Supplementary Information

Three-dimensional graphene scaffold supported thin film silicon anode for lithium-ion battery

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The Ni foil (round, ~1.4 cm diameter) was chosen as the substrate for graphene synthesis, which is bright shown in Fig. S1(a). After treated in hydrogen plasma atmosphere under 650 ^{o}C in CH_4 atmosphere, graphene was grown on the Ni surface, and consequently the color becomes black as shown in Fig. S1(b). In Fig. S1(c), it is found that after deposition of Si, the foil color changes to gray.

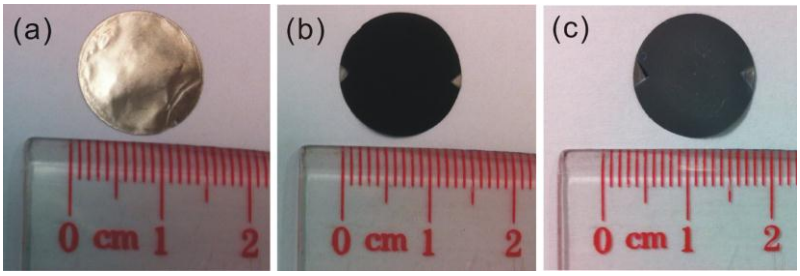

Fig. S1 Photos of (a) Ni foil, (b) after growth of graphene on Ni foil, and (c) after deposition of Si on the top of graphene.

Fig. S2(a) reveals HRTEM images of the graphene sheet scratched from nickel foil. The hexagonal arrangement of atoms can be observed regularly in some areas. However, for the whole area of the graphene sheet, some dislocations appear, suggesting that the synthesized graphene is polycrystalline. This is in agreement with the Raman spectrum of the as-synthesized 3D graphene shown in Fig. 4(a). In addition, the hexagonal structure of the synthesized graphene can be proved from fast Fourier transferred (FFT) image (Fig. 4(b)). In Fig. 4(c), the SAED pattern of the graphene sheet is also demonstrated, which confirms the conclusion of the polycrystalline characteristic.

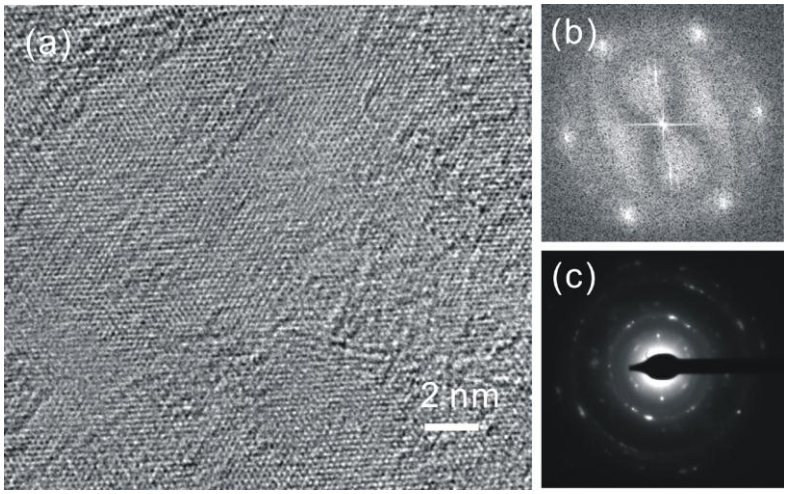

Fig. S2 (a) HRTEM image of graphene sheet, (b) fast Fourier transferred (FFT) image of Fig. S2(a), and (c) SAED pattern of the as-grown graphene sheets.

After 500 cycles at a current density of 797 mA g^{-1} (Fig. S3(a)), the cell was detached in a glove box. GSSSE was washed in acetone, isopropyl alcohol, and deionized (DI) water sequentially. After that, GSSSE was transported in air to SEM system for morphology observation and elemental mapping analysis. Fig. S3(a) shows the SEM image of GSSSE after 500 cycles at a current density of 797 mA g^{-1}. After long cycles, it reveals that the Si

nanostructure has been pulverized and forms a sponge-like structure. It is interesting to note that even the Si electrode has been pulverized into nanoparticles, but the specific capacity after 500 cycles is still 84% of that at the 50th cycle. Then, the sample was sonicated for 2 seconds, which results in the removal of some surface Si layers, and thus the inner structure of the Si anode can be observed as shown in the Fig. S3(b). It demonstrates that the pulverized Si nanoparticles were attached on graphene surface tightly, which suggests that the graphene effectively acts as a scaffold for Si nanostructures. In addition, the high quality 3D flexible graphene also effectively impedes the aggregation of the Si nanoparticles.

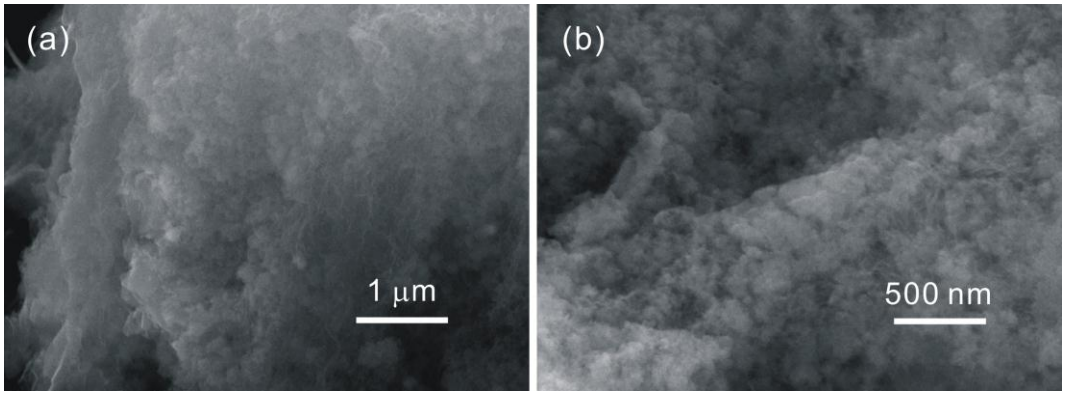

Fig. S3 45^{o} tilted SEM images of (a) GSSSE after 500 cycles at a current density of C/3, and (b) after sonication treatment.

In order to further confirm our observation, energy dispersive X-ray spectroscopy (EDS) elemental mapping for C, O, Si, and Ni in the area shown in the SEM images in Fig. S4 (a) were carried out as shown in Fig. S4(b)-(e). It verifies our conclusion that even the Si electrode was pulverized; the Si nanoparticles are still uniformly attached on the graphene scaffold. Oxygen is also found on GSSSE after chemical reactions during cycling. It should be noted that some

residue of electrolytes may still exit after washing, and GSSSE may be oxidized in some extent during transportation in air. These factors may affect the SEM and elemental mapping results.

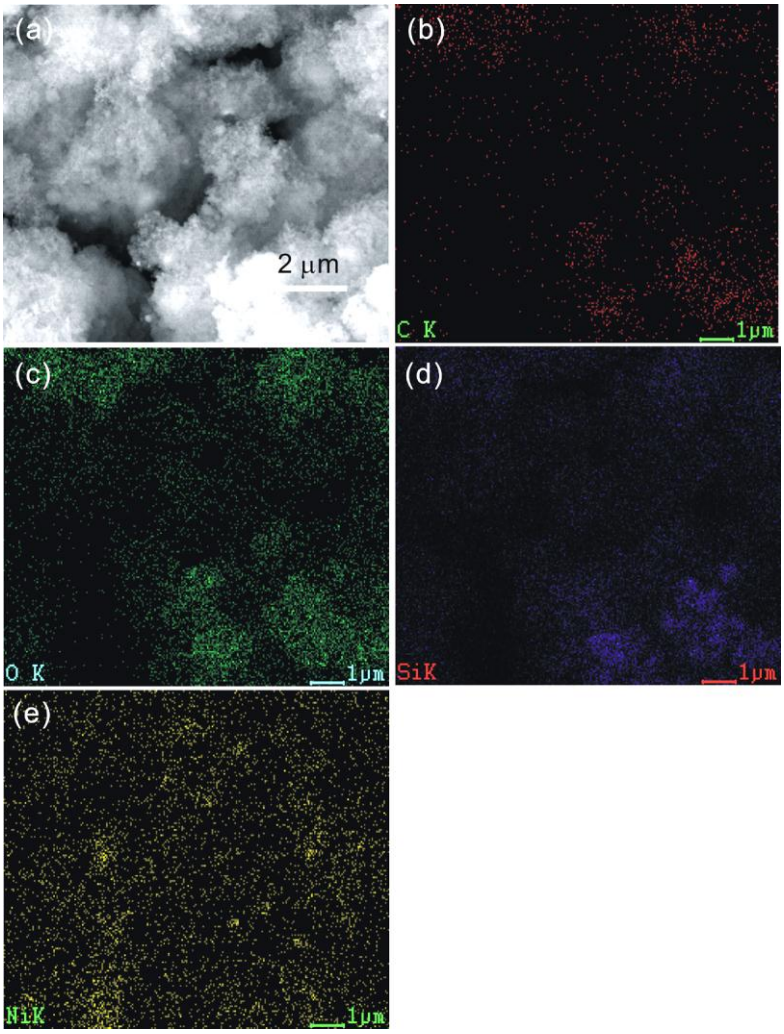

Fig. S4 EDX mapping for C, O, Si and Ni elements in the whole area of one selected region shown in the SEM image in Figure 4S(a).