High Efficiency Energy Storage of Graphene-Based Composites

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Energy storage is vital to meet the challenge of global warming and finite fossil-fuel supplies in modern society [1]. Graphene, a unique two-dimensional carbon material, is predicted to be an excellent electrode material candidate for energy conversion/storage in supercapacitors and lithium ion batteries (LIBs) because of its high specific surface area, good chemical stability, excellent electrical and thermal conductivity as well as remarkably high mechanical strength and Young's modulus [2-4].

Controllable synthesis of graphene sheets (GSs) in a large scale is the prerequisite and essentially important for the energy storage applications of graphene. We proposed a simple and effective strategy to tune the number of graphene layers by selecting suitable starting graphite using a chemical exfoliation method [5], developed a mild oxidation and exfoliation method to prepare large-area graphene oxide with a size up to 200 micrometers and realized the size-controlled synthesis of graphene oxide by simply tuning the content of C-O in graphite oxide [6], proposed a hydrogen arc discharge exfoliation method for the synthesis of GSs with excellent electrical conductivity from graphite oxide [7], and developed a rapid and nondestructive low-temperature reduction method to effectively improve the electrical conductivity of graphene oxide by using HI acid [8].

In order to fully utilize the advantages of GSs for energy storage, we proposed the use of GSs/metal oxide (or conducting polymer) as electrode materials for high performance supercapacitors and LIBs. Based on this idea, we designed and synthesized a series of graphene/metal oxide nanoparticles composites by combining sol-gel and low-temperature annealing processes, including GSs/hydrous RuO₂ composites for high energy supercapacitors [9], GSs/Co₃O₄ and GSs/Fe₃O₄ composites for high energy LIBs [10, 11], and GSs/TiO₂ and GSs/Li₄Ti₅O₁₂ composites for high power LIBs [12]. In order to achieve high energy and power densities, we also developed a high-voltage asymmetric electrochemical capacitor based on graphene as negative electrode and a GSs/MnO₂ nanowire composite as positive electrode in a neutral aqueous Na₂SO₄ solution as electrolyte [13]. Moreover, by incorporating with polyaniline, we fabricated GSs/polyaniline composite paper via *in situ* anodic electropolymerization for high performance flexible supercapacitor electrodes [14].

All the above composites show a greatly improved capacity, cycling stability and rate capability compared to solo graphene and metal oxide, demonstrating the positive synergistic effect of GSs and metal oxide on the improvements of electrochemical performance. We believe that the performance of GSs-based composite electrodes can be further improved by optimizing the composition and structure of GSs and particles, and the architecture and synthesis process of composites, to meet the future requirements for high energy and high power energy storage systems.

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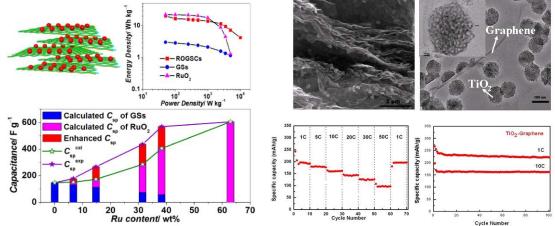
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Figures



 $GSs/hydrous RuO_2$ composites for high energy supercapacitors (left panel) [9] and GSs/TiO_2 composites for high power LIBs (right panel) [12].