



Dual-carbon-confined hydrangea-like SiO cluster for high-performance and stable lithium ion batteries

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ABSTRACT

Silicon monoxide (SiO) is regarded as a strong candidate for next-generation lithium ion battery anode materials because of its high energy density, low cost, and relatively low volume expansion compared to silicon (Si). However, the intrinsic low electrical conductivity and non-negligible volume expansion limit the practical application of SiO. Herein, dual-carbon-confined hydrangea-like SiO clusters are developed via chemical vapor deposition (CVD) growth, followed by a spray drying approach as a novel anode material for high-performance and stable lithium ion batteries. The evolution of the buffer layer along with sufficient void spaces to alleviate the volume expansion, besides the increased electrical conductivity, contributes to the improved discharge capacity of 1071 mAh g⁻¹ after 200 cycles at a high current density of 0.75 A g⁻¹ with a low expansion ratio of 9%. The distinct dual-carbon-confined hydrangea-like structure leads to synergistic improvements in battery performance, which will pave the way for promoting the commercial application of silicon-based anode materials.

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Introduction

With the newly emerging and growing power demands for advanced electrified transportation and large-scale energy storage solutions, it is important to improve the energy density of lithium ion batteries (LIBs), which are regarded as the most important energy storage systems and extensively used in our lives, through the development of advanced anode materials [1,2]. Silicon-based materials are considered strong alternative anode materials for commercial graphite having a theoretical specific capacity of 372 mAh g⁻¹ due to their high specific capacity (~4200 mAh g⁻¹ for Si, ~2400 mAh g⁻¹ for SiO, and ~1680 mAh g⁻¹ for SiO₂), natural abundance, environmental benignity, and relatively low working potential [3–6]. Nevertheless, the severe volume fluctuation (~400%) during repeated alloying/dealloying processes and the low intrinsic electrical conductivity (<10⁻³ S cm⁻¹ at 25 °C) of Si lead to pulverization of the electrode, loss of electric contact, and unstable formation of a solid-electrolyte interphase (SEI) layer, resulting in rapid capacity fading [4,7–9].

Compared to Si, silicon monoxide (SiO) exhibits relatively improved cycling stability owing to the in situ generation of

lithium silicate (Li₄SiO₄) and lithium oxide (Li₂O) during the initial discharge process, acting as a buffer against volume expansion [10]. However, non-negligible volume expansion (~200%) with low electronic conductivity still hampers the practical application of SiO anodes [11,12]. Among the diverse strategies used to improve the structural and interfacial stability of SiO, including pre-lithiation [13,14], using nano-sized porous SiO [15], and selecting optimized binders [16], the designing of SiO-based carbon composites is one of the promising routes, which not only provides a buffer layer against large volume expansion, but can also increase electrical conductivity. Because the carbon types and microscopic structures of SiO-based carbon composites significantly affect the characteristics of interfacial stability and subsequent performance of LIBs [17], the beneficial configuration of SiO and carbonaceous materials ensures electrical contact with the surrounding conducting material with a fluent passage of lithium ions and buffers the volume expansion of materials, which are important [18–20]. In this regard, graphene as a crystalline carbon material with sp² bonded carbon atoms arranged in a hexagonal 2D lattice is considered a remarkable conducting material owing to its unique 2D structure, excellent electrical conductivity, superior mechanical flexibility, good chemical stability, and high surface area [21].

Herein, we report the design and synthesis of crumpled winkle rGO (denoted as CW-rGO)-decorated SiO hydrangea clusters using SiO which is individually coated with a vertically grown carbon

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