Fig. S1. (a) SEM secondary electron image, (b) and (c) Elemental maps of Si and C in Si/CNF, respectively, (d) EDS spectra of Si /CNF.
Fig. S2. XRD pattern of the Si/CNF electrode. No detectable peak for Si was observed. The two peaks can be indexed to the stainless steel (JCPDS no. 33-397).
**Fig. S3.** X-ray diffraction patterns of Si film on Cu substrate (red line) and Cu substrate alone (black line).

**Fig. S4.** Raman spectra of CNFs before and after the Si film deposition (red line: Si/CNF, black line: CNF).
Fig. S5. TEM images of the Si/CNF composite anode after 200 cycles.

Fig. S6. Cycle performance of CNF alone electrode between 0.01 and 2.0 V at a current rate of 0.5 C (or 150 mA g\(^{-1}\)) in the first 3 cycles, and 1 C (or 300 mA g\(^{-1}\)) for the rest of the cycles.
Fig. S7. Cyclic voltammetry (CV) of the Si/CNF anode. The initial 3 cycles were limited in the potential range between 0.01 and 1.5 V, while at the 4th cycle, the potential limit was extended to 3.0 V. The solid electrolyte interphase formation was observed at 0.5 V during the initial cathodic scan. The CV curves are typical for Si anodes, showing two cathodic peaks at 0.23 and 0.06 V and two anodic peaks at 0.33 and 0.49 V; the cathodic shoulder peak at 0.09 V and the anodic peak at 1.7 V are very likely due to the lithiation and delithiation of the CNF, a poorly crystalized carbon.
Fig. S8. CNFs grown on stainless steel foil. (a) CNF-grown stainless steel foil in CVD reactor. (b) CNFs are totally covered on both sides of stainless steel foil size of 15 × 150 mm.