



A graphene coated cotton for oil/water separation



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ABSTRACT

Development of functional materials that can remove oil from water by an energy-efficient way is highly desirable yet still challenging. Herein, graphene coated cotton was fabricated by a self-assemble technique. This graphene coated cotton could be used as an oil absorbent material, due to its superhydrophobicity and superoleophilicity. It was found that the oil absorption capacity of the graphene coated cotton differed slightly after ten cycles of oil–water separation. The graphene coated cotton also possessed excellent mechanical properties, resulting from the special texture combined the advantages of graphene and raw cotton. Most interestingly, when applied in conjunction with a vacuum system, the time needs for oil collection was shortened. We believe that this kind of sorbent is a promising candidate for use in large-scale removal of oils from water.

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1. Introduction

With the increasing awareness of environmental protection and need for water recycling, there is a growing demand for technologies that can efficiently absorb, remove, and transfer oil spills or organic contaminants from water. For example, accidents of ships in the sea can often result in release of spill oil in seawater and rivers can also be contaminated by wastewater from industry. To mitigate this problem, numerous of oil sorbent materials such as foams [1–5], nanocomposites [6–8], and carbon nanotube materials [9,10] have been developed. However, these materials are usually requiring cost consuming and complicated processes to fabricate, while others show poor efficiency in oil removal, exhibiting low sorption capacity and poor selectivity for the uptake of spilled oils. Therefore, demand for finding novel materials with high absorption capacity, high selectivity, easy fabrication, and low cost is highly imperative.

Graphene, a two-dimensional sheet of covalently bonded carbon atoms, has recently attracted tremendous interests owing to its outstanding properties, such as extremely high electrical conductivity, thermal conductivity, mechanical flexibility, large specific surface area [11–17]. In particular, their low density, hydrophobicity, and mechanical flexibility make them attractive candidates for separation and absorption of organic contaminants from water. One important route to employ graphene as effective absorbent material is to construct three-dimensional (3D) hierar-

chical structures. However, the graphene foams generally have the disadvantages of brittle mechanical performances and are easily irreversibly damaged under mechanical deformations [18,19,27]. This limited mechanical durability prohibits its large-scale production for oil–water separation materials and thus severely hinder its use in practical applications.

In this study, we develop the self-assemble approach to fabricate graphene coated cotton. This graphene coated cotton can be applied as a reusable oil sorbent scaffold in water, with high oil absorption capacity and good reusability. Moreover, the surface texture and functionality of this graphene coated cotton was retained even 100 cycles of compression test, indicating its mechanical durability. This graphene coated cotton applied to separate oil from water offers benefits of simplicity, easy scale up production, high oil-absorption capacity, and recyclability. We believe this oil sorbent will have great potential for large-scale oil–water separation.

2. Experimental section

2.1. Material

The raw cotton was purchased from a local store. The graphite powder (diameter in the range of 10–38 μm) and hydrazine hydrate 85% were provided by Sinopharm Chemical Reagent Co., Ltd. Hexane was used as received and dyed with oil red¹ O. The rest chemicals were all of analytical grade and used as received.

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¹ For interpretation of color in Fig. 9, the reader is referred to the web version of this article.

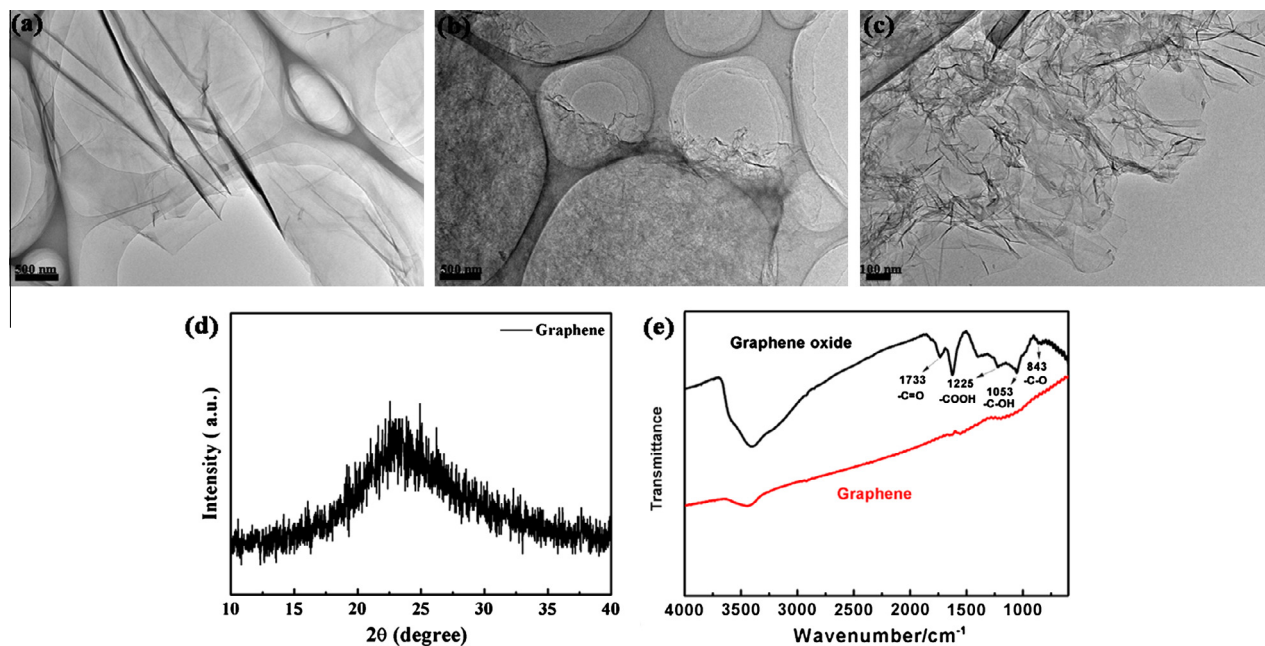


Fig. 1. TEM images of the graphene oxide (a); graphene (b and c); XRD pattern of graphene (d); and FTIR spectra of graphene oxide and graphene (e).

2.2. Preparation of graphene oxide

Typically, graphite oxide (GO) was synthesized from natural graphite powder by a modified Hummers method [20,21]. 2 g graphite powder was added into 120 ml cooled concentrated H_2SO_4 , and then KMnO_4 (15 g) was added gradually under stirring. Subsequently, the mixture was stirred at 35 °C for 2 h, and 230 ml distilled water was slowly added to the solution. The reaction was terminated by adding 0.7 L distilled water and 20 ml 30% H_2O_2 solution. The resulting brilliant yellow mixture was washed with 5% HCl solution and distilled water. Finally, the graphene oxide nanosheets were obtained by drying the suspension in a vacuum oven at 60 °C.

2.3. Preparation of the graphene coated cotton

A piece of cotton was first cleaned with acetone and distilled water using an ultrasonic cleaner, followed by drying. The as dried cotton was then dipped into a dispersion of 30 ml graphene oxide solution (0.4 mg ml^{-1}) and 2 ml hydrazine hydrate. Then, the mixture was poured into a 100 ml autoclave and placed in oven at 90 °C for 12 h. Finally, the obtained graphene coated cotton was removed from the autoclave and placed to dry in an oven at 110 °C to obtain superhydrophobic samples.

2.4. Characterization

FEI Tecnai F30 transmission electron microscopy (TEM) and Bruker IFS66/S Fourier transform infrared (FTIR) spectrometer were employed to investigate the morphology and microstructure of graphene and graphene oxide. The morphology of the surface was observed by field-emission scanning electron microscope (JEOL JSM-6701F FESEM). X-ray power diffraction (XRD) patterns were obtained using a D/Max-2400 X-ray diffractometer. X-ray photoelectron spectra (XPS) were obtained on an ESCALAB250xi spectrometer equipped with a focused monochromatic Al X-ray source (1486.6 eV). Raman spectroscopy (Renishaw in via Raman microscope with 633 nm line of an Ar ion laser as an excitation source) was employed to characterize the microstructure of the

samples. Contact angle (CA) was measured with 5 μL droplets of water using a Krüss DSA 100 (Krüss Company, Ltd., Germany) apparatus at ambient temperature. The optical images were captured by a digital camera (Canon).

3. Results and discussion

Fig. 1a and b shows the TEM images of graphene oxide and graphene. Fig. 1c is the magnified image of graphene. As shown in Fig. 1d, the diffraction peak positioned at 2θ around 23.9° implying that the graphene was comprised of randomly ordered graphitic platelets in a corrugated structure [22]. Fig. 1e shows the FTIR spectra of graphene oxide, and graphene. Furthermore, the peaks of graphene oxide positioned at 1733 cm^{-1} , 1225 cm^{-1} , 1053 cm^{-1} , and 843 cm^{-1} are attributed to C=O stretching vibrations from carbonyl and carboxylic group, C–OH stretching, and C–O vibrations of the epoxy groups, respectively [23]. Raman spectra of graphene oxide, graphene sheets (collected from residual

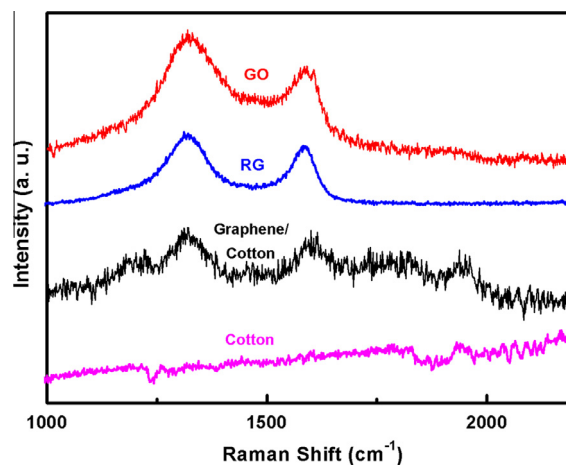


Fig. 2. Raman spectra of graphene oxide, graphene sheets (collected from residual solution after hydrothermal reaction), cotton, and graphene coated cotton under the excitation of 633 nm.

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