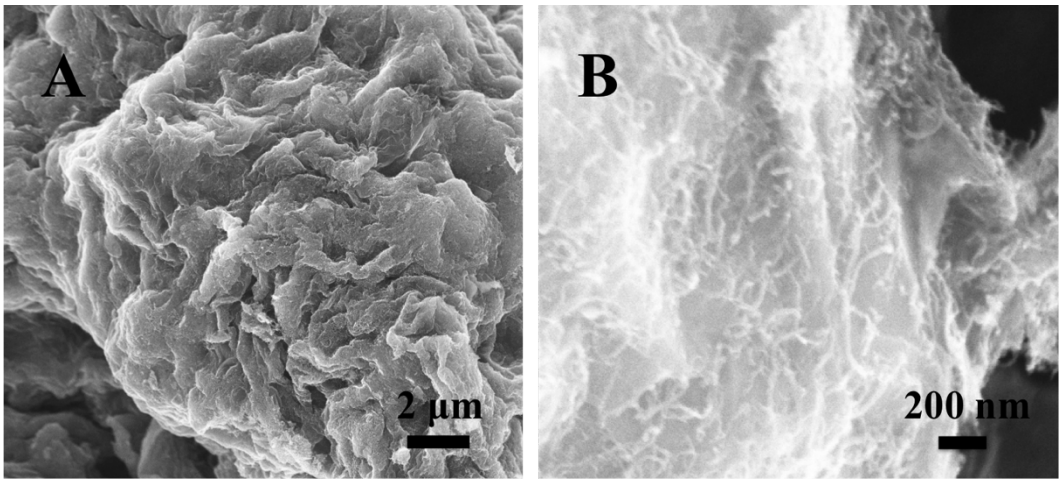

Figure S6. (A) Low- and (B) High-magnification SEM images of pure CNT-graphene networks architecture.

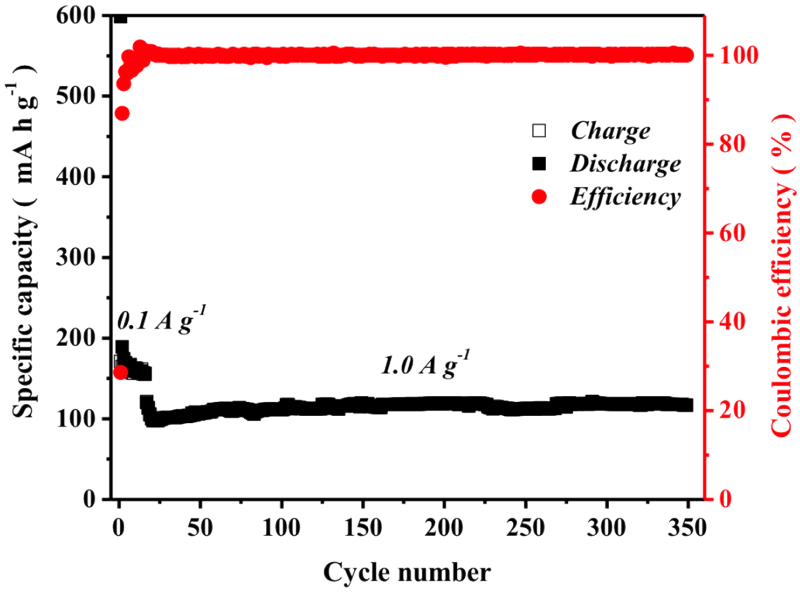

Figure S7. Cycling stability of the pure CNT-graphene electrode at 0.1 A g^{-1} for 16 cycles and at 1.0 A g^{-1} for 340 cycles.

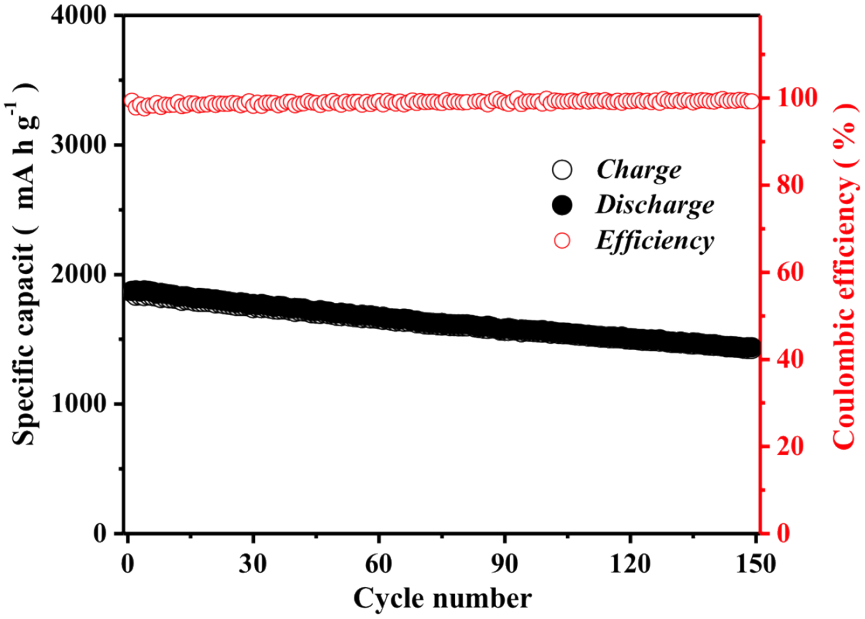

Figure S8. Cycling stability of the composite electrode at 0.1 A g^{-1} for 150 cycles after 10 cycles activation at 0.1 A g^{-1}. A discharge capacity retention of 76.7% can be obtained with high Coulombic efficiency.

References

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[S3] J. R. Dahn, T. Zheng, Y. Liu, J. S. Xue, Science **1995**, 270, 590.