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Air-dried graphene-based sponge for Water/oil separation and strain sensing

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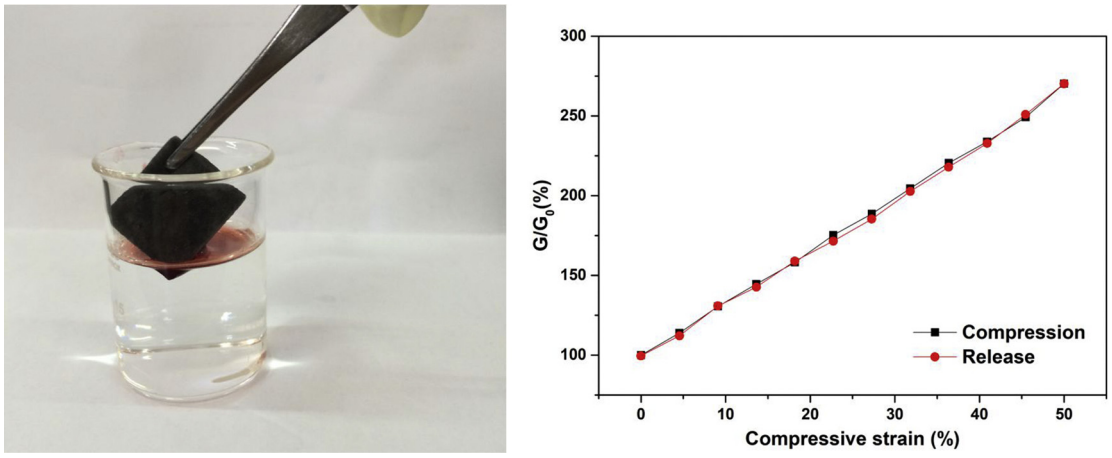
GRAPHICAL ABSTRACT



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ABSTRACT

An environmental-friendly, cost-effective and mild approach to prepare graphene-based sponge (GS) by *in situ* reduction-assembly of graphene sheets on the skeletons of melamine sponge was reported. The fabrication process is facile and no freeze-drying process is required. The structures of the GS were characterized by Raman spectra, X-ray photoelectron spectroscopy (XPS), X-ray diffraction (XRD) and scanning electron microscopy (SEM). The hydrophobic and oleophilic GS exhibited high absorption capacities for organic liquids and excellent recyclability. Moreover, the electrical conductance of the GS is strain-sensitive, which changes in proportional with compressive strain. These significant performances make the GS candidate for potential applications in water/oil separation and strain sensing.

1. Introduction

Graphene, a two-dimensional crystal of monolayer carbon atoms tightly packed into honeycomb lattice with sp^2 hybridization, since its discovery, has attracted world-wide attention and extensive research interest [1–5]. The extraordinary electronic, optical, thermal, mechanical and chemical properties [6–11] endow graphene with a wide range of applications such as electronics [12], energy storage and conversion [13,14], environmental remediation [15], catalysis [16], sensors [17,18], composites [19] and biomedicine [20]. However, the excellent properties of graphene are usually compromised by restacking and aggregation of graphene sheets due to the strong π–π interaction

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and van der Waals force among graphene sheets. In order to solve this problem, numerous studies have been devoted to three-dimensional (3D) graphene architectures such as aerogels and hydrogels because of their large specific surface areas and porosity, lightweight, strong mechanical strength, and high electrical and thermal conductivity [19,21–24].

Up to now, graphene aerogels and hydrogels have been prepared by various methods and exhibit promising potential applications in many areas [25–29]. However, the practical applications of these 3D graphene assemblies are usually limited by their low mechanical strength as well as time-consuming and power-consuming freeze-drying or supercritical-drying process to transfer hydrogel to aerogel. To overcome the first obstacle, graphene-polymer double networks have been developed by absorbing monomer into the prepared graphene hydrogel followed by *in situ* polymerization [30–32]. Even so, the fabrication procedures are complicate and time-consuming and freeze-drying process is still needed to obtain aerogel. Nguyen et al. fabricated graphene-based sponges through dipping sponges into a dispersion of graphene nanosheets in ethanol followed by drying in the vacuum oven at 100 °C for 2 h [33]. Nevertheless, the coated graphene sheets are just physically interconnected and the adhesion fastness of graphene sheets on sponge skeletons is relatively low, which would reduce their recyclability. Li et al. prepared elastic graphene aerogels with the aid of *in situ* polymerization of PAAm during the gelation of a graphene oxide (GO) aqueous solution [34]. Although no freeze-drying was required to make the graphene aerogels, the preparation procedure involves freezing at −75 °C, heating at 70 °C for 20 h and dialysis for at least 48 h.

In this work, we report a green, facile and low-cost method to fabricate graphene-based sponge (GS) through *in situ* reduction-assembly of graphene sheets on the melamine sponge skeletons by mild chemical reduction followed by air-drying. The reduction-assembly process was accomplished by immersing a melamine sponge into GO aqueous suspension and heating the GO filled sponge in vitamin C (VC) solution. VC is a commonly used reduction agent to reduce GO and spontaneously form reduced graphene oxide (rGO) hydrogel with interconnected network [25,35,36]. After coating with hydrophobic graphene nanosheets, the wettability of melamine sponge changed from hydrophilic to hydrophobic and showed large selective absorption capacities to organic solvents and oils. In addition, the electrical conductance of the GS changed in proportional to compressive strain, indicating its potential application of strain sensor.

2. Materials and methods

2.1. Materials

Natural graphite flake (−325 mesh, 99.8%) was bought from Alfa Aesar and used for preparing GO. Sulfuric acid (H_2SO_4, 98%), hydrochloric acid (HCl, 36%), potassium permanganate ($KMnO_4$, 99.5%), sodium nitrate ($NaNO_3$, 99%), hydrogen peroxide (H_2O_2, 30%), vitamin C ($C_6H_8O_6$, 99.7%), and various organic solvents were purchased from Sinopharm Chemical Reagent Co., Ltd, China.

2.2. Preparation of GS

GO was prepared by Hummers' method [37]. Typically, a melamine sponge was immersed in a GO aqueous suspension with concentration of 0.3 g L^{-1}. The GO solution impregnated melamine sponge was then transferred to a 0.03 M VC solution and heated in a water bath at 90 °C for 2 h. After cleaning with deionized water for several times and air-drying in atmosphere under 80 °C, the GS was obtained.

2.3. Characterization

Raman spectra was recorded using a micro-Raman microscope (Horiba Jobin Yvon LabRAM HR-800) with 514 nm laser. X-ray photoelectron spectroscopy (XPS) was carried out on an ESCALAB 250Xi X-ray photoelectron spectrometer. X-ray diffraction (XRD) analysis was performed on a Rigaku RINTTTR III X-ray diffractometer. Contact angle (CA) were measured under ambient condition using a Krüss DSA 100 optical contact angle measuring instrument. Scanning electron micrographs (SEM) were conducted on a field-emission scanning electron microscope (JSM-6700F, JEOL, Japan).

2.4. Organic liquids absorption

The GS was placed into a beaker filled with the organic liquid. After saturated with organic liquid, the GS was removed. Before and after absorption of organic liquids, the GS was weighed as m_1 and m_2, respectively. The absorption capacity of the GS was calculated according to the equation $Q_o = (m_2\text{-}m_1)/m_1$.

2.5. Electrical conductance measurements

The electrical conductance of the GS was measured with CHI66E electrochemical workstation (Chenhua Instruments Co., Ltd., Shanghai, China) by sandwiching with two copper plates. The conductance of each sample was calculated from the slope of I–V curve recorded from 0 to 1 V.

3. Results and discussion

The GS was prepared by an *in situ* reduction-assembly process with melamine sponge as a framework. The melamine sponge is a cheap and commercially available material with ultra-light mass, ultra-loose network structure, high elasticity and stability, and thus was selected as skeleton to deposit graphene sheets (Fig. 1a). In a typical preparation procedure, a melamine sponge block was soaked a GO suspension (0.3 g L^{-1}), and the sponge was impregnated with GO solution immediately. The intermolecular hydrogen-bonding interactions between NH- on

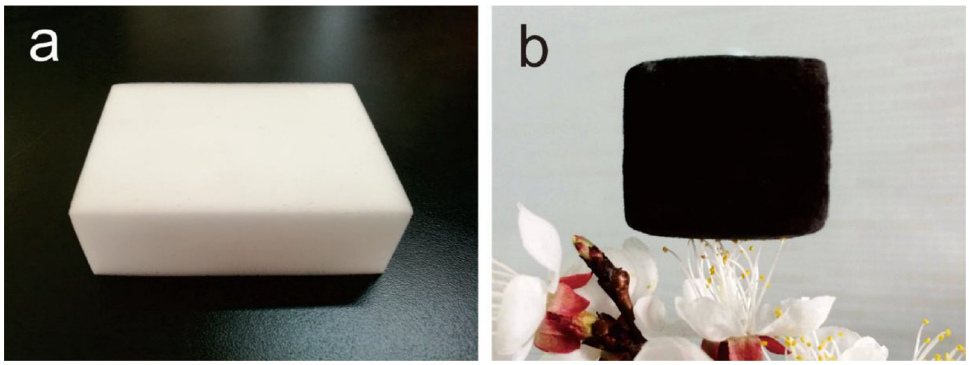

Fig. 1. Photographs of a bare melamine sponge (a) and a GS (b) standing on the androecia of a flower.