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Facile Spray Drying Route for the bath Lily-Like Graphene Sheet-Wrapped Nano-Si composite for Lithium Ion Battery Anode

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Silicon is one of the most promising anode materials on account of its acceptable cost, low alloy potential, and the highest known theoretical capacity of about 4200 mAh g^{-1} for Li storage. However, Si anodes show large volume changes upon lithiation and delithiation, which could cause its pulverization and loss of electrical contact between the active particles, resulting in low power capability and rapid capacity fading. In order to accommodate the volume change of Si during cycling, great efforts have been undertaken, such as the nanonization of Si-based materials and the preparation of various Si/C composite.

Recently, graphene as a kind of novel carbonaceous materials has attracted worldwide attention since it was first reported in 2004. Although graphene can not substitute for graphite as anode material for lithium ion batteries due to its large initial irreversible capacity loss, relatively high charge/discharge plateau and limited cycling stability, it could be an excellent substrate to accommodate active materials

Herein, we make use of commercialized Si powder and a simple spray drying method to prepare a novel bath lily-like graphene sheets-wrapped nano-Si (GS-Si) composite anode material for lithium ion batteries. In our procedure, no surfactant, no filtration and washing processes and no high vacuum condition are required. It is safe and environmentally friendly.The obtained GS-Si composite possesses an open nano/micro structure, in which nanosized Si particles are uniformly dispersed and wrapped in GS matrix. The GS not only constitutes a good conducting network, but also provides enough void spaces to accommodate the volume change of Si and prevent the aggregation of nano-Si particles during cycling.

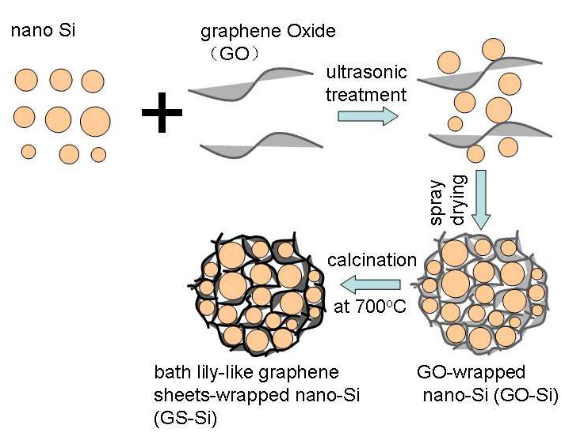

Fig. 1 Schematic of the formation process of bath lily-like graphene sheets-wrapped nano-Si (GS-Si) composite

As shown in Scheme 1, the overall preparation process of bath lily-like graphene sheets-wrapped nano-Si (GS-Si) composite involves three steps: First, nano-Si powder is sonicated with graphene oxide (GO) in deionized water. Second, the mixture is spray-dried to form graphene oxide-wrapped nano-Si (GO-Si) composite microparticles. Finally, the obtained GO-Si composite is reduced in a stream of hydrogen/argon (20:80) mixture at 700 °C for 3 h to convert to the bath lily-like GS-Si composite.

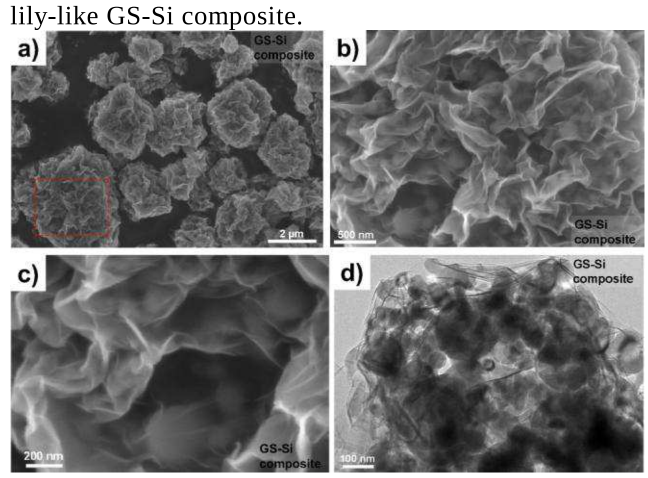

Fig. 2 FESEM images of (a, b, c) GS-Si composite and TEM image of (d) GS-Si composite.

Field emission scanning electron microscopy (FESEM) and transmission electron microscopy (TEM) images clearly display the composite morphology (Fig. 2). The special wrapping structure benefited from the stacking and folding of GO during the spray drying procedure. However, the inner structure of the particles can not be clearly observed due to the crinkled but close stacking of GO. After further thermal treatment, GO-Si composite is reduced to GS-Si composite(Fig. 2a-c). The original particle size of GO-Si composite is retained and no more agglomeration is detected. In comparison with the GO-Si composite, the GS-Si composite present a typical crinkled and rippled morphology of GS and the particle structure becomes more open, in similar to a bath lily. Moreover, it is clearly observed from Fig. 2b that the wrapped Si nano-particles are evenly dispersed in the composite particle. A deep insight into the GS-Si composite microstructure is actualized by TEM analysis. As shown in Fig. 2d, almost all the Si nano-particles are encapsulated by fish net-like GS.

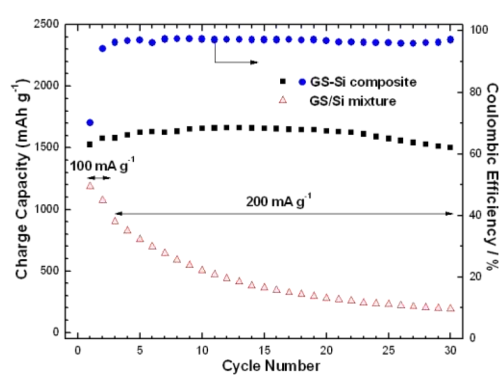

Fig. 3 Cycling performance of GS-Si composite and GS/Si mixture

In summary, a novel bath lily-like graphene sheets-wrapped nano-Si composite was successfully synthesized by a simple spray drying route. The composite based on a commercial Si product exhibits a high reversible capacity of 1525 mAh g^{-1} and superior cycling stability (Fig. 3), which could be attributed to the special open nano/micro-structure, where the advantage of a synergistic effect between highly conductive GS and active nanoparticles is fully taken. This simple but effective nano/micro assembly technology could be applicable for the large-scale production of various graphene-based composite materials with high performance for electrochemical energy storage and conversion.