



Oxygen-containing Functional Groups Enhancing Electrochemical Performance of Porous Reduced Graphene Oxide Cathode in Lithium Ion Batteries



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ABSTRACT

Exploring high performance and environment-friendly electrode materials is highly desirable for the sustainable Li-ion batteries (LIBs) system. In this study, a facile approach of the modified Hummers' method combining with special thermal reduction was proposed to synthesize nanostructured reduced graphene oxide (RGO) with abundant oxygen-containing functional groups. The resultant RGO showed high specific capacity and excellent cyclability as cathode materials for LIBs. The specific capacity of about 220 mAh g⁻¹ at a current density of 50 mA g⁻¹ was achieved after 100 cycles. More importantly, it was demonstrated that the capacity increased with the increase of the amount of oxygen functional groups, highlighting the significant effects of oxygen-containing functional groups of RGO on high lithium storage performance.

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1. Introduction

With the decrease of the use of fossil fuels as well as the increasing demand for clean energy in the twenty-first century, Lithium ion batteries (LIBs), due to their characteristics of high-energy density, high working potentials, and long cycle life, etc [1,2], have been proved to be one of the most advanced battery technology [3]. The current LIB technology highly depends on the use of lithium transition metal oxides or phosphates (e.g., LiCoO₂, LiNi_xMn_yCo_{1-x-y}O₂ or LiFePO₄) as cathodes with the high redox potentials [4], but these cathode materials show very limited energy capacities. Further, these are not environmentally benign, and the mineral resources of Co and Ni are scarce, resulting in high cost LIBs with limited performance. Therefore, the search for new energetic electrode materials, especially cathode materials, for LIBs has been highlighted in battery chemistry [5].

Two dimensional (2D) graphene materials have been one of the most promising miracle materials originating from its unique single-atom-thick sheet of carbon atoms arrayed in a honeycomb pattern [6,7]. Benefiting from remarkable advantages of large specific surface area (2630 m² g⁻¹), excellent electrical conductivity (10⁶ S cm⁻¹), high charge mobility (200000 cm² V⁻¹ s⁻¹), good chemical and environmental stability [6–8], graphene materials have been demonstrated to be a new class of outstanding candidate for electrochemical power source fields. As a result, they have been widely explored for applications in energy-related areas, including solar cells [9,10], LIBs [11–13], supercapacitors [14,15], etc. More importantly, graphene materials have shown great promises to improve the performance of the aforementioned energy systems, which satisfies the urgent demand for renewable energy production and efficient energy storage in modern society [14]. It is well known that graphene materials has been widely researched as LIB anodes [16–18], and show much higher reversible capacity than the commercial graphite, as previously demonstrated by our group [17,19], and various graphene-based composite materials presented enhanced battery performance [20–23]. Recently, differing from the anode side, it was reported

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that reduced graphene oxide (RGO) with oxygen-containing functional surface groups can work as cathode materials of LIBs [24]. During charge/discharge processes, as the redox centers, the oxygen-containing functional surface groups (such as $>C-O$ and $-COOH$) of RGO can rapidly and reversibly capture lithium ions through surface adsorption and/or surface redox reaction. Thus, functionalized graphene exhibit higher energy density with long-lasting cyclability in comparison with the current cathode materials [25,26]. For example, Bor Z. Jang's group proposed a novel LIB using nanostructured graphene as both the anode and the cathode [25]. The obvious difference is that the cathode was functionalized graphene. They obtained an energy density of 160 Wh kg^{-1} and a power density of 100 kW kg^{-1} . In the recent reports, it was indicated that the functionalized graphene with more oxygen-containing groups can deliver high performance as LIB cathodes [24,27]. However, the aforementioned functionalized graphene cathodes with enhanced performance are only based on chemical reduction with tedious and low yield, which shows some difficulties in scale-up, and detailed reactive mechanisms have not been discussed.

In this report, we developed a facile approach of special thermal reduction to design partial reduced graphene oxide (RGO). It is worth noting that during the process of special thermal reduction, the controllable air gas was specially introduced to control oxygen-containing functional groups onto RGO. The as-prepared partial RGO as cathode materials for LIBs are expected to show a superior cycle stability and higher energy performance compared with previous reports [24,27]. It was demonstrated that oxygen-containing functional groups on RGO is the significant key as reaction center with lithium ions. More importantly, this approach shows some promising possibilities for mass production of the proposed cathode material.

2. Experimental

2.1. Materials

Natural graphite powder (purity $\geq 99.95\%$) was supplied by Aladdin Chemistry Co., Ltd. (Shanghai, China). Concentrated sulfuric acid (95%~98%), fuming nitric acid (65~68%), KMnO_4 (99%), NaNO_3 (99%) and H_2O_2 (30%) were used as received from Tianjin Guangfu Chemical Research Institute (China).

2.2. Synthesis of graphite oxide (GO)

Graphite oxide (GO) was initially fabricated by modified Hummers method, as previously reported by our group [19]. The natural graphite (1 g) and NaNO_3 (0.75 g) were uniformly mixed, then 34 mL of H_2SO_4 (98%) was added into the mixture and stirred for 2 h in an ice bath, meanwhile, KMnO_4 (4.5 g) was slowly added to the dispersion. After the mixture was stirred for 5 days at room temperature, 100 mL of H_2SO_4 (5%) was added to the mixture and stirred for another 2 h. Then 5 mL of H_2O_2 (35%) were slowly added to the solution until there was no gas bubbles produced. The resultant graphite oxide was then repeatedly washed with HNO_3 (10%) aqueous solution. The resulting GO was then thoroughly centrifugated with deionized (DI) water to obtain GO suspension. The GO suspension was finally dried at room temperature for 24 h to obtain GO.

2.3. Synthesis of reduced graphene oxide (RGO)

The as-prepared GO was ground into fine powder. After the tube furnace was heated up to 950°C at a rate of $10^\circ\text{C min}^{-1}$, the fine powder was quickly put into the center location, and was thermally

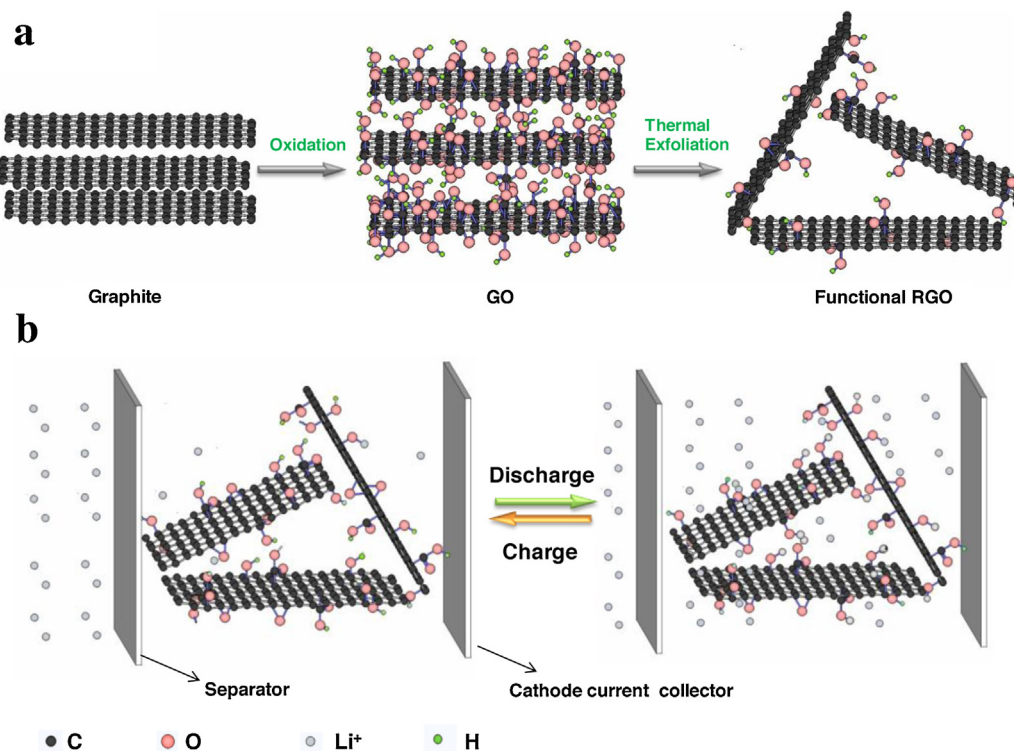


Fig. 1. (a) Schematic illustration of the oxidation mechanism and pore formation and reduction mechanism; (b) Schematic illustration for the lithium-storage mechanism of the reduced graphene oxide (RGO).

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