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Highly-efficient and recyclable oil absorbing performance of functionalized graphene aerogel

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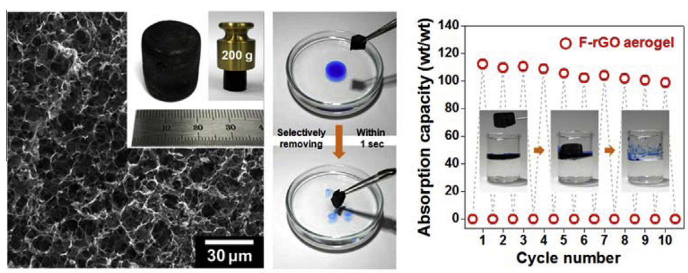
HIGHLIGHTS

- Graphene aerogel is readily achieved *via* self-assembly of graphene oxide sheets.
- Surface modification of graphene aerogel is acquired by a one-step solution method.
- Surface modified graphene aerogel has high porosity and hydrophobicity.
- Absorption test was performed under various kinds of oils and organic solvents.
- Surface modified graphene aerogel shows excellent absorbability and recyclability.

GRAPHICAL ABSTRACT



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ABSTRACT

Functionalized graphene aerogel with high porosity and hydrophobicity is prepared by surface modification of self-assembled graphene oxide aerogels. Fluorinated functional groups are introduced into the surface of three-dimensionally macroporous graphene aerogel through a one-step solution immersion method. Successful fabrication and surface modification of graphene aerogel are confirmed by various techniques. The functionalized graphene aerogel represents superior physical features, including low density (bulk density of 14.4 mg cm^{-3}), high porosity (>87%), mechanical stability (supports at least 2600 times its own weight), and hydrophobicity (contact angle of 144°). By combining the structural features and hydrophobic surface property, the functionalized graphene aerogel not only exhibits excellent absorption performance for various types of oils and organic solvents (capacity up to 11,200% of its weight), but also shows a remarkable regeneration capability (no obvious change in absorption capacity), making them an ideal candidate for eliminating spilled oils and other toxic organic pollutants.

1. Introduction

Oil spill in Gulf of Mexico caused crude oil to leak from oilfield into ocean and near coast in April 2010, and similar environmental disasters have occurred frequently all over the world. These accidents have highlighted environmental and ecological issues. Accordingly, an interest in eliminating pollutants (*e.g.* crude oil, petroleum products, and toxic organic solvents) is increasing with strong social concern. In response, various sorbent materials such as organic [1–4], inorganic nanowire membranes [5,6], macroporous nanocomposites [7,8], and carbon nanotubes [9,10] have been so far investigated for many separation and purification processes.

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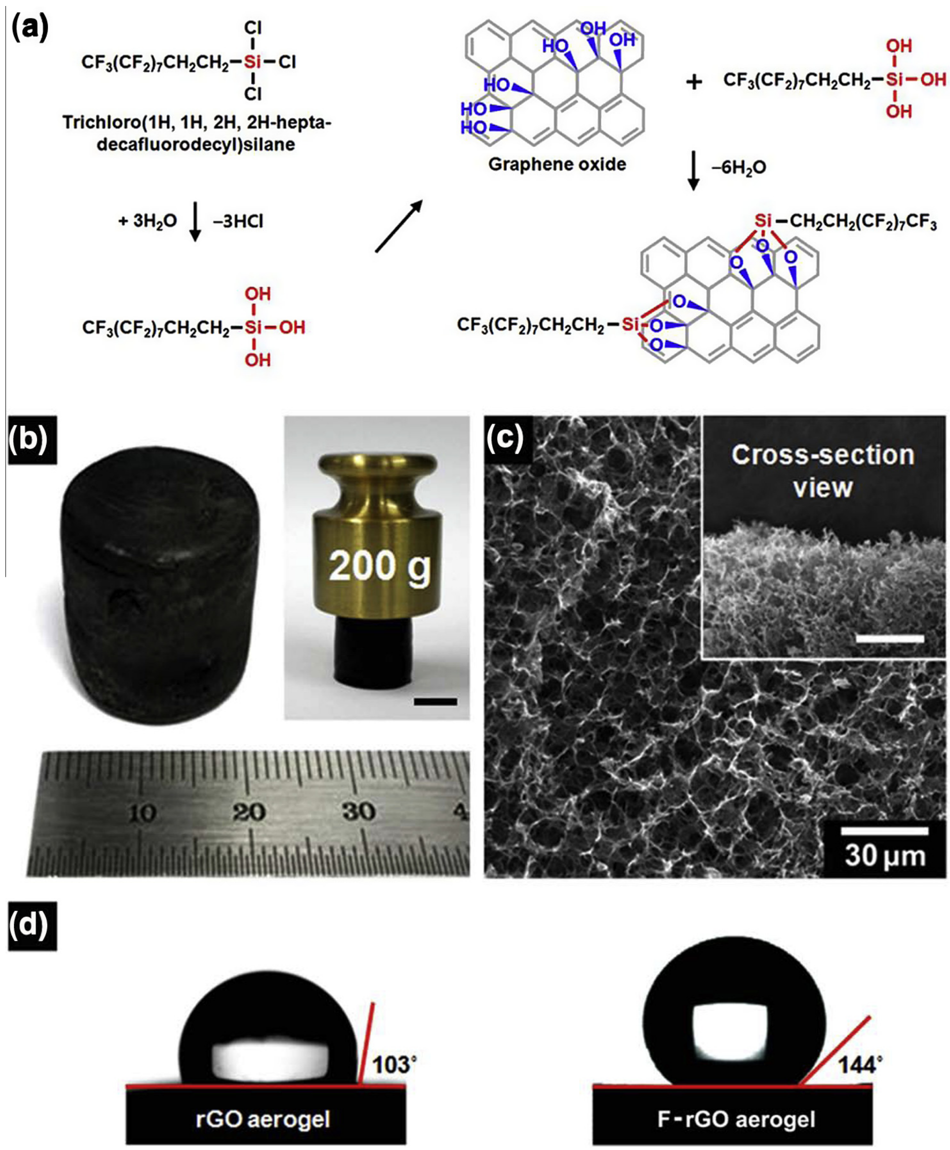

Fig. 1. (a) Schematic illustration for the surface modification mechanism during silanization reaction. (b) Photograph of a centimeter-sized F-rGO aerogel obtained in a single reaction. Digital image (inset) showing that the F-rGO aerogel supports mass loading of 200 g (scale bar: 1 cm). (c) SEM images of the F-rGO aerogel (inset scale bar is 100 μm). (d) Optical images of water droplet on the surface of rGO and F-rGO aerogels.

However, most previous and existing materials possessed the critical drawbacks of high cost, complicated procedures and difficulty in scale up, which could hinder their practical or industrial applications. Recently, three-dimensional (3D) graphene-based macroporous architectures have been intensively explored as a promising absorbent with a remarkable absorption capability resulting from their high porosity and available surface area, excellent chemical and mechanical stabilities, and diverse chemical modification of carbon surface [11–19]. In addition, it has been known that the chemical modifications can tune the surface properties in order to improve chemical affinities for high capacity and selectivity of target guest molecules [20]. Nonetheless, it is difficult to preserve the 3D architecture while effectively modifying the graphene surface. Therefore, it is desirable for developing a rational chemistry to functionalizing graphene-based porous materials with high absorption capacity and chemical inertness for broad range of oils and organic solvents.

Herein, we present a solution-based surface modification approach for synthesizing functionalized graphene-based aerogel that can be utilized as an advanced absorbent of oils and organic solvents. In order to explore the feasibility of surface modified graphene-based aerogel as an absorbent, the hydrophobic effects of functionalization on wettability as well as absorbent performances were evaluated using various oils and organic solvents. It is expected that the functionalized graphene-based aerogel would have high absorption capacity due to a combined or synergistic advantages of the high porosity and available surface area arising from the 3D macroporous networks, as well as the strong oils and organic solvents affinity attributable to the hydrophobic surface. The strong internetworks of the surface-modified graphene aerogel can also provide chemical and mechanical durability for good regeneration capability.

2. Experimental

2.1. Preparation of graphene oxide (GO)

The GO solution was prepared by oxidation of expandable graphite (ASBURY CARBONS) using the (modified) Hummers'