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Corrosion-resistant hydrophobic MFI-type zeolite coated mesh for continuous oil-water separation

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ABSTRACT: Efficient separation of oil-water mixtures has become a critical challenge due to frequent oil-spill accidents and enormous release of oil-containing wastewater by families or catering services. Superhydrophobic coatings on metal meshes have been evidenced as the effective and energy-saving materials for oil-water separation. However, it is difficult to fabricate the separating coatings with long-term superhydrophobicity and the outstanding anticorrosion properties for practical application. In this study, we fabricated the hydrophobic MFI type zeolite coatings on the pristine stainless-steel meshes via in situ crystallization method and subsequently modified with environmentally safe hexadecyltrimethoxysilane compound. The as-prepared coating meshes showed good self-cleaning, anti-fouling, and anticorrosion properties. Furthermore, the coated meshes were capable of continuously and efficiently separating different kinds of oil-water mixtures under gravity-driven pressure and had over 99.0% high separation efficiency. More important, the prepared hydrophobic MFI coated meshes had equivalent separation performance with the superhydrophobic coating meshes. Meanwhile, the hydrophobic MFI coated meshes demonstrated the excellent reusability performance and the long-term stability. These as-prepared meshes thus have great potential for applications in separating oily wastewater from oil field as well as recycling of waste oil from

1 catering services.

2 **Keywords:** hydrophobic coating; stainless steel mesh; MFI; in situ crystallization;
3 oil-water separation.

4

5 **1. INTRODUCTION**

6 Efficient separation of oil-water mixtures has become a critical challenge
7 because of frequent oil-spill accidents¹ and enormous release of oil-containing
8 wastewater by families² or catering services³, which have a destructive influence
9 on our aquatic environment and ecosystem. Collecting the waste oils, especially
10 the cooking oils, has thus attracted increasing our interest because it not only can
11 produce bioplastics⁴ and bio-lubricants⁵ but also can reduce drinking water and
12 groundwater contamination. However, waste cooking oils in the slop are
13 especially difficult to remove due to their complex composition and arduous
14 biodegradability⁶.

15 In the previous decades, great efforts have been made via using miscellaneous
16 physical⁷, chemical⁸, and biological⁹ methods to remove or re-collect oils from
17 wastewater with no additional environmental pollution and to recycle oils in
18 industries. These conventional methods have their own disadvantage, such as less
19 effective, high energy consumption, high maintenance and operating cost, using
20 toxic chemicals, as well as generating secondary pollutants³. The membrane
21 technology is regarded as the most beneficial approach to separate oily
22 wastewater due to its fairly high separation efficiency and comparatively simple
23 operating procedure¹⁰⁻¹⁴. Among them, the metal meshes covered with
24 superhydrophobic / superoleophilic (oil-removing)^{15, 16} or superhydrophilic /
25 underwater-superoleophobic (water-removing)¹⁷, together with switchable
26 superhydrophobic–superhydrophilic¹⁸ coatings have been extensively evidenced

1 to be the remarkably effective and energy saving materials for the oil-water
2 separation process both in industry and in academia.

3 Compared to the superhydrophilic materials, the superhydrophobic materials
4 have competitive advantages in the oil-water separation process, especially in the
5 oily wastewater treatment, although some studies concluded that the
6 superhydrophobic materials were easily fouled by oils ^{17, 19}. The considerable
7 advantages of the superhydrophobic meshes in the oily wastewater separation
8 can be attributed to the following reasons ²⁰: according to the capillary theory
9 applied to the selective permeability of oil and water, the oil phase rather than the
10 water phase tends to adsorb and aggregate on the
11 superhydrophobic/superoleophilic mesh, and then the oil phase has absolute
12 priority to permeate the mesh. The water phase, however, is blocked by the
13 hydrophobic mesh. Since the oil content in the oily wastewater is usually less than
14 50% (v/v) ^{21, 22}, it might be more economical to pass through the mesh by the
15 relatively small amount of oil rather than the water. As a result, less separating
16 mesh area and accordingly smaller separation processing capacity might be
17 needed for the oil-water separation.

18 Compared to hydrophobic materials, however, superhydrophobic material
19 surfaces are susceptible to damage ^{23, 24} because of the fragile hierarchical micro-
20 nano structures on the substrate surface or the surface contamination when used
21 in practice. Recently, our group developed a fluorine-free hydrophobic
22 hexadecyltrimethoxysilane (HDTMS)-TiO₂ coated meshes for oil-water the
23 separation ⁵. the separation efficiency of the as-prepared meshes maintained over
24 97.8% after 35 separating cycles. The result indicated that the
25 superhydrophobicity of the separating mesh was nonessential for the highly
26 efficient oil-water separation.

On the other hand, anticorrosive property of the separation materials is also an important factor to be considered because oily wastewater is usually in acidic, alkaline or high salinity conditions ²⁶. Inspired by this information, our group developed the superhydrophobic and anticorrosive electroless FAS@Ni-P coating meshes for the rapid water-oil separation ²⁷ and the highest separation efficiency of the mesh is 96.8%. Compared with the inorganic oxide coatings ²⁸ and the organic polymer coatings ^{29,30}, the zeolite membranes, specifically the MFI zeolite membranes have attracted considerable interests in both industrial and academic area of oil-water separation because of their high thermal and mechanical stability, excellent chemical stability, as well as high corrosion resistance ³¹⁻³³. For example, Liu et al. ^{34, 35} prepared the hydrophilic MFI-type zeolite coatings on the porous stainless steel meshes by in situ and secondary growth method. The oil rejection rates of the as-prepared meshes are above 96% for various oils. Li et al. ³⁶ fabricated the stainless steel mesh (SSM) supported pure-silica zeolite beta (PSZB) coatings with superhydrophobicity for oil-water separation. The as-prepared coating meshes could maintain high separation efficiencies higher than 99% after abrasion for 80 cycles or exposure to corrosive media.

According to the above analyses, in this study, we fabricated the hydrophobic MFI type zeolite coatings on the pristine stainless-steel meshes via in situ crystallization method and subsequently modified with environmentally safe hexadecyltrimethoxysilane (HDTMS). The surface morphology, the chemical components as well as the functional groups of the as-prepared meshes were characterized with a high-resolution field emission scanning electron microscope (FESEM), an energy dispersive X-ray spectrometer (EDS), and a Fourier transform infrared spectroscopy (FT-IR), respectively. The self-cleaning and the anti-fouling properties were also evaluated. Specifically, the anticorrosion behaviors of the meshes were estimated by Tafel analysis and EIS tests in detail. A home-made

separator was designed to analysis the oil-water separation properties including the intrusion pressure, the separation efficiency, and the separation flux.

2. EXPERIMENTAL SECTION

2.1. Materials and reagents

Pristine stainless steel (named as SS) wire meshes with the mesh screens of 500, 1000, and 2000 were purchased from Shanghai Yan Jin Filter Factory, China. The mesh screen number was provided by the factory. Hexadecyltrimethoxysilane (HDTMS, $\text{H}_3\text{C}(\text{CH}_2)_{15}\text{Si}(\text{OCH}_3)_3$, 96%) was purchased from Guangzhou Longkai Chemical Co., Ltd., China. Tetrapropylammonium hydroxide solution (40%), tetraethoxysilane (TEOS), absolute ethyl alcohol, and acetone were purchased from Chengdu Kelon Chemical Reagent Factory, China. Sodium hydroxide (NaOH) was purchased from Shanghai Chemical Reagent Factory, China. Sodium chloride (NaCl) and hydrochloric acid (HCl, 36.0-38.0 wt.%) were purchased from Chongqing Sichuan East Chemical (group) Co. LTD, China. Six-in-one normal temperature phosphating solution (MY401A, pH 2-3) was purchased from Nanhua Metal Surface Treatment Co., Ltd., China. Senior red ink (855 type) was purchased from Guizhou Doctor Chemical Co. LTD, China. Diesel oil (No. 0) was obtained from China Petrochemical Corporation (Sinopec Group). The dishwashing liquid, and the cooking oils, such as sunflower oil and peanut oil were purchased from a local Yonghui Supermarket, China.

2.2. Preparation of the hydrophobic MFI zeolite coating on the stainless-steel mesh

Scheme 1 illustrates the schematic procedure for preparing the hydrophobic MFI type zeolite coated SS (named as MFI@SS) meshes. This procedure consisted of three main processes including the activation of the SS mesh surface, the preparation of the MFI@SS meshes, and the hydrophobic treatment of the coated

1 meshes.

2 *Cleaning and activating of the pristine mesh surface:* Prior to cleaning, the
3 pristine steel mesh was cut into a rectangular shape of 80 × 80 mm. The mesh was
4 then put into water cleaning solution with dishwashing liquid in an ultrasonic
5 water bath (KQ-400KDE, Kunshan Ultrasonic Instrument Co., Ltd., China) at 40 °C
6 for 10-15 min. The mesh was then sequentially rinsed with plenty of tap water and
7 18.2 MΩ·cm water. After that, the mesh was cleaned with 3.0 wt. % NaOH solution,
8 acetone, absolute ethyl alcohol, and 18.2 MΩ·cm water in the ultrasonic water bath
9 at 40 °C, respectively. For activation, the cleaned mesh was activated in 10%
10 phosphating solution at atmosphere temperature for 15 min. Finally, the activated
11 SS mesh was dried with a hairdryer (HP8200, Philips Electronics Hong Kong Ltd.,
12 China) and then reserved in a clean container.

13 *Preparation of hydrophobic MFI-type zeolite-coated meshes:* The MFI type zeolite
14 coatings on the SS mesh with the mesh screens of 500, 1000, and 2000 were
15 prepared by in situ crystallization method³⁷. Briefly, the synthesis solution was
16 made by adding tetraethoxysilane (TEOS) to a tetrapropylammonium hydroxide
17 (40% TPAOH) solution and later diluted in 18.2 MΩ·cm H₂O with a final molar
18 composition of 1TEOS: 0.82TPAOH: 152H₂O under stirring (Thermostatic
19 magnetic mixer, B11-2, Shanghai Sile Instrument Co., Ltd., China). After the
20 solution was aged for 2 h under stirring, the synthesis solution became a stable
21 solution. The solution was then directly poured into the Teflon cup in the Teflon
22 Lined Stainless Steel Autoclave (100mL, Jinan Henghua Technology Co., Ltd.,
23 China). The activated SS meshes were rolled and vertically put into the Teflon cup
24 and submerged under the solution level. The autoclave was then sealed and placed
25 in a digital blast drying oven (SK101, Shanghai Shengke Instrument Equipment
26 Co., Ltd., China) at 180 °C for 10 h. The curled coating meshes were taken out and
27 unfolded into a flat state, and then thoroughly washed with 18.2 MΩ·cm water.

1 After that, the coated meshes were dried at room temperature, and calcined at
2 600 °C (heating rate of 2 °C/min) and incubated for 2 h in a muffle furnace (SXZ-
3 10-12, Shanghai Essien Electric Furnace Factory, China) followed by ultrasonic
4 cleaning (KQ-400KDE) in 18.2 MΩ·cm water for 10 min, and air-dried for use.

5 *Hydrophobic treatment of the coating:* The MFI-type zeolite coated meshes were
6 hydrophobically modified with hexadecyltrimethoxysilane (HDTMS) solution
7 including HDTMS, 18.2 MΩ·cm water, and absolute ethanol by a ratio of 3:10:87,
8 respectively. To begin with, the HDTMS solution was stirred on the thermostatic
9 magnetic mixer (B11-2) at 30 °C. The solution was then allowed to stand for 1 h to
10 completely hydrolyze the HDTMS to obtain a hydrophobic solution. The MFI-type
11 zeolite coated meshes were subsequently immersed for 3 h into the solution and
12 then baked in the digital blast drying oven (SK101) at 120 °C for 1 h to complete
13 the hydrophobically modified process³⁸.

14

Scheme 1 Schematic fabrication process of the hydrophobic MFI@SS mesh coatings.

2.3. Measurement and characterization

2.3.1. Characterization

The morphology and the structure of the meshes were obtained using the high-resolution field emission scanning electron microscope (FESEM, Zeiss Supra-55) at the electron high tension (EHT) voltage of 15 kV. Chemical compositions of the meshes were explored using the energy dispersive x-ray spectrometer (EDS, Oxford Instruments INCAPentaFET-×3) at an accelerating voltage of 15 kV. Fourier transform infrared spectroscopy (FT-IR) was applied to detect the existence of the functional groups by using the Thermo Scientific™ Nicolet™ In™10 (ThermoFisher Scientific) with a spectrum range of 400–4000 cm^{-1} and a scan resolution of 1 cm^{-1} . The adhesion property of the MFI@SS coating mesh was

1 qualitatively evaluated using the cross-cut test ³⁹ with Adhesion Tester (QFH-
2 HV600, Taizhou Aitest Instrument Co., Ltd., China).

3 2.3.2. Wettability

4 The static WCA were determined using the sessile drop technique at room
5 temperature on the optical contact angle/surface and interfacial tension meter
6 (DropMeter™ A-200, MAIST Vision Inspection & Measurement Co., Ltd, China).
7 Water droplet with 2 μ L was set on the mesh at each WCA measurement. All WCA
8 measurements were independently repeated five times, and the average values
9 together with their standard deviation were subsequently calculated with
10 Microsoft Excel (Excel version in Microsoft Office 2016 for Win 7).

11 2.3.3. Anticorrosion tests

12 Corrosion is a tricky problem for practical application of the coated meshes in
13 oil fields ⁴⁰ and oil-water separation systems ⁴¹. In this study, the anticorrosion
14 properties of the hydrophobic MFI@SS mesh were evaluated by using the PGSTAT
15 128N potentiostat (Metrohm Autolab, Switzerland) in a three-electrode cell
16 (CHX400, Chuxi Industrial Co., Ltd., Shanghai, China) with the corrosive solution
17 containing 3.5 wt. % NaCl at room temperature. A saturated calomel electrode
18 (SCE) acted as the reference and a platinum sheet with 2 mm thickness as the
19 auxiliary electrode. The mesh sample served as the working electrode. The three
20 electrodes were put vertically in the corrosive solution and the distance among
21 the electrodes remained the same for all the measurements. Before each corrosion
22 test was carried out, a defined area (1 cm $\times$ 1 cm) of the sample was exposed to
23 the corrosion solution. Other parts of the sample surfaces were enveloped by a
24 mixture of epoxy resin and polyamide with a volume ratio of 2:1. The covering on
25 the sample surfaces was subsequently solidified at 60 $^{\circ}$ C for 4 h in the digital
26 display blast drying oven (SK101, Shengke Equipment Co., Ltd., Shanghai, China).

1 To ensure effective contact between the working electrode and the SS mesh
 2 substrate for the electrochemical test, the clamping point on the sample was
 3 previously ground with a 400-mesh sandpaper to remove the surface coating.
 4 More important, the contact point of the working electrode on the mesh should be
 5 carefully placed above the corrosive liquid surface to prevent current short circuit
 6 during the electrochemical test.

7 Tafel (potentiodynamic polarization) tests and EIS (Electrochemical Impedance
 8 Spectroscopy) measurements were both investigated to examine the corrosion
 9 resistance of the hydrophobic MFI@SS meshes as well as the pristine SS meshes.
 10 Tafel curves were measured under a scan voltage range of -1.50 to 0.60 V under a
 11 scan rate of 0.01 V · s⁻¹.⁴² The open circuit potential (OCP) was first evaluated to
 12 stabilize the rates of the anodic and cathodic reactions. The corrosion current
 13 density (I_{corr}) and corrosion potential (E_{corr}) of the coated meshes and the pristine
 14 SS meshes were acquired by extrapolating the linear portions of the Tafel plots⁴².
 15 The polarization resistance, R_p , was obtained from the Tafel plots combined with
 16 the Stearn-Geary Equation:⁴³

$$17 \quad R_p = \frac{\beta_a \cdot \beta_c}{2.303 \cdot (\beta_a + \beta_c) \cdot I_{corr}} \quad (1)$$

18 where β_a and β_b are the slopes of the anodic and the cathodic Tafel plots
 19 ($\Delta E / \Delta \log |i|$, V · dec⁻¹), respectively. The corrosion rate (CR, in millimeter per
 20 year) was calculated according to eq 2:⁴⁴

$$21 \quad CR = \frac{3270 \cdot I_{corr} \cdot M}{\rho \cdot D} \quad (2)$$

22 where M and ρ (g·cm⁻³) are the equivalent weight and the density of the steel,
 23 respectively; D is valence.

24 The EIS spectra were collected over a frequency range of 0.01 - 100 kHz and

1 with a 10 mV sinusoidal applied potential.⁴² The spectra were fitted to an
2 appropriate equivalent circuit (EC) using the ZSimpWin Demo V3.60 software
3 (EChem Software Bruno Yeum, PhD. Ann Arbor, Michigan, USA). All experiments
4 were carried out at open circuit potential (OCP) and repeated three times
5 individually to confirm the reproducibility of the results.

6 **2.4. Continuously oil-water separation experiments**

7 To evaluate the oil-water separating properties of the hydrophobic MFI@SS
8 meshes, the separating experiments were performed with a home-made separator
9 consisting of two pieces of plexiglass tubes with inside diameter of 50.1 ± 0.3 mm
10 (Figure 1). The hydrophobic rectangular MFI@SS mesh was firstly cut into a circle
11 shape with a diameter of 60 mm for the separating tests and then installed
12 between one pair of flanges and sealed with two silicone gaskets. The flanges were
13 fastened with four sets of stainless-steel bolts and nuts to prevent leakage.
14 Subsequently, the separator was immobilized on two iron platforms with the tube
15 inclined $10\text{-}15^\circ$ downwards along the outlet end (Figure 1). The offcuts of the
16 meshes were collected for the surface characterization and the anticorrosion
17 performance tests.

18 Diesel oil and $18.2\text{ M}\Omega\cdot\text{cm}$ water (dyed in red with Senior red ink to clearly
19 distinguish the oil-water interface) with a volume ratio of 1:1 as a feedstock were
20 directly mixed with the thermostatic magnetic stirrer (B11-2, Shanghai Sile
21 Instrument Co., Ltd, China) during the oil-water separating experiment. The
22 MFI@SS mesh was prewetted for 5 s with oil before feeding the oil-water mixture.
23 The feedstock was delivered to the inlet of the separator with a peristaltic pump
24 (YZ1515x, four pump head, Baoding Shenchen Pump Co., Ltd., China). The oil
25 phase permeated through the MFI@SS mesh and was collected with a container
26 while water was impeded behind the mesh (See Figure 1). The mass of the oil in

1 unite time was directly measured with an electronic balance (JY502, Shanghai
2 Puchun Measuring Instrument Co., Ltd.) and then the volume of oil was calculated
3 according to the oil density. Accordingly, the separation flux, J ($\text{L}\cdot\text{m}^{-2}\cdot\text{h}^{-1}$), was
4 calculated with eq 3: ⁴⁵

$$5 \quad J = 1000 \times \frac{(m/\rho)}{S \times t} \quad (3)$$

6 where, m is the oil mass penetrating the hydrophobic mesh corresponding to the
7 testing time, kg; ρ is the oil density, $\text{kg} \cdot \text{m}^{-3}$; S is the effective area for oil pathway,
8 m^2 ; and t is the time for testing the separation flux, h.

9 After separating the oil, the residual liquid was synchronously pumped from the
10 outlet of the separator and recycled into the container of the feedstock (Figure 1).
11 The separation efficiency, η , was finally determined for each separating process
12 according to eq 4: ⁴⁶

$$13 \quad \eta = \frac{V_1}{V_0} \times 100\% \quad (4)$$

14 where, V_1 is the volume of the collected oil passing through the mesh, and V_0 is the
15 volume of oil in pristine oil-water mixture, mL. Moreover, the residual oil after
16 each separation was measured using the standard of SY/T 5329-2012 ⁴⁷ (a
17 criterion of China Petroleum Industry) with colorimetric method. Petroleum ether
18 was used to extract oil from water, and the absorbance was measured at 430 nm
19 using a UV-vis spectrophotometer (TU-1901, Beijing Puxi General Instrument Co.,
20 Ltd.). To measure the water content, however, it should be noted that water in the
21 oil-water mixture should not be dyed with red ink.

Figure 1. Schematic diagram of home-made oil-water separator illustrating the continuously separating process.

3. RESULTS AND DISCUSSION

3.1. SEM, FT-IR, and adhesion test

Figure 2 shows the typical FE-SEM micrographs of the pristine SS wire mesh (500-mesh screen) and the as-prepared zeolite-coated mesh on a 2000-mesh screen at low and high magnifications. Figures 2(a-d) show that both the SS meshes with the mesh screens of 500 and 2000 possessed a smooth surface. Furthermore, the pristine SS mesh with the 500-mesh screen had the approximate rectangular pores with the size around $48.6 \times 51.9 \mu\text{m}$, whereas the SS mesh with the 2000-mesh screen was highly dense and no pores could be obviously found on the mesh. Figures 2e and 2f show that the MFI coating on the mesh was dense without cracking. Figures 2f and 2g demonstrate that the MFI-type zeolite crystals

1 had the elongated coffin shape particles⁴⁸ rather than the equiaxed MFI particles,
2 as shown in Scheme 1. The length of the MFI-type zeolite crystals along the *a*, *b*,
3 and *c* axis⁴⁸ was approximate 1.0, 0.4, and 3.2 μm , respectively (Figure 2g, SEM),
4 which indicated the *c*-axis was the longest dimension. A possible explanation
5 might be attributed to a secondary growth of the crystals occurred along the *c*-axis
6 when hydrothermally treated at higher temperature⁴⁹. Both the shape and the
7 size of the MFI zeolite crystals were consistent with those results obtained by
8 Wang et al.⁵⁰ The EDS analysis was utilized to characterize the chemical
9 composition of the MFI@SS coating on 2000 mesh. The elements of Al and Si were
10 found in the MFI coating (Figures 2h and 2i, Table 1). Fe, Cr, Ni elements from the
11 SS mesh substrate could also be found in the EDS spectrum because the detecting
12 depth of electron beam irradiation was several microns which might be larger
13 than the thickness of the MFI coating.⁵¹ The results indicated that the MFI-type
14 zeolite crystals were successfully prepared on the SS mesh. Furthermore, the FE-
15 SEM images clearly showed that the micro/nanoscale rough structure acting as
16 the roughness layer was successfully constituted on the mesh surface (Fig. 2d and
17 2f) which was very beneficial for forming highly hydrophobic mesh surfaces.

1

2 Figure 2. FE-SEM micrographs and EDS analysis of the typical pristine SS mesh: (a) and
 3 (b): 500-mesh screen; (c) and (d): 2000-mesh screen; (e)-(g): MFI zeolite coated mesh
 4 on the 2000-mesh screen; (h) and (i): the test zone selected from (e) for EDS analysis
 5 and the corresponding EDS spectrum.

6

Table 1. Element components of the MFI@SS mesh by EDS.

Element	Al	Si	Cr	Fe	Ni	Total
Mass percentage/%	0.08	16.48	20.50	57.57	5.38	100.00
Atomic percentage/%	0.14	27.86	18.71	48.94	4.35	100.00

Figure 3 presents the FT-IR spectra of a typical hydrophobic MFI@SS mesh with the 2000-mesh screen. The spectrums located at the wave numbers of 2853.2 cm^{-1} and 2916.8 cm^{-1} showed the asymmetric and symmetric stretching vibrations of $-\text{CH}_3$ and $-\text{CH}_2$ bonds which corresponded to the presence of long-chain alkyl hydrophobic groups ⁵² resulting from HDTMS. Furthermore, the peak at 1569.8 cm^{-1} indicated the C-H absorption peak ⁵³. Peaks at 457.5 cm^{-1} , 803.7 cm^{-1} , 1044.2 cm^{-1} corresponded to Si-O-Si vibrations of HDTMS ^{53, 54}. The peak at 537.6 cm^{-1} was attributed to the deformation vibration of Al-O-Si ¹⁵. The absorption peaks at 1643.5 cm^{-1} and 3403.7 cm^{-1} were the bending vibration peak of H-O-H and the antisymmetric stretching vibration peak of O-H. ⁵⁵

Moreover, the adhesion test shows that the adhesion strength grade of the MFI coating to the substrate was 0 according to ISO 2409-2013 classification. The cutting edges of the mesh coating were completely smooth and without peeling off, and none of the squares of the lattice was detached. The result indicated that the adhesion between the MFI coating and the stainless steel mesh substrate was quite strong.

Figure 3. FT-IR spectra of the hydrophobic MFI@SS mesh with the 2000-mesh screen.

3.2. Wettability and self-cleaning property

The wettability of the mesh is a crucial property for separating oil-water mixtures due to the opposite wettability of the hydrophobic mesh to oil and water simultaneously⁵⁶. We hereby investigated the water contact angles of the pristine SS meshes and the MFI@SS meshes. Figure 4a shows that the water drops rapidly penetrated the pristine 500-mesh screen due to the hydrophilic surface property as well as the large aperture sizes of the mesh. Unlike the pristine 500-mesh screen, however, the more compact pristine 1000 and 2000 meshes could not be penetrated by water and accordingly had the water contact angle (WCA) of $43.6 \pm 5.8^\circ$ and $66.3 \pm 1.5^\circ$, which showed hydrophobicity of the meshes (Figures 4b and 4c). The water contact angles of the meshes increased to $125.7 \pm 3.5^\circ$, $130.0 \pm 4.9^\circ$, and $144.8 \pm 3.9^\circ$, respectively, when the pristine meshes were coated with MFI-type zeolite membrane and were then hydrophobically modified with HDTMS. The results, as shown in Table 2, also revealed that the water contact angle of the

1 MFI@SS meshes increased with the increase of the pristine mesh screens.

2 The formation mechanism of the hydrophobic coating could be interpreted as
3 follows: in hydrophobic HDTMS solution, one HDTMS molecular could be
4 hydrolyzed to one hexadecylsilanol molecular which had one $\text{CH}_3\text{-(CH}_2\text{)}_{15}\text{-Si}$
5 group at the non-polar end and three -OH groups at the polar end ²⁵. On the other
6 hand, MFI-type zeolite coating surface adsorbed a large amount of -OH groups
7 because of the hydrophilicity of the MFI-type zeolite ⁴⁸. When MFI-type zeolite
8 coating mesh was dipped into the hydrophobic HDTMS solution, the chemical
9 links could be formed between hexadecylsilanol and the MFI-type zeolite coating
10 because of the dehydration of the -OH groups. As a result, the long carbon chain,
11 $\text{CH}_3\text{-(CH}_2\text{)}_{15}\text{-Si}$, could envelop the MFI-type zeolite coating and the hydrophobic
12 coating surface was obtained. In other words, the water contact angle (WCA) of
13 the MFI-type zeolite coating mesh increased after the hydrophobic treatment with
14 HDTMS solution.

15
16 Figure 4. Water contact angles of different meshes. (a), (b), and (c): the pristine 500,
17 1000, 2000-mesh screen, respectively; (d), (e), and (f): the MFI@SS mesh after

hydrophobically treated with HDTMS.

Table 2. Water contact angles of the meshes with the mesh screens of 500, 1000, and 2000 before and after hydrophobic treatment.

Mesh screen	500	1000	2000
Pristine mesh	penetrating	$43.6 \pm 5.8^\circ$	$66.3 \pm 1.5^\circ$
MFI@SS mesh	$125.7 \pm 3.5^\circ$	$130.0 \pm 4.9^\circ$	$144.8 \pm 3.9^\circ$

Self-cleaning property is one of the specific characters of the hydrophobic materials to make a contamination-free surface. Undoubtedly, the well-known case of the hydrophobic self-cleaning surface is the lotus plant leaves (*Nelumbo nucifera*)⁵⁷. In this study, the self-cleaning property of the hydrophobic MFI@SS coated mesh was characterized by dipping the mesh into the muddy solution as well as by removing the conventional pollutant⁵⁸ of the chalk powder covered on the mesh. Figures 5(a-f) demonstrate the video screenshots of the dipping and flushing process of the mesh into the muddy water (clay content $\sim 300 \text{ kg}\cdot\text{m}^{-3}$). Before dipping, the dust was mixed and magnetically stirred in water to obtain the muddy water. The coated mesh was immersed into the muddy solution for 30 s. After that, the mesh was taken out and repeatedly flushed by $18.2 \text{ M}\Omega\cdot\text{cm}$ water with a wash bottle. The self-cleaning property of the mesh could be qualitatively evaluated from the amount of the muddy washed down from the surface of the mesh (See Supporting information, V1). The results in Figures 5e and 5f indicated that the muddy could hardly adhere to and retained on the mesh, which showed the excellent self-cleaning property of the mesh. Meanwhile, the mesh could maintain stable hydrophobicity after artificial contamination with the muddy solution. Figures 5g and 5h demonstrated that the water stream flowed passed the coated mesh immediately, synchronously imbibed and cleaned up the chalk

1 powder easily and completely due to the reliable self-cleaning property of the
2 hydrophobic mesh (See Supporting information, V2).

3
4 Figure 5. Demonstration of the self-cleaning effect of the hydrophobic MFI@SS coating
5 mesh: (a) the coating before dipping into the muddy solution, (b) dipping time 30 s, (c)
6 after dipping, (c) flushing with 18.2 MΩ·cm water, (e) and (f) after flushing; (g) the
7 coating covered with dusts of chalk powder, (h) the coating flashed with 18.2 MΩ·cm
8 water and the chalk powder completely removed from the coating.

9 3.3. Intrusion pressure

10 The oil-water mixture would be at risk of simultaneous permeation of the mesh
11 as soon as the separating pressure surpassed the intrusion pressure and thus
12 sharply decreased the separation efficiency. Therefore, the intrusion pressure is
13 another crucial capability for oil-water separation. To further quantitatively study
14 the waterproof ability, the maximum height ($h_{\max}$) of the water column that the
15 MFI-type zeolite coated mesh could support was measured and the water
16 intrusion pressure, P , was directly calculated with eq 5:⁵⁹

$$17 \quad P = \rho g h_{\max} \quad (5)$$

18 where, ρ is the density of water, kg·m⁻³; g is the gravitational acceleration, m²·s⁻¹;

and $h_{\max}$ is the maximum water column that the hydrophobic coated mesh can bear, m. Figure 6a shows the schematic home-made device for testing the intrusion pressure of the hydrophobic coated mesh. To prevent leakage, the MFI@SS mesh was horizontally placed and firmly sealed with two silicone gaskets between the flanges of the two vertical plexiglass tubes with the inside diameter of 50.1 ± 0.3 mm. Figures 6b, 6c and 6d demonstrate digital photographs of measuring intrusion pressures of the MFI@SS meshes with the mesh screens of 500 (a), 1000 (b), and 2000 (c), respectively. Dyed $18.2 \text{ M}\Omega\cdot\text{cm}$ water with red ink was carefully injected from the upper inlet and the underneath of the hydrophobic mesh was observed absorbedly. The maximum height of the water column was immediately measured and recorded when the first drop of water leaked through the mesh. The above measurement was repeated three times independently, and the average height as well as its standard deviation was calculated subsequently using eq 5. The results showed that the maximum water-holding height of the MFI@SS mesh with the mesh screens of 500, 1000, and 2000 was 14.4 ± 0.3 , 24.5 ± 0.2 , and 33.1 ± 0.5 cm, respectively, corresponding to the intrusion pressure of 1.41 ± 0.03 , 2.40 ± 0.02 , 3.24 ± 0.04 kPa, respectively. The result also indicated that the intrusion pressure of the MFI@SS mesh increased markedly with the decrease of the mesh screen. These results could be explained according to the following theoretical intrusion pressure eq 6:⁴⁶

$$\Delta P = \frac{2\gamma_{ow}}{R} = -\frac{\gamma_{ow} \cos \theta_l}{(A/C)} \quad (6)$$

where γ_{ow} is the interfacial tension of oil and water, $\text{N} \cdot \text{m}^{-1}$; R is the radius of the meniscus, m; C (m) and A (m^2) are the circumference and the cross-sectional area of the mesh aperture, respectively; θ represents the WCA on the hydrophobic MFI@SS mesh, $^\circ$.

1 As we all know, for rectangular or circular pores of the mesh, the value of A/C
2 is a linear function of the mean pore radius $\bar{r}_p$. As a result, we concluded from eq
3 6 that the intrusion pressure, ΔP , is inversely proportional to $\bar{r}_p$. In this study the
4 mesh with higher mesh screen indicates smaller $\bar{r}_p$. Therefore, the relation
5 between the intrusion pressure and the mesh screen in the current study was
6 consistent with the trend predicted by eq 6. We could also ascertain that the
7 intrusion pressure was positive (negative capillary effect ⁶⁰ in the oil/water/air
8 three phase interface) because the WCA on the hydrophobic MFI@SS mesh surface
9 was $\theta_w > 90^\circ$ (as shown in Figure 4). Therefore, the water could be prohibited by
10 the mesh because of the positive intrusion pressure ($\Delta P > 0$). Consequently, the
11 mesh could withstand some extent of water pressure (Figure 6). On the other hand,
12 the liquid (such as oil) with the contact angle, $\theta_o < 90^\circ$, corresponding to the
13 negative ΔP value made the liquid spontaneously permeate the mesh.

1

2 Figure 6. Measurement of the maximum water intrusion pressures of the MFI@SS
 3 meshes. (a) Scheme diagram of the home-made testing device; (a), (b), and (c) digital
 4 photographs of measuring intrusion pressures of the MFI@SS meshes with the mesh
 5 screens of 500 (a), 1000 (b), and 2000 (c), respectively.

6 3.4. Permeability of diesel oil

7 As discussed above, the negative intrusion pressure resulted in oil
 8 spontaneously passing through the hydrophobic mesh. To further understand the
 9 permeability of the oil on the hydrophobic MFI@SS mesh, the permeating process
 10 was recorded with a mobile digital camera with 3000 pixels to obtain the
 11 permeating time for one drop of diesel oil passing through the hydrophobic
 12 MFI@SS mesh. For example, to obtain a picture sequence of the video (Supporting

1 information, V3-V5), the video was graphically sequenced by the free software of
2 Video to Picture® 4.0 (Copyright © 2019 Watermark-software.com) with the
3 output rate of 25 images per second.

4 Figure 7 demonstrates the selecting graphic sequences of the permeating
5 process of one diesel oil droplet permeating through the MFI@SS meshes.
6 Regarding the MFI@SS mesh with the 500-mesh screen, Figures 7(a-1) and 7(a-2)
7 show one diesel oil droplet was dripping onto the mesh while Figure 7(a-3)
8 illustrates the droplet exactly dipped on the mesh and accordingly the time of this
9 screenshot was set to zero. The result showed that the total time was approximate
10 0.56 s for one diesel oil permeating freely through the MFI@SS mesh with the 500-
11 mesh screen. Figures 7(b-1) - 7(b-6) and 7(c-1) - 7(c-6) exhibit the permeating
12 time of the MFI@SS meshes with the 1000- and 2000-mesh screens was
13 approximate 4.32 and 10.56 s, respectively, which were much longer than that of
14 the MFI@SS mesh with the 500-mesh screen.

15 The difference of the permeating time could be qualitatively interpreted by the
16 classical Hagen-Poiseuille eq 7: ^{61, 62}

17
$$J_v = \frac{\varepsilon \pi r_p^2}{8\mu} \frac{\Delta p}{L} \quad (7)$$

18 where J_v is the flow flux of the mesh, $L \cdot m^{-2} \cdot h^{-1}$; ε is the surface porosity, r_p is the
19 pore radius of the mesh, m; Δp represents the pressure drop through the mesh, Pa;
20 μ is the dynamic viscosity of the fluid passing through the mesh, $N \cdot s \cdot m^{-2}$; L is the
21 path length of liquid running through the mesh (i.e. the thickness of the mesh in
22 this study), m. We could assert that the MFI@SS meshes with 500-, 1000-, and
23 2000-mesh screens had almost equal thickness, L , under the same MFI coating
24 preparation condition. The MFI@SS mesh, however, both had larger surface
25 porosity, ε and pore radius, r_p , with smaller mesh screen of the stainless mesh

1 substrate. Meanwhile, when one oil droplet passed through the meshes, the
 2 pressure drop, Δp , was only affected by gravity. Altogether, the flow flux, J_v , was
 3 directly proportional to εr_p^2 . Furthermore, the permeating time was positively
 4 correlated with the flow flux, J_v . Consequently, the MFI@SS mesh with the 500-
 5 mesh screen had the smallest oil droplet permeating time.

6 Thus, both specific water-repellent and oil-permeable properties achieved
 7 simultaneously endowed the MFI@SS mesh with the 500-mesh screen to be a
 8 highly prospective material for oil-water separation.

9
 10 Figure 7. Video sequence of the permeating behavior of one diesel oil droplet on the
 11 MFI@SS mesh. (a-1) -(a-6), the hydrophobic MFI coating on the 500-mesh screen; (b-
 12 1) -(b-6), the hydrophobic MFI coating on the 1000-mesh screen; (c-1) -(c-6), MFI
 13 coating on the 2000-mesh screen.

14 3.5. Anticorrosive performance

15 3.5.1. Tafel analyses

16 Figure 8 presents Tafel curves of the pristine SS mesh and the hydrophobic
 17 MFI@SS coating mesh measured at a scan rate of $0.01 \text{ V}\cdot\text{s}^{-1}$ in 3.5 wt.% NaCl

aqueous solution at room temperature. The result showed that the corrosion potential (E_{corr}) of the hydrophobic MFI@SS mesh shifted towards more positive potential from -0.99 V to -0.52 V, and the corrosion current I_{corr} decreased sharply approximate two orders of magnitude from $2.19 \times 10^{-5} \text{ A} \cdot \text{cm}^{-2}$ to $1.23 \times 10^{-7} \text{ A} \cdot \text{cm}^{-2}$ compared with those of the SS mesh (Table 3). Meanwhile, the polarization resistance, R_p , of the hydrophobic MFI@SS mesh was more than 300 times higher while the corrosion rate (CR) was approximately 180 times lower than those of the SS mesh. In general, lower I_{corr} or lower CR values suggest lower corrosion dynamic rate while more noble E_{corr} and higher R_p imply lower corrosion thermodynamic trend, which corresponds to better corrosion resistance and lower corrosion rate.^{41, 63} These results indicated that the hydrophobic MFI@SS mesh possessed better anticorrosion behavior under the corrosive condition.

Figure 8. Tafel curves of the pristine SS mesh and the hydrophobic MFI@SS coating mesh recorded at a scan rate of $0.01 \text{ V} \cdot \text{s}^{-1}$ in the 3.5 wt.% NaCl aqueous solution at room temperature.

Table 3. Electrochemical Parameters Achieved by Tafel Extrapolation Method ^a.

Item	E_{corr} (vs. SCE)/V	$\beta_a/\text{V}\cdot\text{dec}^{-1}$	$\beta_b/\text{V}\cdot\text{dec}^{-1}$	$I_{\text{corr}}/\text{A}\cdot\text{cm}^{-2}$	$R_p/\Omega\cdot\text{cm}^2$	CR/mm $\cdot\text{a}^{-1}$
SS mesh	-0.99	0.75	0.30	2.19×10^{-5}	4.26×10^3	0.17
MFI@SS	-0.52	0.70	0.84	1.23×10^{-7}	1.35×10^6	9.34×10^{-4}

^aThe data are normalized by apparent surface area.

3.5.2. EIS measurements

Electrochemical Impedance Spectroscopy (EIS) is another effective tool for characterizing the corrosion resistance of the materials. Figure 9 shows the Nyquist and Bode plots of the EIS data for the SS mesh as well as the hydrophobic MFI@SS coating mesh obtained in 3.5 wt. % NaCl aqueous solution at room temperature. The Nyquist diagram (Figure 9a) demonstrates that the hydrophobic MFI@SS mesh featured a larger diameter semicircle and higher impedances values at low frequencies than that of the SS mesh, which indicated that the MFI@SS mesh had a lower corrosion rate and better anticorrosion performance ⁴². This result was consistent with that obtained from the Tafel analysis (Figure 9). The Bode plot (Figure 9b) illustrated that both the peak value in the phase angle and the $|Z|$ value of the hydrophobic MFI@SS mesh at 0.01 Hz were greatly larger than those of the pristine SS mesh, which also suggested that the resulting mesh exhibited much preferable corrosion resistance. ⁶⁴

To give an insight into the anticorrosion property of the hydrophobic MFI@SS mesh, the EIS spectra were fitted by an electrical circuit (EC) model of $R(QR)(QR)$, as shown in Figure 10. A constant phase element (CPE), Q , is generally applied for the equivalent circuit to substitute for the ideal electrical capacitance in the real corrosion environment. ⁶⁵ R_s in the EC model represents the solution resistance ⁶⁶ between the working electrode (WE) and the reference electrode (RE). The left parallel circuit consists of the constant phase element, Q_c and the corrosion

resistance, R_{pore} of the hydrophobic MFI@SS coating (or the passive film corresponding to the pristine SS mesh as a comparison), respectively. This circuit at low frequency represents the first time constant in the Bode plot and is adapted to the electrical resistance to the ionic current through the pores of the coating or the passive film.⁴² Q_{dl} in the EC model stands for the constant phase element at the double layer coating/substrate interface. R_{ct} represents the charge transfer resistance at the passive film (coating)/electrolyte interface and indicates the permeating resistance of the ions and water through the pores⁴². The right parallel circuit (Q_{dl} , R_{ct}) in the EC model indicates the charge transfer process at coating/substrate interface⁶⁵, as shown in Figure 10.

The schematic diagram of the corrosion process is also shown in Figure 10. Although the prepared coatings appear very dense and compact, in the coatings, there still exists some surface cracks and pitting holes with the size from a few microns to tens of microns resulting from the sintering in a higher temperature.⁶⁷ As a result, the corrosive medium was able to penetrate the coating or the passive film formed on the SS surface and then reacted with the SS substrate which resulted in delamination and corrosion at the coating/SS interface.

The fitting results are both shown in Table 4 and Figure 9 as the black lines. The results of χ^2 tests in Table 4 were less than 2‰ indicating a good overlap between the experimental data (msd) and the simulated line (cal) by fitting the EIS spectra data with this EC model.⁶⁶ The quantitative data shown in Table 4 demonstrated that smaller numerical change of the solution resistance R_s was found suggesting the relative stability of three-electrode electrochemical systems during the EIS tests⁶⁷. Compared with the SS mesh, the high value of R_{pore} and the low value of Q_c indicated a better anticorrosion ability of the hydrophobic MFI@SS mesh. Equally, the value R_{ct} of the hydrophobic MFI@SS mesh coating was 5.3 times greater than that of the SS passive film, which also demonstrated the superior corrosion

1 resistance of the hydrophobic MFI coating.⁶⁵

2

3

4 Figure 9. Nyquist and Bode diagrams of the EIS data for the SS mesh and the
5 hydrophobic MFI@SS coating mesh obtained in 3.5 wt. % NaCl aqueous solution at room
6 temperature: (a) Nyquist plot and (b) Bode plot; msd: measured values, cal: fitted values
7 by using ZSimpWin 3.60.

Figure 10. Equivalent circuit for fitting the EIS data obtained in 3.5 wt. % NaCl aqueous solution at room temperature as well as the corresponding corrosion mechanism of the coating (RE: reference electrode, WE: working electrode).

Table 4. Optimum fit parameters for the SS mesh and the hydrophobic MFI@SS coating mesh using the model in Figure 10.^a

Item	$R_s/\Omega \cdot \text{cm}^2$	$Q_c/\Omega \cdot \text{s}^n \cdot \text{cm}^{-2}$	n_c	$R_{\text{pore}}/\Omega \cdot \text{cm}^2$	$Q_{\text{dl}}/\Omega \cdot \text{s}^n \cdot \text{cm}^{-2}$	n_{dl}	$R_{\text{ct}}/\Omega \cdot \text{cm}^2$	χ^2
SS mesh	24.45	6.60×10^{-5}	0.49	92.00	2.11×10^{-6}	0.80	2.21×10^5	1.70×10^{-3}
MFI@SS	34.56	3.24×10^{-6}	0.79	331.0	1.74×10^{-5}	0.85	1.66×10^6	1.23×10^{-4}

^a The data are normalized by apparent surface area.

3.6. Oil-water separation performance of the hydrophobic MFI@SS mesh

The separating experiments of the oil-water mixtures were carried out in the home-made separator (Figure 1 and Supporting information, V6) to evaluate the feasibility of the application of the hydrophobic MFI@SS meshes. The oil-water interface could be easily recognized because the water was dyed with red ink. When the oil-water mixture was piped into the separator by the peristaltic pump, the oil permeated quickly through the obtained mesh and flowed into the beaker

1 due to the capillarity effect and van der Waals interaction ⁶⁸ while the water was
2 prevented before the mesh and accumulated simultaneously in the left part of the
3 separator (Supporting information, V6). Subsequently, the blocked water was
4 pumped from the separator and recycled with the feedstock for further separation.
5 It was noteworthy that the whole separation process was continuous and
6 instantaneous (Supporting information, V6) and was promoted only by gravity
7 without involving any other external forces²⁷. We found that no visible red water
8 was observed in the final oil container after the oil-water mixture was separated
9 with the MFI@SS mesh. Furthermore, we found that the oil-water horizontal
10 interface almost located in the middle of the mesh section area during the stable
11 stage of the separating process as shown in Supporting information, V6.
12 Accordingly, it was noted that the effective area of S used for estimating the
13 separation mass flux in eq 3 should be the half of the cross-sectional area
14 calculated by the inside diameter of the organic glass tubes.

15 Figure 11a shows the separation flux and the efficiency of the hydrophobic
16 MFI@SS meshes for three different oil-water mixtures. The results indicated that
17 the separation flux was not only affected by the mesh screen, but also by the oil
18 type in the oil-water mixtures. Thus, higher mesh screen, namely smaller mesh
19 aperture size results in lower separation flux. On the other hand, the diesel oil-
20 water mixture had the highest separation flux while the vegetable oil-water
21 mixtures had much lower separation flux.

22 These phenomena could also be interpreted by the Hagen-Poiseuille eq 7. ^{61, 62}
23 On the one hand, from eq 7, we could ascertain that the flow flux varies directly as
24 r_p^2 and inversely as μ under the same pressure drop provided by gravity. In other
25 words, the mesh with larger pore radius, namely smaller mesh screen, leads to
26 higher separation flux for the same oil-water mixture, which is consistent with the

1 result in Figure 11a. On the other hand, interestingly, we found that the dynamic
2 viscosity of the oils using in the oil-water mixture are vastly different, especially
3 between the fuel oil (μ is 0.0030 Pa·s at 26 °C for diesel oil⁶⁹) and the vegetable oil
4 (μ is 0.049 Pa·s for sunflower oil and 0.057 Pa·s for peanut oil at 26 °C⁷⁰).
5 Therefore, we could conclude from eq 7 that the separation flux of the diesel oil-
6 water mixture was significantly higher than those of the sunflower oil- and the
7 peanut oil-water mixtures, while the separation fluxes had no essential difference
8 between two vegetable oil-water mixtures. As expected, the results in Figure 11a
9 were consistent with those deduced by eq 7.

10 Figure 11b shows that the separation efficiencies of all the hydrophobic
11 MFI@SS meshes for the selected oil-water mixtures were greater than 97.0%
12 which were comparable to the oil rejection rates of the hydrophilic or
13 superhydrophilic MFI type zeolite-coated meshes^{34, 35, 48}. Particularly, the
14 separation efficiency of the hydrophobic MFI@SS mesh with the 500-mesh screen
15 for the diesel oil-water mixture reached the highest value of 99.1%, which is
16 higher than or roughly equivalent to those of many superhydrophobic coated
17 meshes^{15, 46, 59, 68, 71, 72}. These results indicated that the prepared hydrophobic
18 MFI@SS coating meshes had equivalent separation performance with the
19 superhydrophobic coating meshes. We could also find that the separation
20 efficiency slightly decreased with increasing the mesh screen of the hydrophobic
21 MFI@SS mesh for all the oil-water mixtures. Similarly, the separation flux
22 decreased by increasing the dynamic viscosity of the oil in the oil-water mixtures
23 under the same hydrophobic MFI@SS mesh (Figure 11b).

24 The residual oil concentration in water after separation was 38.2-44.7 mg·L⁻¹
25 which was higher than the third-level sewage discharge standard of the petroleum
26 pollutants for urban drainage systems with sewage treatment plants in China (GB
27 8978-1996)⁷³. This result indicated that a small amount of oil entered the water

1 phase and formed an O/W emulsion. Therefore, the result suggested that the as-
 2 prepared hydrophobic MFI-type zeolite coated mesh was not suitable for oil-
 3 water emulsion separation. As a result, further work is strongly recommended to
 4 study the separation of the O/W emulsion.

5 Figure 11c shows that the WCA on the hydrophobic MFI@SS mesh had no
 6 remarkable decrease and still maintained the hydrophobic property after 20
 7 separation cycles in the oil-water mixture (by taking the diesel oil-water mixture
 8 as an example), which indicated the superb robustness and favorable stability of
 9 the mesh. Furthermore, as shown in Figure 11d, the separation efficiency, η was
 10 also above 98.0% and the separation flux, J , had no noticeable decline after 20
 11 separation cycles which demonstrated the outstanding reusability performance
 12 and the long-term stability of the mesh.

13
 14 Figure 11. Oil-water separation studies of the MFI@SS mesh: (a) and (b) the separation
 15 flux, J , and the separation efficiency, η , of different oil-water mixtures; (c) durability
 16 performance of the WCAs by taking the diesel oil-water mixture as an example; (d)
 17 reusability performance of the separation efficiency, η and the separation flux, J , by the

1 taking the diesel oil-water mixture as an illustration.

2 **4. CONCLUSIONS**

3 To summarize, we developed the MFI-type zeolite coatings on the stainless-
4 steel (SS) meshes by in situ crystallization method and followed by
5 hydrophobically modified with HDTMS for highly efficient oil-water separation.
6 The hydrophobic MFI@SS mesh showed eminent self-cleaning and anti-fouling
7 properties. The intrusion pressure of the MFI@SS mesh increased markedly with
8 the decrease of the mesh screen. Both specific water-repellent and oil-permeable
9 properties achieved simultaneously endowed the MFI@SS mesh with the 500-
10 mesh screen to be a very perspective material for oil-water separation. Tafel
11 analysis and EIS tests showed that the hydrophobic MFI@SS mesh possessed
12 better anticorrosion behavior under the corrosive condition. The continuous oil-
13 water separation experiments indicated that lower mesh screen, namely larger
14 mesh aperture size, along with lower dynamic viscosity of the oil resulted in
15 higher separation flux. The separation efficiency slightly decreased with
16 increasing the mesh screen of the hydrophobic MFI@SS mesh for all the oil-water
17 mixtures. The separation efficiency of the hydrophobic MFI@SS mesh with the
18 500-mesh screen for the diesel oil-water mixture reached the highest value of 99.1%
19 which was higher than or roughly equivalent to that of many superhydrophobic
20 coated meshes. The hydrophobic MFI@SS mesh demonstrated the excellent
21 reusability performance and the long-term stability after 20 separation cycles.
22 These as-prepared meshes would provide a versatile strategy for industrial use in
23 separating oily wastewater from oil field as well as recycling of waste oil from
24 catering services.

1 **ASSOCIATED CONTENT**

2 **Supporting Information**

3 The Supporting Information is available free of charge on ACS Publications
4 website.

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13 **Notes**

14 The authors declare no competing financial interest.

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20 **Contents of the supporting information**

21 Supporting Information V1: The self-cleaning property of the hydrophobic
22 MFI@SS mesh was qualitatively evaluated with muddy water solution (clay
23 content $\sim 300 \text{ kg}\cdot\text{m}^{-3}$). The result demonstrated that the coating mesh surface
24 could keep clean and the original surface color (AVI).

Supporting Information V2: Chalk powder covered the hydrophobic MFI@SS surface to simulate dust pollution. The result showed that the chalk powder could hardly stick to the mesh surface which demonstrated the self-cleaning and non-sticking performance of the as-prepared mesh (AVI).

Supporting Information V3-V5: The diesel oil drop spread and passed through the hydrophobic MFI@SS mesh with the 500-, 1000-, 2000 mesh screen, respectively. The videos were recorded with a mobile digital camera with 3000 pixels to obtain the permeating time (AVI).

Supporting Information V6: The video recorded the separating experiments of the oil-water mixtures in the home-made separator to evaluate the feasible application of the hydrophobic MFI@SS meshes in oil-water separation (AVI).

Graphical abstract

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