Electronic Supplementary Material (ESI) for Journal of Materials Chemistry A.

Supplementary Information

For the mechanical measurement, the 3D aligned CNT/Si hybrid was cut to form rectangular strips with sizes of 60 mm × 5 mm. The thickness was measured by SEM. The samples were tested on a HY0350 table-top universal testing instrument. The electrodes were loaded in a uniaxial tension at a rate of 1 mm min^{-1} with a gauge length of 40 mm. To test the tensile properties after cycling, the electrode strips were electrochemically cycled in a sealed glass tube with lithium foil as a counter electrode using 1 M of $LiPF_6$ in a mixture of ethylene carbonate and diethyl carbonate (volume ratio of 1/1) as the electrolyte. After cycling, the electrode strips were soaked in acetonitrile overnight to wash the residual electrolyte. They had been finally rinsed with deionized water prior to the mechanical measurement.

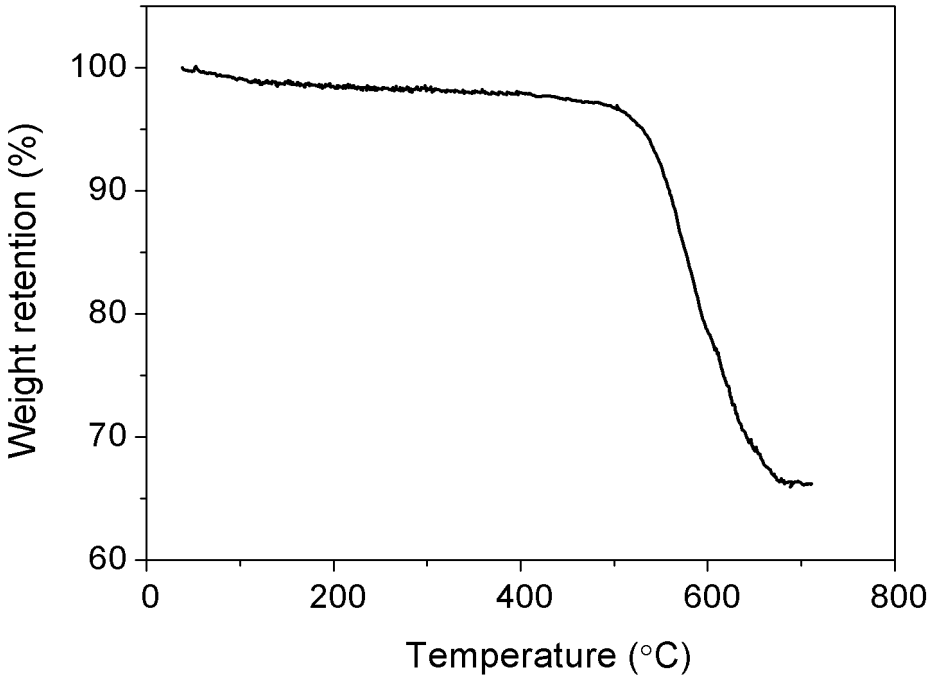

Figure S1. Thermogravimetric analysis of the 3D aligned CNT/Si hybrid in air.

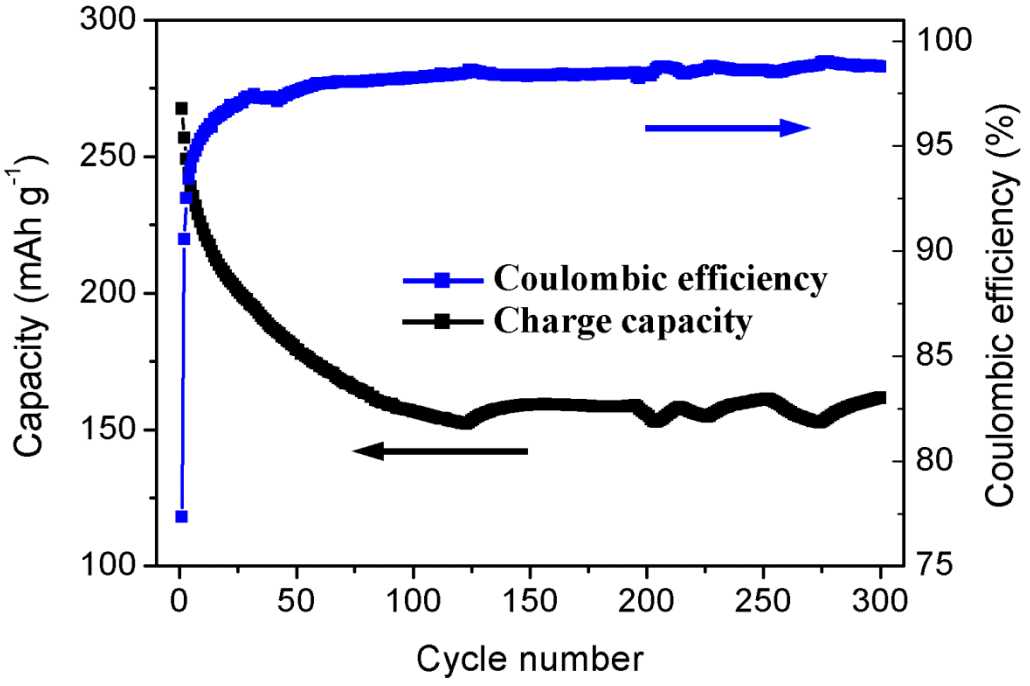

Figure S2. Cyclic performance of the bare CNT at a current density of 1 A g^{-1} at a voltage range between 3 and 0.005 V.

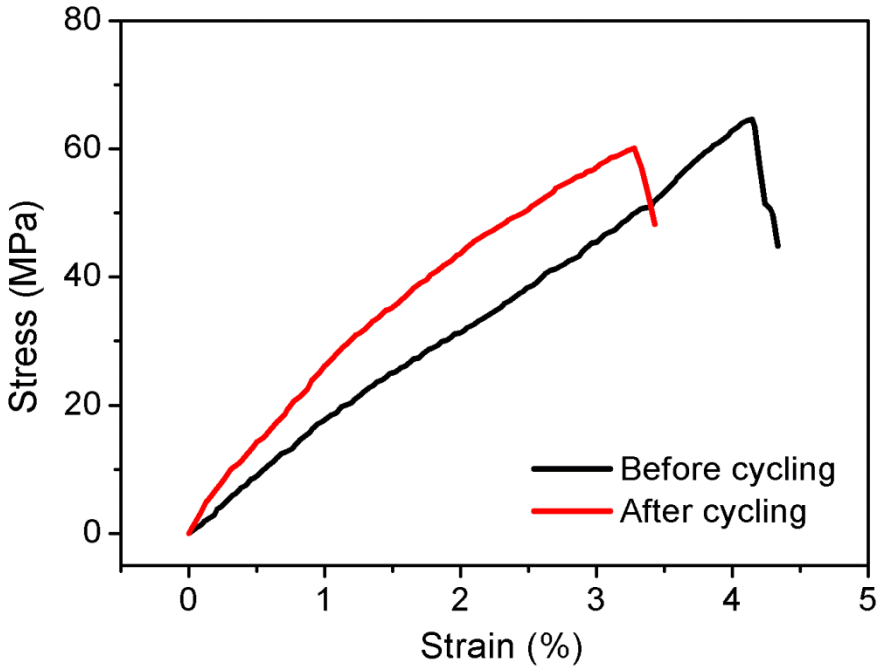

Figure S3. Stress-strain curves of 3D aligned CNT/Si hybrid anode before and after being cycled for 100 cycles at a current density of 5 A g^{-1} at a voltage range between 3 and 0.005 V.

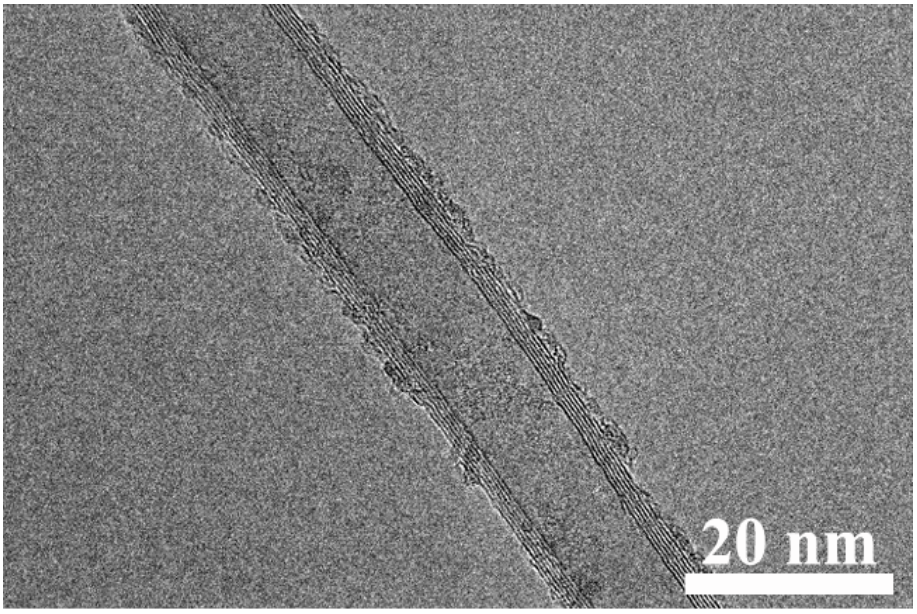

Figure S4. TEM image of a CNT.

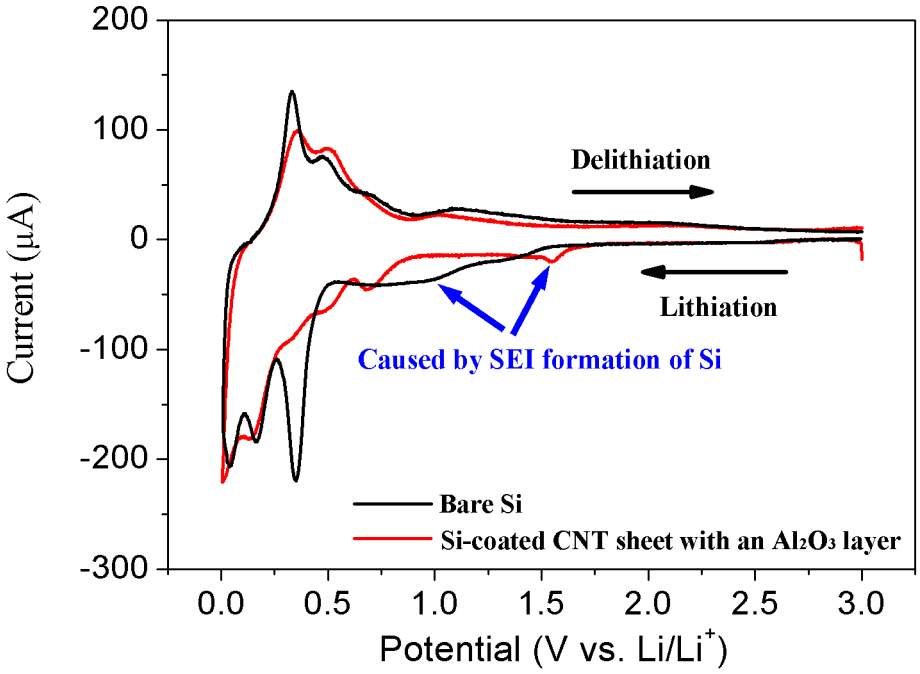

Figure S5. Cyclic voltammograms of the bare Si and Si-coated CNT sheet with an Al_2O_3 layer cycled from 3 to 0.005 V versus Li/Li^+ at a scan rate of 0.1 mV s^{-1}. Here SEI corresponds to the solid electrolyte interface.

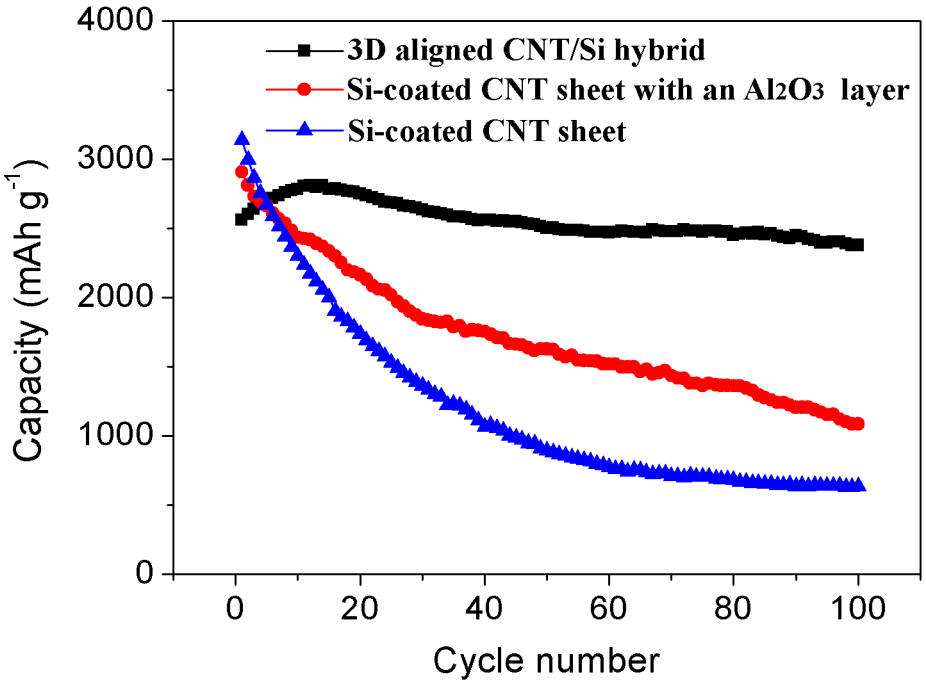

Figure S6. Comparison of the electrochemical performance of the Si-coated CNT sheet, Si-coated CNT sheet with an Al_2O_3 layer and 3D aligned CNT/Si hybrid cycled between 3 and 0.005 V versus Li/Li^+ at 1 A g^{-1} for 100 cycles.

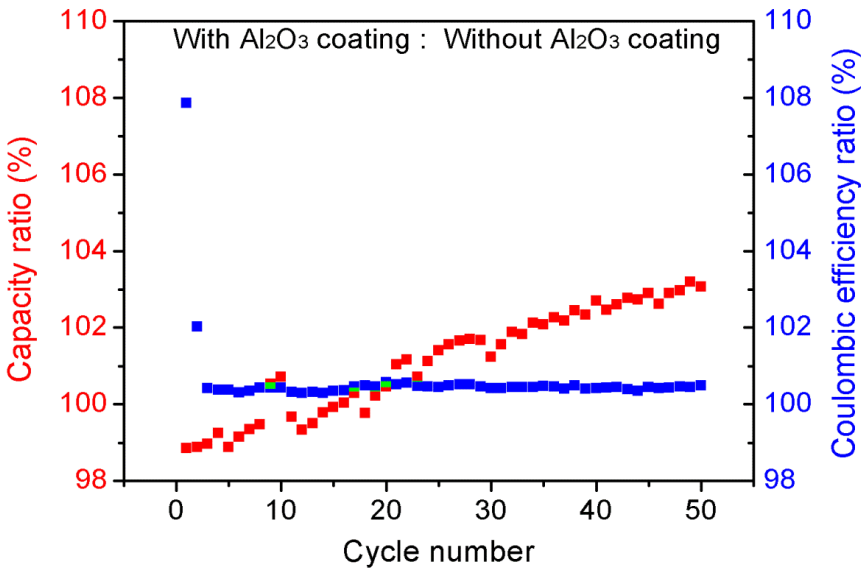

Figure S7. Comparison on the electrochemical performance between 3D aligned CNT/Si hybrid with and without an Al_2O_3 coating. The electrodes were cycled for 50 cycles at a current density of 1 A g^{-1} at a voltage range between 3 and 0.005 V.