



# A simple and fast microwave assisted approach for the reduction of graphene oxide

Qinglong Yan, Qing Liu, Jieqiang Wang\*

*School of Materials Science and Engineering, University of Jinan, Jinan 250022, PR China*

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## Abstract

Herein, a simple and fast method for chemical reduction of graphene oxides (GO) was carried out, the most common monosaccharide (glucose) here served as a green, non-toxic and high-effective reduction reagent. After reacted in an alkaline microwave-assisted environment for several hours, the water-soluble glucose reduced graphene oxide (GRGO) was obtained. Meanwhile, the as-prepared GRGO powders can be easily dispersed in several common organic solvents. And the colloidal solutions show no signs of agglomeration even after a week. The Raman spectra XRD and the TG analysis as well as other tests show the GO sheets were reduced successfully. The electrochemical testing results reveal that GRGO3-based flexible electrode has specific capacitance as high as  $179 \text{ F g}^{-1}$  at the potential scanning rate of  $2 \text{ mV s}^{-1}$ . This environment friendly short-time microwave-assist solvothermal treatment is efficient and has a promising prospect of application.

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**Keywords:** A. Microwave processing; Chemical preparation; Graphene oxide; Flexible electrode

## 1. Introduction

Graphene, a single hexagonally sheet of  $\text{sp}^2$ -hybridized carbon atoms tightly packed into a two-dimensional honeycomb lattice, has excellent electrical mechanical, optical properties and thermal properties as well as its high specific surface areas [1–4]. Over the past few years, due to the unique properties of this material, graphene based flexible thin-film transistors, hydrogen storage materials and supercapacitor electrode materials has attracted great research interests. Thus, large-scale production of graphene is needed. Many techniques have been reported to produce graphene, such as chemical vapor deposition (CVD) [5,6], micromechanical exfoliation of graphite [7], epitaxial growth on electrically insulating (SiC) surfaces [8], and chemical reduction of graphene oxide (GO) [9–11]. Among all these methods, the solution-processable chemical GO reduction approach is considered as the most promising route toward mass production of graphene based materials.

Graphene oxide is a water-dispersible material, with many hydroxyl and epoxide groups on its basal plane, and carbonyl and carboxyl groups on its edges [12,13]. Graphene oxide is usually synthesized by exfoliation of graphite oxide, which is obtained by oxidizing graphite using strong acid and oxidants [14].

Typically, strong reductants such as hydrazine monohydrate [15], sodium borohydride [16,17], and strong alkali [18] are used to reduce aqueous dispersions of GO. However, these hydrazine and sodium borohydride are corrosive, flammable and highly toxic which have hazard to personnel health and the environment. And research shows that trace amounts of hydrazine could be harmful to such applications such as organic solar cells [19]. Recently, some mild non-toxic and environment-friendly reductants have been researched widely. Urea [20], ascorbic acid [21,22], amino acid [23,24] and reducing sugar [25,26] are used as substitutes for hydrazine monohydrate. However, although those methods have advantages of low-cost, non-toxic and large-scale production, the reduced graphene oxide in those methods usually has a low solubility in water and most organic solvents. The reduced graphene oxide sheets can easily agglomerated, this means that for further use of those reduced GO, capping reagents

\*Corresponding author. Tel.: +86 13964112085.

E-mail address: [mse\\_wangjq@ujn.edu.cn](mailto:mse_wangjq@ujn.edu.cn) (J. Wang).

(polymers or surfactants) should be used which may affect the properties of the graphene sheets.

Herein, we describe a new approach for the reduction of graphene oxide using glucose in an alkaline environment with the assist of microwave heating. The main advantage of microwave assist heating over other traditional heating methods is heating the reaction mixture uniformly and rapidly. Microwave assist heating can provide significant enhancement in the transfer of energy directly to the reactants which causes an instantaneous internal temperature rise [27,28]. Thus can shorten the reaction time greatly, and improve the reaction efficiency. The use of glucose as a new effective reductant for the reducing of GO can obviously remove a part of the oxygen-containing functional groups on its surfaces. And the GRGO powders can be dispersed in water as well as other polar solvents and holds stability for a long time (more than 7 d). Thus it is very important for further synthesis of graphene-based materials. The as-prepared GRGO also has a good electrochemical performance. The thin slice electrode made from the GRGO without adding any other active materials can reach a specific capacitance of  $179 \text{ F g}^{-1}$  at a scan rate of  $2 \text{ mV s}^{-1}$ .

## 2. Experimental section

### 2.1. Chemicals

Glucose ( $\text{C}_6\text{H}_{12}\text{O}_6$ , 99%) was purchased from Shanghai Chemical Reagents Company, ammonium hydroxide ( $\text{NH}_3 \cdot \text{H}_2\text{O}$ , 25%) and sodium sulfate anhydrous ( $\text{Na}_2\text{SO}_4$ , 99%) were supplied by Sinopharm Chemical Reagent Co. Ltd. Deionized water used during experiments was freshly prepared by an Ulupure system (18 M $\Omega$ ). All of the chemicals used in this reduction were of analytical reagent grade. Cellulose membrane (50 mm, 0.22  $\mu\text{m}$ ) was bought from Shanghai Xinya Purification Device Factory.

### 2.2. Synthesis of GO and GRGO

GO was prepared from natural graphite powder using the modified Hummers. The GO suspension with a mass concentration of  $0.2 \text{ mg mL}^{-1}$  was prepared by dispersing the prepared GO powders into water with the help of ultrasonication (KQ-500E Ultrasonic Cleaner, KunShan Ultrasonic Instruments Co. Ltd).

Typically, to study the performance of glucose on the deoxygenation of graphene oxide, we first diluted the original graphene oxide solution to a concentration of  $0.2 \text{ mg mL}^{-1}$ . To avoid the agglomeration of the colloidal, the pH of the dispersions was adjusted to 9 by dropping diluent ammonia solution. After that, a certain amount of glucose was dissolved in the dispersion for about 30 min under stirring. In a typical reaction procedure, 50 mL solution was transferred into a double-walled 100 mL vessel, which has an inner liner and a cover made of Teflon PFA and an outer high strength sleeve. The vessel was sealed and maintained in a Microwave Accelerated Reaction System (MARS-5, CEM Corporation, USA) at  $95^\circ\text{C}$  for 0.5 h, 1 h, 2 h and 3 h. To facilitate the distinction, we named the

reduced graphene oxide as GRGO0.5, GRGO1, GRGO2, and GRGO3.

To remove the residual reductant, the reduced dispersions were washed repeatedly by vacuum filtration through mixed cellulose membrane with 50 mm in diameter and 0.22  $\mu\text{m}$  pore size using ultrapure water. Then the filter cake, which was the reduced graphene oxide, was dispersed in a certain amount water again to form a stable suspension or dried at  $80^\circ\text{C}$  for 24 h in vacuum to get powder samples. Both the suspensions and the powder samples were for the further characterization analysis.

### 2.3. Preparation of GRGO-based electrode materials

A certain amount GRGO tightly adhered on the cellulose membrane through a vacuum filtration, after drying for 2 d. The membrane-GRGO hybrid material was cut into a  $1 \times 1 \text{ cm}^2$  as electrode slice.

### 2.4. Characterization

The crystalline structure of samples was characterized with a X-ray diffractometer (XRD, Bruker D8-Advance, Cu K $\alpha$  target,  $\lambda = 1.5406 \text{ \AA}$ ). A field emission scanning electron microscope (FE-SEM, FEI, QUANTA FEG 250, United States) was used to observe the morphology and size of as-prepared samples. Ultraviolet–visible (UV–vis) absorption spectra were recorded on a Hitachi U-4100 UV–vis spectrophotometer at room temperature. The aqueous suspensions of graphene oxide or reduced graphene oxide were used as the UV–vis samples, and the pure deionized water was used as reference. The Fourier transform infrared (FT-IR) spectra were obtained on a FTS-165 (Bio-Rad, United States) spectrometer. The samples for FT-IR measurement were prepared by grinding the dried powder of reduced graphene oxide or graphene oxide with KBr, and then compressed into thin pellets. Raman measurements were performed on a High Resolution Raman spectrometer (LabRam HR Evolution, HORIBA JOBIN YVON S.A.S.). The powders of reduced graphene oxide or graphene oxide were placed on a clean  $\text{SiO}_2/\text{Si}$  substrate for the Raman measurement.

The Cyclic Voltammetry electrochemical measurement was performed on a CHI604D (Shanghai) electrochemical workstation with  $2.0 \text{ mol L}^{-1}$   $\text{Na}_2\text{SO}_4$  solution as the aqueous electrolyte. A Pt foil and a saturated calomel electrode were applied as the counter and reference electrodes, respectively.

## 3. Results and discussion

The reduction of GO with the presence of glucose can be directly observed by the color change of the dispersions, from bright yellow brown to black, as shown in Fig. 1(a). No distinct color change could be observed in the GO without the participation of glucose under  $95^\circ\text{C}$  microwave assisted condition. The dispersion stability of reduced GO is a key problem due to the unique properties of graphene are based on the individual sheet and most of the applications require reduced graphene oxide (RGO) firstly being dispersed in water or some organic solvents. As mentioned in many papers [12,13,29], the

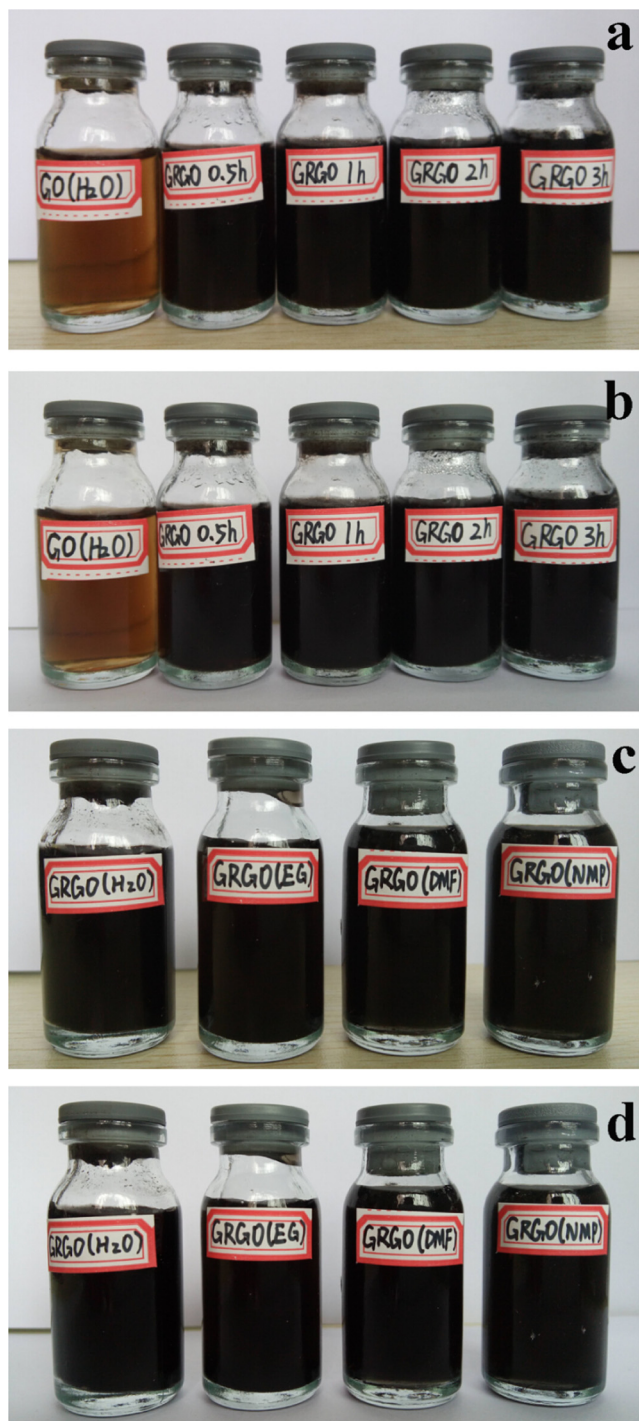


Fig. 1. (a) Images of GO dispersion and different freshly prepared GRGO dispersions immediately after sonication. (b) Images of GO dispersion, and different GRGO dispersions for 7 d after sonication (c) Images of GRGO2 dispersed in different organic solvents immediately after sonication (d) Images of GRGO2 dispersed in different organic solvents for 7 d after sonication. (For interpretation of the references to color in this figure, the reader is referred to the web version of this article.)

GO can be dispersed easily in water and other polar solvents (such as NMP, DMF, EG, etc.) because of the presence of a variety of hydrophilic oxygen groups, such as hydroxyl (–OH), epoxy groups (–O–), and carboxyl groups (–COOH), and

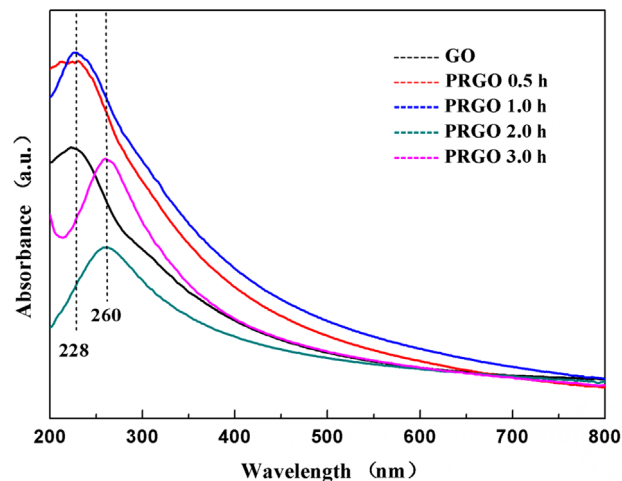


Fig. 2. UV–vis absorption spectra of as-prepared GO and GRGO dispersions. (For interpretation of the references to color in this figure, the reader is referred to the web version of this article.)

remain stable for several weeks without any visible precipitation. Therefore, the dispersibility of RGO is an important parameter during the reduction. In our work, we compared the dispersibility of GRGO in several kinds of commonly solvents: water, DMF, NMP, and EG. The as-prepared GRGO powders were dispersed in those solvents to form a  $0.2 \text{ mg mL}^{-1}$  solution through high power ultrasonication (500 W, 30 min), then those dispersions were placed for a week. As indicated in Fig. 1(a–d), with the help of ultrasonication the as-obtained GRGO sheets show a good dispersibility and stability in those solvents. Even after a week (Fig. 1(b and d)), the GRGO suspensions in those solvents show no signs of agglomeration. The as-prepared GRGO sheets have a very good water-solubility and a good stability in many solvents.

Time-dependent of reducing GO was carried out by UV–vis absorption spectroscopy and the results shown in Fig. 2. A maximum absorption peak at 228 nm corresponding to the  $\pi \rightarrow \pi^*$  transition of aromatic C–C bonds in GO was found to continuously shift to the red region with the increase of reduction time. The maximum absorption peak of reduced GO finally reaches a plateau value of 260 nm as the reaction time increased to 2 h, 3 h, this indicates completion of the reduction, and the formation of highly conjugated structure like that of graphite.

Fig. 3 shows the XRD patterns of the GO and GRGO. The GO shows a sharp diffraction peak at  $2\Theta = 11.2^\circ$ . The expanded interlayer spacing of 0.79 nm in comparison with that of graphite confirms the presence of oxygen functional groups on the GO sheets. After reduction with glucose, the sharp peak disappears, a new broad diffraction peak at  $2\Theta = 24^\circ$  appears, which is much closer to the typical (002) diffraction peak of graphite (d-spacing 0.335 nm at  $2\Theta = 26.6^\circ$ ), confirming the reduction of GO during this reduction process. After being reduced by glucose, a sharp peak appears around  $16^\circ$ , this is mainly due to the ramification of the reaction between the GO sheets and the glucose.

Fig. 4 shows the FTIR spectra of GO and the as-prepared GRGOs. As shown in the figure, the FTIR spectrum of GO

shows an obvious absorption band at  $1720\text{ cm}^{-1}$  due to the C=O stretching. And the vibration and deformation peaks of –OH at  $3422\text{ cm}^{-1}$  and  $1410\text{ cm}^{-1}$ , a C=C characteristic peak at  $1630\text{ cm}^{-1}$ . After being reduced by the presence of glucose and ammonium hydroxide, the C=O stretching vibration peak

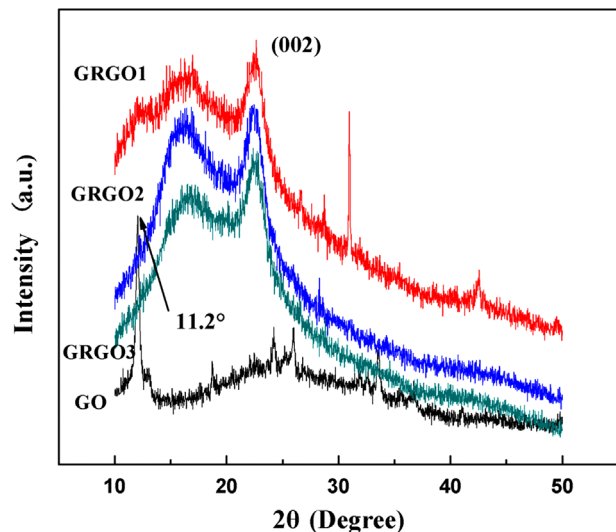


Fig. 3. XRD patterns of GO, and as-prepared GRGO powders.

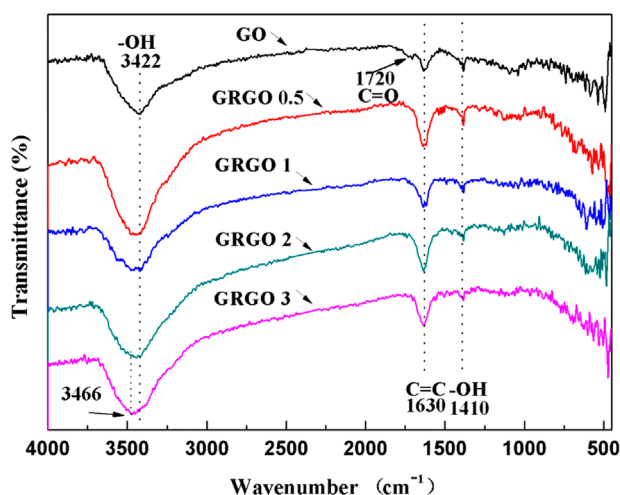


Fig. 4. FTIR spectra of GO, GRGO0.5, GRGO1, GRGO2, and GRGO3.

(at  $1720\text{ cm}^{-1}$ ) disappears while many other oxygen functionalities still retained on the reduced GO. With the increase of reduction time, the spectrum of GRGO3 also exhibits the presence of those oxygen functionalities, but the –OH peak at  $3422\text{ cm}^{-1}$  red shifts to  $3466\text{ cm}^{-1}$ . Those residual oxygen functionalities might form hydrogen bonds with the aldonic acid which was brought by the hydrolysis of glucose. Such interactions can prevent the agglomeration of the reduced GO sheets.

Typical SEM images of the as-prepared GO and GRGO sheets are shown in Fig. 5. The thin gauze-like GO sheets can be observed in Fig. 5(a). The surface of the GO sheets is very smooth and no obviously aggregations are observed. The wrinkled GO sheets are loosely connected to each other. The obtained GRGO sheets randomly aggregate together, as depicted in Fig. 5(b). The wrinkled surface becomes less smoothly. With the reaction time increasing, more aggregated RGO sheets are found, but the wrinkled appearance still can be seen clearly.

Raman spectroscopy was also carried out for the characterization of the as-prepared GRGO sheets. Under normal conditions, the Raman spectrum of graphene is characterized by two main features, the G mode (the  $E_{2g}$  mode of  $sp^2$  carbon atoms) and the D mode (arising from the symmetry  $A_{1g}$  mode).

The variation of the relative intensities of G band (usually observed at  $\sim 1575\text{ cm}^{-1}$ ) and D band (usually observed at  $\sim 1350\text{ cm}^{-1}$ ) in the Raman spectrum of the GO during the reduction usually reveals the change of the electronic conjugation state. With the reduction, the ratio of D/G increased, indicating the reduction take place. And this agrees well with the Raman spectrum of the GO reduced by other reductants. The number of  $sp^2$  hybrid carbon atoms in the GRGO sheets is more than the  $sp^3$  hybrid carbon atoms, which means the average size of the  $sp^2$  hybrid carbon layer plane in the GRGO sheets is larger than that in the GO sheets. Fig. 6 indicates that the ratio of D/G increases with the increase of reduction time, which indicates the reduction becomes more entirely.

TGA was used to examine the thermal stability of the prepared GRGO sheets. Generally, the main mass loss for graphene oxide takes place around  $200\text{ }^\circ\text{C}$ , is attributed to the decomposition of the most labile oxygen functional groups. The removal of adsorbed water accounts for the mass loss measured below  $100\text{ }^\circ\text{C}$ . The slow and steady mass loss observed in the whole temperature range above  $300\text{ }^\circ\text{C}$  can

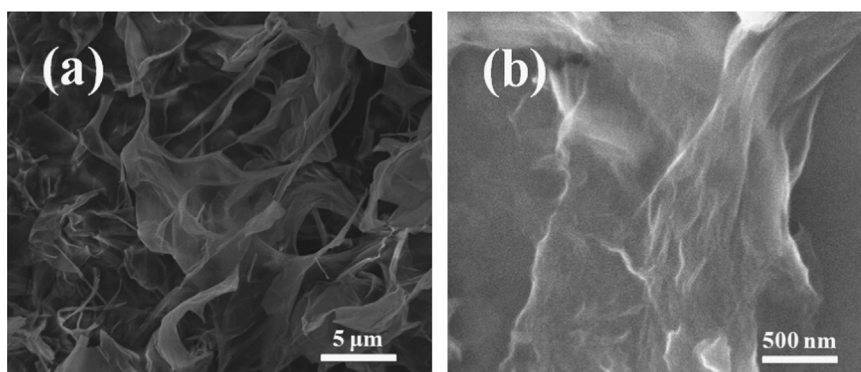


Fig. 5. SEM images of GO and as-prepared GRGO powders.

be ascribed to the more stable oxygen functional groups. As shown in Fig. 7, the GO exhibits a loss of about 45 wt% below 200 °C, which resulted from the labile oxygen-containing functional groups such as C–O and H<sub>2</sub>O vapors. While the GRGO1 exhibits only 20 wt% loss below 200 °C. In contrast, the thermal stability of GRGO1 increases dramatically.

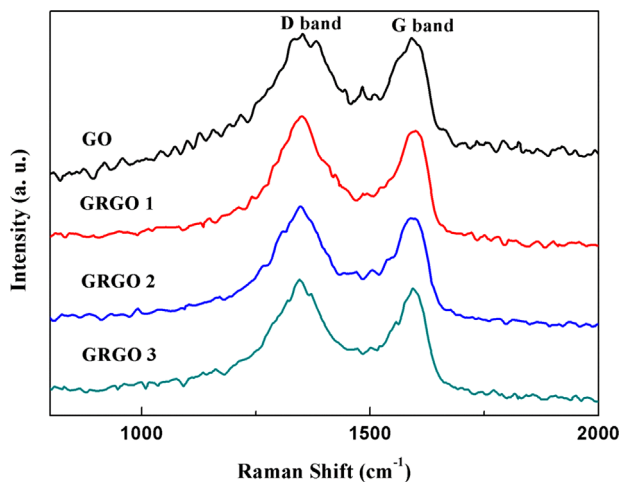


Fig. 6. Raman spectra of GO before and after being reduced by glucose for different reducing times.

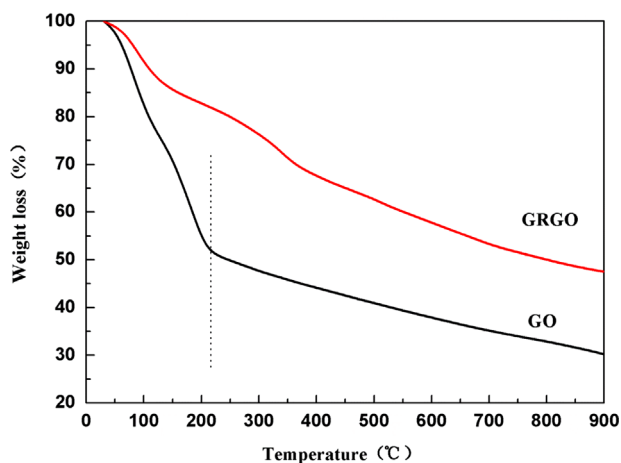


Fig. 7. TGA plots of graphene oxide and GRGO.

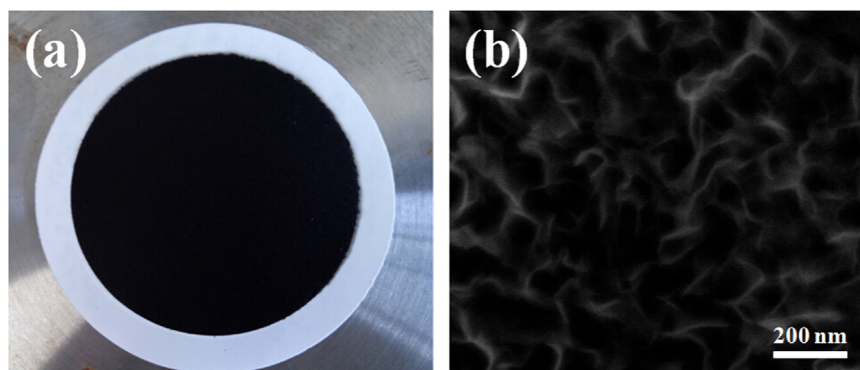


Fig. 8. The photograph of vacuum filtration GRGO film and the SEM image of the GRGO film.

The GRGO sheets tightly adhere on the cellulose membrane through vacuum filtration of a certain amount of GRGO dispersions (for example 20 mL with a concentration of 0.2 mg mL<sup>-1</sup>), and the picture of sample is shown in Fig. 8(a). Fig. 8 (b) shows the SEM image of the surface of the as-prepared GRGO membrane, many wrinkled nanosheets adhere on the surface of the membrane homogeneously. The GRGO-membrane is cut into a 1 × 1 cm<sup>2</sup> as electrode slice (as shown in inset of Fig. 9(a)). Cyclic voltammetry test was carried out based on the GRGO-based electrode. No metal support or current collector was used in the test. The test results are shown in Table 1. With the increase of reduction time, the specific capacitance has a remarkable increase, the capacitance of the GRGO3-based electrode is up to 179 F g<sup>-1</sup>. And it can be seen that these curves exhibited symmetrical and rectangular-like shapes, indicating superior supercapacitor behavior at different areal density (Fig. 9(a)). The CV curves of the as-prepared GRGO3 electrode at different scan rates are shown in Fig. 9(b). No obvious distortion in the CV curves is observed as the potential scan rate is increased. The specific capacitances of GRGO3-based electrode slice are 179, 120, 80.6, and 55 F g<sup>-1</sup> at scan rates of 2, 5, 10, and 20 mV s<sup>-1</sup>, respectively. Obviously, the as-prepared GRGO sheets show a good electrochemical performance.

As a monosaccharide, the glucose has hydroxyl and carboxyl, therefore characteristic response of these functional groups can easily take place. As an alcohol, it can generate ether and ester. As an aldehyde or ketone, reactions such as addition reaction, oxidation and reduction reactions can take place easily. Glucose as an aldohexose, can oxidized to aldonic acid by the presence of GO. And the aldonic acid can further be converted into lacton. Finally, oxidized products with a series of hydroxyl groups and carboxyl groups generated. Those groups on those oxidized products might form hydrogen bonds with the residual oxygen functionalities on the surfaces of reduced GO. The interactions between those oxygen groups and the electrostatic repulsion induced by the residual oxygen functionalities on the surface can disrupt the  $\pi$ - $\pi$  stacking interaction between the reduced GO sheets and further prevent the formation of the agglomerates.

#### 4. Conclusions

In conclusion, we have shown an easy and fast way for the reduction of graphene oxide. Reducing sugar here acts as a green

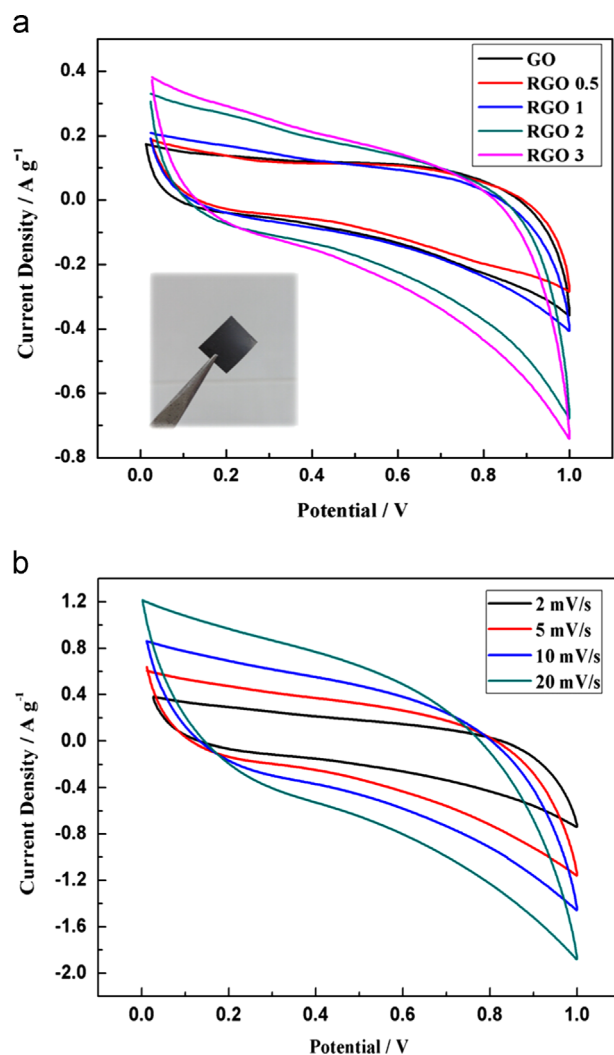


Fig. 9. (a) CV curves of the as-prepared GRGO electrodes. (b) CV curves of the as-prepared GRGO3 electrode at different scan rates.

Table 1

The specific capacitances of the as-prepared different GRGO electrodes at the scan rate of 2 mV/s.

Samples	GO	GRGO0.5	GRGO1	GRGO2	GRGO3
C (F g <sup>-1</sup> )	101	98	104	166	179

non-toxic high-effective reductant. With the assist of microwave, water soluble partly reduced graphene oxide sheets are obtained. The as-prepared GRGO powders also can be dispersed in several common organic solvents. The Raman spectra, XRD as well as TG analysis show the GO powders were successfully reduced by glucose in an alkaline environment. The electrochemical study reveals that GRGO-based flexible electrode has the specific capacitance of 179 F g<sup>-1</sup> at the scan rate of 2 mV s<sup>-1</sup>.

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