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Micro-sized Si-C Composite with Interconnected Nanoscale Building Blocks as High-Performance Anodes for Lithium-ion Batteries

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Silicon has been intensively pursued as promising anode materials for next generation Li-ion batteries owing to its high theoretical capacity (>3500 mAh/g) and abundance.[1] However, Si undergoes a large volume change during the lithiation and delithiation processes, causing severe pulverization of Si particles, an unstable solid-electrolyte interphase (SEI) layer, and structural degradation of the electrode – all of which can lead to poor cycling stability.[2,3]

Numerous efforts have been made to address these issues, with the main focus on development of Si-based nanomaterials, such as nanowires, nanotubes, and nanoparticles.[4-6] Though Si-based nanomaterials offer high gravimetric capacity and long cycle lifetime, they suffer from intrinsically low tap density, leading to low volumetric capacity.[7] Furthermore, preparation of nano-sized Si either requires chemical/physical vapor deposition or involves complicated processes, leading to costly, low-yield synthesis that is difficult to scale up to practical levels.[8,9] To date, the abundance of Si has not been fully capitalized upon due to lack of a low-cost strategy for large-scale synthesis of Si anode materials with superior performance.

Micro-sized materials are favorable for practical battery applications since they often enable higher tap density than nano-sized materials and, as a result, are expected to offer higher volumetric capacity. However, the disadvantages of micro-sized Si materials as anodes are obvious. Micro-sized Si particles are more likely to undergo disintegration upon volume change during lithiation/delithiation compared with nano-sized materials, resulting in severe capacity fading.[10] Micro-sized materials also have long ion/electron transport paths that adversely affect high rate capability.[11] Because of these tradeoffs, it is desirable to develop new materials that combine the advantages of both micro-sized and nano-sized Si materials to improve the cycling performance, rate capability, and energy density of Si anodes.

We report a new, low-cost and large-scale approach to prepare a micro-sized Si-C composite composed of interconnected Si and carbon nanoscale building blocks with excellent performance as an anode material for LIBs (Figure 1a and 1b). All starting materials are commercially available and the composite can be prepared at grams scale in lab. To examine the distribution of silicon and carbon in the Si-C composite particles, energy-dispersive X-ray spectroscopy (EDS) mapping was carried out on a cross section of a single Si-C composite particle. The silicon and carbon was found to be uniformly distributed throughout the cross-section of the particle, indicating that micro-sized Si-C composite particles consist of nano-sized Si and C that are three-dimensionally interconnected at the nanoscale throughout the particles.

The Si-C composite exhibits a reversible capacity of 1459 mAh/g after 200 cycles at 1 A/g with a capacity retention of 97.8% (Figure 1c). Capacities of 1100 and 700 mAh/g can be obtained at high current densities of 6.4 A/g and 12.8 A/g, respectively (Figure 1d). The Si-C composite also has a high tap density of 0.78 g/cm^3 due to the high packing of primary particles within the micro-sized particles, which gives a high volumetric capacity of 1326 mAh/cm^3 at 400 mA/g.

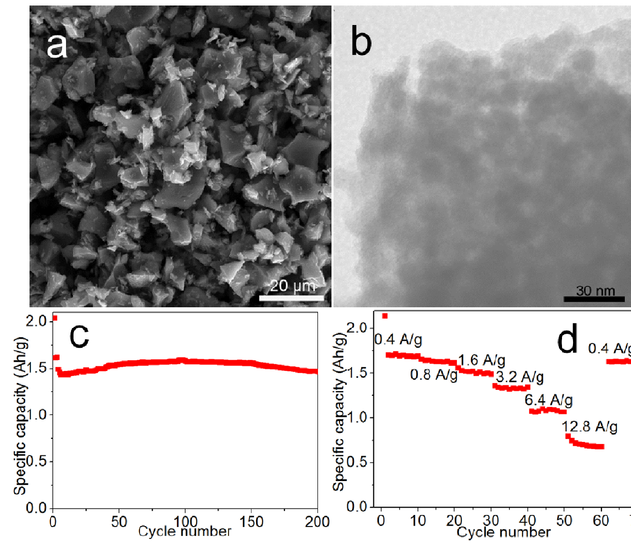

Figure 1. (a) SEM image, (b) TEM image, (c) cycling performance and (d) rate capability of the micro-sized Si-C composite.

The excellent electrochemical performance of the micro-sized Si-C composite benefits from the nanoscale size of the Si building blocks and the uniform carbon filling. The Si building blocks are small enough to avoid significant fracture during volume change, while the uniformly distributed carbon ensures high utilization of silicon even when the micro-sized particles crack upon cycling.

The low-cost and large-scale synthesis of the Si-C composite combined with its excellent performance makes it a promising anode material for practical application in LIBs.

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