



# Nitrogen-containing carbon/graphene composite nanosheets with excellent lithium storage performances

Honghu Wang, Hongrui Peng, Guicun Li<sup>\*</sup>, Kezheng Chen<sup>\*</sup>

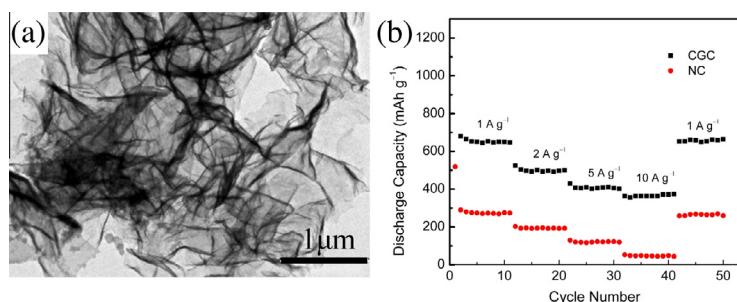
Laboratory of Functional and Biological Nanomaterials, College of Materials Science and Engineering, Qingdao University of Science and Technology, Qingdao 266042, China

## HIGHLIGHTS

- Carbon/graphene composite (CGC) nanosheets are obtained by a facile method.
- The CGC nanosheets consist of nitrogen-containing graphitized carbon and graphene.
- The CGC nanosheets exhibit excellent lithium storage performances.

## GRAPHICAL ABSTRACT

Nitrogen-containing graphitized carbon/graphene composite nanosheets exhibit high specific capacity, excellent rate performance and outstanding cycling stability.



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## ABSTRACT

Nitrogen-containing carbon/graphene composite (CGC) nanosheets have been synthesized by the in situ chemical polymerization of pyrrole on both surfaces of graphene oxide combined with high temperature carbonization. The CGC nanosheets show high specific surface area (106.22 m<sup>2</sup> g<sup>-1</sup>), large pore volume (0.382 m<sup>3</sup> g<sup>-1</sup>), and high nitrogen content (7.86 at.%). The nitrogen-containing carbon coating with low graphitization degree on both sides of graphene can prevent effectively from the aggregation of graphene oxide during its thermal reduction process. When evaluated for lithium storage capacity, the CGC nanosheets exhibit enhanced electrochemical performances in comparison with polypyrrole derived nitrogen-containing carbon (NC) nanoparticles, including high reversible capacity (651.5 mAh g<sup>-1</sup> at 1 A g<sup>-1</sup>), excellent rate capability (363.7 mAh g<sup>-1</sup> at 10 A g<sup>-1</sup>), and good cycling stability (100% capacity retention after 100 cycles at 1 A g<sup>-1</sup>). The high lithium storage performances of the CGC electrodes can be attributed to the high electronic conductivity, abundant lithium adsorption sites, and short diffusion distance of lithium ions arising from the CGC nanosheets with nitrogen-containing carbon on both sides of graphene.

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## 1. Introduction

During the past decades, lithium-ion batteries (LIBs) have gained commercial success in portable devices, and have also been

<sup>\*</sup> Corresponding authors. Tel./fax: +86 532 84022814.

E-mail addresses: [guicunli@qust.edu.cn](mailto:guicunli@qust.edu.cn) (G. Li), [kchen@qust.edu.cn](mailto:kchen@qust.edu.cn) (K. Chen).

regarded as one of the most promising candidates for applications in electric and hybrid vehicles [1–3]. However, the rate performances of the current LIBs cannot satisfy the increasing demands for high energy density, high rate capability, and low cost [4–6]. Graphite with a theoretical capacity of 372 mAh g<sup>-1</sup> has been widely used as a commercial anode material for LIBs, although its capacity and rate performances need to be further improved

[7,8]. Compared to the conventional graphite anodes, carbon nano-materials including carbon nanotubes [9], carbon fibers [10,11], and hollow carbon nanospheres [12,13], have exhibited large the lithium-ion storage capacities due to their high graphitization degree and specific surface area, but their electrochemical properties are not satisfactory for high power LIBs [14,15]. Porous carbon has also been researched as an alternative anode material due to its significant superiority over graphite such as short lithium ion diffusion paths and large electrode-electrolyte interfaces [16,17]. The incorporation of heteroatomic dopants including nitrogen, sulfur, boron, and phosphorous can modify the electronic and crystalline structures of the carbon host [18–21], which have been widely used as electrode materials in supercapacitors and LIBs [22]. Among them, nitrogen dopant is of interest because of the strong interaction between nitrogen-containing carbon and lithium, and more active sites for favourable lithium ion insertion [23]. However, the low graphitization degree of porous carbon limits its electronic conductivity, resulting into the poor rate capability.

In recent years, graphene with a maximum theoretical capacity of  $740 \text{ mAh g}^{-1}$  on the basis of double-layer adsorption configuration [6], has been expected to be good candidate materials due to their ultra-large specific surface area, excellent electrical conductivity, high lithium ion storage capacity, and short lithium ion diffusion distance [24–26]. Lian and co-workers have reported that graphene prepared by thermal exfoliation method exhibits high capacity of  $1264 \text{ mAh g}^{-1}$  at a current density  $0.1 \text{ A g}^{-1}$ , but shows a poor cycling stability because of the aggregation of graphene due to strong  $\pi$ - $\pi$  interactions [27,28]. Coating a thin carbon layer onto graphene to form carbon/graphene composite nanosheets seems to be an effect way to avoid the aggregation of graphene and thus improve the electrochemical energy storage performances due to its high specific surface area and short charge diffusion distance [29–31]. For examples, Wan and co-workers have reported ionothermal synthesis of porous carbon hybridized with graphene using glucose as carbon precursor as superior anode material for LIBs [30]. Li and co-workers have developed a  $\text{H}_2\text{O}$  activation route to synthesize sandwich-type microporous carbon nanosheets using poly(benzoxazine-co-resol) as carbon precursor, which exhibit enhanced double-layer capacitance performances

[31]. Recently, nitrogen-containing carbon synthesized by the direct carbonization of nitrogen-containing polymer precursors such as polyaniline and polypyrrole, have received considerable interest because the intrinsic nitrogen-containing functional groups can facilitate the incorporation of nitrogen active sites into carbon matrix [32]. The aniline or pyrrole monomer can be easily polymerized on both surfaces of reduced graphene oxide [33], which inspires us to find a new route to fabricate nitrogen-containing carbon/graphene composite nanosheets. Herein, we report the synthesis of nitrogen-containing carbon/graphene composite (CGC) nanosheets via a facile in situ chemical polymerization of pyrrole on both surfaces of graphene oxide and subsequent high temperature carbonization, which exhibit excellent electrochemical performances for LIBs, such as high reversible capacity, excellent high rate capability, and good cycling stability.

## 2. Experimental

### 2.1. Synthesis of the CGC nanosheets

Graphene oxide (GO) was synthesized from natural graphite flakes by a modified Hummer method [33,34]. The following procedure was used for in situ chemical polymerization of pyrrole (Py) onto GO surface to prepare the polypyrrole (PPy)/GO composite nanosheets. In a typical synthesis, pyrrole (4 mL) and GO (200 mg) at the weight ratio of 20:1 were dispersed in 900 mL of  $0.1 \text{ mol L}^{-1}$  HCl aqueous solution by ultrasonication for 20 min. Then 100 mL of HCl ( $0.1 \text{ mol L}^{-1}$ ) solution containing ammonium peroxydisulfate (13.6 g) was rapidly added to the above mixture suspension solution at  $0$ – $5 \text{ }^\circ\text{C}$ . The polymerization reaction was carried out for 12 h without any disturbance. Then the black precipitate was filtered off, washed with ammonium hydroxide, deionized water, and ethanol several times, and then dried at  $80 \text{ }^\circ\text{C}$  for 24 h. The pure PPy nanoparticles were synthesized chemically via the similar procedure. The obtained PPy/GO composite nanosheets were heated at  $900 \text{ }^\circ\text{C}$  for 2 h under a nitrogen atmosphere at a heating rate of  $5 \text{ }^\circ\text{C min}^{-1}$  to carbonize the PPy and reduce GO to graphene. For comparison, the nitrogen-containing carbon (NC) nanoparticles were obtained under the same condition using PPy nanoparticles as precursor.

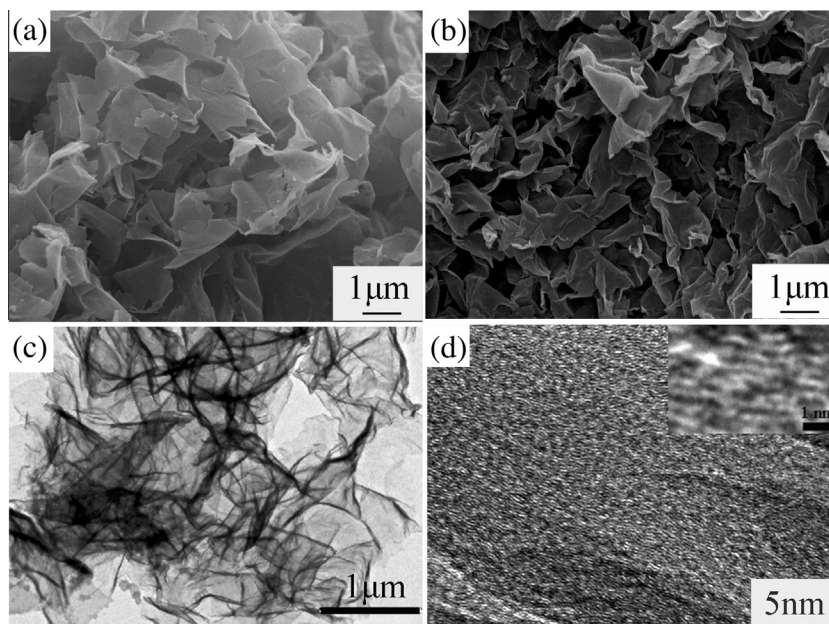


Fig. 1. (a) SEM image of the PPy/GO composite nanosheets, (b) SEM, (c) TEM, and (d) HRTEM images of CGC nanosheets.

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