Supporting Information

Two-dimensional ultra-thin SiO_x ($0 < x < 2$) nanosheets with long-term cycling stability as lithium ion battery anodes

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Electronic Supplementary Information contains Experimental section and Fig. S1-S10.

1. Experimental

1.1 Synthetic procedures

1.1.1 Preparation of ultra-thin SiO_x ($0 < x < 2$) nanosheets

0.15 g of calcium silicide ($CaSi_2$, purchased from Sigma Aldrich) and 1 g of NH_4Cl were added into 6 g of ionic liquid (IL) butyl-3-methylimidazolium chloride (provided by Shanghai Chengjie Chemical Co. LTD., China). The mixture was stirred at 90°C for 2 h to form a nearly homogeneous dispersion. Then, the dispersion was transferred into a Teflon-lined stainless-steel autoclave and heated at 190°C for 3 days. After the autoclave cooled down to room temperature naturally, the reaction products were washed with formamide to remove IL and some inorganic salts. The obtained solids were re-dispersed in CH_3CN and sonicated for several hours. The upper yellow solution was separated and centrifuged to afford SiO_x nanosheets (~80 mg).

1.1.2 Preparation of carbon-coated SiO_x nanosheets

The SiO_x nanosheets (0.2 g) were added in Tris-buffer (50 mL, 10 mM) and sonicated for 20 min. Subsequently, 0.1 g of dopamine hydrochloride was added and the dispersion was stirred for 24 h. The products were collected and washed with water. The dry powders were placed in a tube furnace and heated under Ar at 400°C for 2 h with a heating rate of 2°C min^{-1}, and then at 800°C for 3 h with a heating rate of 5°C min^{-1}. The calcined products were immersed in dilute HF solution for 10 min, filtered and washed with distilled water to yield the SiO_x@C nanosheets.

1.2 Characterization

Powder X-ray diffraction (PXRD) data were collected on a Bruker D8 Advance instrument using a Cu Kα radiation (λ = 1.54056 Å) at room temperature. Scanning Electron Microscope (SEM) images and Energy Dispersive Spectroscopy (EDS) were obtained on a Hitachi S-4800 field-emission scanning electron microscope at an acceleration voltage of 5.0 kV and 20 kV, respectively. Transmission electron microscopy (TEM) characterization was carried out using a JEM-2100 (Japan). Atomic Force Microscopy (AFM) was performed using a SPI3800/SPA400 (Seiko Inc., Japan) in contact mode with a Si CANTILEVER. Raman spectroscopy (InVia-Reflex, Renishaw) was performed with a 633 nm wavelength. X-ray photoelectron spectroscopy (XPS) measurements were recorded with a PHI 5000 VersaProbe. Thermogravimetric (TG) analyses were performed on a simultaneous STA449F3 thermal analyzer under flowing air with a heating rate of 5°C min^{-1}. Fourier-transformed infrared (FT-IR) spectra were measured on a FT-IR spectrometer (Vector22) with the KBr pellet method. Nitrogen sorption isotherms were collected at 77 K (Micrometrics ASAP 2020 analyzer) after vacuum degassing of the sample at 200°C for 8 h.

1.3 Electrochemical measurements

CR2025 cells were assembled in an argon-filled glove box to perform electrochemical experiments. The electrode was composed of 80 wt% of active material, 10 wt% of conductive graphite, and 10 wt% of sodium carboxymethyl cellulose as a binder. The mixture was stirred in water and blade-coated on a piece of Cu foil. After drying at 85°C in vacuum for 12 h, the foil was cut into disks of 12 mm in diameter. The electrolyte consisted of a solution of 1 M $LiPF_6$ in a mixture of 1:1 (vol/vol) ethylene carbonate / diethyl carbonate with 2 wt % vinylene carbonate added. Pure Li foils were served as counter electrodes. The discharge-charge measurements were performed on a Neware battery testing device (Shenzhen, China) at the constant current mode over the range of 0.01−2 V. The specific capacities were calculated based on the total weight of SiO_x@C nanosheet composites. For each electrode, the loading amount of SiO_x@C is around 0.8 mg cm^{-2}.

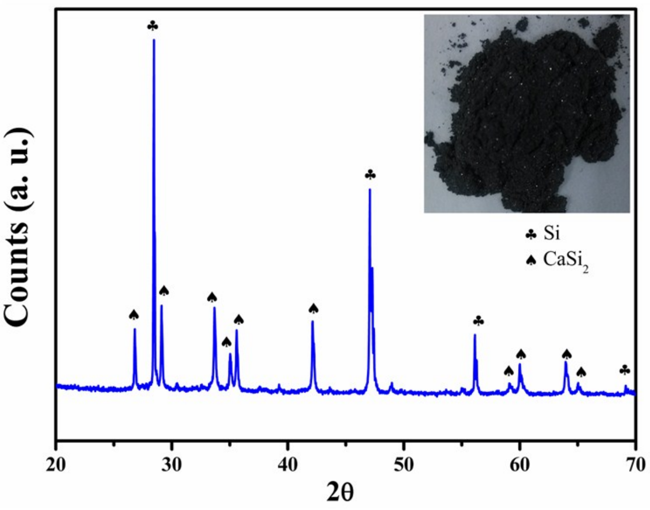

Fig. S1. PXRD of $CaSi_2$ powders (insert shows a digital camera image).

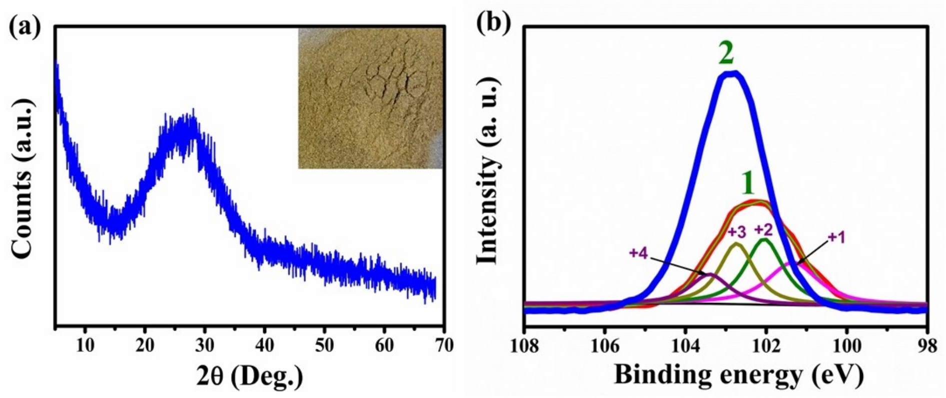

Fig. S2. (a) PXRD pattern of SiO_x nanosheets (Insert shows a digital image); (b) XPS spectra of (1) as-prepared SiO_x nanosheets (including the resolved Si 2p peaks) and (2) SiO_x nanosheets after calcined at 700 °C in air for 2 h.

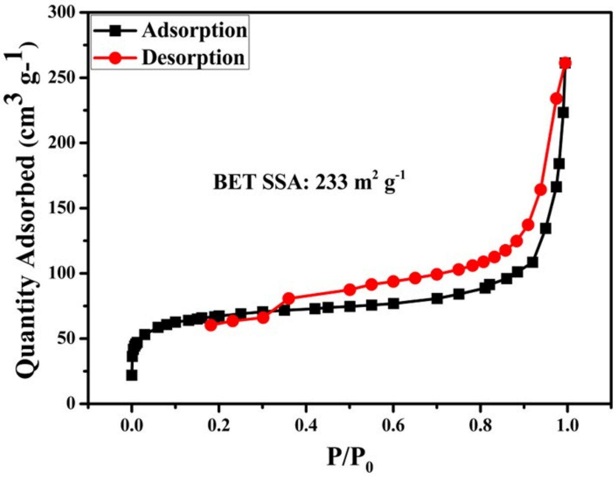

Fig. S3. Typical nitrogen adsorption isotherms of SiO_x nanosheets.

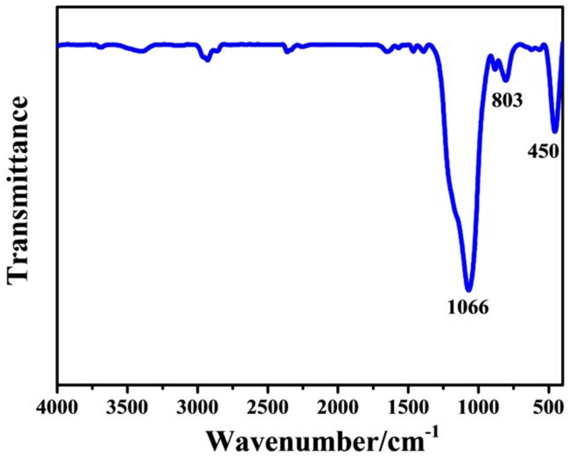

Fig. S4. FT-IR spectrum of the obtained SiO_x nanosheets.

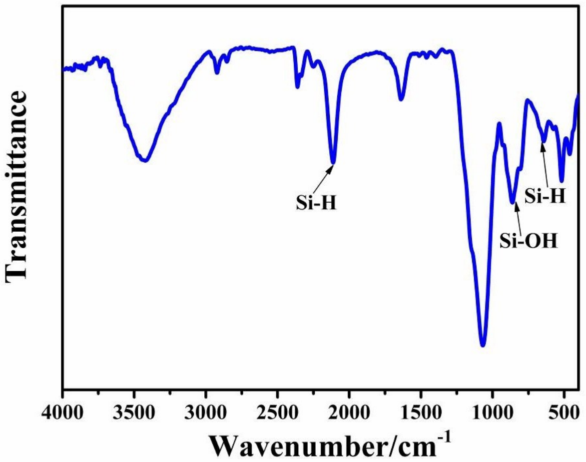

Fig. S5. FT-IR spectrum of the Si_6H_6 nanosheets.

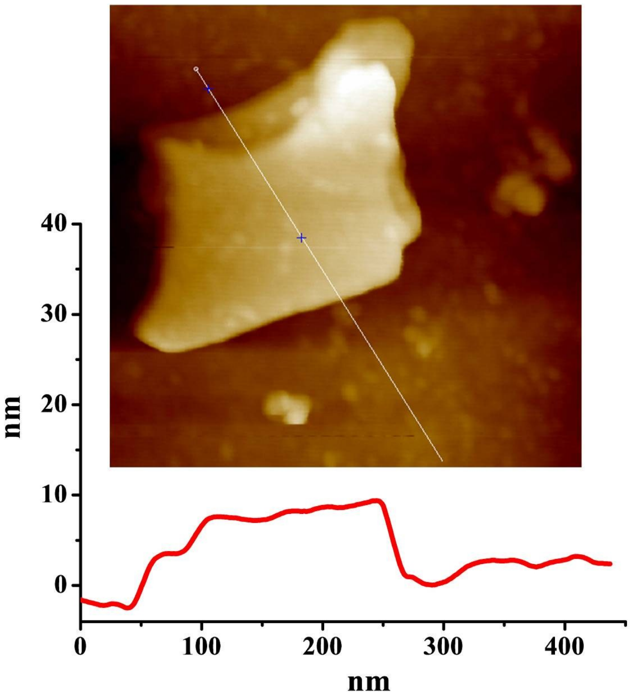

Fig. S6. AFM image of the obtained SiO_x nanosheets.

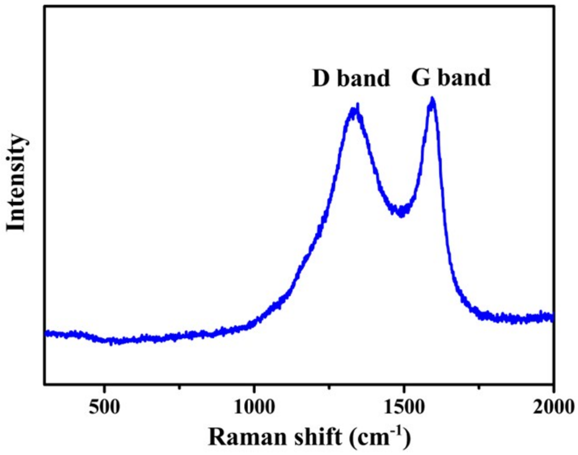

Fig. S7. Raman spectrum of SiO$_x$@C nanocomposite.

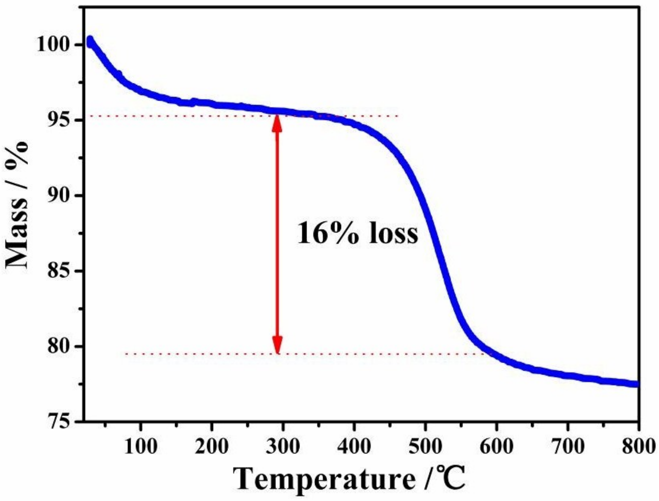

Fig. S8. TG curve of the SiO$_x$@C nanocomposite.

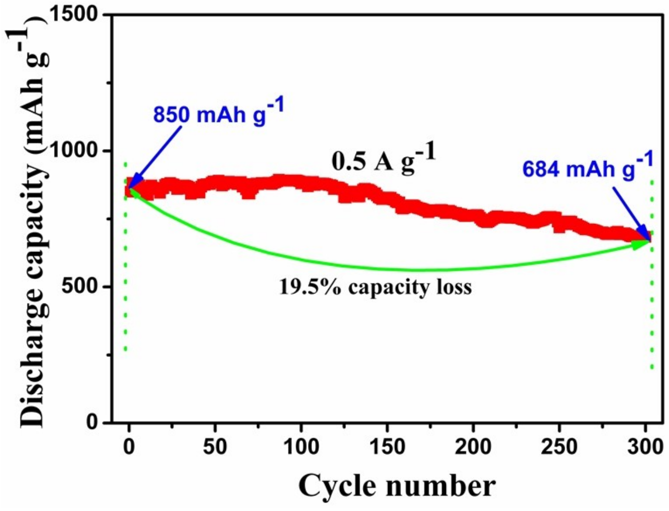

Fig. S9. Cycling performance of SiO_x nanosheet anode.

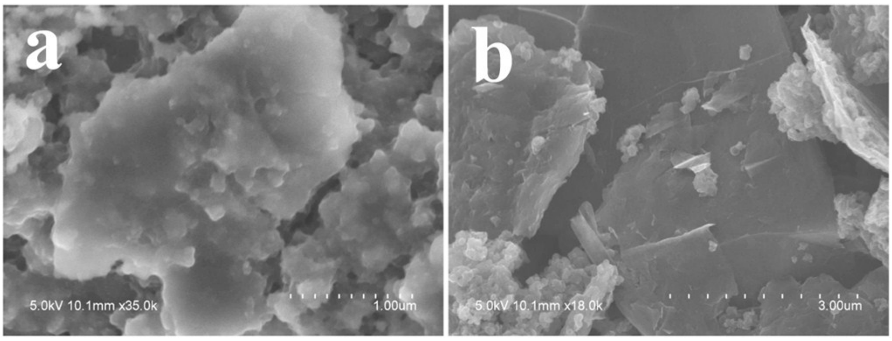

Fig. S10. SEM images of the SiO_x@C nanosheets after charging-discharging for 400 cycles: (a) washed with acetonitrile; and (b) washed with acetonitrile and 1 M HCl solution.