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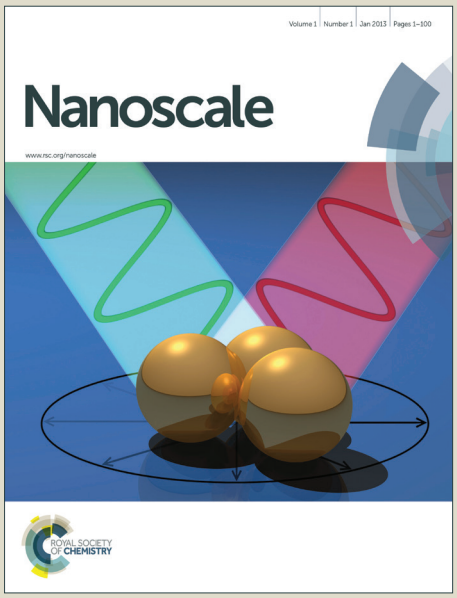

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Synthesis of functionalized 3D porous graphene using both ionic liquid and SiO_2 spheres as "spacers" for high-performance supercapacitors application

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Abstract

In this paper, a high-capacity supercapacitor material based on functionalized three-dimensional (3D) porous graphene was fabricated by low temperature hydrothermal treatment of graphene oxide (GO) using both ionic liquid (IL) and SiO_2 spheres as "spacers". In the synthesis, the introduction of dual "spacers" effectively enlarged the interspace between graphene sheets and suppressed their re-stacking. Besides, the IL also acted as structure-directing agent played a crucial role in inducing the formation of unique 3D architecture. Consequently, fast electron/ion transport channels were successfully constructed and numerous oxygen-containing groups on graphene sheets were effectively reserved, which had unique advantages in decreasing ion diffusion resistance and providing additional pseudocapacitance. As expected, the obtained material exhibited superior specific capacitance and rate capability compared to singly "spacer" designed electrodes, and simultaneously maintained excellent cycling stability. Specifically, there were nearly no loss of its initial capacitance after 3000 cycles. In addition, we further assembled a symmetric two-electrode device using the material which showed outstanding flexibility and low equivalent series resistance (ESR). More importantly, it was capable of yielding a maximum power density of about 13.3 kW kg^{-1} with an energy density of about 7.0 W h kg^{-1} at a voltage of 1.0 V in 1 M H_2SO_4 electrolyte. All these impressive results demonstrate that the material obtained by this approach is greatly promising for high-performance supercapacitors application.

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1. Introduction

Supercapacitors with high power density, excellent charging/discharging rate capability, and long life-cycles have become one of the most intense research focuses in the electrical energy storage field.[1–4] They commonly store energy using either ion adsorption (electrochemical double layer capacitors, EDLCs) or fast surface redox reactions (pseudocapacitors).[3,5,6] It is well accepted that the textural properties of electrode materials play a dominant role in the development of supercapacitors. Naturally, it would be very interesting to develop a hybrid type of electrode material with unique architecture, where EDLCs and pseudocapacitors can concurrently combine to contribute to the high power property and better energy storage performances. In this point of view, recent efforts have been focused on the preparation of high-capacity electrode materials, which may be achieved both by providing desired electro-active species and by creating open porous channels with enhanced specific surface area to improve the accessibility of the ions from the electrolyte to the active regions of electrode materials.

As a promising electrode material, graphene, an atom-thick two-dimensional nanostructure, is receiving growing attention due to their excellent electronic conductivity, good electrochemical stability, high surface area and flexibility.[4,7,8] Various morphologies of graphene or graphene-based composites have been developed as electrode materials for supercapacitors.[9–15] Nevertheless, easily and efficiently reducing of graphene oxide (GO) to reduced graphene oxide (rGO) is still a key topic in this research field. Among all the reduction strategies, thermal exfoliation of GO is conceived to be simple and environmentally friendly in which no hazardous reductant is used. But this process usually requires a rapid heating (>2000 °C min^{-1}) up to high temperature, which means large energy consumption and critical treatment conditions.[16] Recently, hydrothermal treatment of GO has attracted more and more attention because of its outstanding advantages, such as high yield, simple manipulation, easy control, environmentally friendly and so on.[17–19] Particularly, low temperature hydrothermal treatment can remain desired oxygen-containing groups on the surface of graphene which not only enhance the surface wettability of graphene electrodes but also significantly increase the specific capacitance by the