Desalination and Water Treatment
www.deswater.com
doi:10.5004/dwt.2018.22207

111 (2018) 155–164
April

Superhydrophilicity and underwater superoleophobicity graphene oxide-micro crystalline cellulose complex-based mesh applied for efficient oil/water separation

Tao Yan[a], HuiwenMeng[a], Wenjihao Hu[a,b,]*, Feipeng Jiao[a,]*

[a]*Department of Chemical Engineering, Central South University, Changsha 410083, China, Tel. +86- 731-58293557 Fax +86- 731-58293557, email: jiaofp@163.com (F. Jiao)*
[b]*Department of Chemical and Material Engineering, University of Alberta, Edmonton T5K1B9, Canada, Tel. +1-780-716-2168, Fax +1-780-716-2168, email: huwenjihaoluke@163.com (W. Hu)*

Received 26 October 2017; Accepted 9 March 2018

ABSTRACT

With the aggravation of the pollution in our environment, the novel interface materials have gained more and more attention. In this work, we reported an approach to prepare graphene oxide-microcrystalline cellulose complex-based mesh with superhydrophilicity and underwater superoleophobicity. Hydrophilic groups were grafted to enhance the superhydrophilicity and improve the superoleophobicity at the same time. The modified mesh exhibited under water superoleophobicity with oil contact angle more than 150°, while water can quickly permeate the wire mesh surface with a contact angle value of 0°. In the process of oil/water separation tests, water molecules permeate the wire mesh under the force of gravity, and the oil was retained on the mesh. Thus, the oil/water mixtures with different proportions can be separated in a high efficiency up to 99.1%. In addition, our superhydrophilic meshes are robust in harsh water conditions and can be used as an efficient filtration membrane. And our superhydrophilic meshes still have high efficiency separation in poor conditions, such as alkaline,acidic or saline solutions. Thus, this modified mesh could be an ideal choice for practical oil/water separation which can satisfy the need for water body restoration.

Keywords: Superhydrophilicity; Underwater superoleophobicity; Graphene oxide; Microcrystalline cellulose; Oil/water separation

1. Introduction

With the rapid economic growth and industrial development, a great quantity of oily wastewater has been discharged inhuman daily life as well as many industrial processes. Chemical leakage and oil spills accidents occurred frequently, which not only brought on devastating influence on the ecological environment, but also exposed the whole human safety to this kind of danger [1–9]. Effective methods for controlling the oily wastewater are still a challenge at present [10,11]. Thus, there are massive requirements for advanced materials that can effectively separate water from oily wastewater. Surfaces with a special wettability are apparently significant in the separation of water and oil when refers to water circulation, oil purification and oily wastewater treatment [12,13]. It is common knowledge that the wettability of materials toward oil and water can be reasonably transformed by changing its chemical composition and surface topography of surfaces [14,15]. Until now, a series of super wetting materials have been widely used for oil/water separation [16–18]. For instance, Cao et al. synthesized superhydrophobic and oleophilic copper mesh which was modified by dopamine and n-dodecyl mercaptan [19]. The effective oil/water separation of the superhydrophobic and superoleophilic mesh was made by

*Corresponding author.

Chen et al. [20]. Cho et al. prepared superhydrophobic and transparent siloxane-based nano composites for effective oil/water separation [21]. Nevertheless, these super wetting surfaces are easily polluted or blocked by oil because of their oleophilic properties [22,23]. Furthermore, most artificial superhydrophobic surfaces are unable to meet the requirement of self-cleaning coatings [24,25]. In addition, most of this materials could cause secondary pollution.

In order to deal with the problem of separation of oil/water, an opposite super wetting surface is proposed, which is superhydrophilic and superoleophobic. This material allows the water to pass through, leaving the oil on the surface [26,27]. Theoretically, super hydrophilic/superoleophobic surfaces are energy reversible. Generally, oil has a relatively small surface tension than water [28]. As a result, it is still a great challenge to acquire superhydrophilic/superoleophobic materials [29–33].

Recently, lots of researches have been dedicated to the study of superhydrophilic and superoleophobic materials applied to oil/water separation [34–37]. For instance, Zhang et al. synthesized superhydrophilic and underwater superoleophobic mesh with silicate/TiO_2 coated [38]. Xue et al. synthesized a superhydrophilic and superoleophobic copper mesh that used Cu_2S coatings on mesh [29]. Nevertheless, the main obstacle that constrains them to use is their weak environmental adaptability because they can notre main stable in complex environments such as acidic, alkaline, or saline conditions.

In this study, we develop a cost-effective, chemical stable superhydrophilic and underwater superhydrophobic mesh that is coated with MCC-GO/SiO_2 hydrophilic material on wire mesh and also it is environment-friendly material. The modified mesh has excellent superoleophobicty with oils contact angle more than 150°, while water can quickly permeate the mesh with a contact angle value of 0° due to its hydrophilicity. The separation of oil/water mixtures with different ratio was carried out under the driving force of gravity. The result showed that the brilliant oil/water separation efficiency was up to 99.1%. Thus, it could be extensively used in separating many kinds of oils. Furthermore, the presentation stability of the modified mesh by using a variety of corrosive solution (1 mol/L HCl, 1 mol/L NaOH, 1 mol/L NaCl solution) to test it, the result showed that the modified mesh in harsh environments have high stability. The mesh along with the oil/water separation equipment showed in this study may indicate a new method for large-scale oil/water separation.

2. Experiment

2.1. Materials

Wire mesh (300 mesh size) and micro crystalline cellulose (99%) were obtained from Alibaba Enterprise. Dichloromethane (99%), potassium permanganate (99%, Guangzhou Chemical Reagent Factory, China), sodium hydroxide (96%) and SiO_2 nano particles (average diameter ~20 nm) were purchased from Sinopharm Chemical Reagent Co., Ltd., China. Anhydrous ethanol (99.5%, Guangzhou Chemical Reagent Factory, China), sulfuric acid (98%, Zhuzhou starry sky Glass Co., Ltd) and hydrochloric acid (35%, Zhuzhou starry sky Glass Co., Ltd) were all of analytical grade. Hydrogen peroxide (30%, w/w) was selected from Tianjin Fuqi Chemical Co., Ltd., China. Urea (99%) were obtained from Tianjin Heng xing chemical Reagent Co., Ltd. All of the other chemicals were used as received without further purification.

2.2. Fabrication of MCC-GO composites solution

Graphene oxide (GO) was fabricated from graphite powder by modified Hummers' method [39]. Graphene oxide-micro crystalline cellulose composites (MCC-GO) were synthesized by a simple method. Briefly, 0.30 g of GO was dispersed in 60 mL of deionized water and the solution was ultrasonicated for 30 min at room temperature. Then, urea (8 g) and sodium hydroxide (12 g) were dissolved in 90 mL deionized water and the mixtures were kept in ultrasonic bath for 20 min at 25°C. After that, the mixtures were kept at –12°C for 1 h, 1.05 g of micro crystalline cellulose was added and then stirred well for 2 h to obtain the temporarily stable dissolution. Finally, 9 mL of MCC solution was added to GO aqueous suspension with stirring for 6 h at room temperature. The resulting slurry was treated by centrifugation at 9000 rpm for 15 min and washed with deionized water for several times. The whole washing step was repeated until the pH of solution reaches 7 by using pH measuring paper.

2.3. Synthesis of MCC-GO/SiO_2 hybrid solution coatings

A suspension of SiO_2 nano particles (see Supporting Information S1 for characterizations) in deionized water (3 mL, ~1 wt%) was ultrasonically dispersed in MCC-GO-composites solution with stirring for 30 min at 25°C. We refered to this polymer composites as MCC-GO/SiO_2. The synthesized products (MCC-GO/SiO_2) were dispersed under sonication. The resulting suspensions were coated on wire mesh by immersing. The wire mesh was washed at room temperature with ultra pure water and ethanol solution in ultra sonic bath before immersion. The product coatings were dried at 80°C for 10 min. The immersion and drying process was repeated three to five times.

2.4. Oil/water separation ability

The modified meshes were applied to separating oil/water mixtures. A set number of oils (sesame oil, paraffin,and hexamethylene) were added into water to generate oil/water mixtures. The modified meshes were placed in the middle of the glass tube. Subsequently, the oil/water mixtures were ran directly into the surface of the glass tube, and water will go through the pore of mesh into a beaker and then separated oil on the mesh. The separation efficiency (R) of oil/water mixtures were calculated by collecting the quality of water (M_s) after separation and the water quality of the initial oil/water mixtures (M_0):

$$R = M_0/M_s \times 100\%,$$

all the separation efficiency of the work was tested three times, and an average value was recorded.

2.5. Instrumentation and characterization

The surface micro structure of meshes were observed using a scanning electron microscope (SEM) (TESCAN MIRA3 LMU) and the elemental analysis were detected by energy dispersive X-ray spectroscopy (EDS) (SAMX) method. The Fourier transform infrared (FT-IR) spectroscopic analysis was tested by a Nicolet-Avatar 360 FTIR spectrometer within the wave number from 300 to 4000 cm^{-1}. The water contact angle (WCA) was tested by using JC2000D1, Powereach. Co. Ltd at 25°C. 4 µL of water droplet vertically dropped from the syringe and then contacted the surface of the samples. Contact angles on all samples were tested for three times and average value was recorded.

3. Results and discussion

3.1. Chemical composition of the mesh surface

The surface chemical bond of the material was characterized by FTIR. Herein, GO, pristine wire mesh and MCC-GO/SiO_2 spectra were depicted in Fig. 1. In the GO spectrum, the strong band at 3400 and 1734 cm^{-1} were related to the –OH stretching vibrations and the C=O stretching vibrations while the peaks at 1620 and 1141 cm^{-1} were attributed to the C=C stretching vibrations and alkoxy C-O stretching vibrations, respectively. Obviously, there were no peaks in the pristine wire mesh. Compared with GO, MCC-GO/SiO_2 mesh showed the band around 3450 cm^{-1} ascribed to stretching vibration of –OH. A new peak at 2916 cm^{-1} was attributed to a symmetric C-H stretching vibration. A new peak could be observed at 1215 cm^{-1}, which was ascribed to C-O-C stretching vibration of the MCC-GO, confirming that the electrostatic attraction and dehydration condensation reaction between the carboxyl groups of GO and hydroxyl groups of MCC has occurred. Furthermore, the absorption peak at 837 cm^{-1} was characteristics of Si-O bending vibration and the absorption peak at 1033 cm^{-1} is contributed from Si-O-Si asymmetric stretching vibrations. It is indicated that the SiO_2 nano particles was successfully coated on the mesh [21,40–43].

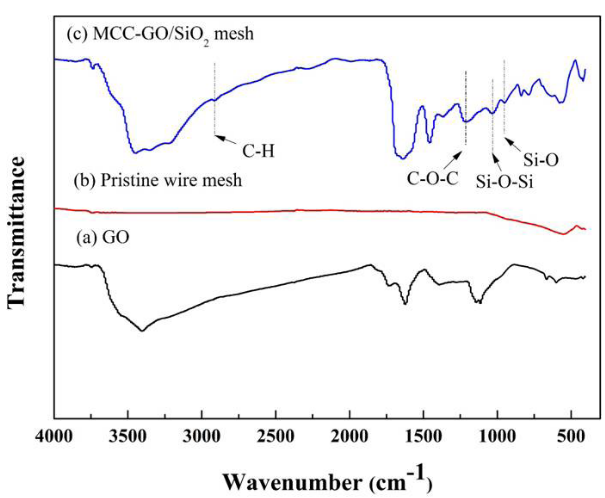

Fig. 1. FTIR spectra for (a) GO, (b) Pristine steel mesh and (c) MCC-GO/SiO_2 mesh.

3.2. EDS and XPS

Figs. 2a and 2b demonstrate the EDS spectra of the pristine wire mesh and MCC-GO/SiO_2 mesh, respectively. In the EDS experiment (Fig. 2a) Ni, Mn, Cr and Fe element scan be discovered in the pristine wire mesh, which were fundamental elements of steel mesh. Compared with pristine wire mesh surface (Fig. 2b), the strong peak area on Si and C can be distinctly found in MCC-GO/SiO_2 mesh, and the table next to the figure was their chemical compositions, which conforms to the chemical compositions of MCC-GO/SiO_2 mesh as expected. Thus, the MCC-GO/SiO_2 was dispensed all over the mesh. The same results are obtained in the XPS spectra (Fig. 2c), and a small amount of C element can be detected in the pristine wire mesh due to different sensitivity. The new element Si was found in MCC-GO/SiO_2 mesh, further indicating wire mesh was modified by MCC-GO/SiO_2 successfully.

3.3. FESEM

The surface topographical images of the pristine wire mesh and MCC-GO/SiO_2 mesh were detected by SEM. Two kinds of meshes had similar structure, indicating that the immersion process of this work enabled the

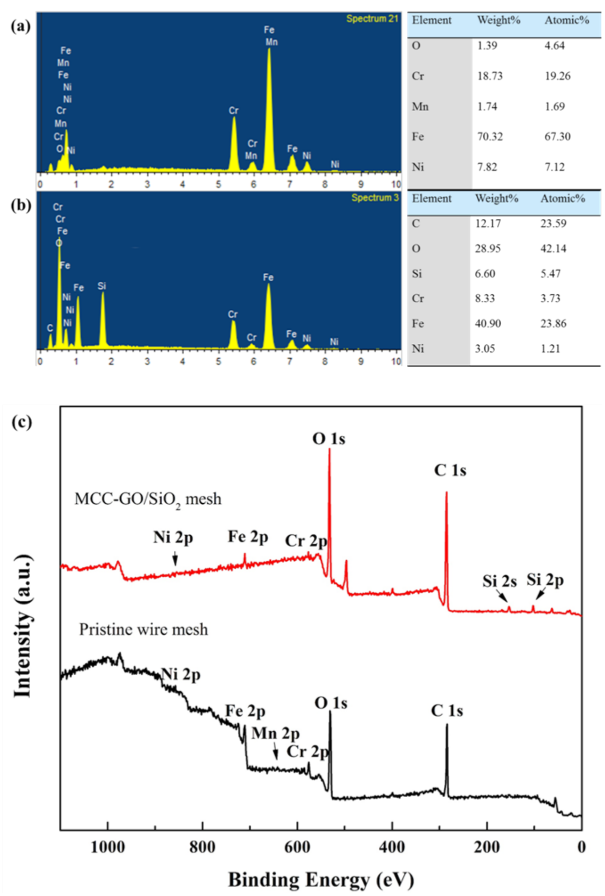

Fig. 2. (a) EDS curves of the pristine steel mesh and (b) MCC-GO/SiO_2 mesh. (c) XPS spectra of pristine wire mesh and MCC-GO/SiO_2 coated mesh.

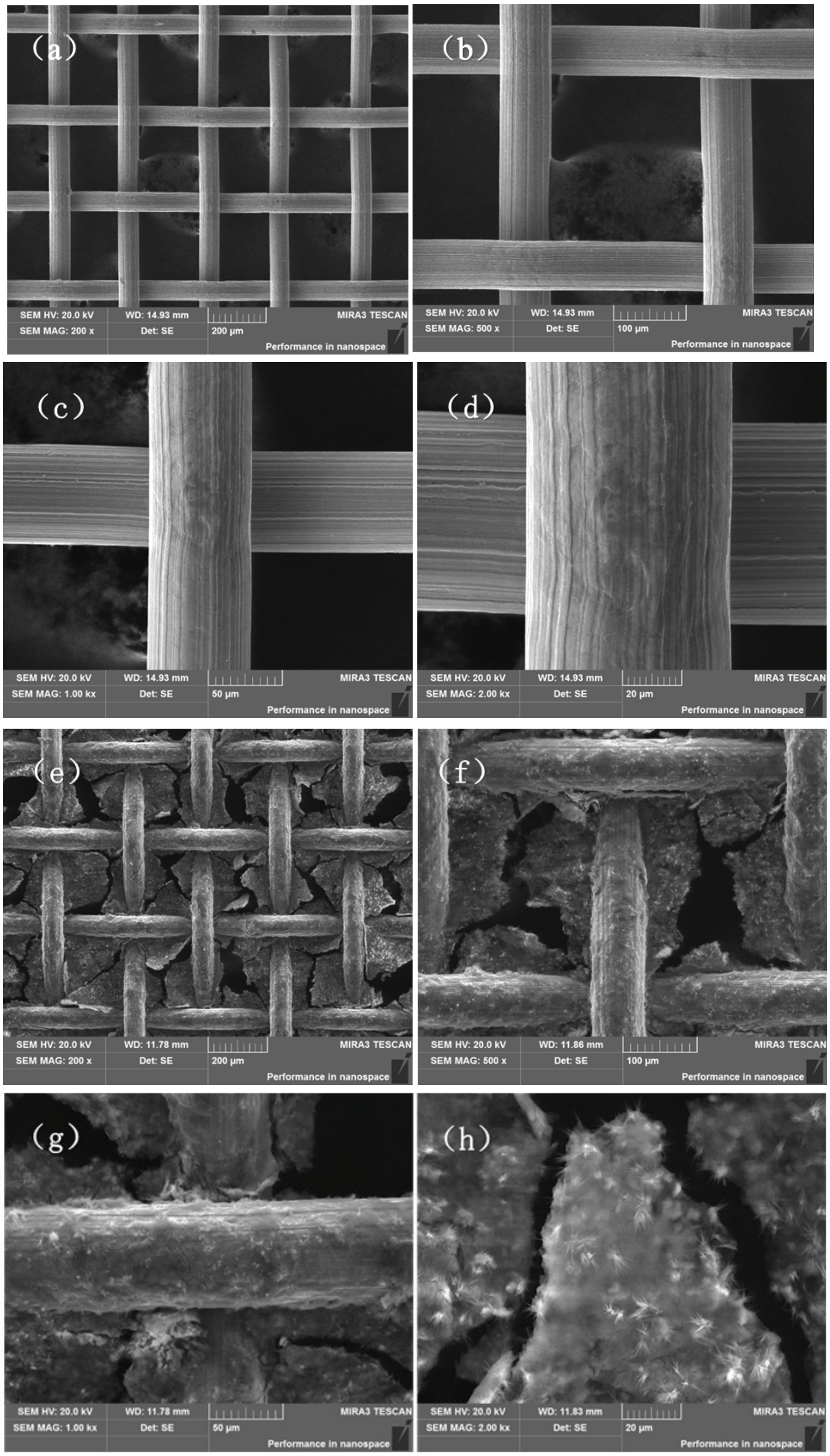

Fig. 3. SEM images with different magnifications for pristine steel mesh(a–d) and MCC-GO/SiO_2 mesh (e–h).

initial structures of the wire mesh to be retained in the MCC-GO/SiO_2 mesh. Figs. 3a and 3b display an image of the wire mesh with an average square-shaped pore size of ~45–55 µm. A thick layer of composite coating material was found on the surface and in the pores of the wire mesh after immersion (Figs. 3e and 3f). The initial wire mesh showed a quite smooth surface even from the magnified image (Figs. 3c and 3d). After modification, the MCC-GO/SiO_2 mesh became rougher and mesh skeletons were wrapped by a composite coating, it can be observed that the wire mesh offered a substrate for the MCC-GO/SiO_2 to be built on the pore surface of the wire mesh. It can be clearly seen that the surface morphology of the mesh changed to porous structure with nano-walls (Fig. 3g). From Fig. 3h, it is obviously seen in the figure that graphene oxide-micro crystalline cellulose complex have been prepared.

3.4. Mesh film wettability

The wettability of the film was constructed by the chemical composition and micro structures of material surface [33,44,45]. On the one hand, compared with the pristine wire mesh, the MCC-GO/SiO_2 mesh showed a microscopic surface structure, suggesting that the surface roughness had been obviously boosted. On the other hand, the hydrophilic SiO_2 doping can be predicted to increase the hydrophilicity. By this means, the properties of superhydrophilic and underwater superhydrophobic were shown in the MCC-GO/SiO_2 mesh as expected (Fig. 4).

In order to further examine the underwater superoleophobicproperty of MCC-GO/SiO_2 toward other organic reagents or oils, we investigated the CAs of a string of oil mixtures and organic solvents such as paraffin, sesame oil and organic reagents of n-hexadecane, n-dodecane, dichloromethane, toluene, carbontetrachloride, and hexamethylene. Obviously, all of these oil and organic reagents droplets have CAs larger than 150°, suggesting the excellent under water superoleophobic properties of MCC-GO/SiO_2 mesh (Fig. 5a). Apparently, the MCC-GO/SiO_2 mesh showed no selectivity for different oils and organic solvents, so it was appropriate for the efficient separation of any oil species that were undissolved in water. Moreover, the superhydrophilic property was shown in Fig. 5b, a drop of water can quickly permeate the MCC-GO/SiO_2 mesh surface within 1.2 s. The wettability of the individual material and the wettability of their mixtures were displayed in the supplementary information (Fig. S2 of the Supporting Information). Their wettability was worse than the MCC-GO/SiO_2 mesh.

3.5. Oil/water separation performance

To further explore the performance of the MCC-GO/SiO_2 mesh, it was applied to the oil/water separation experiment. Paraffin/water mixtures can be easily separated by gravity-driven separation equipment as shown in Fig. 6 (Movie S1, Supporting information). The oil/water mixtures were shown in Fig. 6a. The modified mesh was placed in the middle of two glass tubes, and the oil/water mixtures directly passed through the upper glass tube (Figs. 6b and 6c). Water molecules permeate the mesh due to the force of gravity, while paraffin was left on the mesh because of its superoleophobicity (Fig. 6d). The separation efficiency of the modified mesh was 99.1%, implying that the paraffin/water mixtures were successfully separated with high efficiency. This could be ascribed to the modified mesh surface that demonstrates the underwater superoleophobic property, and the density of paraffin is lower than the water.

To quantitatively evaluate the oil/water separation ability of the MCC-GO/SiO_2, three types of oil/water mixtures (sesame oil/water mixtures; paraffin/water mixtures; hexamethylene/water mixtures) were examined with different volume proportions of water to oil.

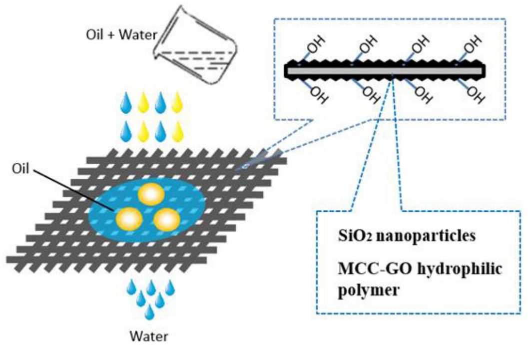

Fig. 4. Schematic illustration of the superhydrophilic and superoleophobic MCC-GO/SiO_2 based mesh for oil/water separation.

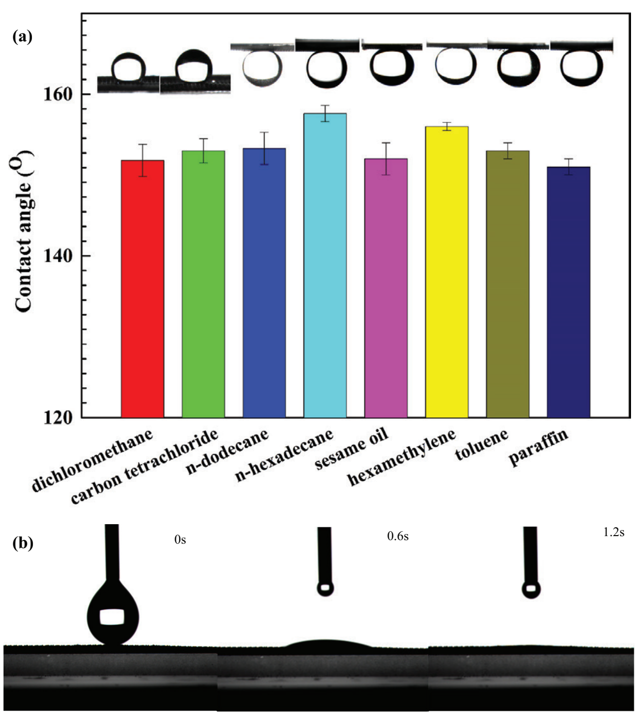

Fig. 5. (a) CAs of different organic solvents and oils on MCC-GO/SiO_2. The insets are photographs of various droplets (b) The processes of a water droplet spread over the MCC-GO/SiO_2 mesh.

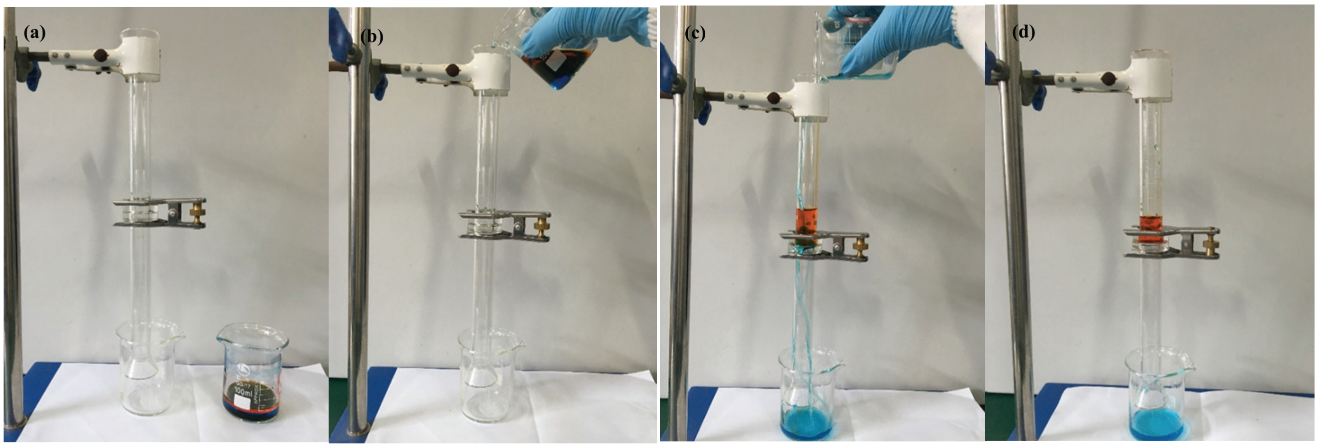

Fig. 6. Experimental process of oil/water separation (water is dyed with methylene blue and oil is dyed with oil red O for clear observation).

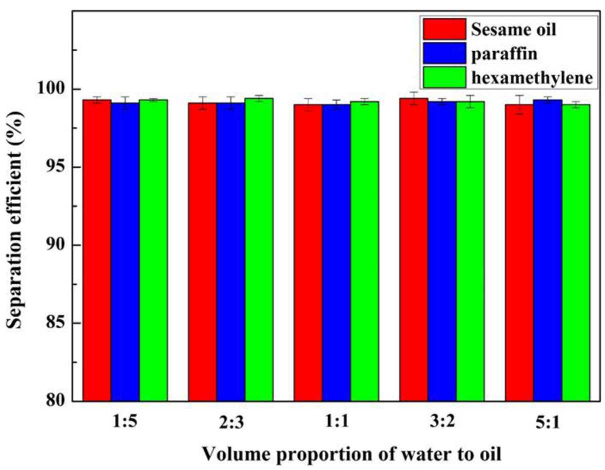

Fig. 7. Separation efficiency of superhydrophilic MCC-GO/SiO_2 for different oil/water mixtures with different oil/water ratio.

As shown in Fig. 7, the separation efficiency of sesame oil/water mixtures with different oil/water proportions were calculated. According to the formula R = M_0/M_s × 100%, the separation efficiencies were 99.3%, 99.1%, 99.0%, 99.4%, and 99.1%, respectively. These consequences showed the brilliant oil/water separation ability of the MCC-GO/SiO_2 mesh, and it indicated that the volume proportion of water and oil had no influence on the separation efficiency. In the case of separating the paraffin/water mixtures and hexamethylene/water mixtures, the same results were also obtained. More importantly, as shown in Fig. 8, in order to examine the performance stability of the MCC-GO/SiO_2 coated mesh,we used several corrosive solutions, involving hydrochloric acid (HCl), sodiumchloride (NaCl), and sodium hydroxide (NaOH). All of them can be successfully separated, showing that the mesh still have a stable separation ability in hostile environment. Furthermore, as shown in Fig. 9, the separation efficiency under different harsh conditions was all above 99.0%, which will provide significant approaches in practical water body restoration.

3.6. Recyclability

The recyclability of the MCC-GO/SiO_2 mesh after ten cycles of separation is exhibited in Fig. 10. The paraffin/water mixtures and hexamethylene/water mixtures were applied to measuring recyclability. After each separation is completed, the polluted mesh was washed by ethanol to get rid of the attached oil and then dried to continue. The clean mesh was used for the next times of separation. In general, the separation efficiency showed excellent ability after ten cycles of separation. The decline is possibly ascribed to the slight effect on the surface structure repeatedly washed by drying process and ethanol. This finding indicated that the MCC-GO/SiO_2 mesh can be recycled for several times.

To further understand the oil/water separation mechanism of MCC-GO/SiO_2 mesh, we have established schematic models in Figs. 11a and 11b. The rough structure and porous structure of the mesh film plays a vital role in acquiring the pattern of liquid permeation. The intrusion pressure (Δp) was illustrated by the following formula:

$$\Delta p = -2\gamma \cos\theta / R$$

where γ is the inter facial tension, θ the contact angle, and R the radius of the pores. When an oil/water mixtures were poured on the MCC-GO/SiO_2 mesh, it was apparent that oil cannot penetrate through the modified mesh with an intrusion pressure $\Delta p > 0$ (negative capillary effect) and the contact angle $\theta > 90°$due to its underwater superoleophobicity (Fig. 11a). Meanwhile,the modified mesh is superhydrophilic and the contact angle $\theta < 90°$, leading to a positive capillary effect ($\Delta p < 0$), and it allows water to pass though the mesh in air (Fig. 11b). Thus, the MCC-GO/SiO_2 mesh possesses a high separation efficiency for oil/water mixtures.

4. Conclusions

In a word, we fabricated a cost-effective, chemically stable MCC-GO/SiO_2 coated mesh with superhydrophilicity and underwater superoleophobicity by immersing. The surface morphology of the mesh with rough structure and

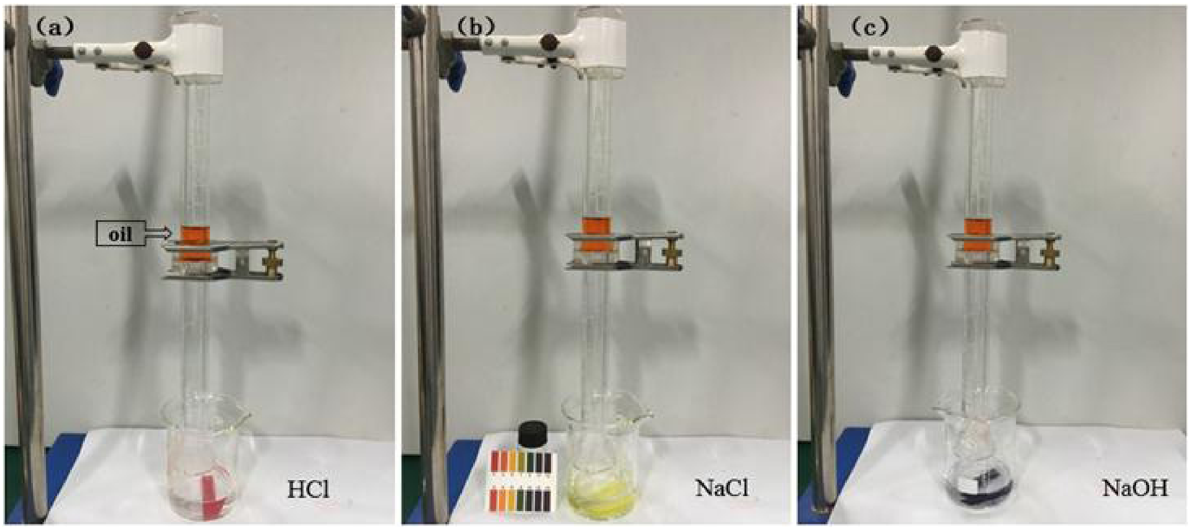

Fig. 8. Gravity–driven oil/water separation in a harsh environment. Mixture of oil and (a) HCl, (b) NaCl, and (c) NaOH aqueous solution as feed.

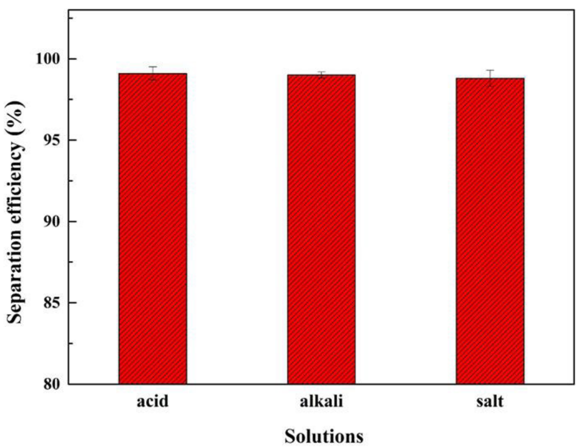

Fig. 9. The separation efficiency of superhydrophilic MCC-GO/SiO_2 for paraffin/water mixture after being immersed into acidic (pH = 1), alkaline (pH = 13), and salty (3.5 wt.%) solutions.

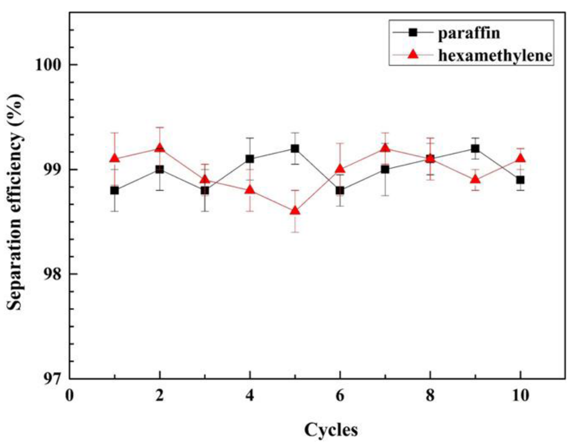

Fig. 10. Recyclabilities of the MCC-GO/SiO_2 mesh.

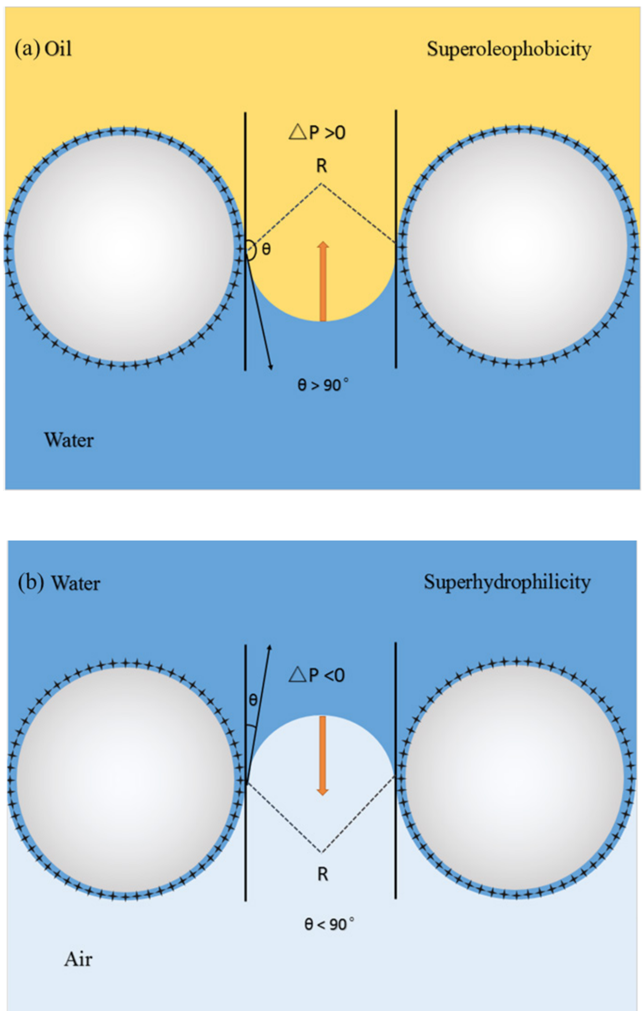

Fig. 11. (a) Oil cannot pass through the mesh as $\Delta p > 0$; (b) water can permeate through the mesh because $\Delta p < 0$.

hydrophilic groups resulted in the mesh that presented superhydrophilicity and underwater superoleophobicity. The contact angle of water is 0° while the contact angle for oil is 157° ± 2°. In oil/water separation tests, water molecules permeate the mesh by the force of gravity and oil was left on the mesh. To evaluate the stability of the mesh, this mesh was used for separating water from the oil/water mixtures in a harsh environment such as acidic, saline or alkaline conditions,and separation efficiency was still very high. These results are useful in the application of oil/water separation to meet the needs for water body restoration.

Acknowledgments

The authors thank National Natural Science Foundation of China (Grant No.21776319) for the financial supports of this work.

References

[1] J. Wang, L. Shang, Y. Cheng, H. Ding, Y. Zhao, Z. Gu, Microfluidic generation of porous particles encapsulating spongy graphene for oil absorption, Small, 11 (2015) 3890–3895.
[2] Y. Yu, H. Chen, Y. Liu, V.S.J. Craig, C. Wang, L.H. Li, Y. Chen, Superhydrophobic and superoleophilic porous boron nitride nano sheet/polyvinylidene fluoride composite material for oil-polluted water cleanup, Adv. Mater. Interfaces, 2 (2015) 1400267.
[3] W. Liang, Z. Guo, Stable superhydrophobic and superoleophilic soft porous materials for oil/water separation, RSC Adv, 3 (2013) 16469.
[4] Z.-X. Wang, C.-H. Lau, N.-Q. Zhang, Y.-P. Bai, L. Shao, Mussel-inspired tailoring of membrane wettability for harsh water treatment, J. Mater. Chem. A., 3 (2015) 2650–2657.
[5] H. Bi, X. Xie, K. Yin, Y. Zhou, S. Wan, L. He, F. Xu, F. Banhart, L. Sun, R.S. Ruoff, Spongy graphene as a highly efficient and recyclable sorbent for oils and organic solvents, Adv. Funct. Mater., 22 (2012) 4421–4425.
[6] Q. Cheng, D. Ye, C. Chang, L. Zhang, Facile fabrication of superhydrophilic membranes consisted of fibrous tunicate cellulose nano crystals for highly efficient oil/water separation, J. Membr. Sci., 525 (2017) 1–8.
[7] W. Ma, Z. Guo, J. Zhao, Q. Yu, F. Wang, J. Han, H. Pan, J. Yao, Q. Zhang, S.K. Samal, S.C. De Smedt, C. Huang, Polyimide/cellulose acetate core/shell electro spun fibrous membranes for oil-water separation, Sep. Purif. Technol., 177 (2017) 71–85.
[8] C.-H. Xue, P.-T. Ji, P. Zhang, Y.-R. Li, S.-T. Jia, Fabrication of superhydrophobic and superoleophilic textiles for oil–water separation, Appl. Surf. Sci., 284 (2013) 464–471.
[9] J. Liu, P. Li, L. Chen, Y. Feng, W. He, X. Yan, X. Lü, Superhydrophilic and underwater superoleophobic modified chitosan-coated mesh for oil/water separation, Surf. Coat. Technol., 307 (2016) 171–176.
[10] Q. Ma, H. Cheng, A.G. Fane, R. Wang, H. Zhang, Recent development of advanced materials with special wettability for selective oil/water separation, Small, 12 (2016) 2186–2202.
[11] G.J. Dunderdale, C. Urata, T. Sato, M.W. England, A. Hozumi, Continuous, high-speed, and efficient oil/water separation using meshes with antagonistic wetting properties, ACS Appl. Mater. Interfaces, 7 (2015) 18915–18919.
[12] Z. Chu, Y. Feng, S. Seeger, Oil/water separation with selective super anti wetting/super wetting surface materials, Angew. Chem. Int. Ed., 54 (2015) 2328–2338.
[13] T. Jiang, Z. Guo, W. Liu, Biomimetic superoleophobic surfaces: focusing on their fabrication and applications, J. Mater. Chem. A, 3 (2015) 1811–1827.
[14] S.S. Latthe, A.L. Demirel, Polystyrene/octadecyltrichlorosilane superhydrophobic coatings with hierarchical morphology, Polym. Chem., 4 (2013) 246–249.
[15] R. Wahi, L.A. Chuah, T.S.Y. Choong, Z. Ngaini, M.M. Nourouzi, Oil removal from aqueous state by natural fibrous sorbent: An overview, Sep. Purif. Technol., 113 (2013) 51–63.
[16] J. Li, D. Li, Y. Yang, J. Li, F. Zha, Z. Lei, A prewetting induced underwater superoleophobic or under oil (super) hydrophobic waste potato residue-coated mesh for selective efficient oil/water separation, Green Chem., 18 (2016) 541–549.
[17] R. Gao, Q. Liu, J. Wang, J. Liu, W. Yang, Z. Gao, L. Liu, Construction of superhydrophobic and superoleophilic nickel foam for separation of water and oil mixture, Appl. Surf. Sci., 289 (2014) 417–424.
[18] J.T. Korhonen, M. Kettunen, R.H. Ras, O. Ikkala, Hydrophobic nano cellulose aerogels as floating, sustainable, reusable, and recyclable oil absorbents, ACS Appl. Mater. Interfaces, 3 (2011) 1813–1816.
[19] H. Cao, W. Gu, J. Fu, Y. Liu, S. Chen, Preparation of superhydrophobic/oleophilic copper mesh for oil-water separation, Appl. Surf. Sci., 412 (2017) 599–605.
[20] Q. Chen, A. de Leon, R.C. Advincula, Inorganic-organic thiol-ene coated mesh for oil/water separation, ACS Appl. Mater. Interfaces, 7 (2015) 18566–18573.
[21] J. Li, D. Li, W. Li, H. Li, H. She, F. Zha, Facile fabrication of underwater superoleophobic SiO_2 coated meshes for separation of polluted oils from corrosive and hot water, Sep. Purif. Technol., 168 (2016) 209–214.
[22] Y. Yang, Z. Tong, T. Ngai, C. Wang, Nitrogen-rich and fire-resistant carbon aerogels for the removal of oil contaminants from water, ACS Appl. Mater. Interfaces, 6 (2014) 6351–6360.
[23] S. Zhou, G. Hao, X. Zhou, W. Jiang, T. Wang, N. Zhang, L. Yu, One-pot synthesis of robust superhydrophobic, functionalized graphene/polyurethane sponge for effective continuous oil–water separation, Chem. Eng. J., 302 (2016) 155–162.
[24] Y. Dong, J. Li, L. Shi, X. Wang, Z. Guo, W. Liu, Underwater superoleophobic graphene oxide coated meshes for the separation of oil and water, Chem. Commun., 50 (2014) 5586–5589.
[25] A. Raza, B. Ding, G. Zainab, M. El-Newehy, S.S. Al-Deyab, J. Yu, In situ cross-linked super wetting nano fibrous membranes for ultrafast oil–water separation, J. Mater. Chem. A, 2 (2014) 10137.
[26] J. Yang, Z. Zhang, X. Xu, X. Zhu, X. Men, X. Zhou, Superhydrophilic–superoleophobic coatings, J. Mater. Chem., 22 (2012) 2834.
[27] J. Yang, H. Song, X. Yan, H. Tang, C. Li, Superhydrophilic and superoleophobic chitosan-based nano composite coatings for oil/water separation, Cellulose, 21 (2014) 1851–1857.
[28] J. Li, L. Yan, Y. Zhao, F. Zha, Q. Wang, Z. Lei, One-step fabrication of robust fabrics with both-faced superhydrophobicity for the separation and capture of oil from water, Phys. Chem. Chem. Phys., 17 (2015) 6451–6457.
[29] Z. Xue, S. Wang, L. Lin, L. Chen, M. Liu, L. Feng, L. Jiang, A Novel superhydrophilic and underwater superoleophobic hydrogel-coated mesh for oil/water separation, Adv. Mater., 23 (2011) 4270–4273.
[30] M.A. Gondal, M.S. Sadullah, M.A. Dastageer, G.H. McKinley, D. Panchanathan, K.K. Varanasi, Study of factors governing oil-water separation process using TiO_2 films prepared by spray deposition of nano particle dispersions, ACS Appl. Mater. Interfaces, 6 (2014) 13422–13429.
[31] C. Zhou, J. Cheng, K. Hou, A. Zhao, P. Pi, X. Wen, S. Xu, Superhydrophilic and underwater superoleophobic titania nano wires surface for oil repellency and oil/water separation, Chem. Eng. J., 301 (2016) 249–256.
[32] L. Feng, Z. Zhang, Z. Mai, Y. Ma, B. Liu, L. Jiang, D. Zhu, A super-hydrophobic and super-oleophilic coating mesh film for the separation of oil and water, Angew. Chem. Int. Ed., 43 (2004) 2012–2014.
[33] J. Yang, L. Yin, H. Tang, H. Song, X. Gao, K. Liang, C. Li, Polyelectrolyte-fluorosurfactant complex-based meshes with superhydrophilicity and superoleophobicity for oil/water separation, Chem. Eng. J., 268 (2015) 245–250.

[34] Y. Li, H. Zhang, M. Fan, P. Zheng, J. Zhuang, L. Chen, A robust salt-tolerant superoleophobic alginate/graphene oxide aerogel for efficient oil/water separation in marine environments, Sci. Rep., 7 (2017) 46379.
[35] D. Zhu, Y. Xia, J. Yang, B. Chen, S. Guo, C. Li, One-step removal of insoluble oily compounds and water-miscible contaminants from water by underwater superoleophobic graphene oxide-coated cotton, Cellulose, 24 (2017) 5605–5614.
[36] C. Ao, W. Yuan, J. Zhao, X. He, X. Zhang, Q. Li, T. Xia, W. Zhang, C. Lu, Superhydrophilic graphene oxide@electrospun cellulose nano fiber hybrid membrane for high-efficiency oil/water separation, Carbohydr. Polym., 175 (2017) 216–222.
[37] F. Sun, W. Liu, Z. Dong, Y. Deng, Underwater superoleophobicity cellulose nano fibril aerogel through regioselective sulfonation for oil/water separation, Chem. Eng. J., 330 (2017) 774–782.
[38] L. Zhang, Y. Zhong, D. Cha, P. Wang, A self-cleaning underwater superoleophobic mesh for oil-water separation, Sci. Rep., 3 (2013) 2326.
[39] J.H. Kang, T. Kim, J. Choi, J. Park, Y.S. Kim, M.S. Chang, H. Jung, K.T. Park, S.J. Yang, C.R. Park, Hidden second oxidation step of hummers method, Chem. Mater., 28 (2016) 756–764.
[40] J. Yang, E. Zhang, X. Li, Y. Zhang, J. Qu, Z.-Z. Yu, Cellulose/graphene aerogel supported phase change composites with high thermal conductivity and good shape stability for thermal energy storage, Carbon, 98 (2016) 50–57.
[41] A. Barati, E. Kazemi, S. Dadfarnia, A.M. Haji Shabani, Synthesis/characterization of molecular imprinted polymer based on magnetic chitosan/graphene oxide for selective separation/preconcentration of fluoxetine from environmental and biological samples, J. Ind. and Eng. Chem., 46 (2017) 212–221.
[42] A. Fernández-Jiménez, A. Palomo, Composition and micro structure of alkali activated fly ash binder: effect of the activator, Cem. Concr. Res., 35 (2005) 1984–1992.
[43] E.-C. Cho, C.-W. Chang-Jian, H.-C. Chen, K.-S. Chuang, J.-H. Zheng, Y.-S. Hsiao, K.-C. Lee, J.-H. Huang, Robust multi functional superhydrophobic coatings with enhanced water/oil separation, self-cleaning, anti-corrosion, and anti-biological adhesion, Chem. Eng. J., 314 (2017) 347–357.
[44] Y.Q. Liu, Y.L. Zhang, X.Y. Fu, H.B. Sun, Bioinspired underwater superoleophobic membrane based on a graphene oxide coated wire mesh for efficient oil/water separation, ACS Appl. Mater. Interfaces, 7 (2015) 20930–20936.
[45] S. Zhang, F. Lu, L. Tao, N. Liu, C. Gao, L. Feng, Y. Wei, Bio-inspired anti-oil-fouling chitosan-coated mesh for oil/water separation suitable for broad pH range and hyper-saline environments, ACS Appl. Mater. Interfaces, 5 (2013) 11971–11976.

Supporting Information

Movie S1: Experimental process of oil/water separation (water is dyed with methylene blue and oil is dyed with oil red O for clear observation). (MP4)

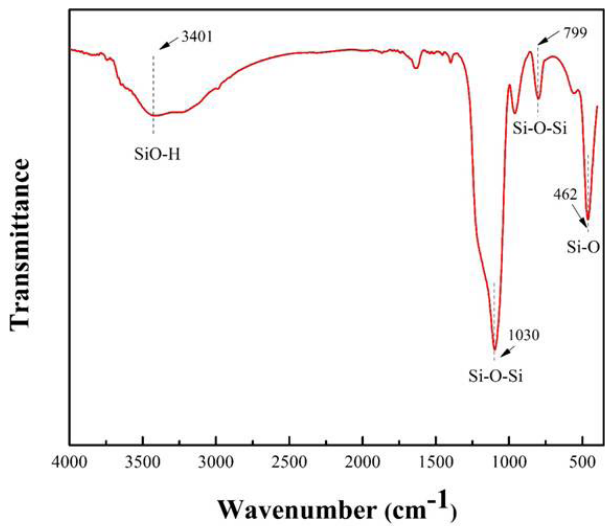

Fig. S1. FTIR spectrum of silica Nanoparticles.

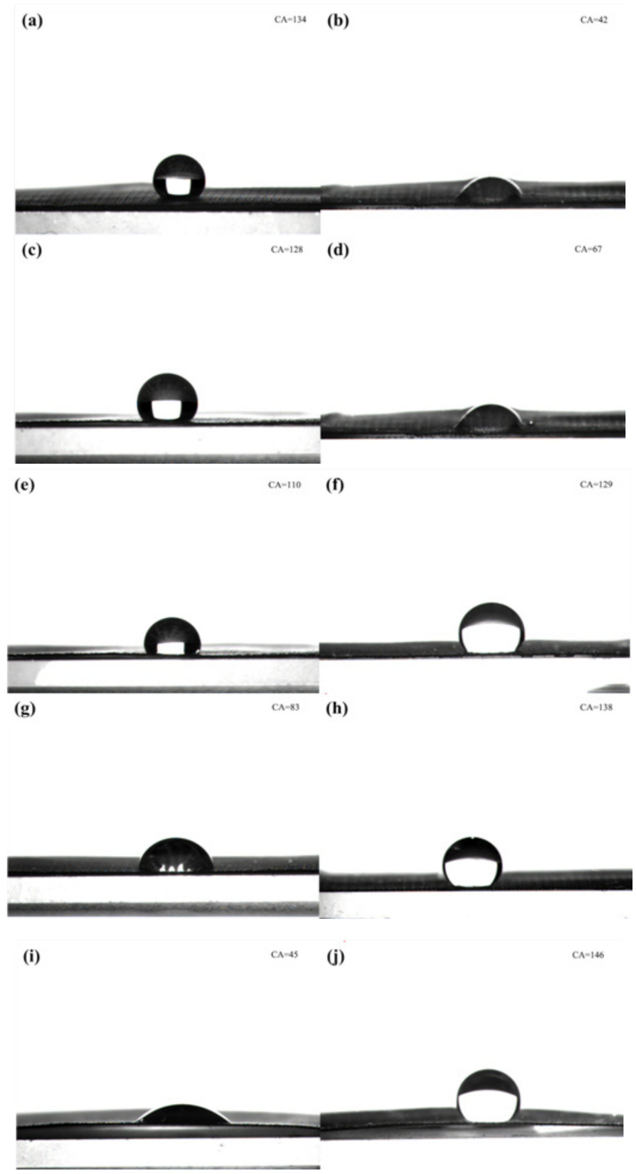

Fig. S2. (a) and (b) are photographs of a water droplet on the original mesh (left) and an oil droplet (dichloromethane) on the original mesh in water (right). (c) and (d) are photographs of a water droplet on the mesh with single microcrystalline cellulose (left) and an oil droplet (dichloromethane) on the mesh with single microcrystalline cellulose in water (right). (e) and (f) are photographs of a water droplet on the mesh with single SiO_2 (left) and anoil droplet (dichloromethane) on the mesh with single SiO_2 in water (right). (g) and (h) are photographs of a water droplet on the mesh with single graphene oxide (left) and an oil droplet (dichloromethane) on the mesh with single graphene oxide in water (right). (i) and (j) are photographs of a water droplet on the mesh with their mixtures (left) and an oil droplet (dichloromethane) on the mesh with their mixtures in water (right).