



Designing Ultrasmall Carbon Nanospheres with Tailored Sizes and Textural Properties by Catalytic Miniemulsion Polymerization for Electrochemical Energy Storage

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To date, a large family of carbon-based materials has been developed for various electrochemical energy storage applications because of their low cost, stable physicochemical properties, and good conductivity. However, kinetic problems still restrict their applications in the high-rate energy storage systems arising from restricted inner-pore ion transport, particularly within micropores ($d < 2$ nm) and small mesopores ($2 < d < 5$ nm). Herein, we demonstrate the efficient design of ultrasmall porous carbon nanospheres with tailored sizes (5–40 nm in diameter) and optimized intra-inter-sphere textural properties for high-rate high-energy lithium-sulfur batteries and supercapacitors. The porous carbon nanospheres are synthesized via a facile, convenient, and scalable catalytic miniemulsion polymerization strategy followed by simple KOH activation. Specially, the intra-sphere micropore dominated carbon nanospheres prepared with single-step activation display the superior performances as conducting reservoirs for sulfur in lithium-sulfur batteries to efficiently alleviate the polysulfide dissolution problems. Furthermore, an optimum dual-step activation facilitates the expansion of intra-sphere micropores into an enlarged hierarchical intra-sphere micro-/meso-pore, rendering ultrasmall carbon nanospheres as high-rate electrode materials for electrical double-layer capacitors for lithium-ion batteries with significantly enhanced ion transports. Moreover, a further increase in rate performance is seen upon a decrease in sphere size from 40 to 5 nm in these applications, confirming the pronounced size effects due to the shortened ion diffusion distances. With their well-defined ultrasmall sizes and controllable intra-/inter-sphere pore structures, we have been able to reveal the important effects of these structural parameters on the electrochemical performances of this class of ultrasmall carbon nanomaterials. Such relationships provide valuable guidelines for the rational design of carbon nanomaterials for high-rate high-density energy storage applications.