



Gamma-ray irradiation-induced reduction and self-assembly of graphene oxide into three-dimensional graphene aerogel



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ABSTRACT

Three-dimensional (3D) graphene aerogel (GA) with honeycomb-like porous structure and high C/O ratio was successfully prepared from graphene oxide (GO) dispersions in isopropanol/water solution by simple γ -ray irradiation and freeze-drying processes. Under irradiation, GO sheets suspending in the mixture were reduced and self-assembled mainly due to the restoration of π - π conjugated structure. The microstructure of GA was observed under different magnifications and the pores are quite uniform with pore size around 10 μm . The reduction effect of GO was confirmed comprehensively by different characterizations. Briefly, γ -ray irradiation is an effective and green method for the reduction and self-assembly of GO sheets.

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1. Introduction

3D graphene/GO architectures with tunable hierarchical morphology and only few-layer stacking porous structure are of great significance considering their low density, high specific surface area, excellent physical and chemical properties [1]. Graphene aerogels have attracted extensive attention in many research fields such as energy storage, adsorption, catalysts supports and sensors [2–5]. Additionally, there are various approaches to integrate individual graphene sheets into advanced multifunctional structures. General approaches include hydrothermal method, chemical reduction, Langmuir-Blodgett assembly, molecular templates, direct chemical vapor deposition, and so on [6–9]. All these methods show their own advantages.

Radiation processing technology, especially γ -ray irradiation, has been regarded as an efficient and environment-friendly method in inducing chemical reactions at any temperature without catalyst [10]. According to the radiolysis of water, water molecules can be decomposed into both oxidative species ($\cdot\text{OH}$) and reductive species ($\text{H}\cdot$ and e_{aq}^-). If the oxidative radicals in water were eliminated by radical scavenger like alcohols and convert into reductive radicals, the resulting reductive radicals can reduce oxidative precursors into reduced species [11,12]. As Chen et al. reported, γ -ray irradiation has been utilized to prepare highly

hydrogenated graphene as a high-performance anode material [13]. It has also been employed to synthesize functionalized graphene in ethanediamine/water or alcohol/water system [14,15]. However, there has been no study utilizing γ -ray irradiation to synthesize GA until now.

Herein, we report a simple method to prepare graphene hydrogel from GO aqueous solution via γ -ray irradiation-induced reduction and self-assembly. After freeze-drying process, the resulting graphene aerogel was investigated in detail.

2. Experimental methods

GO aqueous solution was prepared according to a modified Hummers' method and used directly without further treatment [3]. The whole synthesis procedure of GA was shown in Fig. 1. Firstly, 5 mL GO solution about 4 mg/mL was mechanically stirring and 5 mL isopropanol radical scavenger was added to make a homogenous mixture. The final GO concentration was about 2 mg/mL. Secondly, the mixture went through deoxygenation by bubbling nitrogen into the mixture for 10 min. Then the deoxygenated mixture was sealed in the reaction vial and irradiated for a pre-set absorbed dose (200 kGy) in the ^{60}Co γ -ray radiation source at room temperature. After irradiation, the as-formed graphene hydrogel was washed by ultrapure water to remove unreacted isopropanol molecules. Then the final graphene aerogel was obtained after the freeze-drying process.

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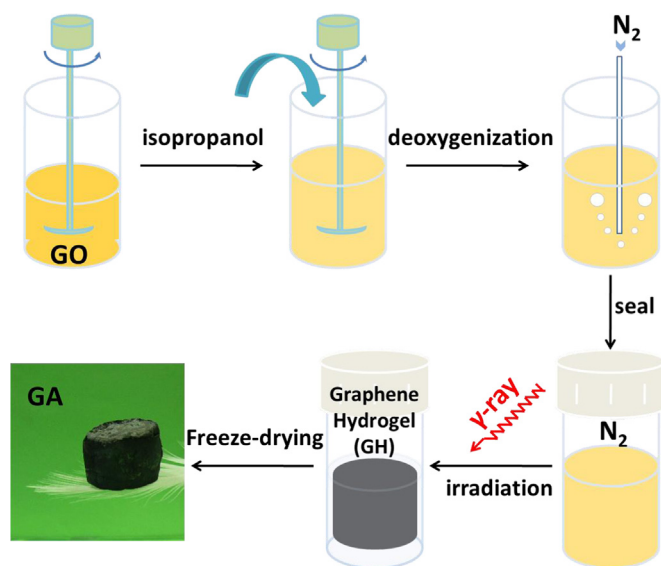


Fig. 1. Schematic illustration of the preparation process of graphene aerogel (GA).

The as-prepared GA was characterized by Scanning electron microscopy (SEM), Fourier transform infrared spectra (FT-IR), X-ray diffraction (XRD), Raman spectroscopy, thermogravimeter (TGA) and X-ray photoelectron spectroscopy (XPS).

3. Results and discussion

The microstructure of GA was investigated by SEM. For the surface of GA in Fig. 2(a), it appears to be quite flat and smooth apart from some wrinkles, of which the contact angle is about 118°

showing its hydrophobic nature. As to the cross-section of GA in Fig. 2(b)–(d), it exhibits that GA has a 3D interconnected porous structure consisted of stacking reduced GO(rGO) sheets. The macropores are uniform with honeycomb-like structures. The average pore size is around $10\ \mu\text{m}$ and the average density of GA is about $3.82\ \text{mg cm}^{-3}$ calculated by simply dividing the weight by the volume. Moreover, the contact angle of the cross-section of GA is around 141° closing to the critical value of superhydrophobicity, resulting from the intrinsic hydrophobicity of rGO sheets and the porous geometric structure. The high porosity and hydrophobicity are of great significance for the practical application of GA.

FT-IR was utilized to investigate the evolution of functional groups from GO to GA, as shown in Fig. 3(a). The characteristic peaks of GO appear for C=O stretching vibrations at $1725\ \text{cm}^{-1}$, O–H bending vibrations at $1410\ \text{cm}^{-1}$, and C–O stretching vibrations at $1220\ \text{cm}^{-1}$, C–OH at $1170\ \text{cm}^{-1}$ and alkoxy groups at $1051\ \text{cm}^{-1}$ [16]. Moreover, the peak at $1620\ \text{cm}^{-1}$ belongs to C=C stretching or skeletal vibrations from unoxidized graphitic domains. After irradiation, the peaks of oxygenous functional groups decrease dramatically except for C–OH at $1170\ \text{cm}^{-1}$. Additionally, some new peaks arise, such as $1523\ \text{cm}^{-1}$ for the bending modes of H bonded to an olefinic carbon, $1080\ \text{cm}^{-1}$ and $980\ \text{cm}^{-1}$ for the bending modes of H bonded to an aromatic carbon, $1295\ \text{cm}^{-1}$ for the asymmetric bending vibrations of alkyl groups. These results confirm the efficient reduction of GO in isopropanol/water system during the gamma-ray irradiation.

XRD patterns of graphite, GO and GA are presented in Fig. 3(b). For GO, it has a wide diffraction peak around 10° with a d -spacing about $8.84\ \text{\AA}$ according to Bragg equation. While after reduction by γ -ray irradiation, there shows a quite broad peak around 23° ($3.86\ \text{\AA}$) which is very close to the sharp diffraction peak of graphite ($3.42\ \text{\AA}$, $2\theta=26^\circ$), suggesting the reduction of GO. Moreover, peak broadening in GA is attributed to poor ordering during the stacking of GO sheets [17].

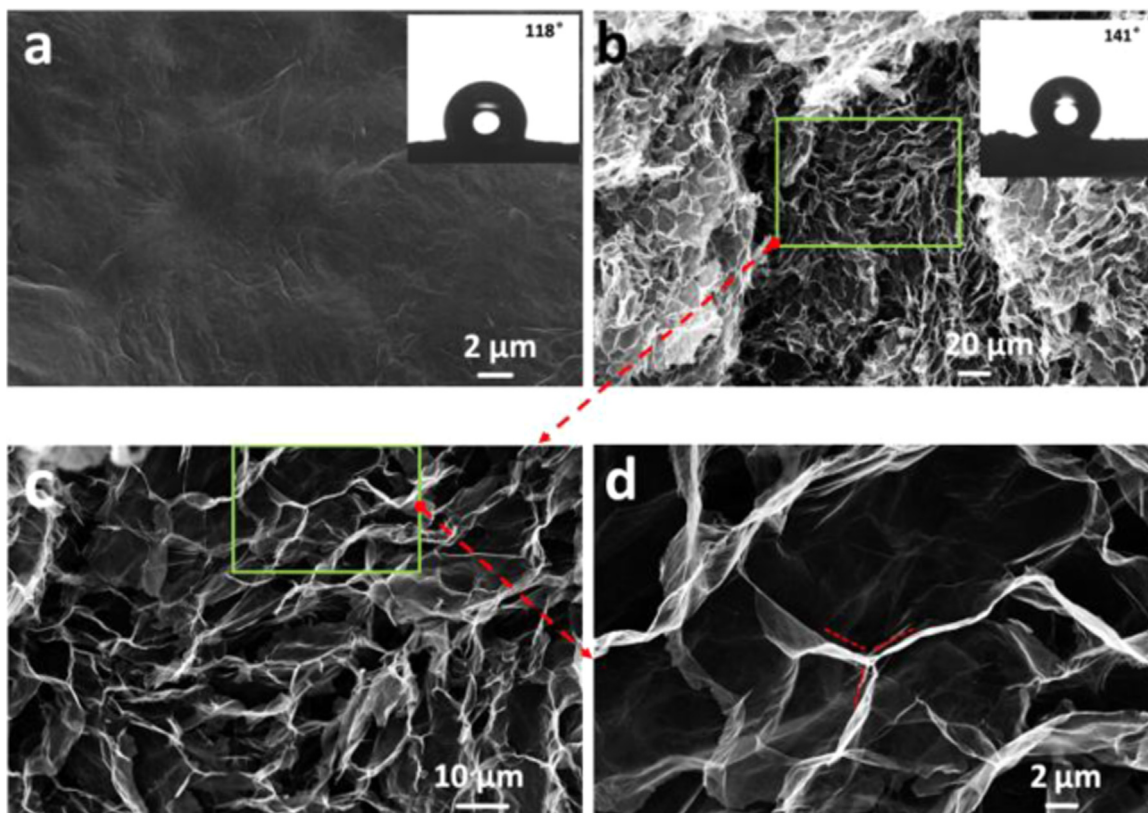


Fig. 2. (a) SEM image of the surface of GA. (b)–(d) SEM images of the cross section of GA with different magnifications.

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