Electronic Supplementary Information for

A Novel Bath Lily-Like Graphene Sheets-Wrapped Nano-Si Composite as a High Performance Anode Material for Li-ion Batteries †

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Experimental details

Preparation of GS-Si composite:

Nano-size silicon (50~200 nm) was supported by Nanjing Emperor Nano Material Co., Ltd. (Nanjing, China). Graphite oxide was synthesized from natural graphite powder (Grade 230, Asbury Carbons) using the modified Hummers method.1 The obtained graphite oxide was exfoliated into deionized water by ultrasonication to form graphene oxide (GO) suspension. Thereafter, a certain amount of nano-silicon powder was added into the GO suspension. The mixture was sonicated for 45 min and then spray-dried at 200 °C to form a GO-Si composite using a B-290 mini spray drier (Buchi, Swiss). The obtained composite was load into a quartz tube in a flow of 20% H2 in Ar at 700 °C for 3 h to form the graphene sheets-wrapped nano-Si (GS-Si) composite. Pure GS samples were also prepared by the same procedure and then mixed with pure Si powder to obtain GS/Si mixture for comparison.

Structural and morphological characterization:

XRD measurements were carried out using a Rigaku D/MAX-2200/PC X-ray diffractometer at 40kV and 20mA, with a Cu Kα radiation source. Raman spectroscopy was used to identify the surface characteristics of the samples using a BRUKER optic SENTERRA (R-200L) Raman spectrometer using a laser with a wavelength of 633 nm at room temperature. Thermogravimetric analysis (TGA) was performed using a STA 449F3 analyzer (NETZSCH Co., Germany) to evaluate the residual level of GS-Si composite after calcination. The morphology and microstructure of the samples were monitored using a FEI Nova SEM 230 ultra-high resolution Field Emission Scanning Electron Microscopy (FESEM) equipped with energy dispersive X-ray spectroscopy (EDS, INCA X-Max 80, Oxford Instruments) and a JEM-2100F Transmission Electron Microscopy (TEM) (JEOL Ltd., Japan). EDS was obtained at four different locations for each sample. The above obtained oxygen content is a mean value.

Electrochemical characterization:

Electrode preparation:
The electrochemical performances of GS-Si composite and GS/Si mixture were evaluated under the same conditions using coin-type half cells. The working electrodes were prepared by pasting a mixture of active material, Super P conductive carbon black (40 nm, Timical) and styrene butadiene rubber/sodium carboxymethyl cellulose (SBR/SCMC, 1:1 by weight) as binder at a weight ratio of 75:15:10. After coating the mixture onto pure Cu foil, the electrodes were dried, cut to Φ14 mm sheets, pressed at 3 MPa, and finally dried at 80 °C in vacuum for 4 h. The active materials loaded on the electrode were about 1.5 mg cm−2.

Cell assembly and electrochemical tests:
CR2016 coin cells were assembled in an argon-filled glove box with lithium metal as counter
electrode and UP3025 separator (provided by UBE Industries, Ltd., Japan). The electrolyte contained 1 M LiPF$_6$ in dimethyl carbonate (DMC) and ethylene carbonate (EC) mixed solvent of 1:1 (LP30 from EM Industries, Inc.). Charge–discharge cycles of the half-cells were evaluated between 0.01 and 1.2 V vs Li$^+/Li$ at room temperature using LAND CT2001A model battery test system (Wuhan Jinnuo Electronics, Ltd.) under constant current condition. The charge-discharge capacities were calculated according to the weight of GS-Si (or GS/Si) material in the electrode. For AC electrochemical impedance spectra (EIS) tests, the cells with GS-Si and GS/Si electrodes were cycled twice at 200 mA g$^{-1}$ and then brought to a standstill for five hours. The open circuit voltage (OCV) vs Li$^+/Li$ of both cells stabilized at about 0.9 V, indicating the same lithiation level. Finally, the EIS of the cells were measured by a Solartron FRA 1250 frequency responses analyzer combined with a Solartron SI 1287 Electrochemical Interface with an amplitude of 10 mV over a frequency range from 100 kHz to 0.1 Hz.

![Fig. S1 TGA curves of pure Si and GS-Si composite.](image)
Fig. S2 SEM images of GO-Si composite.
**Fig. S3** SEM images of GO/Si mixture.

**Fig. S4** The initial two charge-discharge profiles of GS-Si composite and GS/Si mixture at a current density of 100 mA g\(^{-1}\).
References