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A two-step hydrophobic fabrication of melamine sponge for oil absorption and oil/water separation

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ABSTRACT

Hydrophobic sponges have drawn great attention recently as potential absorption materials for oil spill cleanup and water/oil separation due to their excellent elasticity, absorptivity and selectivity. Herein, we demonstrate a facile and cost-effective method to fabricate hydrophobic sponges based on commercial melamine sponge via a two-step process, i.e., silica nanoparticle adsorption and silanization covering procedures. The hydrophobic modification was confirmed by ATR-FTIR and SEM characterization. The modified sponge shows hydrophobicity and oleophilicity and exhibited excellent absorption capacity for various oils and organic solvents (60–109 g/g). Interestingly, this modified sponge displays great endurability towards ultrasonication or at acid/alkali circumstance, and presents remarkable reusability after 12 cycles of absorption-squeezing process. In addition, a piece of modified sponge can be employed for continuous collecting of oil/solvent from water by using a simple collection device. These hydrophobic, elastic, and durable melamine sponges are very promising materials for practical oil absorption and oil/water separation.

1. Introduction

The growth of oil production and transportation has increased the related oil spills and accidental pollution arousing serious environmental and ecological problem over past few decades [1–3]. Notably, cleaning up these oil slicks from the marine surface as well as from the coastline remains a challenge. Several strategies have been introduced to deal with this problem, including in situ burning, [4] skimming, [5] and using chemical dispersant [6]. However, these methods cause additional damage to marine ecosystems. Alternatively, removing oil from water with help of absorbents is considered as an economical, efficient, and ecologically friendly means as the pollutants could be properly discarded and no secondary pollution is created [7–9].

One such absorbable material is sponge, which is porous with a desirable 3D structure and widely used as cleaner and for soundproofing. Superhydrophobic sponge exhibit promising and attractive superiority in terms of high absorption capacity, fast absorption rate, low weight, low cost, and excellent elasticity, which make them an ideal candidate for oil absorption applications [10–13]. As an attractive industrial product, polyurethane (PU) is the most popular material for sponge-based oil absorption applications. Researchers have tried various modification procedures to make polyurethane sponge for practical use in water/oil separation. For example, Zhang et al. fabricated a superhydrophobic oil absorbent by coating commercial polyurethane sponges with polypyrrole through a vapor-phase deposition process. The absorption capacity of the fabricated sponge towards different oils were all above 20 g/g [14]. In another study, polyurethane sponges were modified by grafting with lauryl methacrylate (LMA) in solvent and/or coating with LMA microspheres through a heating and curing procedure. The absorption results showed that the modified PU sponge had an uptake of 50–69 g/g organic solvents based on the solvent [15]. In addition, a variety of materials such as graphene, [16–18] carbon nanotube, [19] polydimethylsiloxane, [8,20] and polytetrafluoroethylene [21] are also used to treat polyurethane sponge for oil absorbing applications.

Other materials such as melamine, [22–25] polyvinyl alcohol (PVA), [26,27] polyester, [28] chitin, [29] cellulose, [30] and graphene [31] could also be fabricated as sponge for oil absorption applications. Among these porous materials, commercial melamine sponges have advantage of low density (4–12 mg/cm^3), high porosity (> 99%), high thermal stability as well as good compressibility. The manufacture process of melamine sponge not only forms a micro/nanoscale

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hierarchical framework but also generates secondary amino groups which make it possible for further modification. Recently, a few publications reported the hydrophobic modification of commercial melamine sponges and indicated them as promising alternatives for effective oil absorption. Shi et al. prepared a fire-resistant, elastic sponge through a two-step polyphenol chemistry strategy [23]. This hydrophobic sponge material exhibits very good absorption capacities of oils/organic solvents up to 69–176 times its own weight. These modification processes provided the melamine sponge a superhydrophobic surface (water contact angle WCA > 150°) as well as a high absorption capacity for oil absorption. Chen and Lu et al. reported a facile method to fabricate a robust and scalable hydrophobic sponge through the thermal reduction of graphene oxide on the skeletons of melamine sponge [25]. The as-fabricated sponge featured with superhydrophobic surface, high adsorption capacity (up to 140 g/g), good recyclability, and excellent oil/water separation efficiency 99.98% for various oils and organic solvents. Dickerson et al. fabricated robust superhydrophobic melamine sponges through silanization via a toluene solution-immersion process [32]. The silanization created self-assembled monolayers on the surface of melamine sponge skeleton which endowed the modified sponge an absorption capacity of 82–163 g/g for various organic solvents and oils.

Inspired by these achievements, we therefore look for a simple and facile method for sponge functionalization. Herein, a two-step hydrophobic fabrication of melamine sponge is developed. First, SiO_2 nanoparticles was synthesized and adsorbed to the surface of the melamine sponge's skeleton to enhance the surface roughness of the sponge. A silanization process with vinyltrimethoxysilane was then carried through to coat the silica-modified sponge and endow the melamine sponge a hydrophobic feature (Scheme 1). These fabricated hydrophobic sponges exhibited excellent absorption capacity for different kinds of oils and organic solvents, showed a robust stability against ultrasonication or acid/alkali treatment. The modification process with regard to melamine sponge is convenient, cost-effective and eco-friendly. These oil absorption materials may provide a fast and selective separation of oils from oil-water mixture and can be used to treat oily wastewater or organic-contaminated water for an environmental protection purpose.

2. Experimental section

2.1. Materials

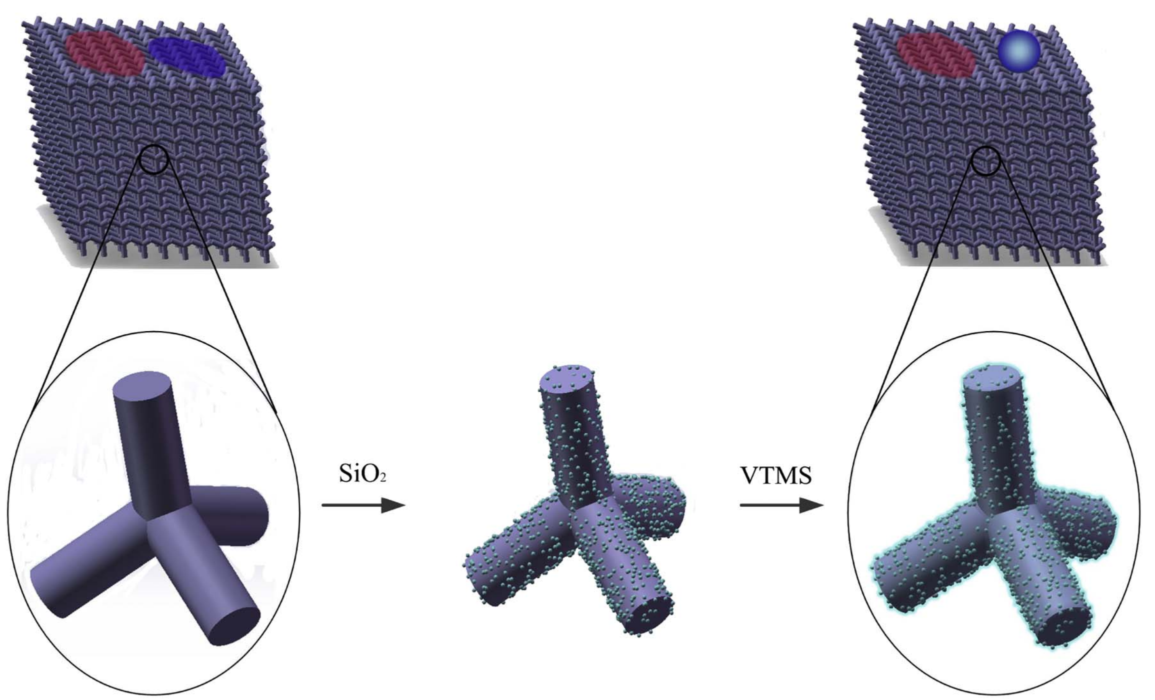
Commercial melamine sponges (MS) were supplied by Freudenberg Household Products Co., Ltd., Suzhou, China. Tetraethoxysilane (TEOS, 98.5%), ammonia (26%), vinyltrimethoxysilane (VTMS, 98%), trichloromethane, Sudan Red III, hydrochloric acid (HCl), sodium hydroxide (NaOH), and organic solvents (ethanol, acetone, carbon tetrachloride, toluene and n-hexane) were purchased from Sinopharm Chemical Reagent Co., Ltd., Shanghai, China. All chemicals are analytical grade and were used as received. Soybean oil was purchased from COFCO Food Marketing Co., Ltd., Tianjin, China. Diesel was purchased from Sinopec, Wuhan, China. Silicone oil was supplied by Hangping Co., Ltd., Beijing, China. Deionized water was used for all experiments.

2.2. Preparation of SiO_2 nanoparticles

Silica nanoparticles were prepared by the conventional sol-gel method. Briefly, 4 mL of TEOS was added into 40 mL of ethanol under stirring at room temperature. Then 1.6 mL of ammonia (25wt%) was added and the solution was kept stirring for 20 min. The mixture was placed without stirring for 2 days to produce homogeneous SiO_2 nanoparticles of 40–80 nm (Fig. 1a).

2.3. Preparation of hydrophobic melamine sponges

The as-prepared SiO_2 nanoparticle solution was diluted in ethanol to obtain a concentration of c.a. 6 mg/mL with the help of an ultrasonication treatment. MS samples with proper size ($1 \times 1 \times 1\ cm^3$) were immersed in the solution for 6 h. The SiO_2-attached sponges MS@SiO_2 was then transferred into the ethanol solution of VTMS and ammonia with appropriate concentrations and the mixture was kept stirring gently for 20 h. The cubes were squeezed to remove the absorbed solution and were washed and dried in a vacuum oven at 60 °C to obtain hydrophobic melamine sponge MS@SiO_2@VTMS.



Scheme 1. Illustration for the fabrication process of the hydrophobic MS@SiO_2@VTMS sponges.