Journal of Materials Chemistry A

PAPER

View Article Online
View Journal | View Issue

Cite this: *J. Mater. Chem. A*, 2016, **4**, 18832

Multifunctional ultrathin aluminum foil: oil/water separation and particle filtration†

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Received 22nd September 2016
Accepted 2nd November 2016

DOI: 10.1039/c6ta08231a

www.rsc.org/MaterialsA

We present here a kind of novel multifunctional ultrathin aluminum foil which consists of large-area regular micropore arrays covered with nanostructures. These multiscale micro/nanostructures show underwater superoleophobic ability (contact angle > 150°) and oil/water separation function. The novel foils were realized by one-step femtosecond laser irradiation, which is a simple and promising method for preparing special micro/nanostructures due to its high precision, excellent controllability, one-step processing and compatible with various materials. In addition, the micropore arrayed aluminum foil also shows robust filtering performance for particles with different sizes, exhibiting multifunctional applications. This work provides a new way for the construction of aluminum foil-based micropore arrays which can be applied in high-efficiency oil/water separation, particle sorting, and other broader fields.

1. Introduction

The separation of oil/water mixtures has become a global concern due to the serious oil pollution caused by discharging industrial oily wastewater, offshore oil accidents and marine transportation.[1–4] Because oil/water separation is an interfacial issue which is controlled by the chemical composition and geometrical structures, designing and constructing functional interfacial materials possessing different wettability to oil and water is proven to be an effective strategy for wastewater treatment.[4–9] Among these functional materials, porous sponges and foams[1,3,6,10–14] have been extensively used for oil/water separation. Although sponges and foams are characterized by light weight, great porosity, good elasticity, and high-efficiency in oil/water separation, these materials are limited to the design and control of the internal structures which are mainly determined by the parent skeletons.[10] In addition, these porous materials have limited adsorption capacity as absorption saturation constrains their ability to absorb more oil.[10] Moreover, their fabrication usually needs complex chemical processing, which brings in serious pollution to the environment. Besides the adsorbent sponges and foams, many meshes have been constructed as alternative strategies and have been successfully applied in oil/water separation.[15–20] For example, Feng and co-workers[15] first prepared a simultaneous superhydrophobic and superoleophilic Teflon (PTFE) coated mesh film for separation of oil and water. Zhang *et al.*[18] reported a nanowire-hair microstructure covered $Cu(OH)_2$ copper mesh which had a high separation capacity for both water-rich immiscible mixtures and dispersed oil–water mixtures. However, their preparation is also carried out under chemical conditions which may cause secondary pollution to the environment. Gao *et al.*[20] developed a dual-scaled porous nitrocellulose membrane which had high oil/water separation efficiency by a facile perforating method. Although this process is scalable, fast, of low-cost, and has high separation efficiency, the formed pores are nonuniform and uncontrollable, which hinder their extensive use.[15–20] Additionally, most of the literature studies do not consider the solution to the continuous separation for the gravity-driven removal of heavy or light oils (compared to the density of water).[15–20] Furthermore, these meshes are restricted to oil/water separation and are not used for other functions.[15–20] Hence, it is of great significance to seek novel functional materials and prepare regular, controllable structures with multifunctional applications in a simple, environment-friendly, and scalable approach.

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† Electronic supplementary information (ESI) available. See DOI: 10.1039/c6ta08231a

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Femtosecond laser fabrication is a new-style method that is widely employed to prepare special micro/nanostructures due to its high precision, precise controllability, one step processing and compatible with various materials.[21–33] Yong *et al.*[32] prepared rough microstructures on polytetrafluoroethylene (PTFE) sheet by femtosecond laser ablation and further generated a penetrating microhole array on the rough PTFE film by a subsequent mechanical drilling process. The resultant surface was successfully applied in the field of oil/water separation due to the inverse superhydrophobicity and superoleophilicity. Here, for the first time, we report on the one-step femtosecond laser irradiation method to induce large-area regular micropore arrays on ultrathin aluminum foils, which have thin thickness (~25 μm), light weight, low cost, good plasticity, and have been commonly applied to packaging, cooking, geochemical sampling, art and decoration and so on. The micropore-arrayed aluminum foils show underwater superoleophobic ability which is responsible for oil/water separation function. By skillfully designing the shape and structure of the aluminum foil oil/water separator, the oil and water can be separated efficiently and continuously. In addition, the uniform micropores were used to filter the particles with different diameters, showing multifunctional applications. This work provides a new pathway for controllable construction of aluminum foil-based micropore arrays with high separation efficiency and extends their applications to broader fields besides water treatment.

2. Experimental

Materials

The aluminum foil was purchased from New Metal Material Tech. Co., Ltd, Beijing, China and the thickness is 25 μm. There are two kinds of oil used in our experiment. The heavy oil is 1,2-dichloroethane ($C_2H_4Cl_2$), and the density is 1.26 g cm^{-3}. The light oil is normal octane (C_8H_{18}), and the density is 0.70 g cm^{-3}.

Fabrication of micropore array aluminum foil

A regenerative amplified Ti:sapphire femtosecond laser system (Legend Elite-1K-HE, Coherent, USA) that generates 104 fs pulses at a repetition rate of 1 kHz with a central wavelength of 800 nm is employed to fabricate aluminum foil with uniform micropore arrays. The focus spot on the aluminum foil surface is about 20 μm. The spacing between two adjacent pores is set to be 20, 40, 60, and 80 μm. The diameters are adjusted from 2.4 to 32 μm by skillfully changing the laser pulse energy and pulse number.

Instrument and characterization

Scanning electron microscopy (SEM) images were obtained using a field-emission scanning electron microscope (JSM-6700F, JEOL, Tokyo, Japan). The contact angles of 5 μL water or oil droplet are measured by using a contact-angle system (CA100D, Innuo, China). The average values are obtained by measuring five drops at different locations on the same surface at ambient temperature.

Oil/water separation experiment

For separating light oil and water, the as-prepared aluminum foil with a pore diameter of 8.2 μm, with a spacing of 40 μm, was horizontally fixed between two glass vessels with a diameter of 25 mm. The oil/water mixtures were prepared by mixing 10 mL oil with 10 mL water. The oil/water mixtures were poured into the filter and the separation was achieved driven by gravity. For separating heavy oil and water, the aluminum foil was pasted at the slot with a width of 5 mm on the upright glass tube with a diameter of 25 mm. The as-prepared aluminum foil is hydrophilic and underwater oleophobic. It can be changed to hydrophobic and underwater oleophilic after modification with a 1.0% ethanol solution of 1*H*,1*H*,2*H*,2*H*-perfluorodecyltriethoxysilane (PFDTES) at room temperature for 12 hours followed by drying at 60° for 20 minutes. For continuously separating oil and water, the hydrophilic and hydrophobic aluminum foils were pasted at the slots on both sides of the glass tube. Before the separation process, a small amount of water was poured onto the aluminum foils to pre-wet the micropores. For clearly observing the separation of the oil/water mixtures, the water was dyed blue color with methylene blue, while the oil was dyed with Sudan IV and showed a red color. The oil content in the separated water was determined by using an IR oil content analyzer (CY-2000, China).

Particle filtration

SiO_2 particles (Huge Biotech Corp, Shanghai) with diameters of 20 and 10 μm, and of 10 and 5 μm were mixed in aqueous solution. Then the mixtures were poured onto the aluminum foil surfaces. The small diameter particles passed through the micropores with the aqueous solution, while the big diameter particles were blocked above the aluminum foil.

3. Results and discussion

Ultrathin aluminum foil with regular micropore arrays were fabricated by the femtosecond laser perforating method, and the schematic fabrication process is shown in Fig. 1a. By setting the laser pulse energy of 50 μJ and perforating space of 20 μm, an area of 40 × 40 mm^2 was obtained (Fig. 1b). The experimental results indicated that the thin aluminum foil can be punched with ~4–5 pulses, and the above-mentioned large area aluminum foil can be processed in ~5.6 hours. The fabricated aluminum foil presents brilliant iridescence under the white light illumination due to the diffraction of light [ESI, Fig. S1†]. From the low-magnification SEM image and the transmission microscope photographs, it is clearly seen that uniform micropore arrays are neatly arranged on the aluminum foil surface. The magnified SEM image exhibited that the pore size is about 2.4 μm, and the rim is covered with rough nanostructures. The uniformity of the micropores is studied by systematically analyzing the pore diameters and the eccentricity, and the results are shown in Fig. S2 in the ESI.†

The unprocessed aluminum foil is hydrophilic and the water contact angle (θ_{WA}) is 53.9°. The microsized pores in combination with nanoscale roughness provide a hierarchical composite structure to the aluminum foil that possess superwetting

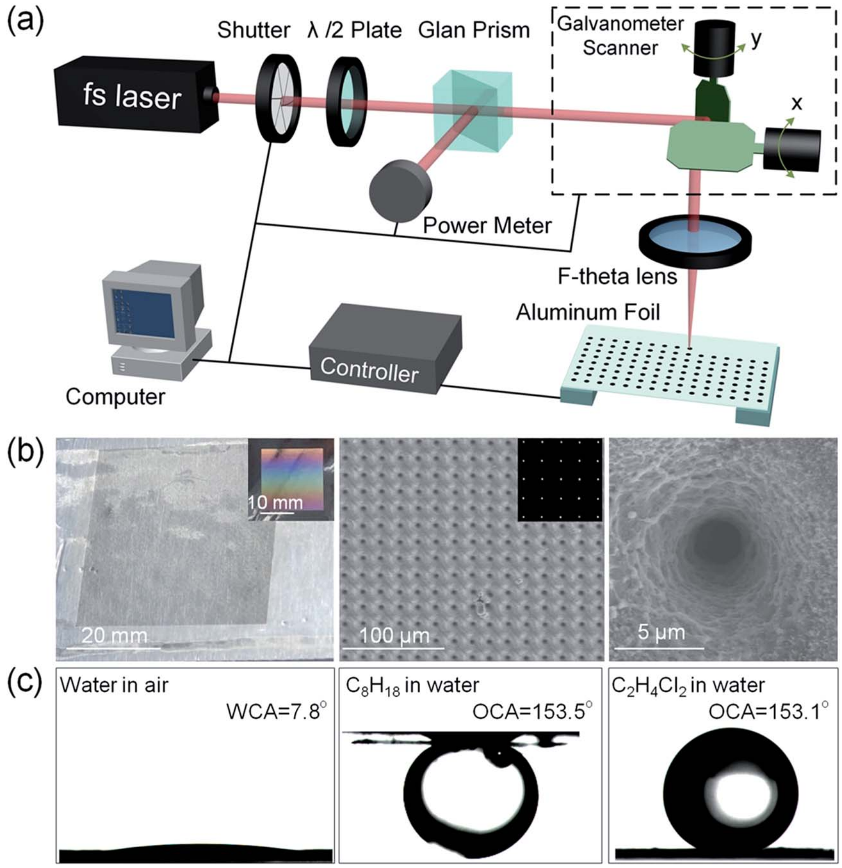

Fig. 1 Fabrication of regular micropore arrays on ultrathin aluminum foil by one-step femtosecond laser perforating. (a) The schematic diagram of fabricating regular micropore-arrayed aluminum foil. (b) Optical and SEM images of the as-prepared aluminum foil: left side of (b) is the optical camera photograph taken under the white light irradiation and the inset image is taken under the inclined incidence of white light showing iridescence. Middle and right of (b) are the SEM images at different magnifications, and the inset is the transmission optical microscope photograph. (c) The contact-angle images of the aluminum foil, demonstrating superhydrophilicity (water contact angle is about 7.8°) and underwater superoleophobicity (contact angle of C_8H_{18} and $C_2H_4Cl_2$ are 153.5°, and 153.1°, respectively).

behaviors,[24,34,35] which is evaluated by contact angle measurements and obtains a contact angle (θ'_{WA}) of 7.8° (Fig. 1c), and can be described by using the Wenzel model as shown in eqn (1)[24,36]

$$\cos \theta'_{WA} = r \cos \theta_{WA} \quad (1)$$

in our case, r is the roughness factor, and the value is larger than 1 [ESI, Fig. S3†]. When the as-prepared aluminum foil is immersed in water, its wetting properties are reverted to be superoleophobic. Typically, for C_8H_{18} whose density is less than water, and for $C_2H_4Cl_2$ which is heavier than water, the contact angles (θ'_{OW}) are 153.5° and 153.1°, respectively. This physical phenomenon can be explained by eqn (2)[18,24]

$$\cos \theta'_{OW} = f \cos \theta_{OW} + f - 1 \quad (2)$$

where f is the area fraction of the solid, which is defined as the ratio of the actual contact area by the oil droplet to the whole area of the rough structure. θ_{OW} is the contact angle of oil on the rough surface in water, and can be expressed as eqn (3)[24,37]

$$\cos \theta_{OW} = \frac{\gamma_{OA} \cos \theta_{OA} - \gamma_{WA} \cos \theta_{WA}}{\gamma_{OW}} \quad (3)$$

where θ_{OA} is the contact angle of oil on the flat surface in air, γ_{OW}, γ_{OA}, and γ_{WA} are the surface tensions of the oil/water, oil/air, and water/air interfaces, respectively.

Fig. 2a shows the representative micropore intervals of 40, 60, and 80 μm, which can be adjusted by altering the perforating space of the laser beam. Additionally, three pores with diameters of 8.2, 19.1, and 32.2 μm are demonstrated in Fig. 2b, and the size distribution is analyzed in Fig. 2c [ESI, Fig. S4†]. The results indicate that the pores are uniformly distributed. In addition, the preparation of a series of micropores with diameters ranging from 2.4 to 32 μm is also studied by changing the laser pulse energy and pulse numbers (Fig. 2d) [ESI, Fig. S5†].

From Fig. 1 and 2, it is clearly indicated that regular micropore arrays with diverse diameters and intervals can be easily prepared on aluminum foil surfaces just by simply adjusting the laser processing parameters without other complex processes. In addition, this process is fast. For example, a 10 × 10 mm^2 membrane with a micropore size of ~15 μm and an interval of 60 μm can be processed in less than 6 minutes.

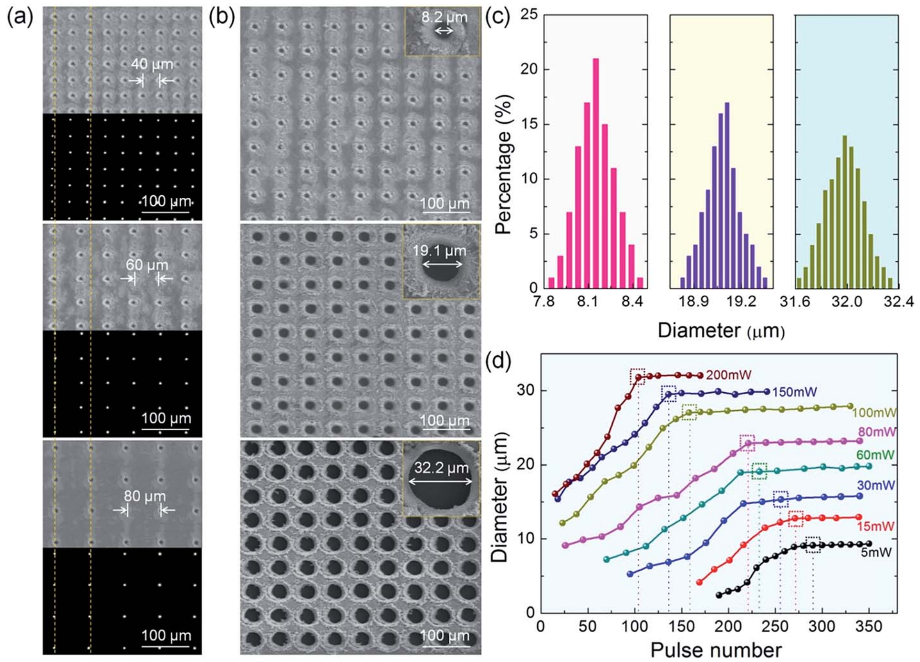

Fig. 2 The precise control of regular micropore arrays with diverse intervals and diameters. (a) The SEM and transmission optical microscope images of the micropores with intervals of 40, 60, and 80 μm, from top to bottom, respectively. (b) The representative SEM images of the micropores with diameters of 8.2, 19.1, and 32.2 μm, respectively. (c) Distribution statistics of the three pore diameters in (b), showing good uniformity. (d) The pore diameters as a function of laser pulse energy and pulse number.

This novel method shows many advantages including high precision, excellent controllability, and environmental friendliness, and thus has wide prospect in many fields.

As mentioned above, the laser-induced micropore arrays intensively change the wettability of the aluminum foil surfaces. The hydrophilicity and underwater oleophobicity of the aluminum foil prepared under different laser processing parameters are systemically studied and the results are summarized in Fig. S6 in the ESI.† It can be clearly seen that aluminum foil becomes more hydrophilic and more underwater oleophobic with the enlarging pore diameters and shrinking intervals. The opposite wetting properties for water and oil are supposed to be applied for controllable oil/water separation. Several proof-of-concept studies were performed to test the oil/water separation capacities of the as-prepared aluminum foil. Here we choose the C_8H_{18} and $C_2H_4Cl_2$ as the target oils, and the photographs of mixtures are shown in Fig. 3a [ESI, Fig. S7†]. Firstly, the C_8H_{18}/water separation experiment is performed, and the schematic diagram is displayed in Fig. 3b. As shown in Fig. 3c, a piece of aluminum foil with a pore diameter of 8.2 μm and an interval of 60 μm was horizontally fixed between a glass tube and conical flask to act as a separating mesh, which is a common installation method reported in previous literature studies. 20 mL C_8H_{18}/water mixture from a beaker was poured onto the aluminum foil, and then the separation was proceeded by the force of gravity. Due to the superhydrophilicity, the water (blue color) rapidly permeates the micropores and then inpours the conical flask, whereas the C_8H_{18} (red color) [ESI, Fig. S7†] is blocked in the upper tube due to the superoleophobicity of the aluminum foil. The C_8H_{18}/water separation is completed in less than 13 s and no external force other than gravity is needed during the separation process [ESI, Movie S1†]. In this way, the 1.54×10^5 L C_8H_{18}/water mixture can be separated by using an aluminum foil with a processing area of 1 m^2 in one hour. This kind of separator can be used to effectively separate a mixture of light oil (whose density is less than water) and water. However, this device is not suitable for the separation of heavy oil (heavier than water) and water mixtures as the heavy oil settles below the water, forming an insulation layer between the water and the aluminum foil which prevents the water from maintaining contact with the aluminum foil. Although some studies have mentioned the separation of heavy oil and water mixtures using this method, it is speculated that additional external forces, such as stirring and shaking, are needed to churn the mixtures to let the water contact with the aluminum foil.[18,20]

For separating the heavy oil and water mixtures in the absence of other external forces, a tube with a side opening which is covered with micropore arrayed aluminum foil is designed (Fig. 3d). When the mixture of heavy oil and water is poured into the upright tube, the water in the oil/water mixture can pass through the side opening although the oil settles below the water, thereby achieving oil/water separation only with the gravity effect. The results show that a 20 mL heavy oil/water mixture can be separated in less than 31 s, which means the accomplishment of separation of 6.5×10^4 L oil/water mixture by per m^2 per h (Fig. 3e) [ESI, Movie S2†]. Furthermore, this separator can separate not only heavy oil/water mixtures but also light oil/water mixtures. Besides, the aluminum foil can maintain its superhydrophilicity and underwater superoleophobicity after 10 separations and can easily be cleaned

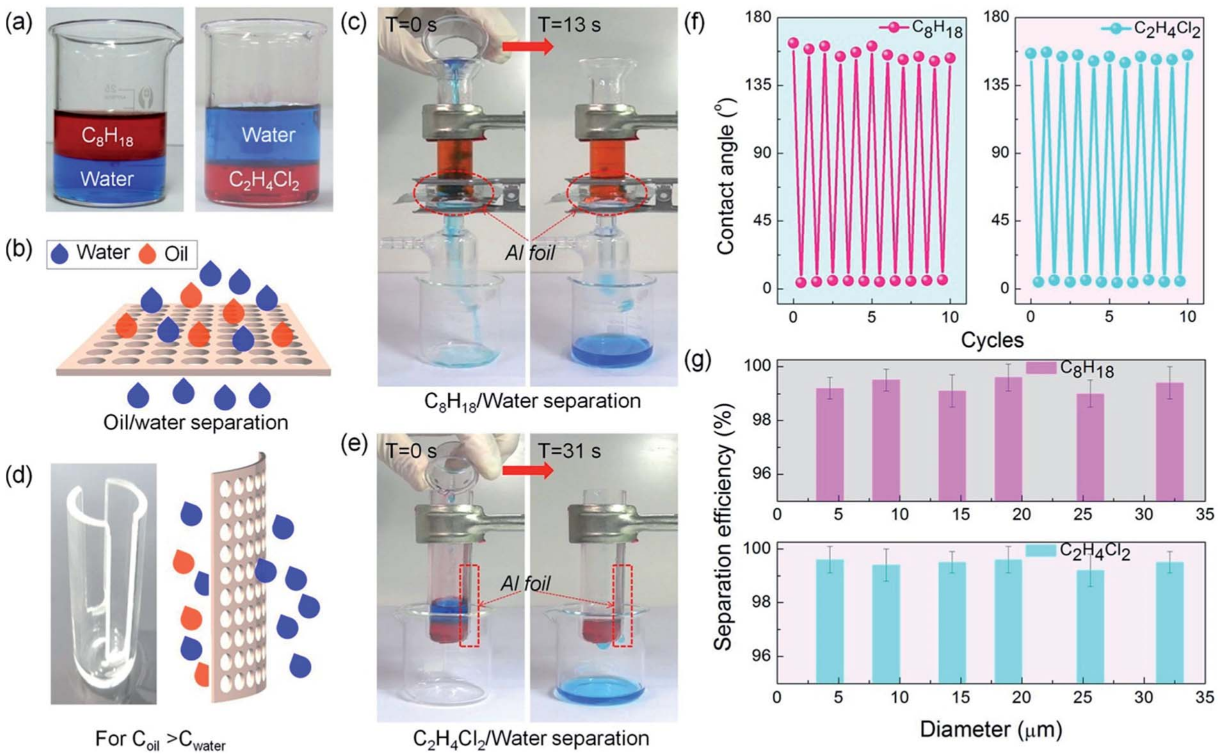

Fig. 3 Oil/water separation of the micropore arrayed aluminum foil for C_8H_{18}/water mixtures and $C_2H_4Cl_2$/water mixtures. (a) The photographs of C_8H_{18}/water mixtures and $C_2H_4Cl_2$/water mixtures. (b) Schematic diagram of C_8H_{18}/water separation. (c) The time-lapse images of the separation of C_8H_{18}/water mixtures. It is indicated that a 20 mL mixture can be separated in less than 13 s. (d) The equipment and schematic diagram of separating heavy oil of $C_2H_4Cl_2$ and water mixtures. (e) Time-lapse images of C_8H_{18}/water separation, and this process is completed in less than 31 s. (f) Cycling performance of micropore-arrayed aluminum foil for repeated use after ultrasonic cleaning. (g) The calculated separation efficiency of the aluminum foil with pore diameters ranging from 2.4 to 32 μm for both C_8H_{18}/water mixtures and $C_2H_4Cl_2$/water mixtures, and the values are all higher than 99%, demonstrating high-efficiency oil/water separation.

using water or ethanol for reuse (Fig. 3f). The separation efficiency of the micropores with pore diameters from 2.4 to 32 μm was quantitatively evaluated by calculating the ratio of the oil before and after separation according to the following equation.[20,38]

$$R(\%) = \left(1 - \frac{C_p}{C_o}\right) \times 100 \quad (4)$$

here, C_o and C_p refer to the oil concentration in the original oil/water mixture and the collected water, respectively. It is indicated that the separation efficiency of this series of micropores was calculated up to 99% for both C_8H_{18} and $C_2H_4Cl_2$ (Fig. 3g). In addition, we found that the aluminum foil can still be used for high-efficiency oil/water separation after being placed for 35 days.

Oil resources are becoming more and more precious as they are rapidly running out. It is vital to develop a novel and simple technique that can achieve oil/water separation and oil collection simultaneously and continuously. To achieve this, only one more step is involved: installing a superhydrophilic aluminum foil and superhydrophobic one on the openings on both sides of the tube. This is because the superhydrophobic micropore-arrayed aluminum foil is underwater superoleophilic, which allows the oil to pass through the micropores and block off the water. For this purpose, the aluminum foil is modified with low surface energy 1*H*,1*H*,2*H*,2*H*-perfluorodecyltriethoxysilane (PFDTES), and the schematic diagram is shown in Fig. 4a. After the low surface energy modification, the original superhydrophilic aluminum foil (CA = 5.1°) is tuned to superhydrophobic (CA = 154.4°) and underwater superoleophilic one (CA = 9.5°). Then the superhydrophilic and superhydrophobic aluminum foils were installed on the openings on each side of the tube. When the oil/water mixtures are poured into the upright tube, the water can only penetrate the superhydrophilic aluminum foil micropores while the oil only pass through the superhydrophobic one, as shown in Fig. 4b. The corresponding experimental results are displayed in Fig. 4c. Just as expected, it is seen from Fig. 4c and Movie 3 in the ESI,† a 60 mL oil/water mixture is rapidly separated in less than 28 s and the oil is collected for recycling. After pouring the oil/water mixtures, the oil and water are purified and guided to the collecting vessel. In the meantime, the separator can free up new space for consecutively separating subsequent oil/water mixtures. Such continuous separating behavior indicates that a small separator can separate and purify a large amount of oil/water mixtures easily and quickly. This novel strategy not only overcomes the disadvantages of traditional methods which

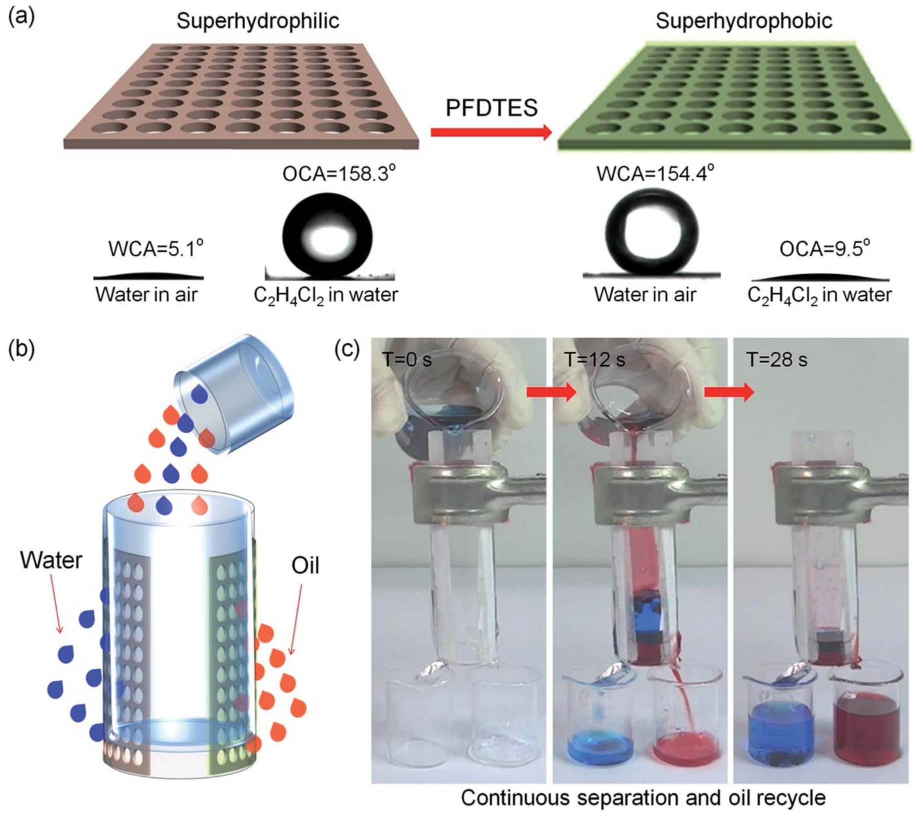

Fig. 4 The demonstration of the continuous separation of oil/water mixtures. (a) Schematic illustrations of fabrication of superhydrophobic aluminum foil by modifying the superhydrophilic one with low surface energy 1*H*,1*H*,2*H*,2*H*-perfluorodecyltriethoxysilane (PFDTES). The modified aluminum foil is superhydrophobic (water contact angle of 154.42°) and underwater superoleophilic ($C_2H_4Cl_2$ contact angle of 9.47°). (b) The home-made devices for continuously separating oil and water mixtures. (c) Time-lapse images of continuous oil/water mixtures separation and the recycle for oil. A 60 mL oil/water mixture is rapidly separated with a foil area of 0.3 × 3 cm^2 in less than 28 s and the oil is collected for recycling. In this way, 23.8 L oil/water mixtures can be separated with 1 m^2 aluminum foil in less than 1 s.

can only separate light or heavy oil and water mixtures in small amounts, but also provides a possibility of recycling water and oil.

In addition, due to the fast and complete discharge of the water and oil from the separator (Fig. 4c), the oil/water separation can withstand the intrusion pressure of oils flowing through the Al foil, which is determined by the oil layer height and can be expressed experimentally and theoretically by eqn (5)[7,20,38–40] and (6)[20,41]

$$P_{\mathrm{exp}} = \rho g h_{\mathrm{max}} \tag{5}$$

$$P_{\mathrm{theor}} = 2\gamma_{\mathrm{OW}} \cos \theta_{\mathrm{OW}}/d \tag{6}$$

in eqn (5), ρ, g, and $h_{\max}$ are the density of the oil, the acceleration of gravity, and the maximum height of the oil layer that the aluminum foil can support. In eqn (6), d refers to the diameter of the micropores [ESI, Fig. S8†]. Hence, this novel separator may open a new channel for the fast, continuous, and high-efficiency oil/water separation.

We also tested the maximum height of the oil that the membrane can sustain. A membrane with a pore size of ∼13.1 μm and an interval of 60 μm was fixed at the lower end of the glass tube. The tested oil is $C_2H_4Cl_2$ and the pressure is exerted by adding water into the upright glass tube. A membrane with a pore size of ∼13.1 μm and an interval of 60 μm was fixed at the lower end of the glass tube. Before the test, the membrane is pre-wetted by spraying water. Due to the underwater superoleophobicity, the oil cannot pass through the membrane. When the height of the oil reaches ∼110.6 cm, the oil can penetrate the superoleophobic membrane and flows into the beaker. This test is also conducted on the membrane with a pore size of ∼24.5 μm, and the obtained maximum height of water is ∼21 cm. As for the membrane with the size of ∼2.4 μm, the maximum water height cannot be measured as the maximum length of the glass tube is 150 cm. Even so, we believe that the maximum water height is far beyond 150 cm.

The regular and uniformly distributed micropore arrays endow the as-prepared aluminum foil with a new function, namely the enhanced particle filtration. In order to illustrate this function, aluminum foils with pore diameters of 13.2 and 7.4 μm, and an interval of 60 μm (Fig. 5a), are applied to filter the particles with different diameters. The filtration of SiO_2 particles with diameters of 20, 10, and 5 μm is illustrated in Fig. 5b and c. When the aqueous solution of particles is dropwise added onto the micropore arrayed aluminum foil, the particles whose sizes are less than the pore diameters can pass

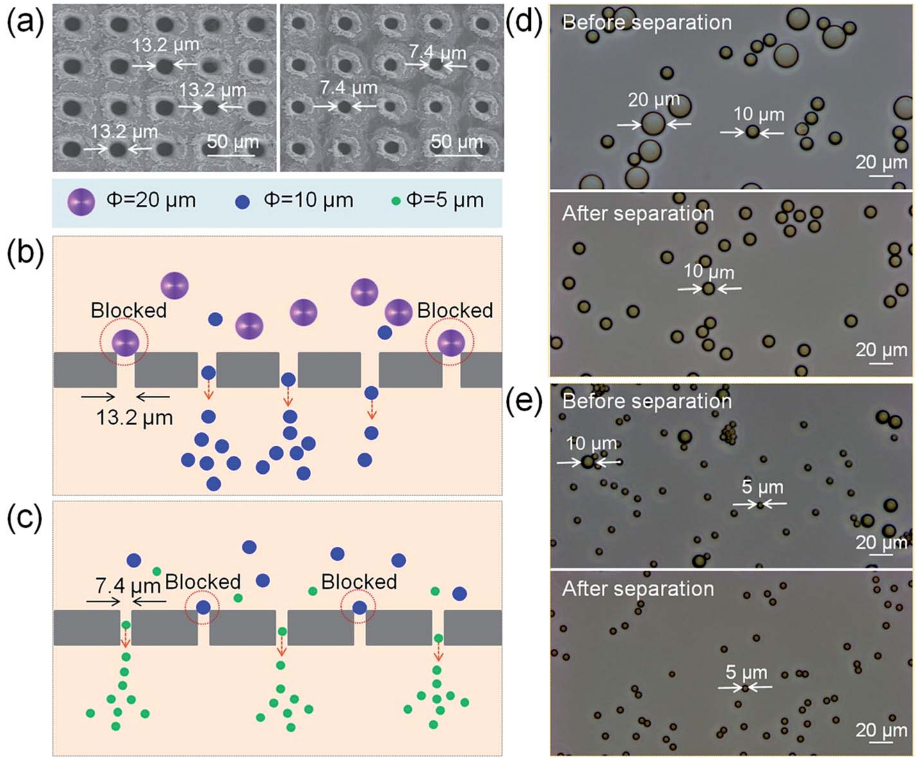

Fig. 5 Multifunctional filtration of different diameter SiO_2 particles with micropore-arrayed aluminum foil. (a) The SEM images of the aluminum foils with pore diameters of 13.2 and 7.4 μm, which are used for filtering particles with sizes of 20, 10, and 5 μm. (b and c) The front-view of the schematic diagram for filtering particles with different sizes. (d and e) The optical microscope images of the particle filtration. By comparing the particles before and after filtration, it is well illustrated that the micropore-arrayed aluminum foil has a high sorting efficiency.

through the micropores under current. However, the particles which are bigger than the micropores will be retained above the aluminum foil. From experimental results exhibited in Fig. 5d and e, it can be seen that the sorted particle sizes are all 10 and 5 μm, respectively, indicating high sorting efficiency. We believe that the simplicity of this approach makes it quite appealing for not only particle filtration with various sizes, but also blood and cell separation, and even more lab-on-a-chip devices.

It is worth mentioning that due to the controllability and flexibility of the femtosecond laser micro/nanofabrication, diverse patterns, such as heart, TaiJi, and star-shaped patterns composed by micropore arrays can be simply and rapidly

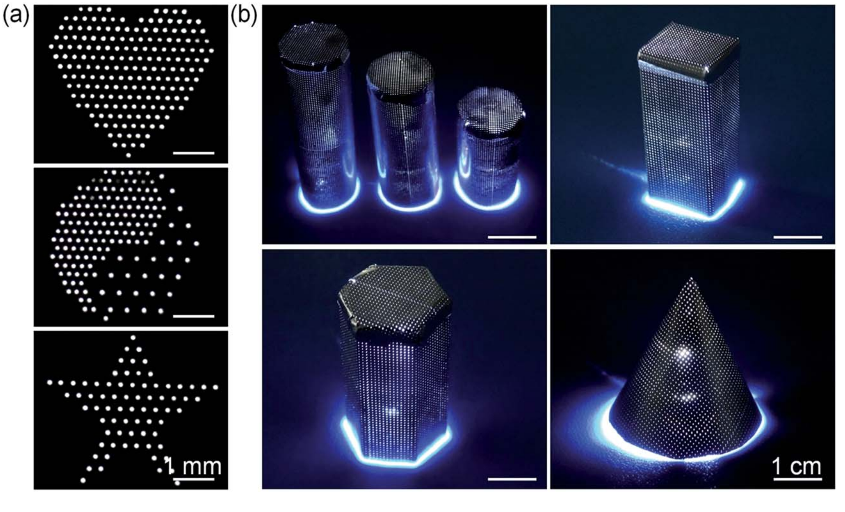

Fig. 6 (a) The microscopic photographs of diverse patterns, such as heart, TaiJi, and star-shaped patterns. (b) The assembly of diverse three-dimensional geometric patterns with the as-prepared aluminum foil due to its good plasticity. All these patterns are irradiated by LED lights from the inside.

fabricated (Fig. 6a and b). Moreover, the good plasticity of the aluminum foil allows it to be folded into diverse three-dimensional geometric patterns. All these advantages are not available to other traditional preparation methods and materials.

4. Conclusion

In summary, we have demonstrated a new oil/water separation device based on micropore arrayed ultrathin aluminum foil fabricated by femtosecond laser perforating. This device can continuously separate not only the light oil and water mixtures, but also the heavy oil/water mixtures with high speed and efficiency, as well as recycling the oil resources for reuse. This novel separation strategy overcomes the shortcomings of conventional methods such as separating only light oil and water mixtures or heavy oil and water mixtures, not being able to continuously separate for retaining oil or water on the separation device, and having a low oil recycling efficiency. The foil displays bright iridescence under the white light irradiation due to the optical diffraction effect of regular micropore arrays. The micropore arrayed aluminum foil also shows robust filtering performance for particles with different sizes, exhibiting multifunctional applications. The combination of the controllable and flexible femtosecond laser micro/nanofabrication technology with the ultrathin, plastic aluminum foil endow the micropore arrays with particular use in the design of sewage treatment and particle filtration equipment, as well as other multifunctional devices.

Acknowledgements

This work is supported by the National Natural Science Foundation of China (No. 51275502, 61475149, 51405464, 51605463, 61675190, and 51675503), Anhui Provincial Natural Science Foundation (No. 1408085ME104), the Fundamental Research Funds for the Central Universities (No. WK2090000006, WK6030000007, and WK2090090018), the China Postdoctoral Science Foundation (No. 2016M590578) and "Chinese Thousand Young Talents Program".

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