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Integrated device based on cauliflower-like nickel hydroxide particles–coated fabrics with inverse wettability for highly efficient oil/hot alkaline water separation

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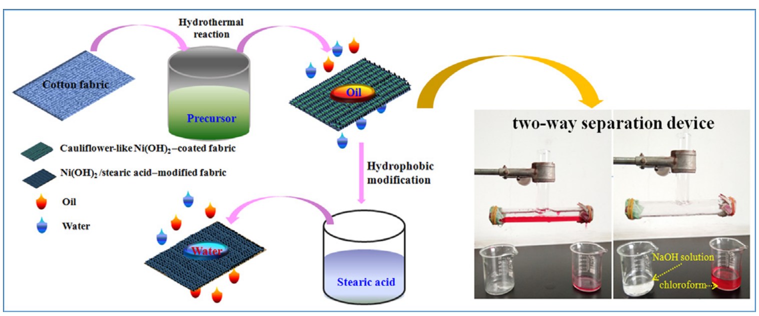
GRAPHICAL ABSTRACT



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ABSTRACT

In this paper, we report for the first time the fabrication of cauliflower-like $Ni(OH)_2$ particles–coated fabrics with inverse special wettability via a facile hydrothermal method. The coated fabric is superhydrophilic in air and superoleophobic underwater, which can be changed into superhydrophobicity after being modified by long-chain fatty acid. After the superhydrophilic fabric is wetted by water in advance, hot alkaline water can be removed from the light oil/water mixture driven by gravity. Also, heavy oil can be removed from the heavy oil/hot alkaline water mixture under the action of gravity using superhydrophobic fabric. The separation efficiencies of the two fabrics for different mixtures of hot alkaline water and light or heavy oil are more than 95.8%. Furthermore, a two-way separation device was designed via integrating a pair of fabrics with opposite wettability. The device can continuously separate oil and water at the same time from oil/hot alkaline water mixture via one-step route with high separation efficiency without consideration of the density of the oil. Moreover, the superhydrophilic fabric is capable of separating various oil-in-water emulsions. We believe that the as-prepared fabrics and the designed device may find wide application in separating oil from alkaline water.

1. Introduction

In recent years, the oil/water separation have drawn great attention due to the occurrence of frequent oil spill accidents and the discharge of a large amount of oily wastewater [1–3]. In consideration of the interfacial process of oil/water separation, immiscible oil/water mixture can be effectively separated using the membranes with special wettability, namely super-repellency and superwetting. So far, a large number of superhydrophobic/superoleophilic membranes, such as polytetrafluoroethylene modified mesh [4], silicone elastomer coated copper mesh [5],

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polycarbonate resin coated mesh [6], MnO_2 nanocrystal decorated mesh [7], polylactic acid nonwoven fabric [8], and Cu nanoparticles coated fabrics [9], have been successfully used for the separation of oil/water mixtures. However, this type of membrane has certain limitation in separating light oil/alkaline water mixtures, because water with higher density than most of oils is liable to form a barrier layer between the oil and the superhydrophobic surface during the separation process, which in turn prevents the permeation of oil through the membrane. In addition, these membranes are easy to be polluted by the oils with high-viscosity and corroded by alkaline water to loss superhydrophobicity during oil/water separation process [10–11]. To handle the membrane fouling by oils, underwater superoleophobic membranes with high separation efficiency, ultrafast separation rate, and good resistance to oil fouling have been successfully developed [12–14]. This type of membrane can efficiently separate oil/water mixture especially without any continuous external stimulus, showing a great potential of application in oily wastewater treatment.

According to the principle that the superhydrophilicity of a surface at air/solid interface is essential to the superoleophobicity at water/solid interface [15,16], a series of underwater superoleophobic surfaces based on various materials have been prepared for efficient oil/water separation. For instance, Shi et al. anchored TiO_2 nanoparticles on the surface of poly(vinylidene fluoride) (PVDF) membrane to obtain underwater superoleophobic membranes that can be used for separating various oil-in-water emulsion [17]. Yang et al. reported a kind of superhydrophilic and underwater superoleophobic PVDF membrane by grafting 3-aminopropyltriethoxysilane, which can separate different oil-in-water emulsions with high permeate flux [18]. Ma et al. fabricated nanofiber-based meshes derived from waste glass, and the resulting meshes exhibit excellent underwater superoleophobicity and can maintain high oil/water separation efficiency under harsh conditions [19]. Song et al. demonstrated an underwater superoleophobic $BiVO_4$ coated mesh being capable of separating oil/water mixture and decomposing organic contaminants in water under simulated solar irradiation [20]. Nevertheless, despite the fact that underwater superoleophobic membranes have been extensively studied, most of membranes still suffer from problems in separation process like alkaline water resistance and thermal stability, which cannot satisfy the demands of continuous oil/water separation. Besides, the regeneration of oil-fouled membranes as polydopamine modified PVDF membrane [21] and tannin-inspired polypropylene membrane [22] is also a great challenge. To resolve the above-mentioned issues, some membranes based on inorganic coating, such as ZnO nanowires coated stainless steel mesh [23], polyethyleneimine/ZnO nanorods modified polyester fabric [24], and mica particles deposited stainless steel mesh [25], have been fabricated and applied in oil/water separation. In particular, Chen et al. prepared a Co_3O_4 nano-needle steel mesh via a hydrothermal synthesis and subsequent calcination method, which realizes the separation of oil-in-water emulsions [26]. Despite this, to the best of our knowledge, membranes with a single wetting performance can only be applied to separate definite oil/water mixtures. For instance, superhydrophobic/superoleophilic membranes can only be used to separate heavy oil/water mixtures, and underwater superoleophobic membranes are suitable for separating light oil/water mixtures [27]. The successive separation of light oil/water and heavy oil/water mixture using the same device designed by inexpensive and biodegradable membranes is extremely rare.

Herein, we report for the first time a cauliflower-like $Ni(OH)_2$ particles–coated fabric fabricated by a facile hydrothermal technique. The as-prepared fabric demonstrates superhydrophilicity in air and superoleophobicity underwater, and the coated fabric can be endowed with superhydrophobicity after modifying with stearic acid. Thus, the as-obtained fabrics can be used to selectively separate oil or highly alkaline water from the oil/water mixtures according to the density of oil. When oil density is lower than alkaline water density, the coated fabric with underwater superoleophobicity can be utilized for removing alkaline water from the oil/water mixture. If oil density is higher than alkaline water density, the modified fabric with superhydrophobicity is able to selectively remove heavy oil from the oil/water mixture. The as-prepared fabrics reveal excellent separation efficiency up to 95.8% for a variety of oil/alkaline water mixtures. More importantly, a special oil/water separation device equipped with a pair of fabrics with opposite wettability (underwater superoleophobicity and superhydrophobicity) is also designed, which can continuously separate both oil and hot alkaline water from the mixtures at the same time via one-step route regardless of the relative density of oil and water. Furthermore, the underwater superoleophobic fabric is able to separate a series of surfactant-free oil-in-water emulsions with high efficiency (>93.0%). To the best of our knowledge, it is the first time to design an oil/water separation device based on cauliflower-like $Ni(OH)_2$ particles–coated fabrics to use for selectively separating the mixture of hot alkaline water and light or heavy oil under gravity.

2. Experimental

2.1. Materials

Cotton fabric was obtained from market, Yinchuan, China. Urea (>98%), nickel sulfate hexahydrate ($NiSO_4 \cdot 6H_2O$, >98%), stearic acid (>99%), n-hexane, toluene, chloroform, kerosene, and ethanol (analytical pure) were received from Sinopharm Chemical Reagent Co., Ltd., China. Gasoline and diesel were purchased from the local gas station, Yinchuan, China.

2.2. Preparation of underwater superoleophobic and superhydrophobic fabrics

Cotton fabrics were rinsed with distilled water and ethanol before use. The fabrication process of underwater superoleophobic and superhydrophobic fabrics with special wettability are displayed Fig. 1a. Firstly, urea (0.5 M) and nickel sulfate hexahydrate (0.2 M) were dissolved into distilled water, and then the solution was transferred into the hydrothermal synthesis reactor. After clean fabric was added into the reactor, the reaction system was sealed, heated to 90 °C, and reacted for 12 h in an oven. Finally, the coated fabric was removed from the solution, washed, and then dried to a constant weight. To achieve superhydrophobicity, the coated fabric was immersed in stearic acid solution (4 mM) for 10 h, and superhydrophobic fabric will be obtained.

2.3. Separation of oil/water mixture

The underwater superoleophobic or superhydrophobic fabric was fixed onto the separation device. A series of oil/water mixtures (V_{oil}:V_{water}, 1:4) were poured into the upper funnel and the separation process was carried out only under the gravity drive. For the fabric with underwater superoleophobicity, it needs to be wetted by water before performing oil/water separation. Water or oil was harvested in the conical flask below the funnel. The separation efficiency (E) was calculated by $E = (w_1/w_0) \times 100\%$, where w_1 is the weight of collected water or oil after separation, w_0 is the weight of the original water or oil before separation. All separation efficiency was measured three times, and an average value was used.