GRAPHENE OXIDE-COATED COTTON FABRICS WITH WETTABILITY FOR CONTINUOUS OIL/WATER SEPARATION

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Abstract

Fabrication of filtration membranes with opposite wettability for the separation of immiscible oil/water mixture has aroused extensive attention owing to the avoidance of oil/water density relationship limitation. Herein, we demonstrate a low-cost, mild and environment-friendly method to prepare underwater superoleophobic graphene oxide (GO) coated fabric through dip coating of GO nanosheets onto cotton fabric, and the super hydrophilic GO-coated fabric can be converted to superhydrophobic reduced graphene oxide (rGO) coated fabric after chemical reduction of the coated GO. The GO and rGO-coated fabrics show excellent efficiencies for the unidirectional separation of various light oils and heavy oils from water, respectively. Both types of fabrics could maintain their original wettability in harsh environments, and demonstrate high efficiency and recyclability for the separation of oil/water mixtures containing hot water, acid and salt. Furthermore, a T-shaped bidirectional separation device was devised by integrating the two fabrics with opposite wettability, which can realize highly efficient continuous separation of various oil/water mixtures regardless of the oil density.

Keywords: cotton fabric, oil/water separation, graphene oxide, reduced graphene oxide, opposite wettability.

Introduction

The filtration membranes for water/oil separation can be divided into two types according to their wetting natures. One kind is the superhydrophobic/superoleophilic membrane, which is an “oil removal” material and only applicable for separating heavy oil ($\rho_{oil} > \rho_{water}$) from water [1-5]. The other kind is called superhydrophilic/underwater superoleophobic membrane, which can be utilized in discharging water from the mixture of light oil and water (ρoil < ρwater). Nevertheless, such oil-repellent and water-permeable material are unfit for unidirectional separation of heavy oil from water since water will be blocked by the oil layer with higher density sandwiched between the water phase and membrane. In a word, both types of separation membranes are not universally applicable. Therefore, it is necessary to devise a couple of filtration membranes with opposite wettability based on the same materials and assembled them into a separation device to achieve continuous separation of the oil/water mixture regardless of the oil density [6-11].

Graphene oxide (GO), the oxidation state of graphene, is exfoliated from graphite oxide. Possessing carboxyl, hydroxyl, and epoxide functional groups on both sides and borders, GO is super hydrophilic and can be used for constructing underwater superoleophobic surfaces. On the other hand, after removing the oxygen functional groups via chemical, thermal or electrochemical treatment, GO can be converted to reduced graphene oxide (rGO). The lack of hydrophilic oxygenated groups and the restoration of graphitic domains endow rGO with superhydrophobicity and superoleophilicity. Previously, we prepared rGO-based aerogel and foam through in situ reduction assembly of GO and demonstrated their superhydrophobicity and superoleophilicity as well as recyclable absorption of oil from water. Therefore, GO and rGO could be a pair of candidate materials for the construction of filtration membranes with opposite wettability for effective oil/water separation of any oil density [12-17]. Herein, we demonstrate a facile, mild, inexpensive, and green route to fabricate flexible membranes with super hydrophilicity/underwater superoleophobicity and superhydrophobicity/superoleophilicity via dip-coating of GO nanosheets onto cotton fabric and chemical reduction of the GO-coated on fabric, respectively. The GO and rGO-coated fabrics exhibit high efficiencies for unidirectional separation of various light oil/water and heavy oil/water mixtures, respectively. Both fabrics also show excellent heat and corrosion tolerance as well as high efficiencies and recyclability for separating the oil/water mixtures containing hot water, acid, and salt. In addition, we also design a T-shaped bidirectional separation device equipped by the two fabrics with opposite wettability to achieve continuous oil/water separation without consideration of the oil/water density relationship.

Experimental part

Fabrication of GO and rGO-coated cotton fabrics.

Cotton fabrics were first to cut into strips with a width of 30 mm and then washed with water. The GO-coated fabric was prepared by dipping the cotton fabric into a GO suspension with a concentration of 2 mg mL− 1 and subsequent drying it by hanging it in the air. The obtained GO-coated fabric was cut into squares (30 mm × 30 mm) before use. The chemical reduction of GO nanosheet coated on fabric was accomplished by heating the GO-coated fabric in a 0.3 M Vitamin C (VC) aqueous solution under a 95 ◦C water bath for 2 h. After washing with deionized water to remove the redundant reactants and then air-drying, the rGO-coated fabric was obtained [14-18].

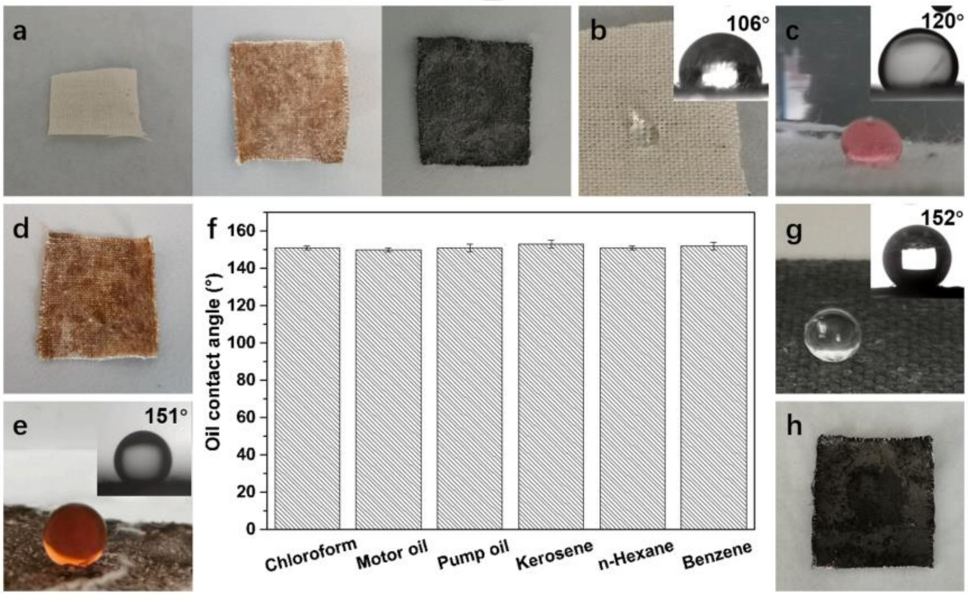

Fig. 1. Photographs of original cotton fabric (left), GO-coated fabric (middle), and rGO-coated fabric (right) (a); water droplet in the air (b) and chloroform droplet in water (c) on the original cotton fabric; water dripped on GO-coated fabric (d); chloroform droplet on GO-coated fabric in water (e); underwater OCAs of the GO-coated fabric for various oils (f); water (g) and chloroform (h) dripped on the rGO-coated fabric.

Water/oil separation

The unidirectional separation device was constructed by fixing the GO or rGO-coated fabric between two quartz tubes using a clip. The separation was accomplished by decanting the mixture of oil and water (m_{oil}: m_{water}, 1: 1) onto the tube above the fabric followed by collecting the permeated phase driven by gravity in a baker blow. The bidirectional T-shaped separation device was assembled by clamping the GO and rGO-coated fabrics at both horizontal sides of a T-tube, respectively. The oil/ water mixture was put into the upward opening of the T-tube, and the permeated oil and water were collected by two beakers beneath the two outlets of the separation device, respectively. The GO-coated fabrics were prewetted by water before oil/water separation. The separation efficiency was computed according to the equation $\eta = m^2/m1$, where m1 and m^2 are the weight of oil or water before mixing and after separation, respectively.

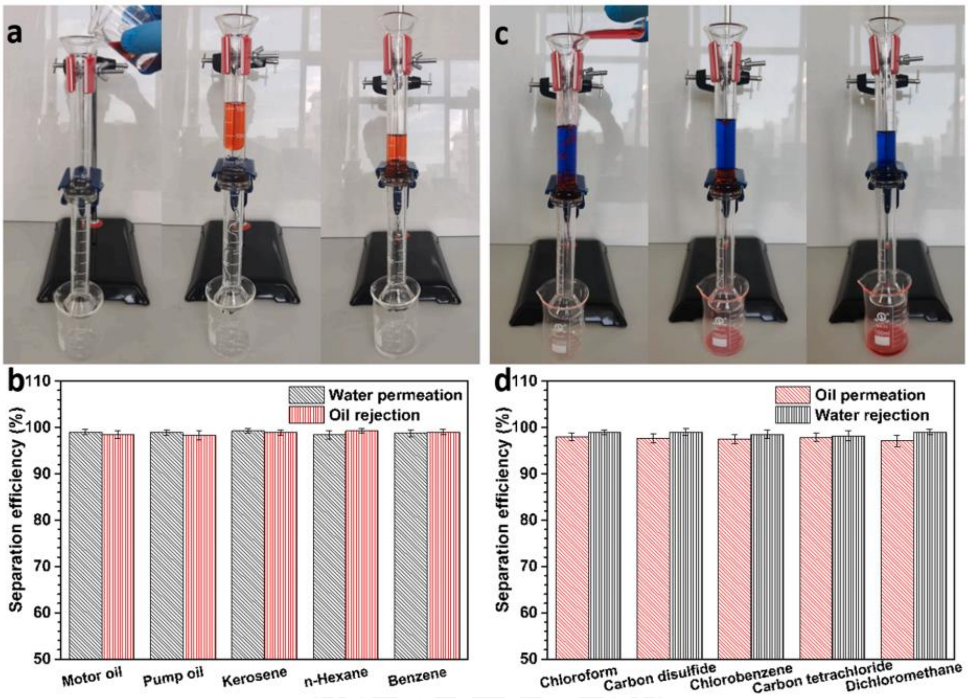

Fig. 2. Unidirectional separation process of the mixture of motor oil and water using GO-coated fabric (a); separation efficiency of the GO-coated fabric for the mixtures of various light oil and water in terms of water permeation and oil rejection coefficient (b); unidirectional separation process of the mixture of chloroform and water using rGO-coated fabric (c); separation efficiency of the rGO-coated fabric for the mixtures of various heavy oil and water in terms of oil permeation and water rejection coefficient (d).

Conclusions

In summary, GO and rGO-coated fabrics with underwater superoleophobicity and superhydrophobicity were prepared through dip-coating of GO sheets on cotton fabric and chemical reduction of the GO-coated fabric, respectively. The underwater superoleophobic GO-coated fabric exhibits separation efficiencies above 98.3% for removing water from various light oils, and the superhydrophobic rGO-coated fabric can remove various oils from water with separation efficiencies over 97.1%. The two fabrics also maintain their original wettability in harsh environments and show high efficiencies for the separation of oil/water mixtures containing hot water, acid, and salt. In addition, the T-shaped bidirectional separation device equipped with the two fabrics presents excellent capability for continuous separation of various mixtures of oil and water without consideration of their density relationship, demonstrating its potential application in practical oily wastewater treatment.

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