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Separation of binary and ternary oil/water mixtures from a highly hydrophobic metal mesh

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ABSTRACT

A highly hydrophobic metal mesh has great potential for its application in oil/water separation due to its special wettability. However, most current oil/water separation devices are simple with limited separation capacity. A separation device based on a highly hydrophobic metal mesh was constructed for different types of oil/water mixtures. Experimental results show that the device not only can be used for the continuous separation of binary oil/water mixtures of any density ratios but also can realize the simultaneous separation of heavy oil/water/light oil ternary mixtures. This achievement is meaningful for practical applications, which will gain great interest in the future.

Key words: continuous separation, highly hydrophobic metal mesh, oil/water mixture

HIGHLIGHTS

- A universal oil/water separation device based on a highly hydrophobic metal mesh was constructed.
- The device can be used for sustainable separation of binary oil/water mixtures and heavy oil/water/light oil ternary mixtures.
- The constructed device allows simultaneous recovery of light and heavy oils in different channels.

1. INTRODUCTION

The frequent oil spills and incessant discharge of industrial oily wastewater have caused great damage to water resources and the ecological environment, which seriously threaten human health (Zhang *et al.* 2017; Li *et al.* 2018). Traditional methods for the treatment of oil/water mixtures, such as gravity sedimentation, centrifugation, and biological and chemical methods (Xue *et al.* 2014; Xu *et al.* 2021) seem workable. However, most of them are high in cost and low in efficiency, and some even cause secondary pollution, impelling researchers to explore more effective countermeasures.

Recently, membrane separation technology based on special wettability has offered a promising and cost-effective route for treating oily wastewater (Liu *et al.* 2017; Wang *et al.* 2022; Shi *et al.* 2021). Through constructing rough surface on the originally amphiphilic (oil-wet and water-wet) metal meshes, 'oil-removing' (super) hydrophobic meshes can be obtained for selective oil/water separation (Li *et al.* 2016; Wang & Wang 2019; Sun *et al.* 2022; Zhang *et al.* 2022). However, little attention has been paid to the separation device. Most researchers simply sandwich the mesh between two containers, which is normally only suitable for the removal of heavy oils ($\rho_{oil} > \rho_{water}$) rather than light oils ($\rho_{oil} < \rho_{water}$) from the binary oil/water mixtures (Xue *et al.* 2011; Gao *et al.* 2016, 2019; Wang & Wang 2019). By tilting the mesh, the removal of light oils seems possible, but this tilting strategy is limited to separating light oil/water mixtures with a small amount of water (Cao *et al.* 2013, 2017). To solve the above problems, we have proposed 2D → 3D conversion of the mesh ('Taylor cone' container) together with the pumping technology to achieve continuous separation of large-volume oil/water mixtures of any density with high efficiency and high purity (Lei *et al.* 2020). However, this device requires good control of the outlet pipe during the separation and it cannot be applied to separate heavy oil/water/light oil ternary mixtures and the disturbing binary oil/water mixtures.

In this work, we construct a universal oil/water separation device based on a highly hydrophobic (water droplet contact angle $\geq 120°$) metal mesh, which not only can be used for the continuous separation of binary oil/water mixtures of any density ratios both in static and disturbing states but also for the simultaneous separation of heavy oil/water/light oil ternary mixtures.

2. EXPERIMENTAL

2.1. Materials

Polyvinylidene fluoride (PVDF, $M_W \approx 625{,}000$) powder was purchased from Sunshui Chemical Co., Ltd. Analytically reagents, including N,N-dimethylformamide (DMF), acetone, hexadecane, and chloroform were obtained from Sinopharm Chemical Reagent Co., Ltd. All chemicals and solvents were used directly without further purification.

2.2. Preparation of a highly hydrophobic metal mesh

The cocktail solution was prepared by pouring 3 g of SiO_2 powder and 2 g of PVDF into mixed solvents of 60 mL of DMF and 40 mL of acetone with continuous stirring at 30 °C for 12 h. Before coating, the commercial brass mesh (200 mesh) was tailored and cleaned by acetone and ethanol. The tailored mesh was then dip-coated in the PVDF–SiO_2 cocktail for 5 min and air-dried for further use.

2.3. Characterization

The surface morphology and elemental distribution of the as-prepared samples were studied using field-emission scanning electron microscopy (FE-SEM, Zeiss, Sigma-HD-01–36, Germany) combined with energy-dispersive X-ray spectroscopy (EDS, 51-XMX1003, Oxford Instruments, UK). The wettability characterization was conducted on a contact angle analyzer (JC2000D3, Shanghai, China) using a droplet (9 μL) of water or hexadecane as an indicator at room temperature. The contact angle data were figured out based on the ellipse fitting method.

The separation efficiency (η) of oil/water mixtures was calculated using the following equation (Lei *et al.* 2020):

$$\eta = \frac{m_P}{m_0} \times 100\%$$

where m_0 and m_P are the mass of the oil before and after separation, respectively.

3. RESULTS AND DISCUSSION

Figure 1 shows schematic diagram (a), CAD model (b), and 3D printed model with the coated mesh (c) of the universal oil/water separation device. As illustrated in Figure 1(a), the device has three zones for the discharge of heavy oil ($\rho_{oil} > \rho_{water}$),

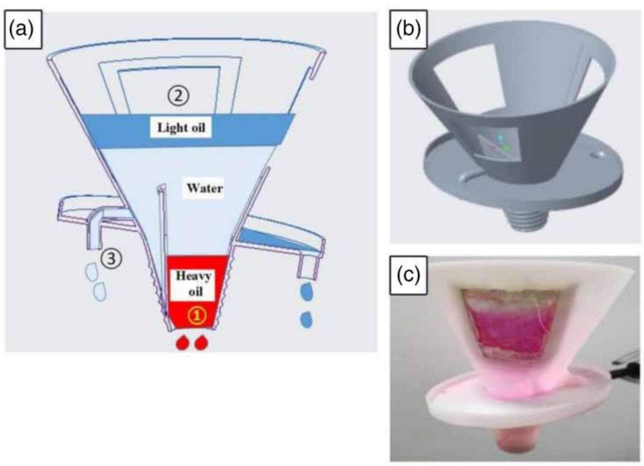

Figure 1 | (a) Schematic diagram of the universal oil/water separation device, (b) CAD model, and (c) 3D printed model (with the mesh) of the corresponding separation device in (a).

light oil ($\rho_{oil} < \rho_{water}$), and water, respectively. Specifically, the highly hydrophobic meshes are fixed tightly at the bottom (zone 1) and the top side (zone 2) of the device for filtering heavy and light oils, respectively. To automatically discharge the cumulative water in the device, a passageway is created on the side of the middle of the device (zone 3), and a baffle plate is set to avoid permeating light and heavy oils.

To demonstrate the feasibility of the proposed device constructed in Figure 1, we have prepared a highly hydrophobic and super oleophilic brass mesh by the dip-coating method, as evidenced by water contact angle (WCA) of 144° (close to super-hydrophobic) and oil contact angle (OCA) of 0°, as shown in Figure 2(a). Figure 2(b) displays the morphology of the dip-coated PVDF–SiO_2 mesh, where the coating not only adheres well to the surface of the brass wire but also fills the gap in between. The EDS from the coating surface shows that four main elements, including Si, C, O, and F are contained (Figure 2(c)). It should be noted that before the dip-coating process, the meshes were tailored according to the different requirements of heavy and light oil filtration zones of the device. As such, the coated meshes can be well installed on the device without damage to the coating.

For the continuous separation of light oil/water mixtures and the convenience of calculating the separation efficiency, a peristaltic pump, a digital electronic balance, and pipes were assembled with the constructed device. Figure 3(a) shows the separation process of static light oil/water mixtures from the constructed system, whereas for the separation of dynamic light oil/water mixtures, a magnetic stirrer was also included for continuous stirring (Figure 3(b)). Figure 3(c) displays the recovery of light oil from the same oil/water mixtures with hexadecane of 50 g and water of 200 g at a pumping rate of 300 mL/min. It demonstrates that the separation of the mixture with/without stirring can be achieved by our proposed system; however, it is noted that the mixture under static conditions shows better separation performance in both the recovery of oil content and the rate of the oil. This may be due to a certain degree of emulsification of the oil/water mixture under agitation (Figure 3(b)), which reduces the filtration rate of the oil/water mixture.

Figure 3(d) shows the effect of the light oil amount on the separation efficiency under static conditions. It is found that increasing the oil amount in the mixture can bring a higher efficiency and our experiments demonstrate that η of 98.5% or higher can be realized when the oil amount exceeds 80 g. This is due to the fact that the coated mesh and pipes in the separation device adsorb (or adhere) a small portion of the oil, and the larger the starting amount of light oil, the smaller

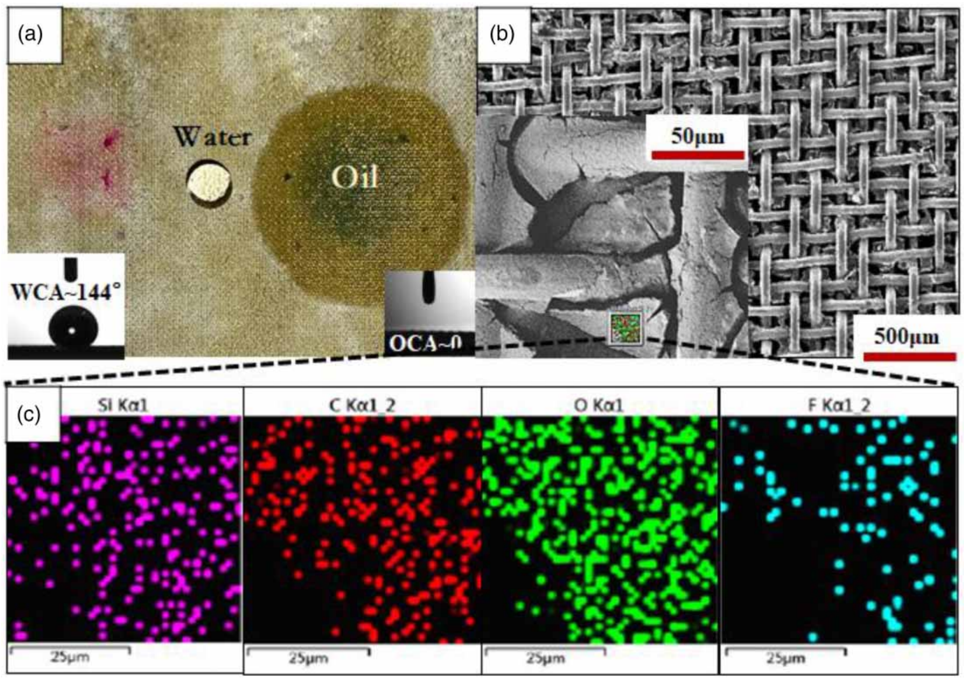

Figure 2 | (a) Wetting behavior of a PVDF/SiO_2-coated brass mesh via placing water and oil droplets on the mesh and measuring water and oil contact angles (inset), (b) SEM images of PVDF/SiO_2 coated brass mesh, and (c) EDS images of the coating (adapted from our previous work Lei *et al.* 2020).

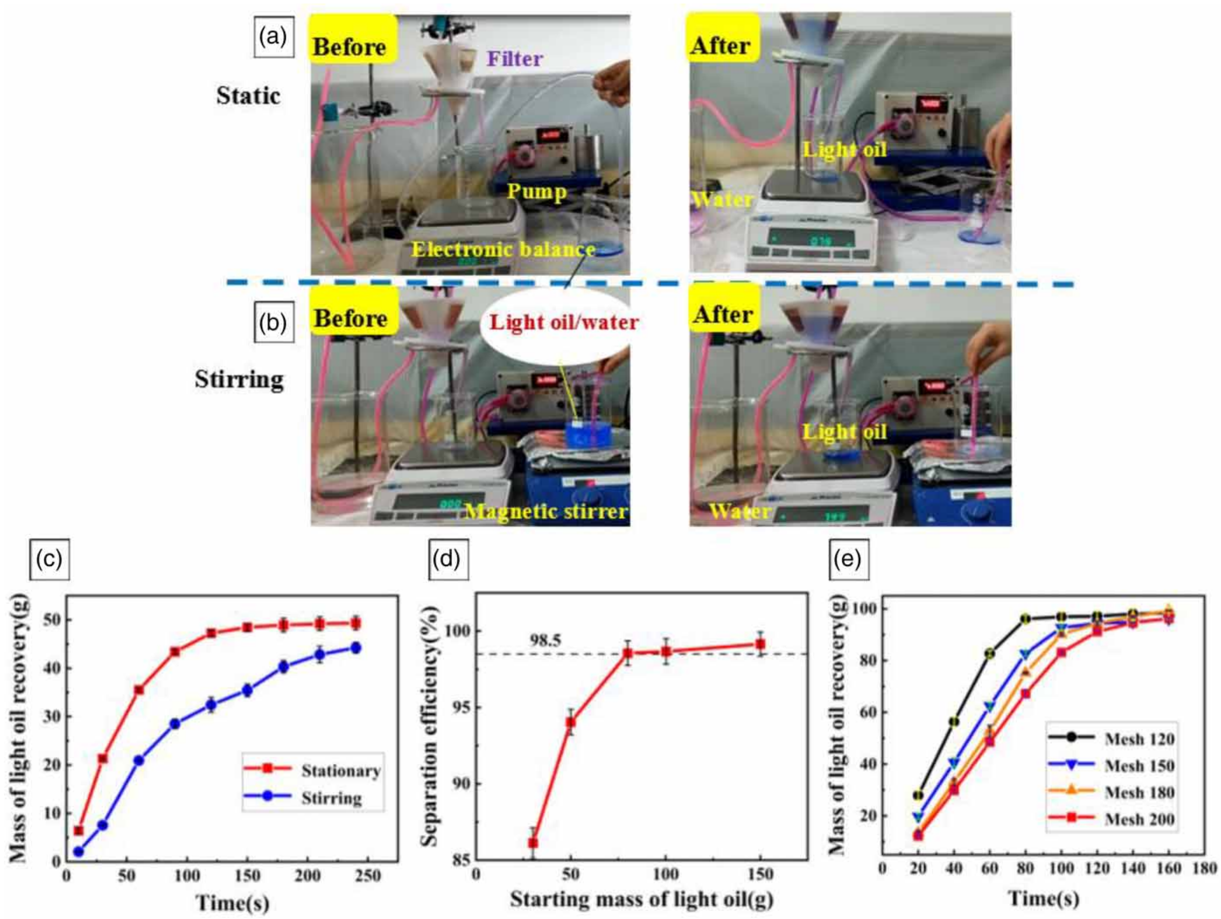

Figure 3 | Snapshots of the separation of light oil/water mixtures under (a) static and (b) stirring conditions, (c) light oil recovery under different conditions (hexadecane: starting amount of 50 g), (d) separation efficiency of light oil/water mixtures with different amounts of hexadecane and 200 g water using the as-prepared PVDF-SiO_2 mesh, and (e) effect of brass mesh number on light oil recovery under static conditions (the oil starting amount of 100 g).

the effect (Lei *et al.* 2020). Figure 3(e) shows the effect of the mesh number on the light oil recovery from the oil/water mixture under static conditions. It can be seen that when the starting amount of the oil is the same as 100 g, the oil recovery rate from 120 mesh is significantly faster than that from other mesh numbers, while for 180 and 200 mesh ones, the oil recovery rates of both are very similar. This is due to the fact that the large number of mesh facilitates the adhesion filling of the coating material, which in turn decreases the passage rate of the wettable liquid (Wang *et al.* 2015; Lei *et al.* 2020).

The snapshots of the separation process of heavy oil/water mixtures under static and stirring conditions are shown in Figure 4(a) and 4(b), respectively. Similar to the separation of light oil/water mixtures, the same pumping rate (300 mL/min) was used and heavy oil/water mixtures with chloroform of 50 g and water of 200 g were experimented. As shown in Figure 4(c), a much better separation performance is achieved for the mixture under static conditions as compared with the mixture under stirring, which may be due to easier emulsification of the heavy oils in the mixture with continuous stirring (Figure 4(b)). In terms of oil recovery from the static mixture with the same amount (50 g) of light and heavy oils, the recovery of heavy oils is much faster than that of light oils but the final recovery amount is less, as shown in Figures 3(c) and 4(c). The above difference comes from the oil property and the separation device. Since the heavy oil (chloroform) used in the experiment is more volatile with a higher density (1.48 g/mL) than the light oil (hexadecane, 0.77 g/mL), the loss of heavy oils during separation should be greater. Thus, the final recovery amount of heavy oils is less. As illustrated in Figure 1, the separation mesh for heavy oils (zone 1) is at the bottom (vertical to the gravity), such that gravity can be fully used to drive the passage of the heavy oils; while for light oils the separation mesh (zone 2) only a component of gravity is used to assist the passage of light oils (Li *et al.* 2016; Lei *et al.* 2020; Xu *et al.* 2021).

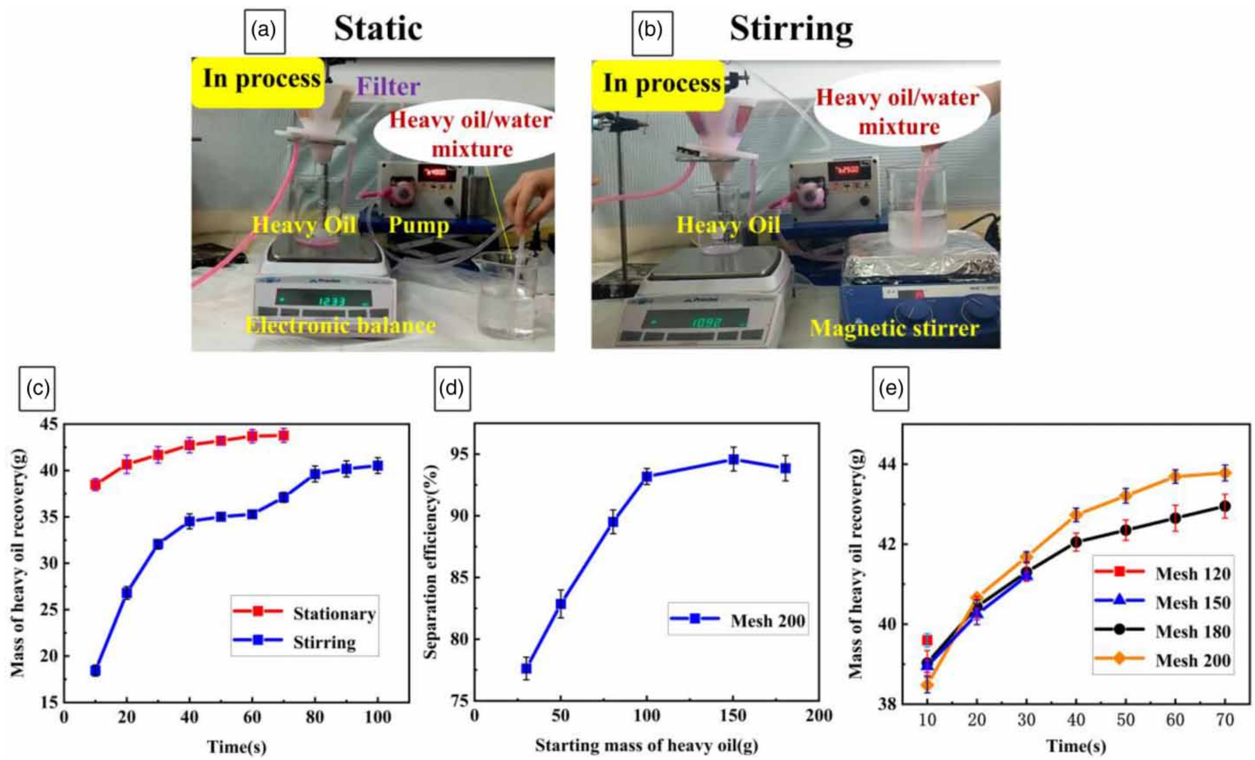

Figure 4 | Snapshots of the separation process of heavy oil/water mixtures under (a) static and (b) stirring conditions, (c) heavy oil recovery under different conditions (chloroform: starting amount of 50 g), (d) separation efficiency of heavy oil/water mixture with different amounts of chloroform and 200 g water using the as-prepared PVDF–SiO_2 mesh, and and (e) effect of brass mesh number on heavy oil recovery under static conditions (the oil starting amount of 50 g).

Figure 4(d) shows that the separation efficiency of heavy oil from the static oil/water mixture increases with increasing the starting oil amount in the mixture, the reason of which has been mentioned above. It is noted that the separation efficiency for the heavy oil/water mixture is much lower as compared with the light oil/water mixture, especially noticeable when the starting oil amount is less than 100 g. This difference should arise from the volatility and density of the oils. Figure 4(e) plots the influence of the mesh number on the recovery of heavy oil from the chloroform/water mixture under static conditions. Unlike the separation of light oil/water mixture, the mesh seems more vulnerable to heavy oils as evidenced by the damage of meshes with small numbers of 120 and 150 during the separation (Figure 4(e)). The failure reveals that the coating (Figure 2(b)) on the mesh with a small number (120, 150) is more vulnerable to the corrosion of the heavy oils (chloroform) (Ping & Lu 2019). Therefore, in the following separation experiment of heavy oil/water/light oil ternary mixtures, the mesh with a big number of 200 was used.

To demonstrate the feasibility of using the constructed device to separate heavy oil/water/light oil ternary mixtures, 50 g chloroform, 50 g hexadecane, and 250 g water were carefully mixed to form the ternary mixtures (Figure 5(b)). The heavy and light oils separated from the mixture were recorded by two electronic balances, as shown in Figure 5(a). Figure 5(c) shows the separation efficiency of the device to continuously separate heavy oil/water/light oil ternary mixtures: the separation efficiency for the light oils maintains nearly 95%, while for the heavy oils, the efficiency is low at about 80%. In our repeated experiments, it was normally found that after seven cycles of separation, the amount of collected 'heavy oil' increased visibly for the permeation of water that probably results from the partial damage of the as-prepared mesh due to erosion of heavy oil. Thus, the separation efficiency after seven cycles is not presented. It should be noted that our constructed device based on a single highly hydrophobic mesh is superior to previous reports (Dunderdale *et al.* 2015; Zhou *et al.* 2018; Das *et al.* 2019), where the combination of two antagonistic wetting membranes is required to achieve the separation of heavy oil/water/light oil ternary mixtures, and the separated oils (heavy and light) are normally mixed together due to the discharge from the same channel.

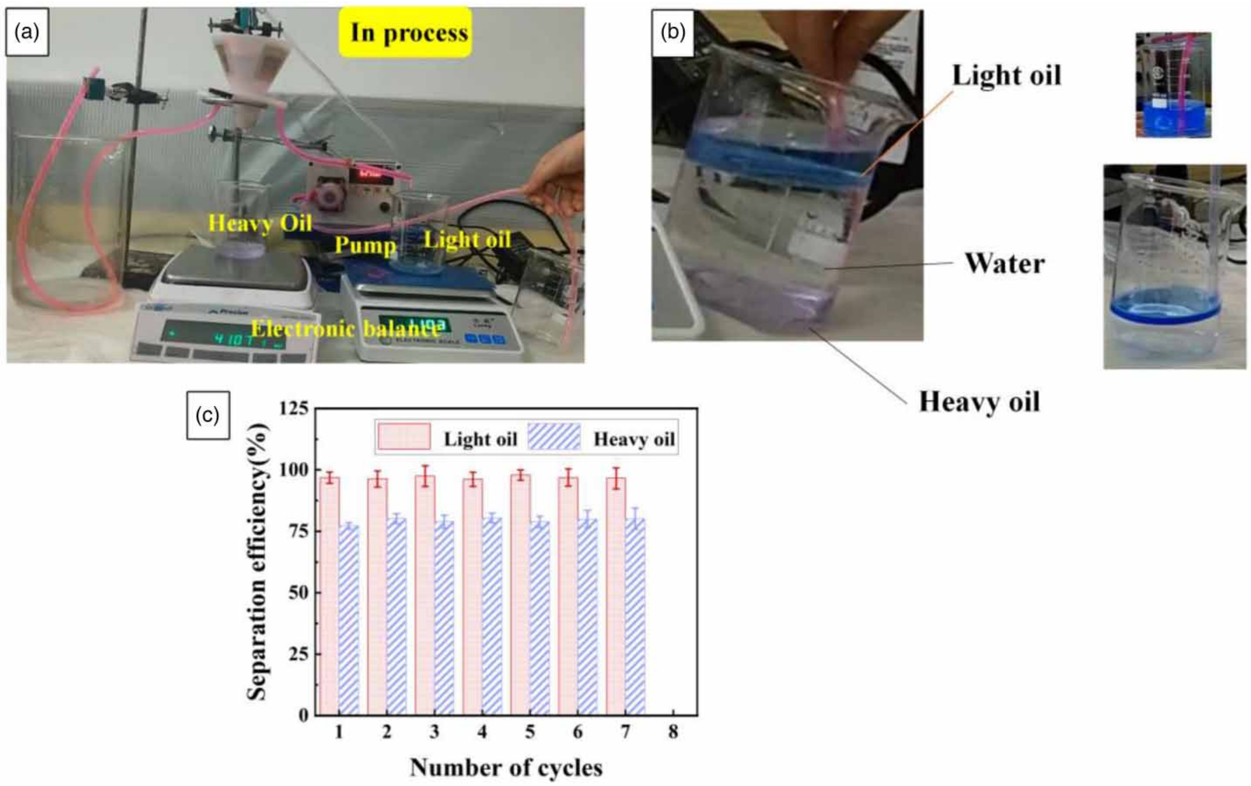

Figure 5 | (a) Snapshot of the separation of heavy oil/water/light oil ternary mixtures, (b) stratification of heavy oil/water/light oil ternary mixtures, and (c) comparison of different oil recovery within eight cycles (chloroform 50 g, hexadecane 50 g, and water 250 g).

4. CONCLUSIONS

In summary, a universal oil/water separation device based on the highly hydrophobic metal mesh has been constructed for sustainable separation of binary and ternary oil/water mixtures. Results from the separation of binary oil/water mixtures show that the separation efficiency under static conditions is better than that under stirring for both light and heavy oil/water mixtures. As compared with heavy oil/water mixtures, light oil/water mixtures normally show higher separation efficiency although with slow recovery rate. In terms of the separation of heavy oil/water/light oil ternary mixtures, the constructed device also allows the simultaneous recovery of light and heavy oils in different channels.

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DATA AVAILABILITY STATEMENT

Data cannot be made publicly available; readers should contact the corresponding author for details.

CONFLICT OF INTEREST

The authors declare there is no conflict.

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