

## Research Article

# Synthesis of a Superhydrophobic Polyvinyl Alcohol Sponge Using Water as the Only Solvent for Continuous Oil-Water Separation

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Few cases of hydrophobic materials synthesized in water have been reported. In this work, water, as the only solvent, is used to prepare a superhydrophobic sponge via a facile and environment-friendly route. The as-prepared sponge, namely silylated polyvinyl alcohol (PVA) sponge, exhibits superhydrophobic and superoleophilic characters. It has the static water contact angle (WCA) of  $152 \pm 1$  and the static oil contact angle (OCA) of  $0^\circ$ , which can lead to excellent selectivity for oil-water separation. Besides, the methyltriethoxysilane (MTES) can form a stable mixed structure with the PVA skeleton, resulting in the rare shedding of polymethylsiloxane nanoparticles and the long-term stability for oil-water separation. Furthermore, the silylated sponge shows a high separation efficiency ( $>99.6\%$ ), removing oil up to 6200~14000 times of its own mass. The findings demonstrated that the silylated superhydrophobic sponge can be a promising candidate in water treatment application.

## 1. Introduction

Along with increasing offshore oil extraction and transportation, oil contamination problems are becoming serious, posing a threat to the living environment and human health [1, 2]. Therefore, cleaning oil from water is an urgent and challenging task [3]. A wide variety of strategies [4] have been developed to eliminate oil pollution, such as burning [5], mechanical extraction [6], coagulation [7], and absorption [8]. Among these technologies, the application of absorbents is considered to be the most attractive for oil removal because of the high efficiency and ease of operation [9]. Till date, a variety of absorbing materials, including commercial resins [10], fibers [11], and mineral products [12], have been developed and widely applied in oil-water separation field. However, these traditional absorbents exhibit poor selectivity, low absorptivity, or difficulty in scale-up. Moreover, many of them are undesirable due to the poor abilities in regeneration and recycling. Recently, a large variety of novel, efficient absorbents are becoming a hotspot in oil-water separation, such as carbon nanotubes (CNTs) [12–14], graphene-based materials [15–17], nanocellulose

[18], polyurethane (PU) [19–21], and PVA-based absorbents [22].

Among them, commercial PU and PVA sponges are viewed as the most potential materials in oil-water separation, because of their low cost, high porosity, and high absorption ability, as well as their easy mass production. At present, the PU- and PVA-based materials with surface hydrophobicity modification have made remarkable achievements in oil-water separation. For example, Pan et al. [23] reported a magnetic polymer-based graphene foam with high oil wettability for oil-water separation; nevertheless, the complicated synthesis route limited its application. Pan et al. [23] and coworkers prepared hydrophobic polyvinyl formaldehyde sponges by one-step surface modification, but massive pyridine, a strong virose organic reagent, is involved as the solvent. Yuan [24] and coworkers designed a hollow  $\text{SiO}_2@ \text{MnO}_2$  modified PU foam for high-efficiency oil-water separation; however,  $\text{MnO}_2$  may introduce heavy metals ( $\text{Mn}^{4+}$ ) and lead to secondary pollution. Given limits of the cases discussed above, we raised a facile and green one-pot synthesis of silylated PVA sponge for oil-water separation, possessing advantages (e.g., high porosity, stable structure,

and low cost). The raw materials, i.e., PVA and MTES, are biocompatible and environment-friendly. The solvent, other than commonly used organic solvent, is only deionized water. The as-made modified PVA sponge exhibited superhydrophobic and superoleophilic properties [25]. Furthermore, the modified PVA sponge could remove oil from water under the pressure of a pump, featuring in high separation capacity, excellent efficiency, and reusability. To sum up, this work provides a facile and green one-pot strategy to prepare modified PVA sponge, which can be applied for practical oil spill treatment and environment protection.

## 2. Materials and Methods

**2.1. Materials.** The reagents, including methyltriethoxysilane (MTES,  $\geq 98\%$ ), hydrochloric acid (HCl, AR), and ammonia ( $\text{NH}_4\text{OH}$ ,  $\geq 28\%$ ), were bought from Sinopharm Chemical Reagent Co., Ltd (Shanghai, China). Sudan III and Methyl blue were purchased from Sigma chemistry Co. Ltd (Shanghai, China). Organic solvents, including dichloromethane (AR), chloroform (AR), *n*-hexane (AR), toluene (AR), and chlorobenzene (AR), were obtained from Beijing Chemical Reagents Co. (Beijing, China). Gasoline was purchased from Sinopec Corp. Commercial PVA sponge was supplied by Alibaba Enterprise. The deionized water was used to make up all mixed solutions and throughout the experiments.

**2.2. Preparation of Silylated PVA Sponge.** A facile and green technological approach was carried out to produce the silylated PVA sponge. Firstly, methyltriethoxysilane (3 ml), with a certain amount of deionized water, was added into a beaker successively, and 0.2 ml of hydrochloric acid catalyst (w/w 1% HCl) was added into the flask in a moment, and then stirred for 0.5 h. Observing a uniformly continuous phase of the mixture, a certain amount of dilute ammonia (0.5 mol/L) was added rapidly and was then kept stirring for a minute at room temperature. Secondly, soak the pre-prepared dried PVA sponge in the mixture for one minute, and then ageing was done at  $50^\circ\text{C}$  for 3 hours. Subsequently, the wet sponge was washed with deionized water for several times. Finally, the wet sponge was freeze-dried for 24 h at room temperature to get the silylated PVA sponge. The preparation process and mechanism of the silylated sponge were shown schematically in Figure 1.

**2.3. Characterization.** The morphologies of the pure and silylated PVA sponge were obtained by a scanning electron microscope (SEM S-4800 HITACHI). The Fourier transform infrared (FTIR) spectra of the two samples were observed by a VERTEX 70 FTIR spectrometer (Bruker, Germany). All spectra were recorded between  $4000$  and  $500\text{ cm}^{-1}$  with a  $4\text{ cm}^{-1}$  and 16 scans per sample. The crystal structures of them were measured by MSAL-XD2 X-ray diffractometer in the range of  $5$ – $40^\circ$  ( $2\theta$ ). Static contact angle measurements of the two samples were performed by the contact angle measuring system (JC2000C1, POWEREACH).

**2.4. Oil-Water Separation Experiments.** First of all, a silylated PVA sponge was prepared with the size of  $3 \times 3 \times 3\text{ cm}^3$ . After that, a simple vacuum suction device is established, as shown in Figure 2. The silylated PVA sponge is connected to a pipe and the other hand of the pipe is connected to a micropump. The silylated PVA sponge is placed at oil-water interface, for the purpose of continuous oil removal from water. When the micropump works, the oil will be pumped to the pipe through the sponge and then collected into an oil collector. At the same time, water will be blocked by the hydrophobic sponge and remain. All the separation tests were operated at room temperature. Through continuous oil-removal tests, we can obtain the separation efficiency ( $R$ ).  $R$  was calculated according to the following equation:

$$R\% = \left( \frac{M}{M_0} \right) \times 100\%, \quad (1)$$

where  $M_0$  is the weight of the original oil mass in the mixture liquid and  $M$  is the weight of the collected oil after separation.

## 3. Results and Discussion

### 3.1. Characterization

**3.1.1. SEM.** The surface morphologies of the pure and silylated PVA sponge were observed by the scanning electron microscopy at different magnifications (Figure 3). SEM images of Figures 3(a) and 3(b) reveal that pure PVA sponge exhibits porous three-dimensional interconnected network structure and smooth surface morphology. After MTES coating on the PVA skeleton surface, the PVA skeleton surface exhibits a relatively rough texture. Meanwhile, the high-magnification SEM image indicates that lots of polymethylsiloxane nanoparticles are successfully modified to the PVA skeleton surface. Nevertheless, there are not any large bulks and deep grooves on the surface, as depicted in Figures 3(c) and 3(d). As a result, nanoscaled polymethylsiloxane nanoparticles were uniformly deposited on the PVA skeleton surface. It can be seen from the SEM photographs that a layer of nanostructured particles is attached to the surface of the three-dimensional microporous skeletons of PVA sponge. This typical micronanostructure is the key to superhydrophobic of the hybrids [26, 27].

**3.1.2. XRD.** In addition, Figure 4 shows the XRD diffraction patterns of PVA sponge and the silylated PVA sponge. As shown in Figure 4(a), the XRD patterns of the PVA sponge demonstrate a typical diffraction peak of PVA at  $2\theta = 19.5^\circ$  [28]. As displayed in Figure 4(b), a broad peak appeared at  $10^\circ$  of the silylated PVA sponge, which corresponded to the amorphous peak of  $\text{SiO}_2$ , which also indicates the existence of the polymethylsiloxane nanoparticles coatings on the PVA skeleton surface.

**3.1.3. FTIR.** The surface chemistry of pure PVA sponge and the silylated PVA sponge can be confirmed from the FTIR spectra, as shown in Figure 5. Firstly, Figure 5(a) reveals the

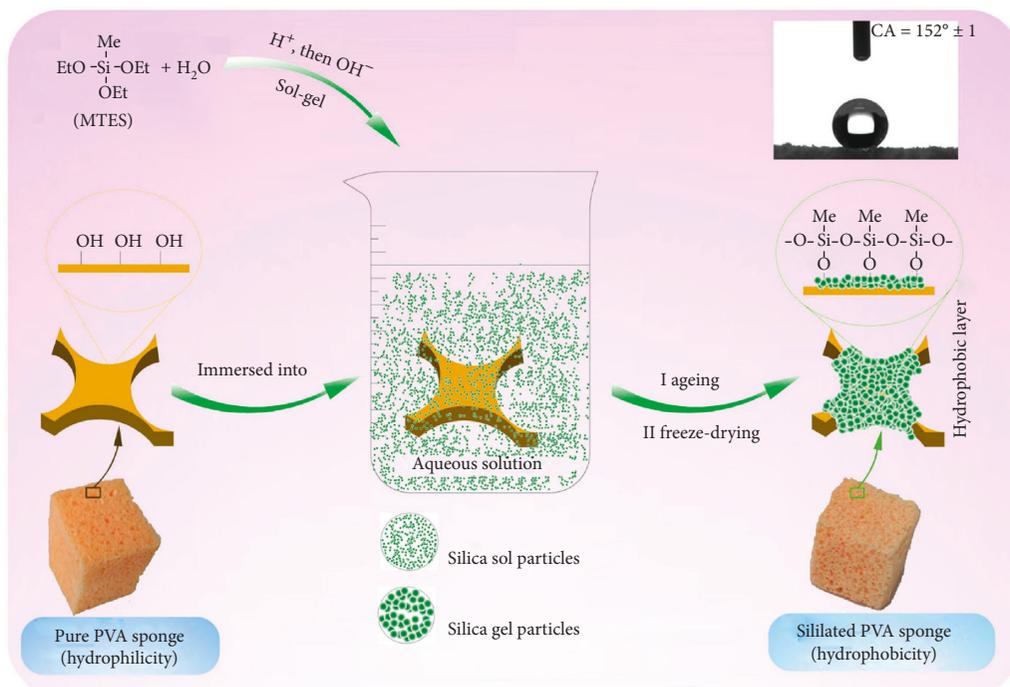


FIGURE 1: Schematic illustration of preparing the silylated PVA sponge by one-pot method.

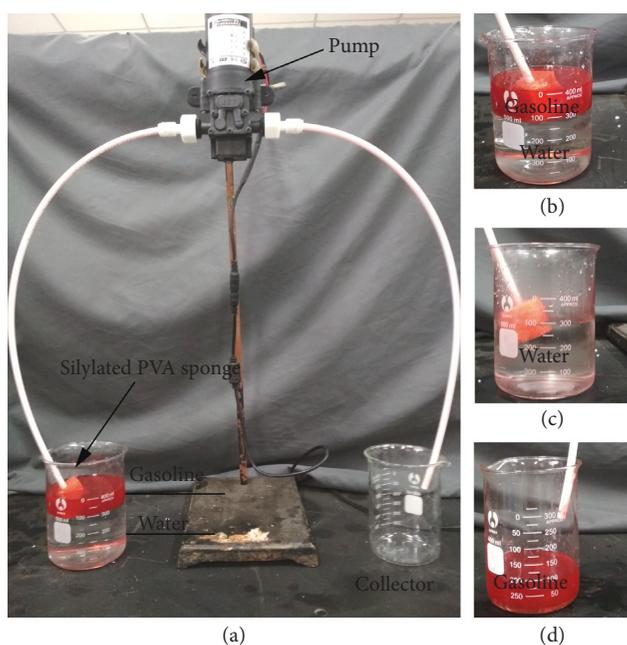


FIGURE 2: Photographs of (a) the continuous oil-water separation device, (b) oil-water mixture image, (c) water left in the beaker after continuous adsorption, and (d) oil collected during continuous adsorption.

FTIR figure of pure PVA sponge. As expected, the FTIR spectrum of PVA shows a broad and strong band at among 3550~3200 cm<sup>-1</sup>, which corresponded to O-H stretching vibration. Furthermore, the stretching vibration absorption peaks of C-H appear at between 2980 and 2950 cm<sup>-1</sup>. Comparing the FTIR spectrum of the silylated PVA sponge (Figure 5(b)) [29, 30], besides the typical peaks of PVA sponge, the peak at 1080 cm<sup>-1</sup> is attributed to the asymmetric

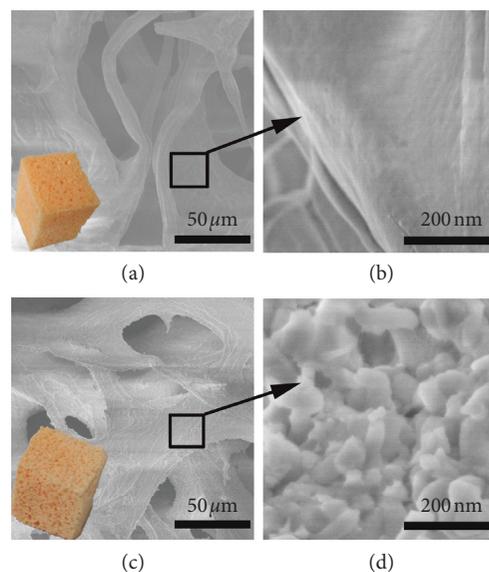


FIGURE 3: SEM images with different magnifications for pure PVA sponge (a, b) and the silylated PVA sponge (c, d). Insets are their corresponding digital photographs.

stretching vibration of Si-O-Si, and the characteristic peak at 800 cm<sup>-1</sup> corresponds to the Si-O-Si symmetric stretching vibration [31–33]. Moreover, due to the successful modification of MTES on the surface of PVA sponge, the O-H stretching vibration of PVA is significantly weakened. This series of data indicate that the MTES has successfully connected onto the surface of the porous PVA sponge.

3.1.4. *Wettability.* The wetting property of the pure and silylated PVA sponge was studied by contact angle

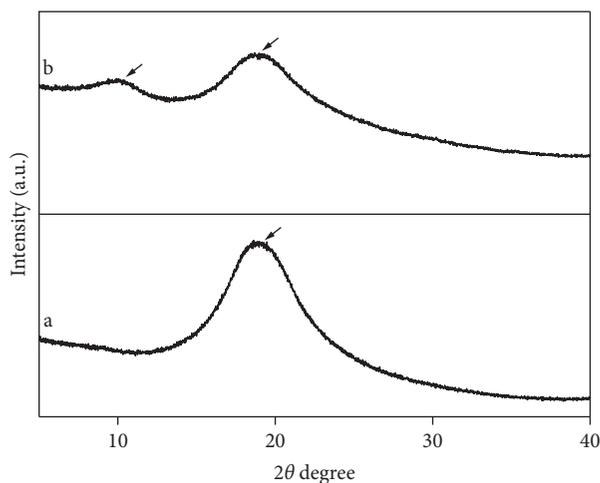


FIGURE 4: XRD spectra of pure PVA sponge (a) and the silylated PVA sponge (b).

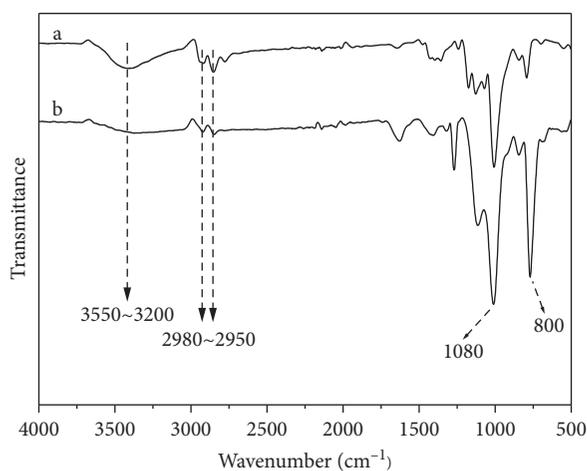


FIGURE 5: FTIR spectra of pure PVA sponge (a) and the silylated PVA sponge (b).

measurement, and the static WCAs were measured at room temperature. As shown in Figure 6, droplets of water and gasoline were deposited onto the pure and silylated PVA skeleton surface. Obviously, the pure PVA sponge showed a strong lyophilic property, as both water and gasoline droplets were absorbed instantaneously (Figure 6(a)). In Figure 6(b), the gasoline immediately penetrated into the silylated PVA sponge, while water remained at the surface, demonstrating that the silylated sponge has combined hydrophobic and oleophilic properties. Figure 6(c) indicated that the static contact angle of the pure PVA sponge is  $0^\circ$ . On the contrary, the silylated PVA sponge displayed a high contact angle, up to  $152^\circ \pm 1$ , as shown in Figure 6(d). The wettability of material depends on both chemical composition and geometrical microstructure of the solid interface, on the basis of Wenzel's theory [34–36]. According to thermodynamic equilibrium, the apparent contact angle and the roughness factor have the following relationship:

$$\cos \theta^w = r \cos \theta, \quad (2)$$

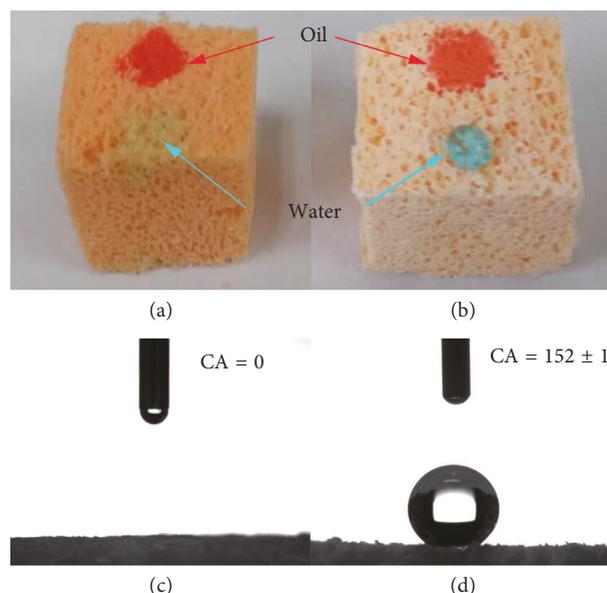


FIGURE 6: Water (colored blue) and oil (colored red) dropped onto the surface of pure PVA sponge (a) and the silylated PVA sponge (b). WCAs of pure PVA sponge (c) and the silylated PVA sponge (d). Plant oil is colored red with Sudan III and water is colored blue with Methyl blue in advance, respectively.

where  $\theta^w$  refers to the apparent contact angle,  $r$  corresponds to the roughness factor of the given interface, and  $\theta$  represents Young's angle. Herein, the surface energy of the sponge was effectively decreased by the polymethylsiloxane groups. Besides, the PVA sponge coated with polymethylsiloxane nanoparticles exhibits superhigh surface roughness. Hence, the composites displayed excellent hydrophobicity with a surprisingly high contact angle ( $152^\circ \pm 1$ ). In this study, the surface of silylated PVA sponge consists of disorder and dispersive nanometer-sized polymethylsiloxane particles; therefore, it owns high roughness and exhibits superhydrophobic property.

**3.2. Absorption Selectivity.** In our case, in order to verify the super oil-removal ability, oil-water separation experiments were implemented with two different densities of oil (Figure 7). Gasoline and chloroform were dropped onto the surface and bottom of aqueous phase, respectively. The silylated PVA sponge can easily remove the gasoline and chloroform from water. The result shows that the silylated PVA sponge has excellent absorption selectivity, whether used on the surface or bottom of the water. In addition, we investigated the selectivity of separation of hydrophobic solvents (various types of oil). However, it showed no selectivity toward the mixture of various hydrophobic solvents. As shown in Figure S2, the static contact angle of the prepared material was zero for all kinds of hydrophobic solvents and there was no difference in the contact angle among different kinds of hydrophobic solvents. Therefore, except for water drops, other different kinds of oil can be well absorbed by the prepared material quickly. This is the reason why reported materials can remove various types of oil pollution from water quickly and efficiently.

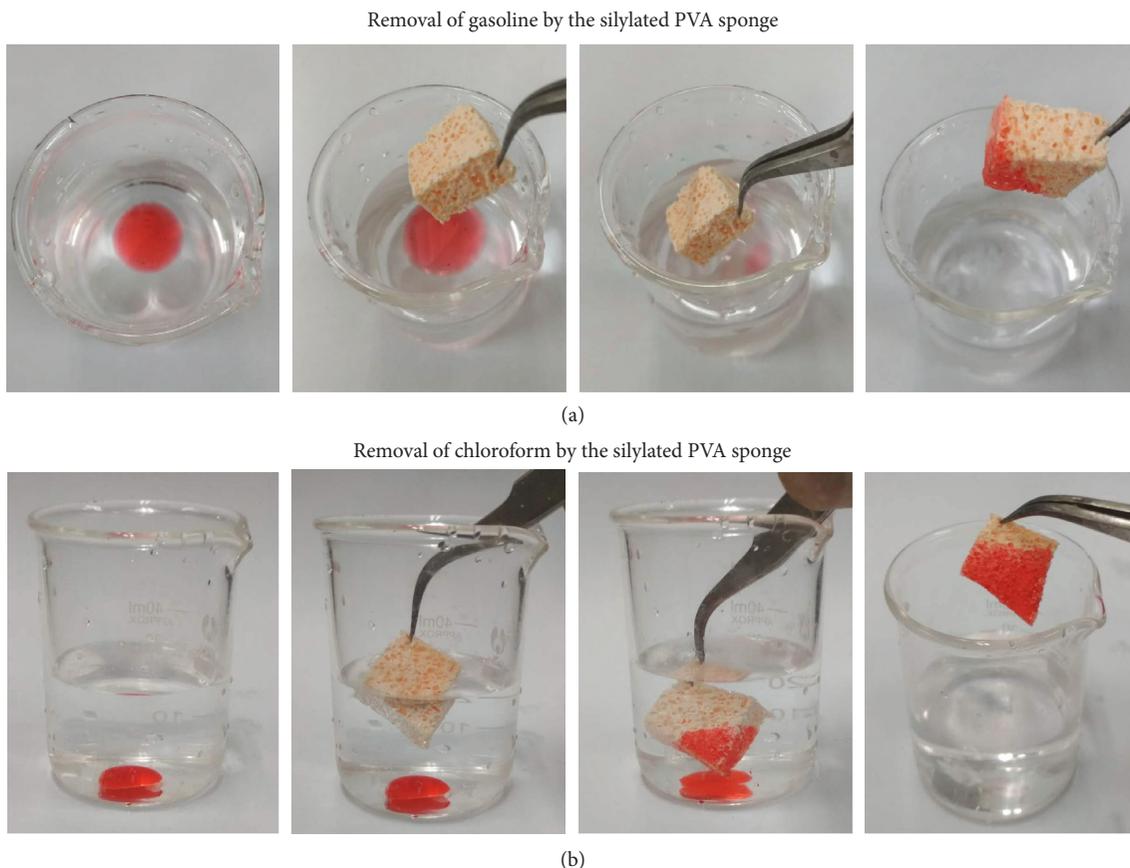


FIGURE 7: Selected removal of gasoline (a) and chloroform (b) from water with the silylated PVA sponge. Gasoline and chloroform are colored red with Sudan III in advance.

### 3.3. Continuous Oil Removal and Absorption Capability.

Because of the superhydrophobic properties, the silylated PVA sponge may become a potential oil-water separation material. In this study, a continuous oil-water separation device was established to evaluate the absorption capacity of the synthetic absorbent. In the experiment, a functionalized PVA sponge (specification:  $3 \times 3 \times 3 \text{ cm}^3$ , 2.21 g) was placed in a container with oil-water mixture (300 ml water and 100 ml oil). The functionalized PVA sponge was connected to the micropump through a pipe (Figure 2). With the aid of the negative pressure generated by the micropump, oil easily passed through the silylated PVA sponge, while water was blocked to achieve selective separation. The whole experiment was completed within 1 minute, leaving clean water in the container.

In order to examine the oil-removal capacity of the silylated PVA sponge, various types of oil and organic solvents are selected to implement continuous separation experiments for two hours. Then, the oil mass flux and separation efficiency are listed in Figure 8. Oil and organic solvents used in this study included dichloromethane, chloroform, *n*-hexane, toluene, chlorobenzene, and gasoline. As shown in Figure 8(a), the silylated PVA sponge exhibits oil mass fluxes ranging from 353 to  $480 \text{ L}\cdot\text{m}^{-2}\cdot\text{min}^{-1}$ . Moreover, the separation efficiency of the absorbent still reaches up to 99.6% during an hour separation. Such high mass fluxes meant the silylated PVA sponge could remove

oil from the water up to 6200~14000 times of its own mass, which is significantly higher than all the materials mentioned in Table 1. As a comparison, the unmodified PVA sponge was used for separating oil from water. As shown in Figure S3, the unmodified PVA sponge is placed at the oil-water interface. Under the pressure of the pump, water is absorbed together with gasoline; so, the unmodified PVA sponge shows no effect on oil-water separation. This is because the PVA sponge exhibits amphiphilic property before hydrophobic modification, which is also confirmed by Figure 6(a). Therefore, the PVA sponge before modification has no selectivity for oil-water separation.

Moreover, we investigated the durability and repeatability of the silylated PVA sponge. As shown in Figure S1, the silylated PVA sponge did not suffer from obviously damage and any loss in separation capacity even after 10 cycles. This is because that MTES could form a stable mixed structure with the PVA skeleton in synthesis, resulting in the rare shedding of polymethylsiloxane nanoparticles and the long-term stability for oil-water separation. The results demonstrated the as-made sponge presented excellent separation capacity and reusability.

3.4. Comparison of Various Absorbents. Saturated absorption capacity depends mainly on the parameters such as the

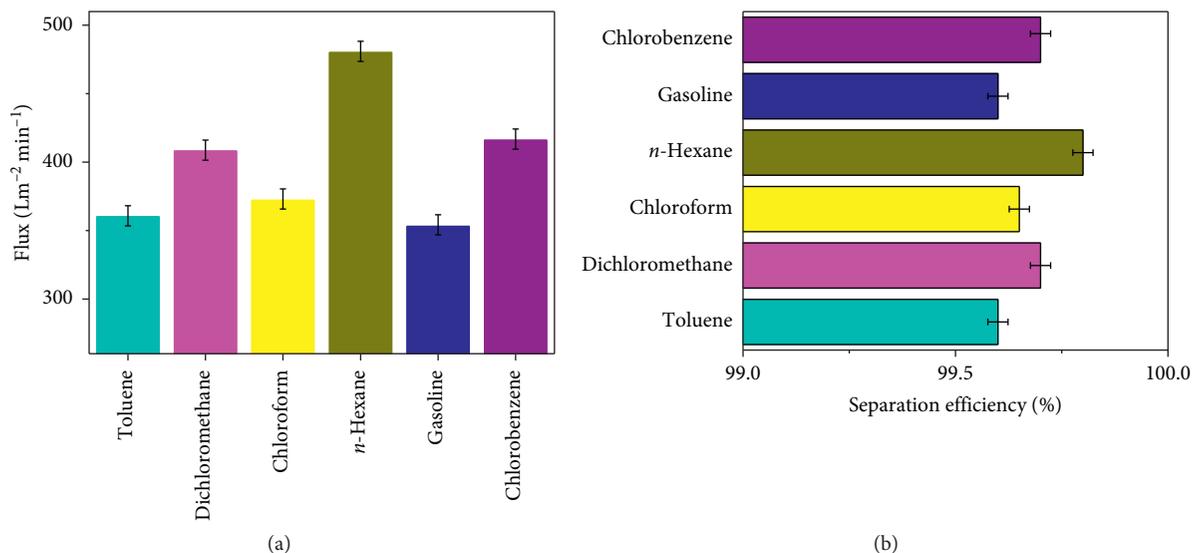


FIGURE 8: (a) The absorption flux and (b) the separation efficiency of the silylated PVA sponge for oil and organic solvents during 1-hour separation.

TABLE 1: Comparison of the absorption capability of various absorbents.

Absorbent	Cost	Oil	Sorption capacity, g/g		Oil recovery method	Ref.
			Once	Total		
Silane-modified nanocellulose	+	Toluene	~170	1020	Distillation	[37]
Functional bacterial cellulose	++	Diesel	120	1200	Squeezing	[19]
Grapheme sponge	++	Dodecane	~20	200	Burning	[38]
CNTs hybrids	++	Chloroform	~95	570	Acetone extraction	[39]
Attapulgite coated PU sponge	—	Kerosene	—	3200	Continuous separation	[40]
Silylated PVA sponge	—	Toluene	—	7600	Continuous separation	Present work

Note: "once" means saturated absorption capacity; "total" is calculated through saturated absorption capacity multiplied by the number of cycles, or mass flux multiplied by the running time.

absorbent's own density and the type of absorbed oil. It is inevitably biased to evaluate the capability of absorbents only according to their saturated absorption capacity. Because the service life of absorbent (cycle number or accumulated use time) is also an important factor used for evaluation of the capacity of absorbent. The separation efficiency of adsorbent often decreases after repeated recycling, which may be due to the loss of functional groups and the decrease of material roughness. Hence, in this work, the total absorption capacity will be calculated according to the number of cycles and the saturated absorption capacity of a part of absorbents reported in recent years. The capability of absorbents was evaluated with comparison of the total absorption capacity. A variety of absorbents and their performances are listed in Table 1. Carbonized materials and absorbents, such as graphene or carbon nanotubes, can be used as the representatives of novel absorbents, revealing excellent absorption performance. Thus, the complexity of the preparation process and high cost limit their practical application. Nanocellulose fiber-based absorbents exhibited excellent absorption capacity for oil and organic solvents, because of its low density. Besides, functionalized 3D PU-based absorbents have become a kind of hot material

pursued by scientists, due to its low cost and compressibility. In the present work, the silylated PVA sponge could be used to separate oil up to 6200~14000 times of its own mass, which is significantly higher than all the absorbents mentioned above. This means that it is an excellent superabsorbent for oil removal and collection. The raw materials of the silylated PVA sponge are really cheap and can be produced on a large scale. Moreover, MTES and PVA are classified as environmentally friendly materials. Besides, the preparation of the silylated PVA sponge is carried out in an aqueous solution, and the process is nonpolluting. These results are encouraging and suggest that the silylated PVA sponge has great potential in practical application.

#### 4. Conclusions

In summary, a facile and environment-friendly synthetic route has been proposed to successfully prepare the silylated PVA sponge with superhydrophobicity and superoleophilicity, without using any organic solvent and multistep reaction. SEM photographs revealed that the surface of interconnected pores in the original PVA skeleton was coated with polymethylsiloxane nanoparticles through an

environment-friendly synthesis method, forming a firm substructure. But, at the same time, the silylated PVA sponge retained original PVA structural integrity. Owing to the silylation of the skeleton surface and such characteristic nanomicro substructures, the silylated PVA sponge possessed superhydrophobicity and superoleophilicity, with the static WCAs running up to  $152^\circ \pm 1$ . This functionalized sponge could be used as a selective filter to realize continuous and effective oil-water separation with the help of a micropump. The absorbent was chemically robust, and the oil-absorption flux reached ranged from 353 to 480 L·m<sup>-2</sup>·min<sup>-1</sup> with excellent separation efficiency (>99.6%). Hence, the silylated PVA sponge, as a promising superabsorbent, may become one of the most potential candidates in water treatment applications.

### Data Availability

The data used to support the findings of this study are available from the corresponding author (Junhui Xiang; xiangjh@ucas.edu.cn) upon request.

### Conflicts of Interest

The authors declare that they have no conflicts of interest.

### Acknowledgments

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### Supplementary Materials

Figure S1: (a) separation cycles of oil-water mixture; (b) SEM image of the silylated PVA sponge before toluene-water separation; (c) SEM image of the silylated PVA sponge after 10 separation cycles of toluene-water. Figure S2: photograph of water (colored blue) and kinds of hydrophobic solvents (colored red) dropped onto the surface of the silylated PVA sponge. Hydrophobic solvents are colored red with Sudan III and water is colored blue with methyl blue in advance, respectively. Figure S3: photographs of (a) the continuous oil-water separation device and (b) oil-water mixture image after continuous adsorption. Gasoline is colored red with Sudan III in advance. (*Supplementary Materials*)

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