



Superoleophilic and under-oil superhydrophobic organogel coatings for oil and water separation

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ABSTRACT

The development of an effective and mechanically robust organogel coating that is both superoleophilic and under-oil superhydrophobic is presented. The organogel was shown to absorb and retain oil, and as a consequence render the surface superhydrophobic. The coating showed high repellence against water while letting oil pass through without the need to apply pressure. Utilization of the coating in oil and water separation was investigated and exhibited $\geq 99\%$ separation efficiency for a wide variety of commercial oil. The coating was also shown to be mechanically robust and can withstand shear tests of different strengths and duration.

1. Introduction

The demand for petroleum has increased tremendously in proportion to the increase of ownership of vehicles. Global vehicles usage has increased by more than 34 million units from 2005 to 2014 [1]. Furthermore, petroleum is a useful raw material in creating petrochemical plastics [2]. In offshore oil and gas production platforms, separation of oil and water is necessary for disposing produced water to the environment. However, mishandling of petroleum products and accidents like oil spill can have huge environmental, economic, and social impacts. For example, the Deepwater Horizon oil spill that occurred in the Gulf of Mexico in 2010 is one of the worst oil spill in history of United States. It was estimated that 4.9 million barrels of oil was released into the ocean [3,4]. Another tragic incident happened when over 1 million gallon of crude oil was spilled into Israel's Evrona Nature Reserve in December 2014 because of a burst pipeline [5]. In the same year, an accident between a bulk carrier and an oil barge resulted in 168,000 gallons of oil spill at Galveston Bay [6]. Oil spills are so frequent that it is crucial to come up with solutions that are effective, efficient, and cheap. There have been a number of ways to remedy oil spills, however most of them involve use of expensive and complicated equipment or requires burning of oil that can further harm the environment [7,8]. It is very critical to develop an affordable and efficient technique for oil

and water separation to minimize the impact of future oil spills [9].

Superhydrophobicity is characterized by high water repellence [10]. A surface can only be classified as superhydrophobic if the contact angle between a water droplet and the surface is at least 150° [11–13]. On the contrary, superoleophilicity is defined as a property that implies a high affinity to oils. A surface can only be categorized as superoleophilic if the contact angle between an oil droplet and the surface is less than 10° [14]. When a coating is said to exhibit both superhydrophobicity and superoleophilicity, the former will cause the surface to selectively repel water molecules while the latter will cause the surface to allow oil to be absorbed [15]. Polymers that are both superhydrophobic and superoleophilic have remarkably high selectivity with respect to its affinity towards liquids of varying polarity, which makes them suitable for oil and water separation [16–18]. Aside from separating a mixture of oil and water, superhydrophobic and/or superoleophilic surfaces have been used as anti-fog, anti-corrosion, and self-cleaning coatings [19–23].

Currently, there are a number of practices to treat oil spills, such as oil containment booms, and in situ combustion. Furthermore, oil-absorbing materials, including polypropylene-based felts, zeolites, activated carbon, organoclays, straw, hair, and wool fibers, have been utilized [24–27]. Some of these treatments have high oil and water separation efficiency but poor recyclability. Others require expensive

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materials to prepare and involve many complicated steps. As a result, there is still a need to find a simpler, more robust, and cheap alternative to treat large-scale oil spill. Xue et al. fabricated a superhydrophilic and underwater superoleophobic hydrogel-coated mesh that selectively let water pass through the mesh while retaining the oil [28]. The stability of the coated mesh was established after undergoing 50 separation cycles. The use of hydrogel, however, may cause a number of problems such as the ease for the water to evaporate, sensitivity of the hydrogel towards water with high salinity, and dependence to pH. It is, therefore, attractive to develop a coating that is unaffected by the salinity and pH. Li et al. coated a mesh with superhydrophilic-underwater superoleophobic ZnO [29]. Separation efficiency of 99% was achieved for kerosene-water mixture, however, it did not perform as well for other systems (petroleum ether, hexane, toluene and rapeseed oil). Lastly, Wen et al. reported a zeolite-coated mesh film for efficient oil-water separation [30]. However, their fabrication process is complicated and their separation efficiency was not reported.

In this study, we demonstrated that an organogel-coated mesh can separate oil and water from its mixture. The organogel imparts both superoleophilicity and under-oil superhydrophobicity that selectively repels water while letting oil pass through. The resulting system proved to be highly selective, robust, and can be easily fabricated. To the best of our knowledge, this is the first time that an organogel has been used as a selective gate that lets oil through but not water. Aside from wettability, the organogel-coated mesh has also been studied for separation efficiency, intrusion pressure, thermal stability, and mechanical stability.

2. Experimental

2.1. Materials

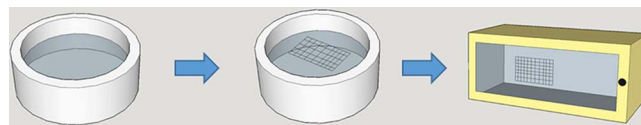
Chemical reagents were purchased from Sigma Aldrich and were used without further purification. Lauryl methacrylate, ethylene glycol dimethacrylate (EGDMA), 3(trimethoxysilyl) propyl methacrylate (TMS-MA), 2,2'-Azobis(2-methylpropionitrile) (AIBN), toluene, silicon oil, kerosene, crude oil, cooking oil, and dichloromethane.

2.2. Characterization

FTIR was done using Cary 600 by Agilent Technologies in ATR mode. Survey and high resolution XPS were performed using PHI Versaprobe 5000 X-ray photoelectron spectrometer with Al K α radiation and was referenced to internal SiO₂. Contact angles were measured using CAM 200 optical goniometer by KSV Instrument, Ltd. Thermogravimetric analysis was done using TGA 2050 system by TA Instruments set at 10 °C/min.

2.3. Coating preparation

Our coating was prepared by mixing 3.323 mL (0.144 mol) of lauryl methacrylate, 139.2 μ L (7.365×10^{-4} mol) of ethylene glycol dimethacrylate (EGDMA), 139.2 μ L (5.839×10^{-4} mol) of 3-(trimethoxysilyl) propyl methacrylate (TMS-MA), 29 mg (1.766×10^{-4} mol) of 2,2'-Azobis(2-methylpropionitrile) (AIBN) and using 5.8 mL of toluene as solvent. The mesh was then immersed in the solution for 10 min, dried and polymerized/crosslinked in an oven set at 100 °C for 24 h.



Scheme 1. Schematic procedure for the coating process: dip-coating, polymerization, and curing of the superoleophilic and under-oil superhydrophobic organogel on stainless steel mesh.

3. Results and discussion

3.1. Fabrication of organogel

The key to having a cost-effective method for oil and water separation is the simplicity and repeatability of its preparation. To introduce the superoleophilicity and under-oil superhydrophobicity onto a stainless mesh, we formulated a coating that contains a monomer (lauryl methacrylate) that can polymerize, absorb oil, and render the surface superhydrophobic, a cross-linker (ethylene glycol dimethacrylate) that can improve the mechanical integrity of the coating under high pressure and shear stress, and an adhesive molecule (3-(trimethoxysilyl)propyl methacrylate) that can react with the monomer and cross-linker and ensure the coating stays on the mesh (Scheme 2). AIBN was also added into the mixture to initiate the free radical polymerization of the acrylate and methacrylate monomers, thus, forming a highly cross-linked network. Stainless steel mesh of varying mesh counts (120, 145, 150) was soaked in the formulation for few minutes to have the components adsorb on the surface and a silane covalently attach through the hydroxyl groups of the steel surface. The higher the mesh counts, the smaller the area of mesh openings. Using meshes with higher mesh counts improves the performance of oil/water separation because it is more difficult for water droplets to pass through the smaller openings since the organogel coating can better block water molecules from passing through smaller openings compared to that of larger openings. The coating is subsequently dried, and the polymerization and curing were initiated by heating the coated mesh at 100 °C for 24 h (Scheme 1).

3.2. Characterization of organogel

To confirm the successful polymerization of the organogel on the stainless steel mesh, infrared spectroscopy (FT-IR) was performed. The infrared spectrum (Fig. 1 (a)) of the pure organogel is characterized by a sharp peak at 1728 cm⁻¹ (ester C=O stretching), and at 1150 cm⁻¹ (C–O–C stretching). Moreover, peaks at 2960 cm⁻¹ and 2861 cm⁻¹ correspond to the CH₃ asymmetric and symmetric stretchings, respectively. On the other hand, the infrared spectrum of uncoated stainless steel shows a peak at 600 cm⁻¹, which corresponds to the vibration of Fe–O. Infrared spectrum of the organogel-coated mesh confirms the successful polymerization of lauryl methacrylate in the presence of cross-linker and adhesive molecule. This result was further confirmed by performing survey (Fig. 1(b)) and carbon high resolution (Fig. 1(c)) X-ray photoelectron spectroscopy (XPS) scans of the coated mesh. The coated mesh was cut to smaller pieces, which were then used as the samples for the survey. The survey scan shows peaks at 284.4 eV for C, 531.5 eV for O, and 153.2 eV for Si2s. Moreover, the high resolution carbon scan shows the presence of at least three oxidation states of carbon, namely C–C at 284.4 eV, C–O at 286 eV, and C=O at 289 eV. Thermogravimetric analysis (TGA) was performed on the organogel to study its thermal

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