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Facile Fabrication of a Polyethylene Mesh for Oil/Water Separation in a Complex Environment

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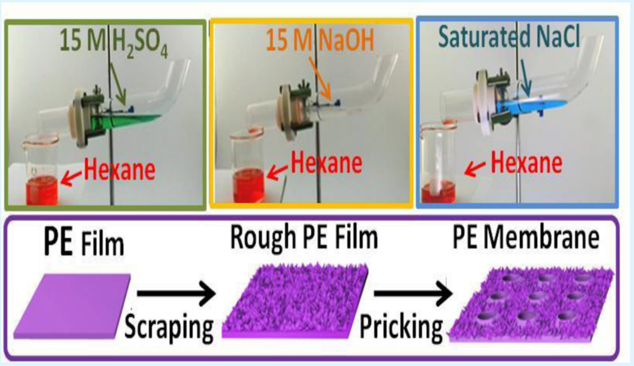

ABSTRACT: Low cost, eco-friendly, and easily scaled-up processes are needed to fabricate efficient oil/water separation materials, especially those useful in harsh environments such as highly acidic, alkaline, and salty environments, to deal with serious oil spills and industrial organic pollutants. Herein, a highly efficient oil/water separation mesh with durable chemical stability was fabricated by simply scratching and pricking a conventional polyethylene (PE) film. Multiscaled morphologies were obtained by this scratching and pricking process and provided the mesh with a special wettability performance termed superhydrophobicity, superoleophilicity, and low water adhesion, while the inert chemical properties of PE delivered chemical etching resistance to the fabricated mesh. In addition to a highly efficient oil/corrosive liquid separation, the fabricated PE mesh was also reusable and exhibited ultrafast oil/water separation solely by gravity. The easy operation, chemical durability, reusability, and efficiency of the novel PE mesh give it high potential for use in industrial and consumer applications.

KEYWORDS: *superhydrophobicity, oil/water separation, recyclability, durability, complex environment*

1. INTRODUCTION

Oil/water separation is an urgent worldwide challenge, because of the upsurge of water pollution caused by industrial oily wastewater and/or frequent oil spills.[1−9] Traditional techniques for oil/water separation have been perfected in the past decades, including gravity separation,[10−12] membrane filtration,[13−16] air flotation,[17−19] adsorption,[17,20−22] coagulation, and flocculation.[23] However, new approaches based on special wettability have been considered to be more effective and more advantageous for use, because oil/water separation is essentially an interfacial matter.[24,25] Various superwetting interfacial materials fabricated by designing superhydrophobic or superoleophobic surfaces have generated great attention and have been applied for industrial, oily wastewater treatment, including chemical etching, electrospinning, self-assembly processes, and others.[12,26−30] Use of the special wettability materials can be generally categorized by the following two strategies: "oil-removing" type materials with superhydrophobicity and superoleophilicity properties that can filter or absorb oils from water and "water-removing" type materials with superhydrophilicity as well as underwater superoleophobicity that can selectively isolate oils from mixtures.[6] Although versatile methods have been successfully developed to fabricate special wettability surfaces, it is still of great importance to develop simple and low-cost techniques that are suitable for large-scale industrial fabrication of these materials using relatively mild and eco-friendly processing conditions.[5,31]

The common properties of industrial oily wastewater, including high acidity, alkalinity, and salt, present a greater challenge for the separation of oil and water where the separation materials must be resistant to harsh conditions.[32−36] Unfortunately, a large number of earlier oil/water separation materials were fabricated using a metallic mesh or porous polyester as substrates, which are generally employed in gentle environmental conditions but have poor durability in harsh environments, such as seawater or highly acidic or alkaline industrial oily water.[37,38] Therefore, novel membrane materials with good stability are needed for practical oil/water separation in complex environments.

In this report, a polyethylene (PE) mesh was prepared that could separate oil from water under harsh conditions using a simple process of pressing, scratching, and pricking of PE powder. The prepared mesh exhibited superhydrophobic, superoleophilic, and ultralow water-adhesive properties, which resulted in the highly effective separation of oil−water mixtures by permitting oil to pass through the mesh while blocking water

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penetration. The high separation efficiency was maintained after 30 uses and after immersing the mesh in highly acidic, alkaline, or salty solutions for a long period. Therefore, the easy, low-cost, and eco-friendly fabrication process, together with the high separation efficiency, make this superhydrophobic PE mesh suitable for large-scale industrial production as a practical method for treating oily wastewater.

2. EXPERIMENTAL SECTION

2.1. Materials. Anhydrous alcohol, sulfuric acid, sodium hydroxide, sodium chloride, 1,2-dichloroethane, and hexane were all analytical grade, purchased from Beijing Chemical Works, and were used without further purification. Low-density polyethylene (LDPE) (Alfa Aesar Chemical Co., Ltd., China), carbon tetrachloride (Tianjin Aoran Fine Chemical Research Institute, Tianjin, China), low-viscosity silicone (PMX200-5CS, Dow Corning), methylene blue (Sinopharm Chemical Reagent Co., Ltd., Beijing, China), and oil Red O (J&K Co. Ltd., Beijing, China) were used as purchased. Gasoline, plant oil, sewing needles, and sandpaper were commercial products.

2.2. Characterization. The SEM images of the PE films were obtained using a field-emission scanning electron microscope (JSM-7500F, JEOL, Japan). Water and oil contact angles were measured on an OCA20 machine (Data-Physics, Germany) at ambient temperature. The water or oil droplets were all about 2 μL and placed carefully onto the materials in the air or immersed in water. The average value of five measurements performed at different positions on the same sample was reported as the contact angle. The water-adhesion forces were measured by a high-sensitivity microelectromechanical balance system (Data-Physics DCAT11, Germany) as follows: a water droplet (5 μL) suspended on a metal cap was slowly moved toward the PE surface at a constant speed of 0.005 mm·s^{-1}. As soon as the droplet contacted the surface, it immediately left in the opposite direction. The forces were recorded during this process, and the adhesion force equals the maximum force minus the force when the droplet left the PE surface. The average value of five measurements performed at different positions on the same sample was adopted.

2.3. Oil/Water Separation. The prepared porous PE mesh was installed between two Teflon holders with an angle of 15°. Both of the holders were attached to a glass tube with the diameter of 30 mm. The oil/water mixtures (50 v/v %) were carefully poured onto the PE mesh, and separation was achieved by the force of gravity. An infrared spectrophotometer oil content analyzer (CY2000, China) was used to measure the oil concentration of the original oil/water mixtures and the collected water after separation. CCl_4 was used as the solvent to extract the oils from the water. The absorbances at 2930, 2960, and 3030 cm^{-1} were measured. The absorption intensity value and the correction coefficient were calculated to obtain the oil content.

The separation efficiency was calculated by the oil rejection coefficient (R(%)) according to

$$R(\%) = (1 - C_p/C_0) \times 100 \tag{1}$$

where C_p and C_0 are the oil concentrations of the collected water after one time separation and the original oil–water mixtures, respectively.

3. RESULTS AND DISCUSSION

3.1. Fabrication and Surface Morphology of PE Mesh. The chemically durable PE mesh was prepared via a simple and easily operated process. Detailed preparation steps are sketched in Figure 1a, which can be generally divided into three steps. First, the low-density polyethylene powder was pressed at 120 °C, 20 MPa for 10 min, then cooled and peeled from the metal mold, to form a PE film. Second, the flat PE film was cut into 3.5 × 3.5 cm^2 pieces and then rubbed with an abrasive paper for five times. The rubbed PE film was further ultrasonically cleaned in ethanol for 5 min and then dried under nitrogen. Third, the PE film was pricked using a commercially available needle, which was polished and cleaned in advance. During the pricking process, the rubbed PE film was first pasted onto a 1 cm thick low-density PE template, which was fixed onto a pricking machine to ensure that the pricking process produced through-holes, and then the PE film was pricked by the vertically fixed needle to obtain PE mesh with arrays of pores.

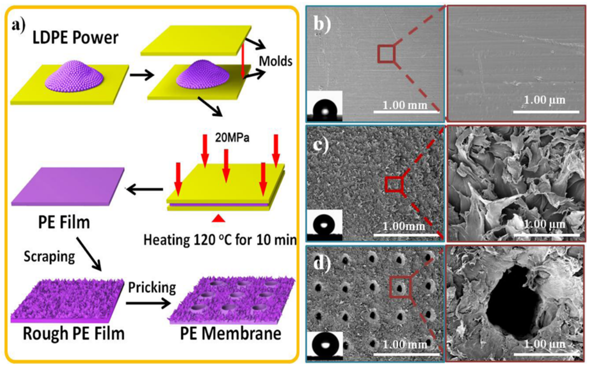

Figure 1. Fabrication process and morphology of the PE mesh. (a) Schematic of the preparing processes of the PE mesh. The SEM images of each step: (b) the flat surface of the PE film fabricated by thermal pressing; the inset is the CA of the PE film; (c) the roughness surface of PE film after scratching; the inset is the CA of the rubbed PE film; (d) porous PE membrane after pricking process, with an average pore diameter of approximately 900 nm.

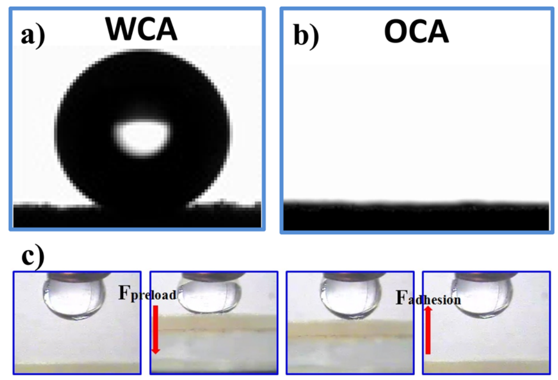

Figure 2. Porous PE mesh exhibited special wettability performances of (a) superhydrophobicity, (b) superoleophilicity, and (c) low water adhesion, respectively.

The morphologies of thermal-pressed PE film, rubbed PE film, and pricked PE mesh during each preparation step are shown in Figure 1b–d. It can be seen from these figures that the thermally pressed PE film exhibited a smooth, flat structure (Figure 1b) based on both the normal and the magnified SEM images with a water contact angle (WCA) of 110° ± 2.8° (inset in Figure 1b). This hydrophobicity degree was probably due to the chemical composition of the PE molecule. After being roughened with the abrasive, the PE surface exhibited micro/nanoscale composite morphology (Figure 1c) where the WCA increased to 154.3° ± 3.2° (inset in Figure 1c). This superhydrophobic degree was due to the roughened structure of the film, which can trap air in comparison to the flat PE substrate. The rough PE film was further pricked to form a porous array structure (Figure 1d), with the average pore diameter of approximately 900 nm. The morphology results revealed that the micro/nanoscale two-tier structure remained after pricking, and together with the pricked microscaled pores, the WCA exhibited a superhydrophobic degree of 152.9° ± 2.6°. Therefore, the chemical property of the PE molecule as

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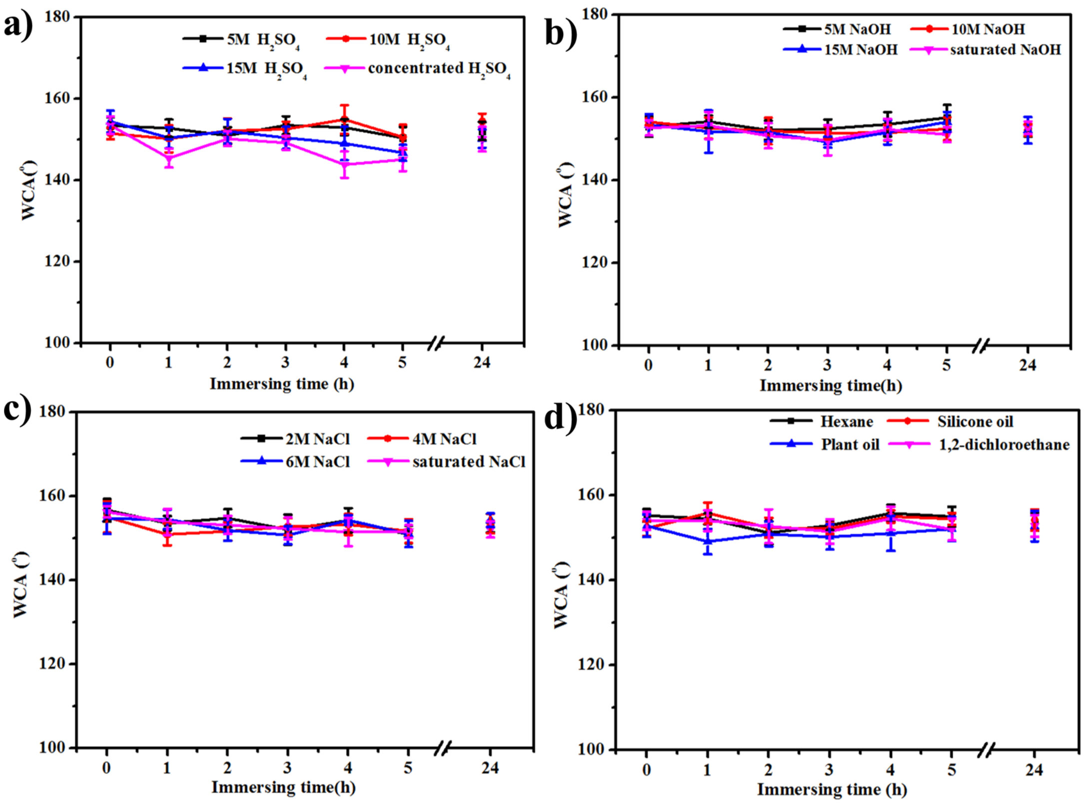

Figure 3. Water contact angle of the PE mesh after immersion in (a) highly acidic, (b) alkaline, (c) salty, and (d) oil environments for several hours.

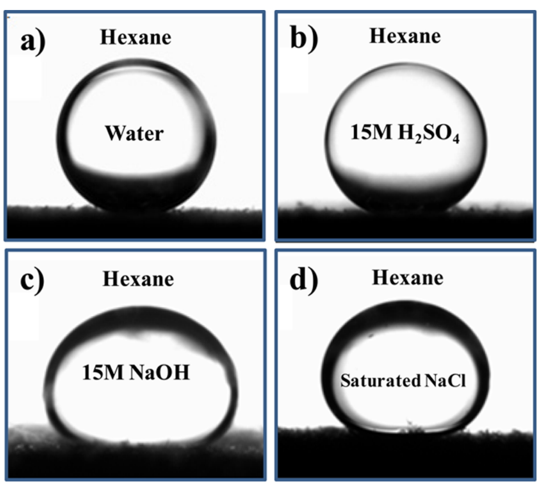

Figure 4. Wettability of the PE mesh under a hexane environment using a droplet of (a) water, (b) 15 M H_2SO_4, (c) 15 M NaOH, and (d) saturated NaCl, respectively.

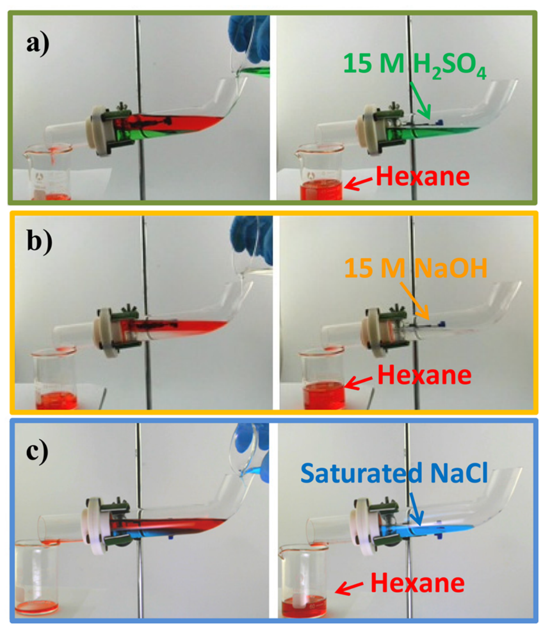

Figure 5. Typical separations of (a) hexane/15 M H_2SO_4 (50:50 v/v %), (b) hexane/15 M NaOH (50:50 v/v %), and (c) hexane/saturated NaCl (50:50 v/v %) mixtures.

well as the roughness structure of the surface led to the superhydrophobicity of the PE mesh, which enabled water to be trapped above it.

3.2. Wettability Performance of the Porous PE Mesh.

The water contact angle, underwater−oil contact angle (OCA), and water-adhesion properties of the porous PE mesh were further evaluated. Since the micro/nanoscale two-tier structure remained after pricking, the porous PE mesh not only was superhydrophobic (Figure 2a) but also showed very low water adhesion (about 20 μN) (Figure 2c). By contrast, when oil droplets were applied (for example, hexane) to the PE mesh, the oil droplets spread quickly and permeated the mesh (Figure 2b). On one hand, this oil wetting behavior can be attributed to the hydrophobic and oleophilic properties of PE material, but on the other hand, when the porous PE mesh contacted water, air was trapped in the micro/nano composite structures to form a water/air/solid composite interface. This decreased the contact area between the water droplet and the solid surface, causing the mesh to be superhydrophobic with low water

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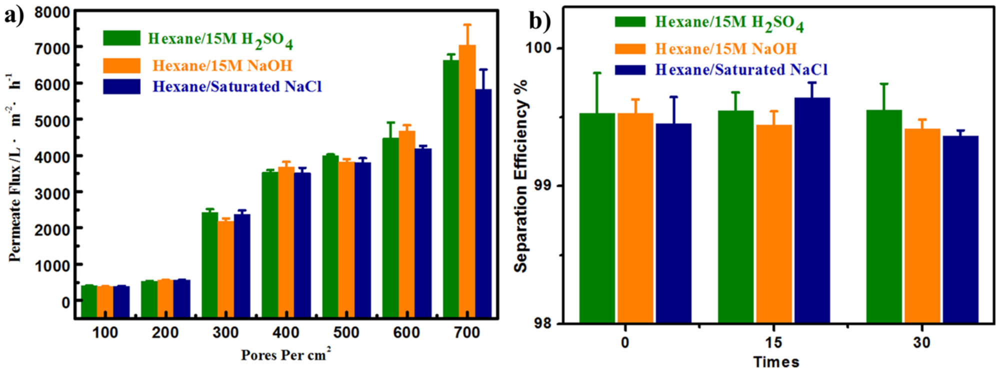

Figure 6. Oil/water separation of the prepared PE mesh. (a) Relationship between the pore number and the permeate flux. (b) Separation efficiency of the prepared PE mesh for separating hexane/15 M H_2SO_4 (50:50 v/v %), hexane/15 M NaOH (50:50 v/v %), and hexane/saturated NaCl (50:50 v/v %) mixtures, after 0, 15, and 30 recycles.

adhesion. These superhydrophobicity, superoleophilicity, and low water-adhesion properties suggested that this mesh would be useful for oil/water separation.

3.3. Wettability Performance of the Porous PE Mesh in Highly Acidic, Alkaline, and Salty Environments. To demonstrate the capacity of the oil/water separation of the novel mesh in a complex environment, highly acidic, alkaline, and salty solutions were used as the droplets to investigate their contact angle on PE mesh. The wettability stability is characterized by the CA measurement after immersing the porous PE mesh in acidic, alkaline, and salty solvents with different concentrations and then rinsing the mesh with water, followed by drying at room temperature. As shown in Figure 3, after immersing the porous PE meshes for hours (from 0, 1, 2, 3, 4, 5 h to 24 h) in different solutions with various concentrations (from 5, 10, 15 mol/L to saturated solution), the porous PE meshes still exhibited identical superhydrophobicity with WCAs near 150° (Figure 3a−c). Moreover, this superhydrophobic property was also maintained after immersing the porous mesh in a wide variety of oils, such as gasoline, plant oil, hexane, silicon oil, and 1,2-dichloroethane (Figure 3d). Therefore, the stable superhydrophobic property of the subject PE mesh not only was resistant to acid, alkali, and salt but also exhibited adequate tolerance to various oils. Such high durability of corrosive liquids should probably be due to the combination of the natural property of the PE molecule and the micro/nano hierarchical structures. On one hand, the PE molecule has the natural property of corrosion resistance; on the other hand, the micro/nano hierarchical structure of the PE mesh provided the surface a typical "Lotus" state with superhydrophobic and low adhesion properties. Therefore, the corrosive liquids can hardly wet the mesh surface and the excellent anticorrosion natural property further enhanced the corrosion resistance of the PE mesh.

The contact angles of water, saturated salt solution, 15 mol/L sulfuric acid solution, and 15 mol/L sodium hydrate solution were also investigated while the mesh was soaked in *n*-hexane. As shown in Figure 4, the porous PE mesh exhibited an underoil superhydrophobic property with contact angles larger than 150°, which suggested that oil can smoothly pass through the PE mesh, but the complex aqueous phase was blocked. The stable superhydrophobic property in complex environments as well as the high resistance of the corrosive solvent suggested that the PE mesh has potential industrial and consumer applications.

3.4. Oil/Water Separation. A series of oil/water separation experiments were conducted to investigate the oil/water separation efficiency of the porous PE mesh in highly acidic, alkaline, and salty environments. As shown in Figure 5, despite the change in the mixtures from hexane/15 M H_2SO_4, to hexane/15 M NaOH or hexane/saturated NaCl, the hexane easily permeated the PE mesh and collected in the beaker below the mesh while the water solution was retained above the PE mesh, indicating a significant separation in extreme oil/water environments affected by gravity. In addition, the PE mesh can be reused after a simple clean up and it maintained a high separation efficiency (>99.5%) after 30 uses (Figure 6a), which was a testament to its ability to resist highly acidic, alkaline, and salty environments.

Apart from the separation efficiency, the permeate flux is yet another factor used in the evaluation of separation; higher permeate flux in a mesh leads to faster separation. Therefore, a series of PE meshes were prepared with various pore numbers and the oil permeate fluxes of these meshes were measured for oil mixtures in various aqueous solutions. For each PE mesh, the average value of five replicate measurements was reported as the permeate flux. As shown in Figure 6b, as the pore numbers were increased from 100/cm^2 to 700/cm^2, the separation flux increased accordingly. At a pore number of 700/cm^2, the average permeate flux of the PE mesh for separating hexane in 15 M H_2SO_4, 15 M NaOH, and saturated NaCl was as much as 6624 ± 161.1, 7066 ± 527.1, and 5842 ± 514.2 L/m^2·h, respectively. These values were much higher than the results reported for other systems, including a cross-linked poly(ethylene glycol) diacrylate coated membrane (<100 L/m^2·h),[39] a superhydrophobic polyvinylidene fluoride membrane (700−3500 L/m^2·h),[40] a SiO_2−carbon composite nanofibrous membrane (>2000 L/m^2·h),[12] and a mineral-coated polypropylene microfiltration membrane (>2000 L/m^2·h).[41] However, because the PE film can be seriously damaged after intensively pricking, the pore numbers cannot be more than 700/cm^2. Therefore, 700/cm^2 was chosen as the standard pore number to use for all of the tests in other parts of this report. This high separation efficiency, reusable, and high permeate flux PE mesh appears to be a good candidate for use in practical industrial separation of oil and corrosive solutions.

4. CONCLUSIONS

In summary, a superhydrophobic, superoleophilic, and low water-adhesion PE mesh was fabricated using an easily operated process of pressing, scratching, and pricking. The resulting PE

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mesh exhibited special wettability even in harsh conditions, such as highly acidic, alkaline, and salty environments, and can directly separate strong acid, strong base, and saturated brine from oil with high efficiency. The separation process can be repeated at least 30 times with consistent separation efficiency. In addition, the high flux allowed the mesh to be applied in ultrafast and gravity-driven oil/water separation. Thus, the easily prepared PE mesh has great potential for use in large-scale oil/water separation in industry or for consumer use.

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Author Contributions

§D.Z. and T.Z. contributed equally to this work.

Notes

The authors declare no competing financial interest.

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■ NOTE ADDED AFTER ASAP PUBLICATION

This paper was published on the Web on September 2, 2016, with minor errors in Figure captions 5 and 6. The corrected version was reposted on September 14, 2016.