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Article in *Advanced Materials* · June 2017

DOI: 10.1002/adma.201701494

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Antipulverization Electrode Based on Low-Carbon Triple-Shelled Superstructures for Lithium-Ion Batteries

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The realization of antipulverization electrode structures, especially using low-carbon-content anode materials, is crucial for developing high-energy and long-life lithium-ion batteries (LIBs); however, this technology remains challenging. This study shows that SnO₂ triple-shelled hollow superstructures (TSHSs) with a low carbon content (4.83%) constructed by layer-by-layer assembly of various nanostructure units can withstand a huge volume expansion of $\approx 231.8\%$ and deliver a high reversible capacity of 1099 mAh g⁻¹ even after 1450 cycles. These values represent the best comprehensive performance in SnO₂-based anodes to date. Mechanics simulations and in situ transmission electron microscopy suggest that the TSHSs enable a self-synergistic structure-preservation behavior upon lithiation/delithiation, protecting the superstructures from collapse and guaranteeing the electrode structural integrity during long-term cycling. Specifically, the outer shells during lithiation processes are fully lithiated, preventing the overlithiation and the collapse of the inner shells; in turn, in delithiation processes, the underlithiated inner shells work as robust cores to support the huge volume contraction of the outer shells; meanwhile, the middle shells with abundant pores offer sufficient space to accommodate the volume change from the outer shell during both lithiation and delithiation. This study opens a new avenue in the development of high-performance LIBs for practical energy applications.

As one of the current major energy storage technologies, lithium-ion batteries (LIBs) are widely expected to power emerging portable electronics and electric vehicles (EVs).^[1-4] To meet this demand, it is necessary to develop high-energy-density LIBs with both long cycling life and high power output. However, LIBs have long suffered from the severe pulverization of anode materials induced by huge volume changes upon Li-ion insertion/extraction, which causes irreversible capacity loss as well as poor cycling stability and rate capability.^[5-17] To solve this problem, in most cases, graphitic carbons with good conductivity and excellent mechanical properties were employed as flexible matrices for the anode materials to improve the cycling performance of batteries.^[18-29] However, the high carbon content (usually greater than 30%) often results in a capacity decrease due to the low theoretical lithium storage capacity of graphite carbons (≈ 372 mAh g⁻¹), substantially limiting their practical applications

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DOI: 10.1002/adma.201701494