



# One-pot hydrothermal preparation of graphene sponge for the removal of oils and organic solvents



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## ABSTRACT

Graphene sponge (GS) has found applications in oil removal due to the hydrophobic nature of graphene sheets. Current hydrothermal preparations of GS use toxic reducing reagents, which might cause environmental pollution. In this study, we reported that graphene oxide (GO) could be hydrothermally reduced by glucose to form GS for the adsorption of oils and various organic solvents. Graphene sheets were reduced by glucose during the hydrothermal treatment and formed 3D porous structure. GS efficiently adsorbed organic solvents and oils with competitive adsorption capacities. GS was able to treat pollutants in pure liquid form and also in the simulated seawater. GS could be easily regenerated by evaporating or burning. After 10 cycles, the adsorption capacity still retained 77% by evaporating and 87% by burning. The implication to the applications of GS in water remediation is discussed.

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## 1. Introduction

Since its discovery, graphene has attracted tremendous interest and efforts due to the unique structure and fantastic properties [1–3]. Among the attractive applications, graphene has been used as adsorbents for various pollutants [4–6]. For instance, Yang and coworkers [7–11] developed graphene adsorbents for the adsorption of heavy metals, dyes and antibiotics. The adsorptions of pesticides, organic molecules and nonmetal ions on graphene adsorbents were also widely concerned [12–14]. During the evaluations, graphene adsorbents showed competitive performance to other adsorbents.

More recently, graphene adsorbents found their applications in oil removal [15–20]. Due to the hydrophobic nature of oils, graphene should be in the reduced forms for oil removal. Porous graphene could accommodate more oils, thus, is the main form in the oil–water separation. There are several types of graphene for oil remediation. The first category is graphene sponge (GS). Hydrothermal methods are widely adopted in preparing GS that had impressive adsorption performance in oil adsorption [15,16]. Bi et al. [17] annealed graphene oxide (GO) aerogel to produce GS for oil removal. Pourmand et al. [18] reported that nanoporous

graphene prepared by chemical vapor deposition (CVD) had high adsorption capacities for oils. The second one is graphene amended polymer sponge [19–21]. Typically, polyurethane (PU) sponge was immersed in GO and then reduced to prepare graphene coated PU sponge for oil and organic solvent adsorption. The third one is graphene amended cotton. Ge et al. [22] coated cotton with graphene for oil/water separation. The fourth category is porous graphene composites. Graphene and other inorganic materials and/or polymers formed composites for oil adsorption [23–25]. Among the aforementioned categories, the preparation of GS is easier and more reproducible. In particular, hydrothermal reduction of GO is regarded as the most facile method to prepare GS for oil removal. However, current studies used reducing reagents that might cause environmental pollutions, such as thiourea and ammonia [15,16]. Therefore, the application of environmental friendly reducing reagents for the preparation of GS should be pursued.

Herein, we reported the one-pot hydrothermal preparation of GS for oil and organic solvent removal by using glucose as the reducing reagent (Fig. 1). GS was characterized to confirm the porous structure and effective reduction. The adsorption of dodecane by GS in pure liquid form and in the simulated water was achieved. The adsorption capacities of diverse oils and organic solvents on GS were quantified. The regeneration of GS was performed by squeezing or burning. The implication to the applications of GS in water remediation is discussed.

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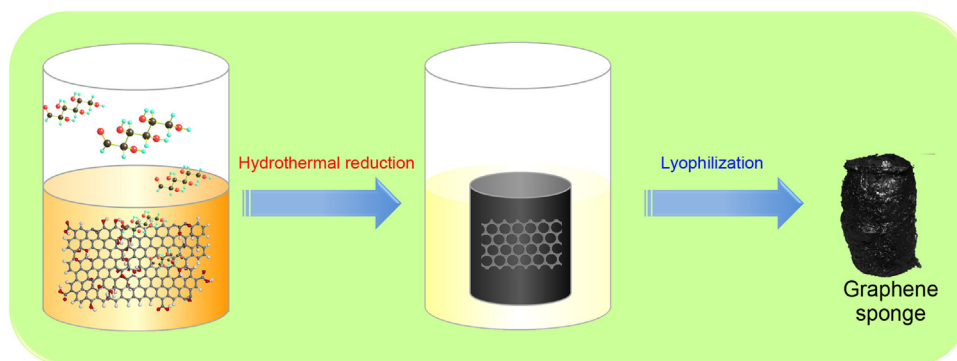


Fig. 1. Schematic illustration of the hydrothermal preparation process of GS.

## 2. Materials and methods

### 2.1. Materials

Graphite was purchased from Huayi Co., Shanghai, China. Glucose was obtained from Yili Fine Chemicals Co., Ltd., Beijing, China. Dodecane was purchased from Kelong Chemical Co., Chengdu, China. Rap oil and machine oil were obtained in local market. Gasoline was bought in local gas station. Crude oil was kindly provided by PetroChina Southwest Oil and Gasfield Company, China. Other chemicals were all of analytical grade.

### 2.2. Preparation of GS

The preparation of GO was following modified Hummers method [26]. Briefly, graphite (3 g) was pre-oxidized by potassium persulfate (2.5 g) and phosphorus pentoxide (2.5 g) in 12 mL of concentrated sulfuric acid. The reaction was taken in water bath at 80 °C for 4.5 h. The residue was collected and dried after pouring the mixture into 500 mL of deionized water.

The pre-oxidized residue was added into 120 mL of concentrated sulfuric acid, and potassium permanganate (15 g) was added slowly under stirring. The mixture was stirred at 35 °C for 2 h, and added into 250 mL of deionized water following by another 2 h stirring. Then 500 mL of deionized water was added and 20 mL of 30% hydrogen peroxide was added dropwise. The yellow product was filtered and washed with HCl aqueous solution (1 mol/L). The yellow solid was dialyzed for 3 days to give graphite oxide. Graphite oxide (10 mg/mL) was sonicated for 1 h to generate GO dispersion.

GO dispersion (10 mg/mL) was mixed with glucose at the mass ratio of 1:1 and sonicated for 30 min. The mixture was sealed in 100 mL Teflon-lined autoclave and maintained at 160 °C for 6 h. After cooling to room temperature, the black rod was washed with deionized water and lyophilized to produce GS.

### 2.3. Characterization

GS samples were characterized by scanning electron microscopy (SEM, Quanta 200FEG, FEI, Netherlands), transmission electron microscopy (TEM, JEM-200CX, JEOL, Japan), X-ray photoelectron spectroscopy (XPS, Kratos, UK) and infrared spectrometer (IR, Magna-IR 750, Nicolet, USA).

### 2.4. Adsorption of oils and organic solvents

To visualize the adsorption, dodecane (2 g) was stained by Sudan red 5B and then placed in a glass plate. After photographing, GS (0.4 g) was added. Photographs were taken at each time interval of 20 s. At the end of photographing, the plate was wiped with the GS to achieve the complete removal.

To evaluate the adsorption ability of GS in seawater, 25 g of sodium chloride, 1.14 g of calcium chloride and 0.7 g of potassium chloride were added into 1 L of ionized water to simulate the seawater. Dodecane (2 g) was stained by Sudan red 5B and added to the simulated seawater. Then, GS (0.9 g) was used to adsorb the dodecane, where GS was drifted by a pipette. The complete removal was achieved after 5 min adsorption.

To measure the adsorption capacity of GS for oils and organic solvents, GS was weighted ( $m_0$ ) and added to 20 mL of liquid (solvent or oil) in a beaker. After 24 h adsorption, GS was taken out and

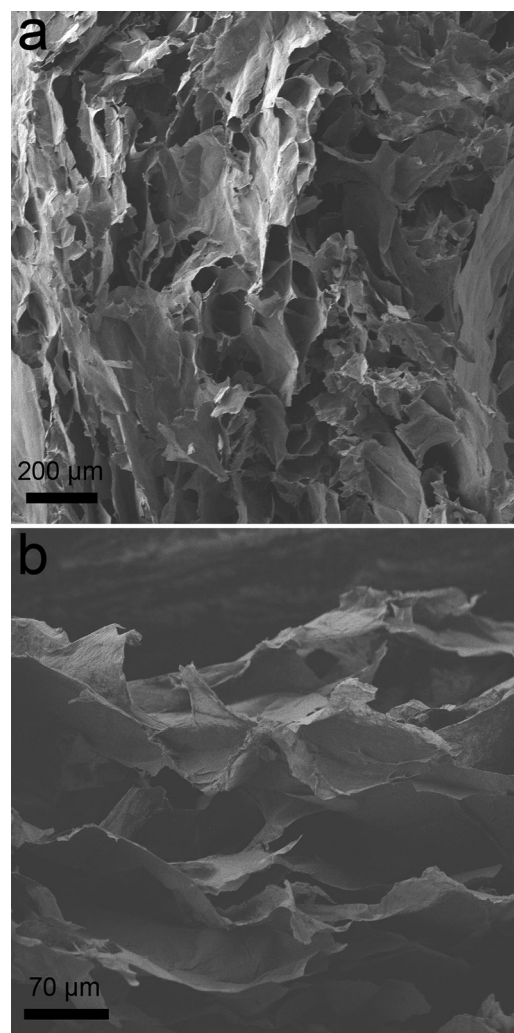


Fig. 2. SEM images of GS at lower (a) and higher (b) magnifications.

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