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Research article

Fabrication of highly hydrophilic filter using natural and hydrothermally treated mica nanoparticles for efficient waste oil-water separation

Udara Bimendra Gunatilake, Jayasundera Bandara*

National Institute of Fundamental Studies, Hantana Road, CP 20000, Kandy, Sri Lanka

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ABSTRACT

For the effective oil/water separation, a novel superhydrophilic (underwater superoleophobic) filter is fabricated with the naturally and hydrothermally treated mica particles. To fabricate a double layered filter, hydrothermally treated mica particles were initially electrodeposited on a stainless steel mesh and a natural mica particles were sprayed on the first hydrothermally deposited mica layer. The double layered mica coated membrane showed superamphiphilic and superhydrophilic/superoleophobic (contact angle >159°) characteristics in air and underwater respectively. The membrane can separate range of oil-water mixtures with oil/water separation efficiency over ~99%. Properties of double layered mica membrane were investigated and noted that the surface adhesion properties of mica is enhanced by the hydrothermal treatment of mica and the higher roughness of the mica layer is maintained by the natural mica.

1. Introduction

In the modern world, water which is one of the scarcest commodities is polluting by several ways in every second. Among the different methods of water pollution, oil spill is one of the major negative contributions to the environment from large number of industries, especially in automobile service stations (Fried et al., 1979; Mazumder and Mukherjee, 2011; Reed et al., 1998). It is known that the prompt water pollution occurs when oil passes in to the water sources, rivers and sea water and the water pollution by oil is considered to be a greater threat to the humans, marine life, animals and to the entire ecosystem (Fosberg, 1974; Nomack, 2010).In past few decades, the following water treatment techniques have been used; (a) *physical treatment methods* such as adsorption of dissolved organics on activated carbon (Gur-Reznik et al., 2008), organoclays (Doyle and Brown, 2000), copolymers and resins, (Jan and Reed, 1989) sand/stone filters, organics evaporating methods (Fakhru'l-Razi et al., 2009), electrodialysis methods (Fakhru'l-Razi et al., 2009), and filtering through oil/water separating membranes, (Kota et al., 2012); (b) *chemical/electrochemical treatment methods* such as coagulation and flocculation methods (Bratby, 1980), advanced chemical oxidation methods (Bautista et al., 2008), electrochemical methods (Ma and Wang, 2006) and photocatalytic treatment methods (Chong et al., 2010; Teng et al., 2015; Gunatilake et al., 2017) and (c) *biological treatment methods* (Fakhru'l-Razi et al., 2009; Sugano et al., 2008; Teng et al., 2015). The major disadvantages of most of these conventional techniques are that these methods are not economical as well as they cannot separate oil/water emulsions efficiently. As a result, the scientists are looking forward to capitalize the cheap and efficient methods to remove oil spills from water.

Contrary to the conventional oil/water separation techniques mentioned earlier, extremely high special wetting solid material surfaces can be used to separate oil and water more selectively and easily (Feng and Jiang, 2006; Li et al., 2016; Sun et al., 2014; Xue et al., 2014). If wetting properties are considered, materials having different affinities towards oil and water would be the best choice for designing of efficient oil/water separation filters. In this respect, materials with both hydrophobic/oleophilic and oleophobic/hydrophilic properties were actuated a broad attention in last few years to invent novel oil/water separation systems (Bellanger et al., 2014; Darmanin and Guittard, 2014; Xue et al., 2014). However, superhydrophobic materials are unsuitable for the separation of oil/water under gravity due to formation of a water barrier layer on superhydrophobic layer preventing

* Corresponding author.
E-mail addresses: bandaraj@ifs.ac.lk, jayasundera@yahoo.com (J. Bandara).

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penetration of oils as water has higher density than most of the common oils (Zhang et al., 2014). On the other hand, underwater superoleophobic and superhydrophilic solid surfaces allow water to penetrate through the membrane while repelling the oil outwards from the membrane allowing separation of oil/water effectively. Going through these theories, various materials such as polyacrylamide hydrogel coated mesh (Xue et al., 2011), $Cu(OH)_2$ covered mesh (Liu et al., 2013), photo-induced TiO_2 thin films, Sawai et al., 2013) polyelectrolyte/clay hybrid film (Xu et al., 2013), PANI (Polyaniline) nanowire film (Ding et al., 2012), polypyrrole nanowire mesh (Ding et al., 2012), have been fabricated to separate oil/water.

Here, we report the fabrication of a novel superhydrophilic and underwater superoleophobic filter to repel and separate oil under water efficiently by using natural mica mineral particles coated stainless steel mesh. The superhydrophilicity nature and micro hierarchical rough particles layered arrangement of the mica particles leads to enhance the underwater superoleophobicity of the filter. The fabricated filter selectively and effectively repel not only low dense oils but also high dense organic oils from oil/water mixtures such as engine oil, crude oil, gasoline, diesel, vegetable oil and forth on. The oil-water separation filter described in this investigation is a low cost method as the main chemical substrate (mica) is taken from a natural source directly. Interestingly, the double layered electrodeposition and spray techniques makes the mica coated mesh more stable under high peeling off forces. Additionally, the cleaning of the mesh is very easy with the help of non-polar solvent at the high contamination levels.

2. Experimental

2.1. Materials

Stainless steel meshes were purchased from Benny the Stoog, USA. Acetone, sodium hydroxide pellets, absolute ethanol, 1,2-dichloroethane,1-octadecene, hexane, and potassium bromide were purchased from Sigma Aldrich chemicals Pvt. Limited, USA. Silicon oil (commercial grade), kerosene (commercial grade), engine oil (SAE 10W-30), rapeseed oil (commercial grade) was purchased commercially and mica flakes were obtained from mica deposit found in Matale, Sri Lanka.

2.2. Fabrication of mica coated stainless steel mesh

Natural flake mica (from Matale, Sri Lanka) was finely ground from ring mill crusher and sieved to collect <50 μm size mica particles. For electrodeposition of mica, mica particles were hydrothermally treated to make them negatively charged. For hydrothermal treatment, 5.0 g of mica was stirred with 55.0 ml of 10 M NaOH for 30 min and the mixture was transferred to a 73.0 ml capacity Teflon chamber with a stainless steel hydrothermal vessel and treated at 160 °C for 12 h. The highly basic (pH ~13) hydrothermally modified mica particles were separated at room temperature by washing with water several times till pH of the solution reaches ~ 10.0–11.0. Stainless steel meshes (#125/100 μm opening width, size 2" × 2") were ultrasonically cleaned in acetone and then rinsed with ethanol and deionized water. For electrodeposition of hydrothermally modified mica, 20 ml of methanol was mixed with 40 ml of hydrothermally modified mica suspension solution. Then two cleaned stainless steel meshes were used as anode and cathode with one inch space difference and 30 V was supplied for 3 min each for both sides of anode mesh. On top of the electrodeposited mica layer, a second mica layer which was prepared by dispersing 1.0 g of natural mica particles in 30 ml of ethanol followed by ultrasonication (15 min) and stirring for 1 h, was deposited by spray method. Finally, the mesh was sintered at 650 °C for 1 h and cooled to room temperature. Finally the mica coated mesh was rinsed with deionized water to remove excess loosely bound mica particles. In this study, three different underwater superoleophobic filters were fabricated; (1) natural mica was sprayed on stainless steel mesh (filter-a), (2) hydrothermally treated mica was electrophoretically deposited on stainless steel mesh (filter-b) and (3) bi-layer filter containing electrophoretic deposited hydrothermally treated mica on stainless steel mesh and sprayed mica layer on the electrodeposited mica layer (filter-c).

2.3. Oil/water separation experiments

The oil/water separation apparatus was made by fixing mica coated stainless steel mesh into a PVC union setup. The oil/water mixture was prepared by mixing 10.0 g of engine oil and 35.0 g of water. The mica mesh was pre-wetted with water and the water/oil mixture (3.5:1 w/w) was poured to the filtration set up (1.0 inch diameter filtering area) at the rate of 1.5 ml/s under the gravity. Water was collected by a beaker under the PVC union setup. To check the stability of the mesh, the filtration was done continuously for several days. After every filtration step, the mica mesh was rinsed by distilled water to remove the trapped oil droplets from the mesh. The separation efficiency was calculated by $\vartheta = \frac{m\ remains}{m\ initial} \times 100\%$ where, $m_{\ remains}$ and $m_{\ initial}$ is the mass of the oil before and after the separation. For the calculation of $m_{\ remain}$ in the filter, oil remain in the filter was collected to a beaker and the oil residues on the column was wiped with a solvent carefully and weight of the oil was taken after evaporation of the solvent. This procedure was repeated for several oils to get the efficiency of each and the recycle efficiency was checked for 20 cycles with engine oil (SAE 10W-30).

2.4. Instrumentation and characterization

Scanning electron microscope (SEM) images and EDX were taken by Carl Zeiss EVO LS15 scanning electron microscope, Fourier transform infrared spectroscopy (FTIR) was recorded from Thermo Nicolet 6700 FTIR machine. Contact angle images were captured from 8-megapixel iSight camera and contact angles were measured using simple geometric angle measuring Protractor. Crystallographic data was taken from Ultima IV x-ray diffractometer.

3. Results and discussion

3.1. Characterization of natural and hydrothermally modified mica

Mica is a phyllosilicates mineral which forms as layer silicates comprising of Si–Al–O tetrahedral sheets between M-O and OH octahedral sheets, where M is usually Al^{3+}, Fe^{2+}, Fe^{3+} or Mg^{2+}. It mostly exists as Biotite $K(Mg,Fe)_3(AlSi_3O_{10})\ (OH)_2$, Lepidolite $K(Li,Al)_3(AlSi_3O_{10})\ (O,OH,F)_2$, Phlogopite $KMg_3(AlSi_3O_{10})\ (OH)_2$ and Muscovite $KAl_2(AlSi_3O_{10})\ (OH)_2$ structures (Laoot et al., 2011; Rickwood, 1981). High in silica, the mica group sheet silicate minerals include several closely related materials having nearly perfect basal cleavage which can be split or delaminated into thin sheets usually causing foliation in rocks. Mica is a lightweight mineral having dielectric and hydrophilic characteristics. More importantly, mica is chemically inert material and can resist nearly all mediums like chemicals, acids, gasses, alkalis and oils which makes it an ideal material for the fabrication of oil-water separation filters (Christenson, 1993).

Strong bonding between mica and steel mesh is a prerequisite for the fabrication of durable filter for the separation of water and oil. However, it was noticed that a filter made of natural mica by