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Superhydrophilic and underwater superoleophobic MFI zeolite-coated film for oil/water separation

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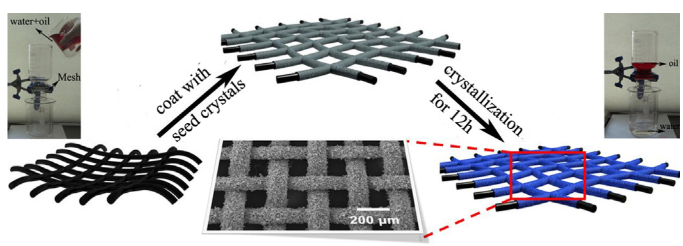
HIGHLIGHTS

- MFI-type zeolite membranes were synthesized by the seed growth hydrothermal method.
- The separation of oil/water mixture was demonstrated by the contact angle and movie.
- The film can be used for several times keeping the high separation efficiency.

GRAPHICAL ABSTRACT



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ABSTRACT

Silicalite-1 (structure type MFI), an important type of zeolite, was prepared on the porous stainless steel wire by seed growth hydrothermal synthesis, forming an amazing film. The coated film consists of well-intergrown crystals with hexagonal prismatic shape (coffin shape), which shows superamphiphilic in air and superoleophobic underwater. This prepared film can effectively collect the oil from oil/water mixture underwater driven by gravity for several times, showing good durability and high separation efficiency, which is very helpful in the promising application of energy-efficient membrance for reducing the environmental impacts of oil spills. This work provides an alternative solution to current separation mesh based-on the surface wettability.

1. Introduction

Recently, increasing attention has been drawn to find solutions for tackling oily wastewater in the environment after oil spillages [1,2]. Separation of oil/water mixture is always difficult and challenging to clean up oil spills or skim from the surface of water. Many methods reported currently are employed to improve this problem, including burning in situ and use of oil containment booms, oil skinners, oil-absorbing materials [3,4]. But most of them are not ideal for the reasons that most spilled oil is wasted or becomes pollutant in the environment [5]. One of the conventional anti-oil pollution is based on the oleophilic materials which can absorb the oil pollutions. Although such materials are always one-off for the absorbed oils which are hard to remove, some modified sponges with both superhydrophobic and superoleophilic properties can be employed for separation and absorption of organic contaminants form water [6]. Moreover, the efficiency of separation will decrease more or less due to the residual oils inside the polyporous sponges.

Another common material to separate the mixture is a membrane with the surface of special wettability. It can avoid the difficulty to remove the absorbed oil. Especially, superhydrophobic–superoleophilic or superoleophobic–superhydrophilic materials can realize the need of separation effectively. For example, the use of superhydrophobic-superoleophilic meshes can

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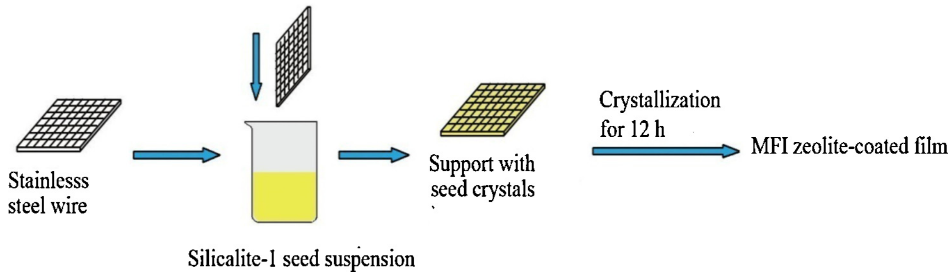

Fig. 1. Schematic map for the fabrication of continuous and compact MFI zeolite-coated film on stainless steel wire.

separate the oil/water mixtures by allowing the oils to fall through the pores in the mesh whist retaining the water on the surface [7]. Cricka et al. [8] reported a strategy to fabricate superhydrophobic polymer-coated mesh via aerosol assisted chemical vapor deposition. Jin et al. [9] fabricated the organosilane surface which is superamphiphobic in air and superoleophilic under water by imitating the "desert beetle effect" underwater. Despite a lot of researches have been done on the materials with selective absorption or organic solvents while repelling water completely, creating membrane-based materials for practical applications in oil–water separation is still a challenge.

Zeolite is commonly used as a kind of catalyst in various applications with special composition and nanoporous structures. The earliest reports about zeolite membrane are mainly their applications in ion-selective sensing [10–12]. Hereafter, about their applications extended to other important fields such as substance separating, membrane reactors [13], chemical sensor, ion-exchange electrodes, light-harvesting. Besides, zeolite membranes with excellent separation performance in various fields can be applied in gas separation, catalysis, liquid separation. Among a range of zeolites, the structure-type MFI gets more and more attention as a model of zeolite system, which has selective separation properties for hydrocarbon isomers, organic/water and other permeate gas mixture [14–20]. Zeolite coatings are in general prepared either by a direct one-step hydrothermal synthesis in situ method [21,22] or the seeds growth [23,24]. The latter method needs non-silicious solid surfaces (e.g. stainless steel, alumina) as substrates which are pre-seeded with crystals. Liu et al. [19] prepared a functional stainless-steel tube coated with silicalite zeolite by pervaporation, being used for the separation of organic/water mixture. Sano et al. [25] fabricated a pure silicalite membrane with a high permselectivity for ethanol to separate the ethanol/water mixture. In addition, zeolite membranes have attracted increasing attention for their excellent chemical resistance to acids and salt solutions. Recently, the zeolite membranes for separation of oil/water mixture were also reported for their resistant to corrosion [26]. So, these membranes with special separation underwater can be used in harsh condition, possessing the promising applications to tackle the oil-spilled problems. Among them, the zeolite membranes synthesized are almost focused on the use of structure-directing agents (SDA). However, the major drawback of these syntheses is that alkali-free tetrapropylammonium hydroxide (TPAOH) templates are too costly to be used in industrial application. In our work, we chose the organic amine-tetrapropylammonium bromide

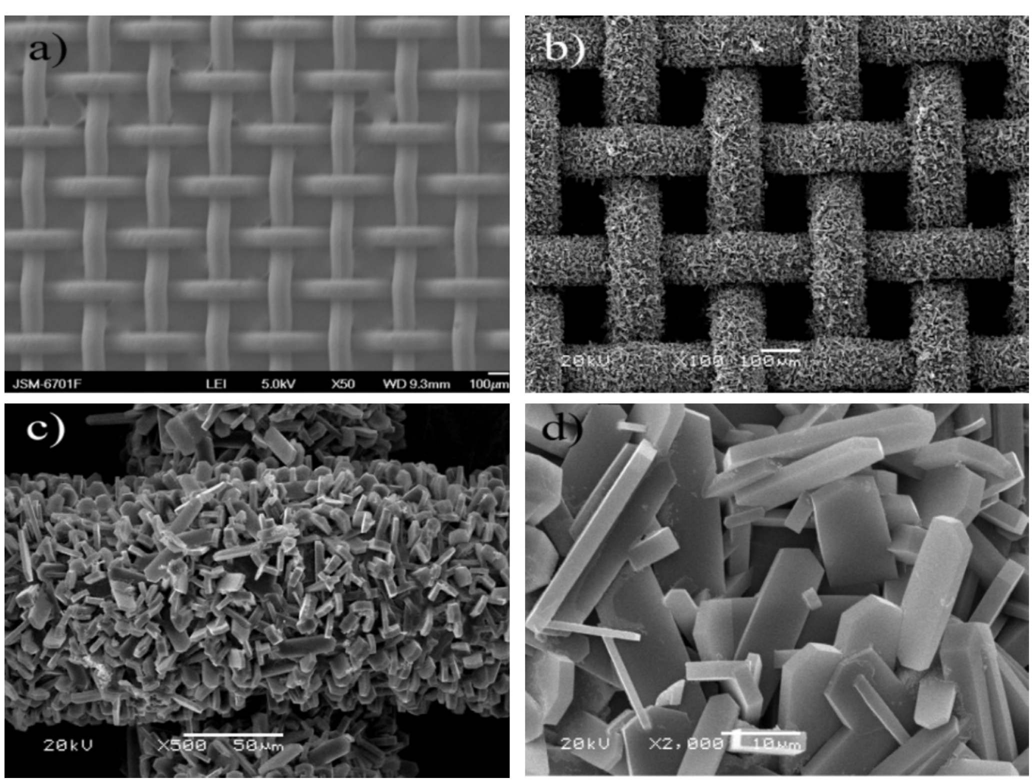

Fig. 2. (a) Large view of bare stainless steel mesh, (b) large scale image of zeolite-coated stainless steel mesh, (c) top images of zeolite on a stainless steel wire, (d) high-magnification zeolite crystals on a stainless steel wire.