High-Flux and Robust Co$_3$O$_4$ Mesh for Efficient Oil/Water Separation in Harsh Environment

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ABSTRACT: Material with special wettability for oil/water separation has drawn more and more attention, since the oil spill accidents and industrial processing are growing in frequency and in volume. A superhydrophilic and underwater superoleophobic mesh was prepared by introducing Co$_3$O$_4$ on a stainless steel mesh, through a simple hydrothermal process and subsequent calcination. The as-prepared Co$_3$O$_4$ mesh can not only separate various oil/water mixtures with high efficiency and high flux, but also work effectively in harsh environment such as highly acidic, alkaline, and salty solutions. Moreover, the Co$_3$O$_4$ mesh can still retain good separation performance after 40 abrasion cycles with sandpaper. The outstanding anticorrosion and antibrassion behaviors make the Co$_3$O$_4$ mesh promising for oil/water separation even in harsh environment.

1. INTRODUCTION

In the past decades, more and more oily wastewater is generated from industrial processing and frequent oil spill accidents, causing tremendous damages to the social and economic activities, as well as the ecological environment. To make things worse, the oily wastewater is normally highly acidic, alkaline, and salty solutions, these harsh environment conditions present more challenges for the oil/water separation. Traditional technologies such as coagulation, oil-absorbing materials, and air flocculation are limited by low separation efficiency and huge operation cost. Thus, how to effectively perform oil/water separation in harsh environment has attracted broad attention.

A novel strategy that utilizes special wettability to separate oil/water mixture is considered as efficient and low cost. According to their different affinities toward water and oil, this kind of materials can be classified into hydrophobic and oleophilic (oil-removing), or hydrophilic and oleophobic (water-removing). The hydrophobic and oleophilic materials cause oil to spread, be absorbed, or pass through them while blocking water penetration, thus they are more easily fouled by oil, showing poor recyclability. Conversely, the hydrophilic and oleophobic materials, especially the hydrophilic and underwater oleophobic, allow water to filter through and repel oil. So, they have better oil resistance and become more feasible in dealing with large-scale oil/water separation.

The first reported superhydrophilic and underwater superoleophobic polyacrylamide hydrogel-coated mesh by Jiang can efficiently separate oil/water mixtures and be fully recyclable at the same time.

Up to now, multiple methods have been developed to fabricate hydrophilic and underwater oleophobic coating, such as lithography, laser processing, spray deposition, sol–gel processes, electrospraying, and hydrothermal method. Especially for the hydrothermal method, the products are commonly of high purity and uniformly dispersed. Moreover, the morphology of the products can be well controlled. Since the surface chemical compositions and surface architectures are crucial to the superwetting behavior, these advantages make hydrothermal method an effective technique to create special wettable materials. For example, Wang et al. prepared an underwater superoleophobic TiO$_2$ coating with a flower-like structure on the fluorine doped tin oxide glass substrate through hydrothermal method. To increase the surface roughness, many other inorganics such as ZnO, CaCO$_3$, Cu(OH)$_2$, and graphene oxide have also been widely used. They can promote the wettability according to the Wenzel model, and further enhance the hydrophilicity due to their high surface energy. However, most of these materials are only reported to be effective in gentle environment, there are few studies on their separation performance in harsh environment. In practical applications, the components of oily wastewater are complex, thus the separation materials need to have good stability over a wide pH range and high mechanical durability to deal with the possible abrasion. Besides, a facile route for the fabrication of these materials can make them more promising in practical oil/water separation.

Co$_3$O$_4$ has attracted much interest due to the excellent physical and chemical properties, thus exhibiting well tolerance toward corrosive solutions. Herein, we propose a Co$_3$O$_4$-coated stainless steel mesh for effective oil/water separation in harsh environment.
harsh environment. In this study, a high-flux and robust mesh was prepared by introducing Co₃O₄ onto a stainless steel mesh via a simple and cost-efficient method. The prepared Co₃O₄ mesh showed excellent hydrophilicity and oil repellency under water, which can separate various oil/water mixtures with high efficiency (>99.4%) and high flux (>75 000 L·m⁻²·h⁻¹). Additionally, the Co₃O₄ mesh can still retain good separation performance not only in corrosive liquids such as acidic, alkaline, and salty solutions but also after sandpaper abrasion, exhibiting outstanding chemical and mechanical durability.

2. RESULTS AND DISCUSSION

2.1. Preparation and Characterization of the Co₃O₄ Mesh. The stainless steel mesh has been widely chosen as a high-performance substrate for the oil/water separation because of its mechanical strength, high filtration flux, and reactive metal surface. In this study, a facile hydrothermal method was used to prepare the Co₃O₄ mesh. The cleaned stainless steel mesh was put into an autoclave containing Co(NO₃)₂ and H₂NCONH₂ solution. After the hydrothermal reaction, the mesh became pink due to the formation of cobalt precursors. Then, the sample transformed into the black Co₃O₄ mesh after calcination. The X-ray diffraction (XRD) patterns of the as-prepared mesh are shown in Figure 1. The peaks can be observed at 2θ values of 19.03, 31.32, 36.90, 59.45, and 65.34°, corresponding to the reflections of (111), (220), (311), (511), and (440) crystalline planes of Co₃O₄, respectively, which indicate the formation of Co₃O₄. Besides, additional peaks at 43.58, 50.72, and 74.56° are a result of the original stainless steel mesh.

Figure 2b–h displays scanning electron microscope (SEM) images of the Co₃O₄ mesh at different hydrothermal reaction time. As can be seen in Figure 2a, the original stainless steel mesh exhibits a smooth surface. After 1 h of hydrothermal reaction and following calcination, some needle-like Co₃O₄ was deposited on the surface. With the increase of reaction time, the needles became denser, and some sheets started to grow on them. These needles were covered with a layer of uniform Co₃O₄ sheets at 6 h, and the sheets started to fall out with further increase in reaction time. As shown in Figure 2e, the length of the Co₃O₄ sheets was 2–4 μm and grew vertically on the surface of the Co₃O₄ needle. A low-magnification image of the Co₃O₄ mesh shows that this unique structure of Co₃O₄ can be well loaded on the stainless steel mesh without blocking its original pore.

Generally, the methods to obtain special wettable materials are time-consuming and complicated, requiring multiple processes and special treatment. Compared with the previous study, our two-step fabrication is facile and cost-effective without special instruments and complex treatment. In addition, the stainless steel mesh and reagents used are all commercial and easy to obtain. More importantly, we avoided using fluorine-containing reagents, which are toxic and common in the preparation of special wettable materials.

2.2. Wetting Behavior and Separation Performance of the Co₃O₄ Mesh. The wettability of solid surface plays a critical role in separating the oil/water mixture. The optical images of water and 1,2-dichloroethane droplets (5 μL) on the original stainless steel mesh and Co₃O₄ mesh are shown in Figure 3. The original mesh shows a water contact angle (WCA) of 126.86 ± 1.56° and an underwater oil contact angle (OCA) of 138.45 ± 1.45°, revealing its intrinsic wettability is hydrophobic in air and oleophobic under water. After covering the Co₃O₄ layer, the mesh became superhydrophilic and showed enhanced underwater oleophobicity. Water can spread quickly on the Co₃O₄ mesh and the underwater OCA reached up to 151.19 ± 0.19°, realizing superoleophobic under water. As shown in Figure 4, the Co₃O₄ mesh also showed low...
oil adhesion for other kinds of oil (petroleum ether, cyclohexane, and hexane). This special wettability of the Co₃O₄ mesh is due to its surface chemistry and unique morphology. Due to the high surface energy of Co₃O₄,38 water would be locked in the nanostructure, form a water membrane on the surface, thus providing a repulsive force to oil phase, and prevent oil from permeating through.

Next the Co₃O₄ mesh was applied to separate immiscible oil/water mixtures; the separation was conducted in a homemade setup, which is shown in Figure 5. When petroleum ether (dyed with oil red O) and water (dyed with methylene blue) mixture was poured onto the prewetted mesh, water can pass through the mesh quickly just driven by gravity, whereas the oil was rejected on the mesh surface and collected in the upper tube. Because the as-prepared Co₃O₄ mesh is superhydrophilic and underwater superoleophobic, water can pass through easily, whereas oil would be repelled due to the repulsive force provided by the water trapped in its surface. No obvious oil droplets can be observed in the collected water, indicating the Co₃O₄ mesh can separate the oil/water mixture effectively. Other oil/water mixtures, including cyclohexane and hexane, were also separated successfully. As shown in Figure 6, the separation efficiencies for different mixtures could all reach above 99.4%, showing excellent separation performance.

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Besides separation efficiency, flux is also an important factor when considering the separation performance. The separation fluxes of different mixtures were also tested, and the results are shown in Figure 6. The fluxes at ordinary pressure of different oils were all above 75 000 L·m⁻²·h⁻¹, which were several times higher than those of the reported separation materials.28,39,40 The high flux of the Co₃O₄ mesh is caused by the hydrophilicity of Co₃O₄ and the large pore size of the mesh. The Co₃O₄ mesh is superhydrophilic, so water can spread on the surface quickly. According to the study by Wang et al., the flux of membrane is greatly influenced by the pore size. By increasing the pore size, flux would improve simultaneously.41 Actual situation usually requires a large number of oil/water mixture to be separated in short time, hence higher flux means better handling capacity.42

2.3. Chemical Durability of the Co₃O₄ Mesh. Figure 7a shows the underwater OCAs of the Co₃O₄ mesh after being immersed into different pH solutions for 24 h. At pH 1, the OCA is a little lower than 150°, indicating the Co₃O₄ mesh may be affected slightly in the strong acidic condition. For pH ≥ 3, there is little change on OCAs and the OCAs are all above 150°; hence, the Co₃O₄ mesh can maintain the underwater superoleophobic property in most instances. Furthermore, the separation efficiencies of the Co₃O₄ mesh under some corrosion environment are shown in Figure 7b. After being immersed into 1 M HCl, NaOH, and NaCl for 24 h, the separation efficiencies for petroleum ether are still all above 99.9%. There is no obvious difference in the efficiency under normal environment (immersed into deionized water). Loading material with good physical and chemical stability on the surface is a common way to enhance the separation material’s anticorrosion performance.43 The stainless steel
mesh itself has better tolerance toward many kinds of corrosive solutions. When the stable Co₃O₄ covered the mesh densely, it further enhanced the chemical durability. The actual industrial wastewater and the seawater are often very complex; they may be high-concentration acidic, alkali, or salty solutions; and these liquids directly contact with the material during separation. So, outstanding chemical durability of the as-prepared Co₃O₄ mesh expands its scope of practical application in oil/water separation.

2.4. Mechanical Durability of the Co₃O₄ Mesh. It is still a challenge for many special wetting surfaces to achieve good mechanical durability. The commonly used method to evaluate the mechanical resistance is the abrasion test. The underwater OCAs were recorded after each abrasion cycle to test the mechanical durability of the Co₃O₄ mesh, and the results are shown in Figure 8a. With the increase of abrasion cycle, the OCAs would gradually decrease. After 40 cycles, the OCA still maintained at more than 148°. Many technologies have been used to construct robust superhydrophilic surfaces, such as adding inorganic adhesives or laser treatment. The as-prepared Co₃O₄ can adhere to the mesh firmly by the hydrothermal process and the subsequent calcination. The desired mechanical durability is also attributed to the unique architecture of the twill weave mesh. Unlike the commonly used plain weave mesh, only part of its coating will be worn away when being rubbed. Figure 8b shows the SEM image of the Co₃O₄ mesh after 40 abrasion cycles. The remaining structures still form water membranes to prevent oils. The mechanical durability of the as-prepared Co₃O₄ mesh can deal with the abrasion in daily use and extend its service life.

3. CONCLUSIONS
In summary, a Co₃O₄-coated stainless steel mesh has been prepared by facile hydrothermal process and subsequent calcination. The obtained Co₃O₄ mesh exhibited excellent hydrophilicity and oil repellency under water. It can separate various oil/water mixtures with a high efficiency of more than 99.4% and a high flux of 75,000 L·m⁻²·h⁻¹, which enables the mesh to be applied in large-scale oil/water separation under ordinary pressure. Due to the desired chemical durability, the Co₃O₄ mesh also showed high separation efficiency even in highly acidic, alkaline, and salty solutions. Besides, the mesh also displayed a steady performance after 40 abrasion cycles with sandpaper, which allows it to avoid mechanical damages. Facile synthesis and excellent performance make the as-prepared Co₃O₄ mesh a promising separation material for practical oil/water separation in various harsh environments, such as the treatment of marine oil leakage and industrial oily wastewater.

4. EXPERIMENTAL SECTION
4.1. Materials. The stainless steel mesh (type: 316 L material, twill weave and 300 mesh) was purchased from Guangzhou Ming Wan Screen Mesh Co., Ltd. (China). Co(NO₃)₂·6H₂O, H₂NCONH₂, acetone, ethanol, HCl (36–38 wt %), and NaOH were all purchased from Sinopharm Chemical Reagent Co., Ltd. (China). NaCl was purchased from Xilong Scientific Co., Ltd. (China). All chemical reagents were analytical grade and used without further purification. All aqueous solutions were prepared with deionized water.

4.2. Preparation of Co₃O₄ Mesh. First, the original stainless-steel mesh was respectively cleaned in acetone, ethanol, 1 M HCl, and deionized water for 15 min and dried at 50 °C. Three millimolar Co(NO₃)₂·6H₂O was dissolved in 60 mL deionized water with 15 mM H₂NCONH₂ and vigorously stirred for 1 h. Then, the resulting solution was transferred into autoclave and the cleaned stainless steel mesh was immersed vertically into the solution. The autoclave was kept at 120 °C for 6 h. When the temperature cooled down to
room temperature, the mesh was washed by water and ethanol, and followed by dried at 50 °C. Finally, the obtained mesh was annealed at 300 °C under air for 2 h.

4.3. Characterization. The X-ray diffraction patterns were collected to confirm the crystal phase of the sample on a X-ray diffractometer (XRD, Rigaku Ultima IV, Japan) with Cu Kα radiation. The surface morphology was observed by using a scanning electron microscopy (SEM, Zeiss Sigma, Germany). The values of water contact angle (WCA) and oil contact angle (OCA) were measured by a contact angle measuring device (DSA100, Data Physics, Germany) at room temperature. The oil concentration was tested by UV−vis−NIR spectrophotometer (Varian Cary 5000).

4.4. Oil/Water Separation. The oil/water separation experiment was conducted by gravity in a filtration apparatus with a diameter of 25 mm. First, the Co3O4 mesh was prewetted by water and then placed between two glass tubes. Three types of oils were used in this separation, including petroleum ether, cyclohexane, and hexane. The immiscible oil/water mixture (1:1, v/v) was poured slowly onto the Co3O4 mesh. The corresponding separation efficiency (η) was calculated according to the following equation

\[ \eta = \left(1 - \frac{C_f}{C_o}\right) \times 100\% \]  

where \( C_f \) and \( C_o \) are the oil concentrations of the filtrate and the original oil/water mixture, respectively. After pouring 20 mL oil onto the mesh, the permeation flux (\( F \)) was determined by calculating the time after collecting 30 mL of water according to the following equation

\[ F = \frac{V}{St} \]  

where \( V \) is the volume of the filtrate, \( S \) is the area of the contacted mesh, and \( t \) is the testing time.

4.5. Durability Tests. The chemical durability was investigated by immersing the Co3O4 mesh into various corrosion liquids for 24 h and then measuring the underwater OCAs. These corrosion liquids included solutions with different pH (from 1 to 14), strong acid (1 M HCl), strong alkali (1 M NaOH), and high concentration salt (1 M NaCl) solutions. Besides, separation efficiencies of the Co3O4 mesh for 1 M HCl, NaOH, and NaCl were also tested.

The mechanical durability was evaluated by measuring the underwater OCAs after several sandpaper abrasion cycles. The Co3O4 mesh was placed between a 100 g weight and a silicon carbide sandpaper and then moved 20 cm in the horizontal direction and returned; the whole process was defined as 1 abrasion cycle.

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Notes
The authors declare no competing financial interest.

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