Supplementary Information

**Flower-like Carbon with Embedded Silicon Nano Particles as Anode Material for Li-ion Battery**

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Experimental

Material preparation

The mixture for making the F-Si/C composite was prepared by dissolving 0.5 g SiNPs (D<100 nm, Aldrich), 0.5 g NaCl (Sinopharm Chemical Reagent Co., Ltd, China), and 0.8 g Polyvinyl pyrrolidone (PVP, Aldrich, $M_w=24000$) in 100 ml DI water and subsequently sonicating for 6 hours to form a homogeneous suspension. The suspension was then spray dried to form a solid precursor with the following conditions: the suspension delivery rate was 15 mL min$^{-1}$; the inlet temperature of the spray dryer was 140 °C; the outlet temperature was 80 °C. This precursor was then carbonized at 900 °C for 2 h in a quartz tube furnace under N$_2$ atmosphere. After the carbonization step, the obtained material was mixed with DI water with 1:100 mass ratio in a beaker and stirred with magnetic stirring for 6 hours at room temperature, then filtered with extraction filtration. To completely remove the NaCl, the washing and extraction filtration processes were repeated one more time and the solid product was dried at 60 °C. The final product is the F-Si/C composite. For comparison, a Si/C composite made with identical fabrication process without using NaCl was also prepared.

Material characterization

The morphologies of the composites were observed and compared using a scanning electron microscope (SEM, JEOL JM3-6390). Elemental analysis of the composites was performed using energy-dispersive X-ray spectroscopy (EDX). A transmission electron microscope (TEM, FEI Tecnai G2 F20 S-TWIN) was employed to characterize the microstructures. The crystal structures of Si/C composites were determined by X-ray diffraction (XRD, Bruker D8 ADVANCE using Cu Kα radiation scanning from $10^\circ<\Theta<80^\circ$). The weight content of SiNPs in the Si/C composite was determined from the weight loss curves measured by thermogravimetric analysis under atmosphere (TGA, SDT Q 600, TA) with a heating rate of 5 °C min$^{-1}$. The N$_2$
absorption and desorption isotherms were measured at 77K using a gas sorption analyzer (Quantachrome, Autosorb IQ). The specific surface area was calculated using Brahnauer-Emmett-Teller (BET) method and the relative pressure range of P/P_0=0.1-0.3 was used for multipoint BET calculations.

**Electrochemical characterization**

The electrochemical performance of the composites was evaluated by galvanostatic cycling using coin cells with the Si/C composite as the working electrode and lithium foil as the counter/reference electrode. The electrodes were prepared with the following procedure: 1) grounding 80 wt.% Si/C composite, 10 wt.% carbon black (Sinopharm Chemical Reagent Co.,Ltd, China), and 10 wt.% polyvinylidene fluoride (Sinopharm Chemical Reagent Co.,Ltd, China) to form a homogeneous mixture; 2) adding 1-methyl-2-pyrrolidinone (NMP) to the mixture to make a slurry; 3) spraying the slurry onto a thin copper foil and drying at 120 °C for 12 h. The electrolyte was prepared by dissolving 1M LiPF₆ into a solvent containing 98 vol.% of ethylene carbonate (EC)/dimethyl carbonate (DMC) (in 1:1 volume ratio) and 2 vol% vinylene carbonate (VC) to improve the cycling stability. The polypropylene (PP) micro-porous film (Cellgard 2400) was used as the separator. The test cells were assembled in an argon-filled glove box.

The galvanostatic charge-discharge test was carried out on Neware CT-3008-S4 station with a voltage range of 0.02 V to 1.0 V (versus Li/Li⁺) at different current densities. Cyclic voltammetry (CV) test was performed using an electrochemical station (Zahner, complete CIMPS-4 system IM-6) with a scan rate of 0.005 mV/s in the same voltage range of 0.02 V-1.0 V. The electrochemical impedance spectroscopy (EIS) was measured at a half discharge state using Zahner IM-6 electrochemical work station in the frequency range of 0.1 Hz to 100 kHz at a room temperature.

The charge/discharge rate was calculated assuming the theoretical capacity of 4200 mAhg⁻¹. The Coulombic efficient was calculated by using the ratio of C_{dealloy}/C_{alloy}, where C_{dealloy} and C_{alloy} are the capacity of the anodes during Li extraction and insertion, respectively. After 100 cycles, cells were charged to 1.0 V and opened for material characterization. In order to remove SEI, electrodes were
soaked in 0.5 M HCl and rinsed with deionized water.

Figure S1 The EDX image of F-Si/C composite after being carbonized and removing NaCl reagents. NaCl was removed completely. As a result, C, O, and Si element levels in the composite appear to be higher than in Figure 2(e). The Oxygen and Carbon backgrounds around the particle are from the conducting tape consisting of Oxygen and Carbon.

Figure S2 $N_2$ sorption isotherms (77K) on F-Si/C and S-Si/C.
Figure S3 Cycling performance of bare SiNPs measured at C/2 (1C = 4200 mAg⁻¹)

Figure S4 SEM images of Si/C composite with higher weight content of SiNPs (72.98%).

Figure S5 Thermogravimetric curves of Si/C composites with different weight content of SiNPs. The tests were conducted under an air atmosphere to get thermogravimetric curves.
Figure S6 Cycling performance of Si/C composite with higher weight content of SiNPs (72.98%) measured at C/20 (1C = 4200 mAg⁻¹). The cycling performance is not as good comparing to Si/C composite with 29.49% weight content of SiNPs. This is because, with weight content of SiNPs at 72.98%, there are not enough carbon framework to cover the SiNPs, which can be seen in Figure S4(b). More SiNPs are exposed to the electrolyte, and the SEI film would form and rupture on the surface of SiNPs during the cycling, leading to the failure of the structure.