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Recycle and reusable melamine sponge coated by graphene for highly efficient oil-absorption

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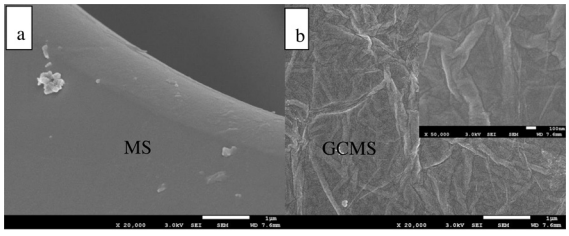
HIGHLIGHTS

- The GCMS was prepared by one step method.
- The GO reduced by Am/Eth is more ecofriendly and higher quality.
- Absorption capacity for gasoline and diesel oil were higher than 105 g g^{-1}.
- Absorption capacity reaches up 95% after it was reused 10 times.

GRAPHICAL ABSTRACT



a Melamine spong b Graphene coated melamine sponges

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ABSTRACT

Graphene coated melamine sponges (GCMS) were fabricated by one step method. Graphene oxide (GO) was treated with mixtures of ammonia and ethanol (Am/Eth) with different proportions. The structures of GO and GCMS were characterized by Fourier transform infrared spectroscopy (FT-IR), X-ray diffraction (XRD) transmission electron microscopy (TEM) and scanning electron microscopy (SEM). Am/Eth mixture has both dispersive and reductive effects on graphene oxide. The GCMS are hydrophobic and oleophilic and show extremely high absorption for oil liquids. The absorption capacities for gasoline and diesel oil were higher than 105 g g^{-1}. In addition, graphene-based sponges are demonstrated to have excellent recyclability. The absorption capacity of the GCMS even reach up to 95% after it was reused 10 times by squeezing. So the GCMS has great application potential in the field of oil spillage cleanup.

1. Introduction

In recent years, oil spill accidents not only lead to energy loss but also have catastrophic impact on marine and aquatic ecosystems [1]. Once oil spills happened, numerous sea birds, fish and shrimp are killed and aquatic plants have sustained serious damage. The crude oil and oil-products spilled in water bodies is hard to remove by conventional sewage process. In practice, oil spills are cleaned by a variety of methods, such as mechanical collection [2], chemical dispersants [3], bioremediation [4], in situ burning [5] and using absorbent materials [6–8]. Especially in complex region or coast, using absorbent materials to deal with spilled oil is a good approach. Traditionally, there are three kinds of oil-absorbing materials including inorganic materials (zeolites, bentonite, coal ash and activated carbon et al.) [8,9], organic synthetic materials (polypropylene fiber, alkyl acrylate copolymers and polyurethane foam et al.) [10] and natural organic material (straw wool fibers, wood chips, reeds and corn stalks et al.) [6,8,9]. Though most of the inorganic materials are cheap, they have low oil absorption capacities and poor recyclability. Natural organic materials absorb not only spilled oils but also water because they contain a number of hydrophilic groups such as hydroxyl and carboxyl. Organic synthetic materials are lipophilic and hydrophobic, but

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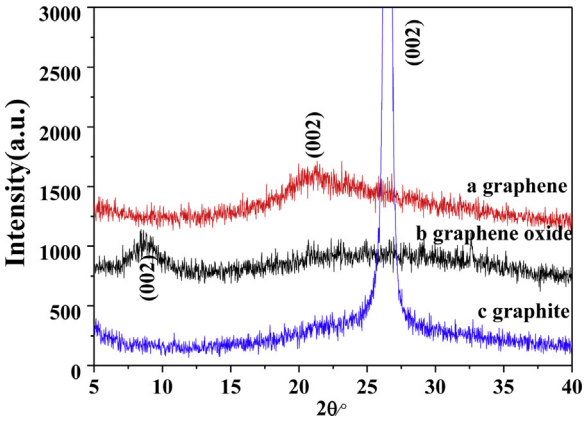

Fig. 1. XRD spectra of the (a) TGO, (b) GO, (c) GT.

these absorbents are incompatibilities with environment and have poor recyclability [11].

Recently, materials with super-oleophilic properties have stimulated much interest because of their high oil absorption capacity while repelling water completely [12,13]. Many effective oil absorbent materials such as carbon nanotubes [14], carbon nanofiber [15], filter paper [16] and graphene aerogels [17] have been developed extensively. However, these materials have limitations for practical application because they are high cost with complicated processing and poor recyclabilities.

Melamine sponge (MS) is a kind of 3D porous material with high porosity (>99%), low densities, good thermal stabilities, environmental protection and low cost. However, MS is a strong absorbing media for not only oils but also water because of its porous structure and having some hydrophilic groups, such as hydroxyl groups, aldehyde groups and ether bonds introduced during synthesis. If MS are changed from hydrophilic to hydrophobic, they will be potential efficient substrates for oil absorption.

Graphene (G) has attracted tremendous attentions because of their intriguing physicochemical and environmentally friendly properties [18,19]. It can be obtained by reducing graphene oxide (GO) [20]. The reduced GO changes from hydrophilic to hydrophobic because the polar functional groups such as hydroxyl, carboxyl and epoxy groups in GO are removed [21]. So combining the advantages of MS and G, the melamine sponge (MS) coated with hydrophobic graphene nanosheets were prepared to deal with oil spills. The prepared graphene-based sponges will be a high efficient and reusable absorbent for spilling oil because of its excellent oil absorption capacities, good recyclability and environmental friendliness.

2. Materials and methods

2.1. Materials

Graphite (GT) was obtained from China national medicines corporation Ltd. MS were purchased from Beijing kelinmei high and new material Co., Ltd. Ammonia, absolute ethanol and hydrazine were purchased from Tianjin Chemical Reagent Co. All the chemicals were used as received.

2.2. Preparation of GO

GO was prepared from graphite by Hummer's method [22]. 0.12 g of GO was dispersed in 60 mL mixture of $NH_3{\cdot}H_2O/C_2H_5OH$ solutions with different volume ratio of 0:60, 4:1, 6:1, 10:1, 60:0,

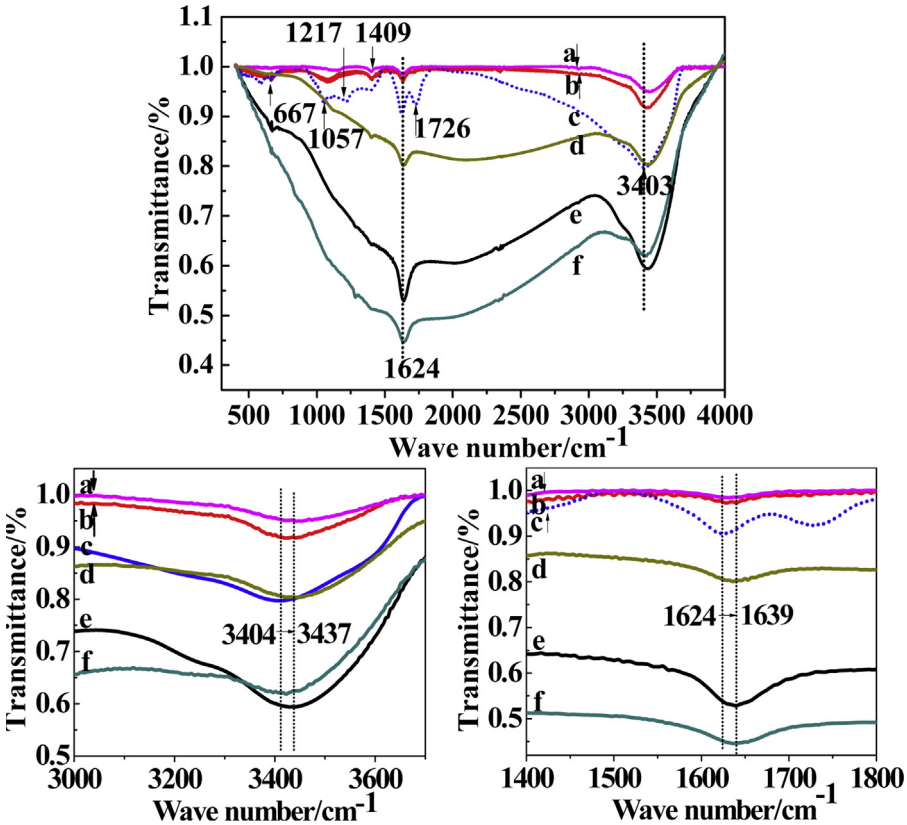

Fig. 2. FT-IR spectra of GO treated with different ways (a) Am/Eth volume ratio, 0:60 (b) hydration hydrazine reduced GO (c) untreated GO (d) Am/Eth volume ratio, 60:0 (e) Am/Eth volume ratio 4:1 (f) Am/Eth volume ratio, 8:1.