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RAPID COMMUNICATION

Electrodes engineering of high power, long life and excellent cycling stability for rechargeable lithium batteries

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Long lifespan

Abstract
Although widely used, the current Li-ion battery technology still suffers from a lack of suitable electrodes with enhanced energy and power density, cycling stability, energy efficiency and cycling life. So far no reliable methods can satisfy all these requirements. There is therefore a need to seek novel electrodes that would combine all the advanced performances and satisfy the increasing demands for energy storage worldwide. Herein, we demonstrate a large-scale bottom-up assembly route for porous bubble hybrid electrodes with excellent electrochemical properties by creating composites based on nanomaterials uniformly dispersed on the outer and inner surfaces of a porous creased carbon bubble host, which serves to hold them tightly by the pores and creases during battery operation and sandwiches them between rapid ion and electron transport pathways. Such integrated electrodes exhibit ultrahigh specific capacity and excellent cycling stability at various rates. Long lifespan of 1000 cycles in half cells retaining more than 90% of their reversible capacity and large rates up to 327*C* for ensample electrodes are achieved with batteries' high energy density and supercapacitors' impressive power density (where 1*C* rate represents a 1-h complete charge or discharge).
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Introduction

The major challenge towards the advancement of current Li-ion battery technology includes inadequate capacity relative to the theoretical one, insufficient energy and power density for intended applications, and limited cycling

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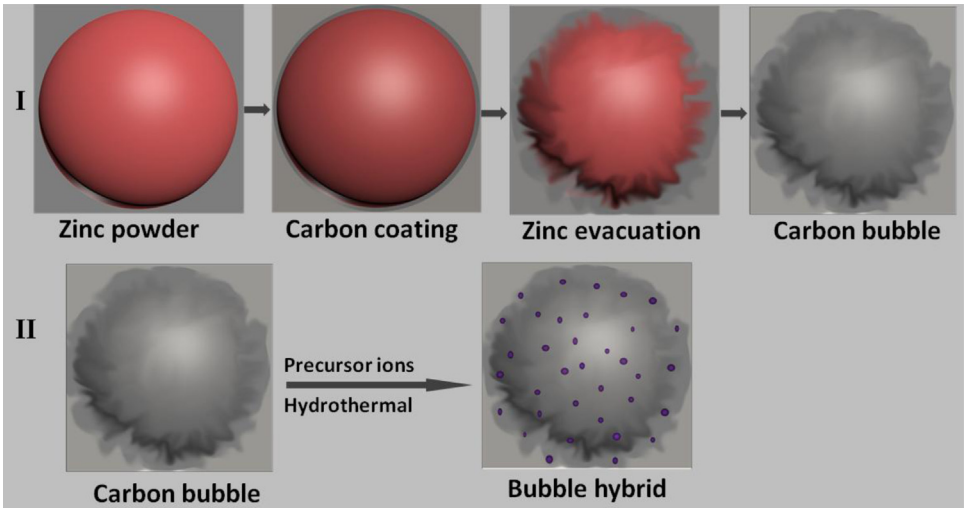

Scheme 1 Schematic illustration for fabricating bubble hybrids: I carbon bubble evolution and II formation process of the bubble hybrid.

life and unsatisfying efficiency [1,2]. These disadvantages should be attributed to relevant electrodes suffering from kinetic problems linked to the solid-state diffusion of Li and electron transport, huge specific-volume changes and the quality of interfaces [3–7]. Thus, the search for novel electrodes to alleviate the above problems has been an urgent task in building next generation lithium-ion batteries, so as to meet the ever-growing requirements for high power and long life and excellent cycling stability. Hybrid nanostructures are a promising class of materials to provide large active surface area, fast electron transport and ion diffusion. Specifically, some elegant strategies based on hybrid nanostructures, for instance, core–shell or yolk–shell structures [8–11], coaxially branched architectures [5,12], and 'self-wound' nanomembranes [13], have been already proposed to enhance electrodes performance. However, the cyclability and cycling life for metal oxides nanostructures have never been satisfactory (less than 300 cycles) relative to the commercial standard (>500 cycles). As indicated above, it is difficult to concurrently obtain electrodes with high power, long life and cycling stability based solely on the design of active materials itself disregarding electrode structures.

Herein, we present a class of novel porous bubble hybrid electrodes characterized by (i) nanomaterials uniformly dispersed on the outer and inner surfaces of porous, creased carbon bubble hosts that hold the active material tightly by the pores and creases during battery operation, (ii) ion and electron conductive carbon bubbles that supply rapid ion and electron transport pathways, (iii) interconnected electrolyte-filled porous networks that benefit rapid ion transport and electrode durability, (iv) highly dispersed nanomaterials that avoid particulates aggregation upon cycling, and (v) nearly all active material concentrating on the conductive surfaces that would improve the rate capabilities. The bubble hybrid electrodes would restrain (i) morphology and microstructure changes (by pores and creases immobilizing the active materials) that may result in undesirable re-distribution of active materials, and (ii) volume change of active materials due to the very small size (quantum dots) that may induce stress and strain in the electrodes, which eventually result in pulverization of active materials. Two examples representative of alloying type and conversion type active materials were chosen to verify the efficiency of the designed electrodes in obtaining high power, long life, and excellent cycling stability in both anodes (tin oxide and iron oxide) and cathodes (iron oxide) for half cells of lithium ion batteries [14–16].

The porous bubble hybrid electrodes were fabricated as illustrated in Scheme 1 (for details, see Supplementary information and methods). First, carbon bubbles with pores and creases were synthesized by a zinc powder templated physical vapor deposition method (Scheme 1I and Figure S1(a)). Then, carbon bubbles dispersed in water/ethanol solvents readily absorbed the water molecules, which would facilitate the hydrolysis of precursor ions (e.g. $Fe^{3+}$and Sn^{4+}) in the hydrothermal process. To get over a smaller energy barrier, these precursor ions would readily assemble into small seeds or crystal nuclei, which have comparable size as that of nanopores in the carbon bubbles and are apt to embed in them. As the seeds evolve into nanocrystals, they will be immobilized by the pores and creases. Eventually, a unique hybrid structure of oxide quantum dots and porous carbon bubbles was attained (Scheme 1 II). As discussed above, the bubble hybrid electrodes promise superior electrochemical properties in comparison with those of other common electrodes.

Material and methods

Carbon bubbles synthesis

150 mg zinc micropowder dispersed in 20 mL absolute ethanol is uniformly coated on a rectangular alumina bar at 55 °C. The bar on one side with 10 mg fullerene powder (Alfa Aesar, 99%), the rest part with uniform thick zinc film (Fig. S1(a)), is set in a horizontal tube of fullerene located in the highest temperature zone. The tube is first pumped below 300 Pa before being heated from room temperature to 350 °C. After holding for 30 min at 350 °C, the tube is then heated to 800 °C and kept for 5 min; thence it naturally cools to room

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