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Preparation of superhydrophobic and superoleophilic polypropylene fibers with application in oil/water separation

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A novel type of fiber material for the separation of oil/water mixed liquids has been prepared by the chemical vapor deposition of hydrophobic silanes on commercially available polypropylene (PP) fiber. To achieve this, the PP fiber materials were first etched by concentrated H_2SO_4. Then, the as-prepared PP fiber materials were immersed in highly concentrated suspension of graphene oxide modified by amine-functionalized SiO_2 nanoparticles (SiO_2–GO). Finally, the as-prepared PP fiber materials were treated by the chemical vapor deposition of hydrophobic silane (octyltrichlorosilane) to form a superhydrophobic surface with a water contact angle more than 156°. The hydrophobic lightweight PP fiber materials are almost instantly filled with the oil phase when selectively absorbing oil from water. The oil can also be drained from the absorbed PP fiber materials. Moreover, the PP fiber material shows the good performance for repeated use.

Keywords: absorption; fiber; chemical vapor deposition (CVD) (chemical reaction)

Introduction

Removal or collection of the organic pollutants from water surfaces has attracted worldwide attention (Ceylan et al., 2009; Yuan et al., 2008). The conventional methods used to solve these problems including collection of oil from the water surface; mixing of oil with water using dispersing agents to facilitate natural degradation; and *in situ* burning of the oil spills (Korhonen, Kettunen, Ras, & Ikkala, 2011). Among existing techniques for the removal of oil from waters, the use of sorbents is generally considered to be an efficient and facile way because of their special wettability (Lahann, 2008). The main requirements for an ideal sorbent material for oil spill cleanup include high hydrophobicity, high uptake capacity, and high rate of uptake, buoyancy, retention over time, durability in aqueous media, reusability or biodegradability, and recoverability of the absorbed oil. Currently, oil sorbent materials can be classified into inorganic mineral materials, synthetic organic products and natural organic materials. The inorganic materials include vermiculite, zeolites, bentonites, and so on. Natural organic materials mainly include wood fiber, cotton fiber, wool fiber, and kenaf fiber. Synthetic organic products mainly include vesicant stuff (Abdullah, Rahmah, & Man, 2010). Although widely applied in practical applications, these absorbent materials still have limitations such as environmental incompatibility, low absorption capacity, poor recyclability, and so on. In particular, these materials absorb not only oils but also water, which reduces the separation selectivity and efficiency. Therefore, novel absorbent materials combined with high absorption capacity, high selectivity and efficiency, low cost, excellent recyclability, and environmental friendliness are important for the development of advanced oil/water separation technology (Wang et al., 2011).

Recently, with the development of nanotechnologies, more and more materials can be endowed with superhydrophobic and superoleophilic properties (Chu & Pan, 2012; Wang et al., 2009; Zhang et al., 2009; Zhang, Pu, Steven, & Severtson, 2010). Superhydrophobic materials (Hitoshi, Jun, & Tetsuo, 2011; Li, Zhang, & Wang, 2008; Wang, Chen, Cheng, & Fu, 2010) have extended applications to the research area of theirs modification. Cheng et al. (2011) prepared functionalized sponges by combining electroless metal deposition with self-assembled monolayers; its super-

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hydrophobicity was obtained by immersing ethanol solution of n-dodecanethiol overnight. Choi et al. (2011) obtained polydimethylsiloxane sponges for the selective absorption of oil from water. Zhu, Pan, and Liu (2011) prepared superhydrophobic and superoleophilic sponges by combining electroless deposition of copper film with immersing ethanol solution of n-dodecanethiol. Li et al. (2012) obtained water-repellent sponges applied for oil absorbing by using metal nanoparticles deposition.

In this paper, the treated PP fiber material was immersed into SiO_2–GO suspension for 5 min. Finally, the dried PP fiber material was modified by silane (octyltrichlorosilane) through chemical vapor deposition (CVD) to obtain water repellency. The GO sheets were mainly used as the substrates to facilitate the fixation of SiO_2 nanoparticles. Furthermore, oily molecules could be easily permeated into layer gap of GO sheets by physisorption, due to the high specific surface area of GO and its chemistry which may help in improving oil absorption (Sun, Yu, & Fugetsu, 2012; Zhu et al., 2010). Moreover, the as-obtained bio-inspired surface shows good durability towards hot water. Interestingly, we also found that the functionalized PP fiber material could be reused with high absorptivity and retaining the superhydrophobicity.

Experimental procedure

Materials

All chemicals were of analytical grade and used as received. Graphite flakes were purchased from Qindao Chenyang Graphite Co., Ltd. (Qindao, China). The chemicals $NaNO_3$ and N-hexane were obtained from Shanghai Zhenxing Chemical Reagent Co., Ltd. (Shanghai, China). Concentrated H_2SO_4 was purchased from Zhejiang Sanyin Chemical Reagent Co., Ltd. (Hangzhou, China). The compounds $KMnO_4$ and ammonium hydroxide (> 30 wt.%) were obtained from Hangzhou Gaojing Fine Chemicals Co., Ltd. (Hangzhou, China). Hydrogen peroxide was obtained from Zhejiang Xinhua Chemical Industry Co., Ltd. (Hangzhou, China). Tetraethylorthosilicate (TEOS), heptane, and toluene were obtained from Tianjin Kermal Chemical Reagent Co., Ltd. (Tianjin, China). Ethanol, pentane, oil red, and benzene were purchased from Hangzhou Changjiang Chemicals Co., Ltd. (Hangzhou, China). The compound (3-aminopropyl)triethoxysilane (> 97 wt.%) was obtained from Nanjing Forward Chemical Co., Ltd. (Nanjin, China). Trichloro(octyl)silane (> 97 wt.%) was obtained from Wuhan Jingnuo Chemical Industry Co., Ltd. (Wuxi, China). Silicone oil was purchased from Fangzhou Chemical materials Co., Ltd. (Fuzhou, China). PP fiber materials were obtained from Yancheng Henggu Fiber Co., Ltd. (Fuzhou, China).

Methods

Preparation of SiO_2 sol modified by (3-aminopropyl)-triethoxysilane

Silica sol modified by (3-aminopropyl)triethoxysilane with the particle size of about 8 nm was synthesized by the hydrolysis and condensation of TEOS in ethanol solvent, similar to Stober's method (Costa, Leite, & Galembeck, 2003; Rossi, Shi, Quina, & Rosenzweig, 2005). TEOS amounting to 1.5 mL was added to a flask containing 1.5 mL of concentrated ammonium hydroxide and 50 mL of ethanol while stirring. Then after 30 min, 0.1 mL of (3-aminopropyl)triethoxysilane was added, the stirring was continued overnight, and after that the sol needed to settle for 4 days to ensure that the rest of TEOS in SiO_2 sol had been hydrolyzed completely.

Preparation of SiO_2–GO sheets

The GO was synthesized using a modified Hummer's method (Ang et al., 2009; Xu, Song, Sun, Lu, & Yu, 2011) with all reagent materials used as received. An amount of 100 mg of GO was dispersed in 100 mL of ethanol; the suspension was sonicated for 1 h. Then, the suspension was transferred into the flask with the temperature heated up to 70°C; next, 50 mL SiO_2 sol modified by (3-aminopropyl)triethoxysilane was added to the suspension, stirring for 24 h. The SiO_2–GO was separated by filtration using nylon membrane (0.22 μm) and finally washed with ethanol to remove the excess functional SiO_2 sol.

Preparation of fiber material with superhydrophobic and superoleophilic features

Pieces of PP fiber material with dimensions of $1.5 \times 1.5 \times 0.6\,cm^3$ (length, width, and thickness) were first washed by dipping in alcohol three times, 10 min each time, and dried carefully under 60°C. Then, they were treated with concentrated H_2SO_4 ($100\,g\,L^{-1}$) for 90 s to increase the defect structure of these surfaces which contributes to the adhesion of SiO_2–GO particles to the PP fiber surface, and finally washed with water carefully. The as-prepared PP fiber material was immersed in high-concentrated SiO_2–GO suspensions for about 10 min at room temperature. The PP fiber material absorbed large numbers of SiO_2–GO particles and were immersed in anhydrous ethanol again, taken out and dried under 60°C. The dried PP fiber material coating SiO_2–GO particles were further treated by octyltrichlorosilane

through chemical vapor phase deposition to obtain superhydrophobicity. The temperature of CVD is 80°C and the time of treatment is 30 min.

Characterization

The microstructures of PP material and functional PP fiber material were investigated by ULTRA-55 field-emission scanning electron microscopy (FE-SEM). The SiO_2–GO particles were analyzed by JSM-2100 transmission electron microscopy (TEM) at an accelerating voltage of 200 kV. The Fourier transform infrared (FT-IR) spectra were recorded on a Nicolet 5700 spectrophotometer using. The wetability of functional PP fiber material was analyzed by measurement of the water angles using SL200B contact angle system at ambient temperature. Water droplets were dropped carefully onto the samples. The average contact angle was obtained by measuring at three different positions of the same sample; the accuracy of measurement is $\pm 1^{o}$. The weight of PP fiber material was measured by electronic balance. The size distribution of particles in the silica sol was measured by dynamic light scattering device.

Results and discussion

A facile and low-cost fabrication technique applied to prepare the functionalized PP fiber material with highly efficient oil/water separation capability was started with commercially available PP fiber material, which is widely used in daily life for cleaning because of its

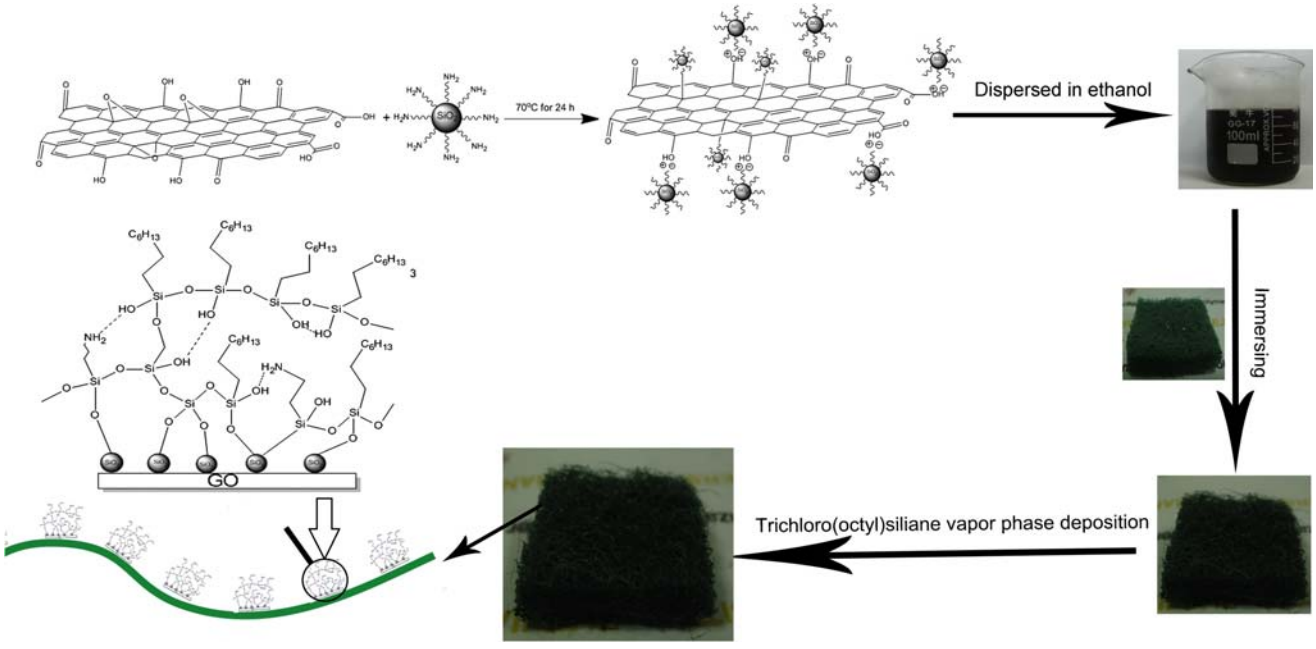

Figure 1. The scheme for the fabrication process of functionalized PP fiber material.

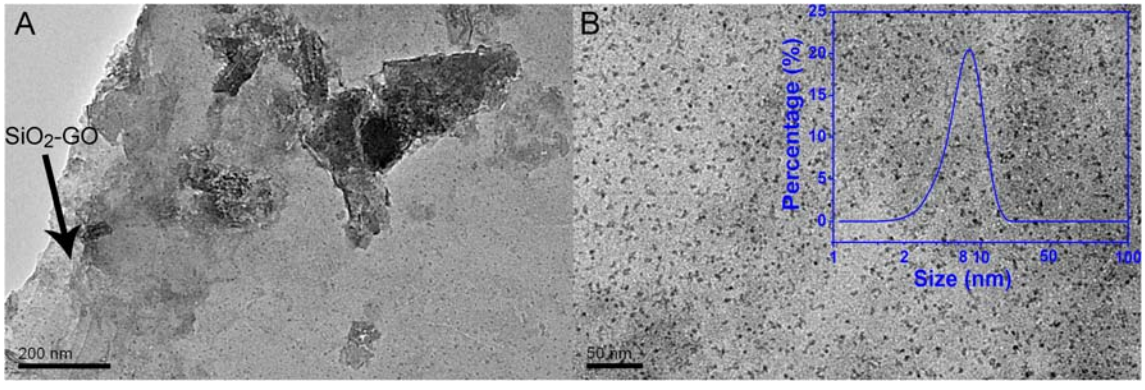

Figure 2. (A) The TEM image of GO modified by SiO_2, (B) the high magnification of (A); the inside is the size distribution of functionalized SiO_2 particles on the GO sheets.

low cost. However, with the development of materials to multifunction, the disadvantage of single function PP fiber material is obvious in the environment protection. So the functionalized PP fiber material needs to be created.

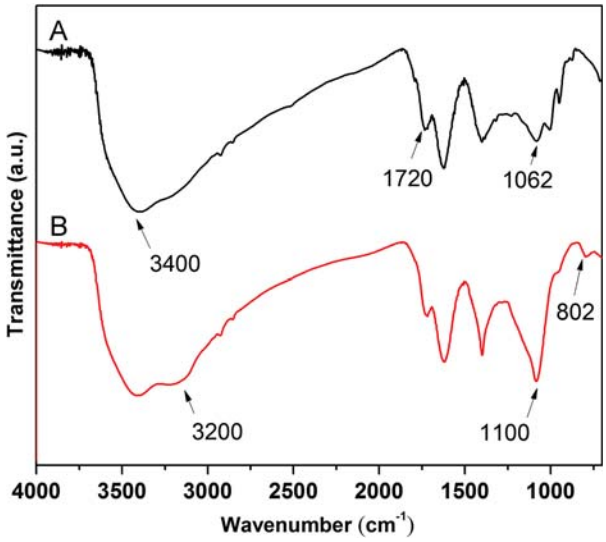

Figure 3. FT-IR spectra of GO (A) and SiO_2–GO (B).

The functionalized PP fiber materials are prepared based on SiO_2–GO particles. Firstly, the PP fiber material was treated with concentrated H_2SO_4. Then, the treated PP fiber material was immersed into SiO_2–GO suspension. Finally, the resulting PP fiber material was modified by hydrophobic silanes to obtain superhydrophobicity. Figure 1 shows the fabrication process of functionalized PP fiber material. The functionalized SiO_2 particles dispersed on the GO sheets by the reaction with epoxy groups in GO and electrostatic force. Figure 2(A) and (B) shows the TEM images of SiO_2–GO. A large amount of functionalized SiO_2 particles were uniformly dispersed on the GO sheets indicating that the SiO_2–GO sheets have been synthesized successfully. In addition, some amine-functionalized SiO_2 particles have agglomeration to some extent due to the existence of hydrogen bond (Roy et al., 2010). The inside of Figure 2(B) shows the functionalized SiO_2 particles on the GO sheets with the size of about 8 nm.

The FT-IR analyses were carried out to develop an understanding of the reaction mechanism of function-

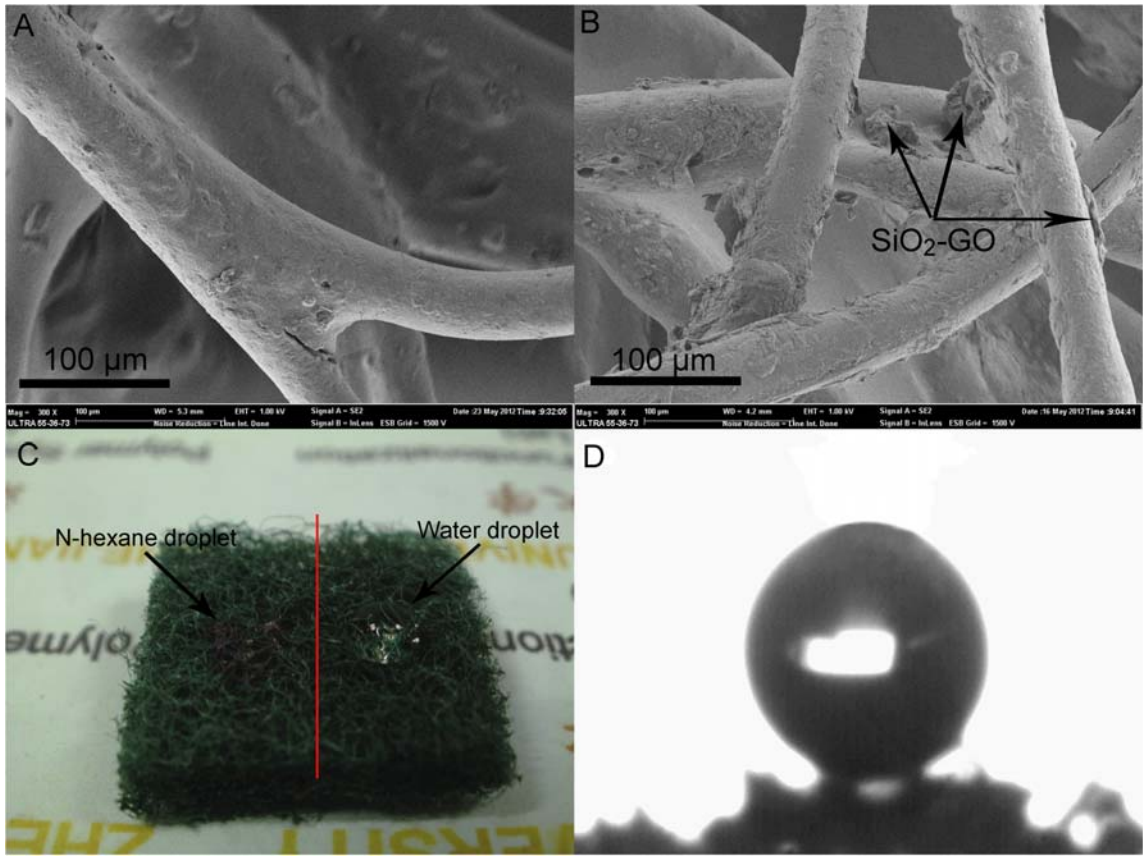

Figure 4. SEM images of the surface of PP fiber before (A) and after (B) treatment; (C) photograph of contact angle measurement with water droplets and red oil droplets; and (D) water contact angle image.

alized SiO_2 particles and GO sheets. Figure 3 shows the FT-IR spectra of GO sheets and SiO_2–GO sheets. The spectrum of GO in Figure 3(A) shows a broad band with a maximum around 3400 cm^{-1} which is due to the O–H stretching. The weak peaks around at 1720 cm^{-1} and 1062 cm^{-1} are associated to the stretching vibration of C=O bond and C–O–C bond (Ma et al., 2012; Xu et al., 2012), respectively. Figure 3(B) shows the spectrum of SiO_2–GO sheets. A broad band around 3200 cm^{-1} is attributed to the N–H stretching of functionalized SiO_2 (Huang, Yang, Chinn, & Munson, 2003; Roy et al., 2010). The C–O–C vibrations of epoxy groups in GO disappeared at 1062 cm^{-1} due to the reaction with NH_2 (Yang et al., 2009). A strong peak appears at 1100 cm^{-1} due to the presence of Si–O–Si asymmetric bond stretching (Tseng & Wu, 2009). A weak peak appears at 802 cm^{-1} due to the presence of Si–O asymmetric bond stretching (Zhang, Zhang, & Guo, 2012).

The surface morphologies of PP fiber were characterized by FE-SEM. The surface of PP fiber untreated by SiO_2–GO particles and trichloro(octyl)silane was smooth (Figure 4(A)). The PP fiber appeared more rough (Figure 4(B)) after the treatment of the surface of PP fiber with SiO_2–GO particles and trichloro(octyl)silane. There were some pieces of SiO_2–GO particles which were adhering on PP fibers due to electrostatic force and hydrogen bond, mainly through the interaction between the hydroxyl groups on the treated PP fiber and the hydroxyl and amino groups on the SiO_2–GO sheets (Tao, Gong, Lu, Sue, & Bergbreiter, 2001).

In order to clarify the superhydrophobicity and superoleophilicity of the as-prepared PP fiber material, contact angle measurements were carried out with water droplets and N-hexane droplets colored with oil red. As shown in Figure 4(C), 20 μL of water forming a droplet presents a ball-like shape on the surface of PP fiber material. However, a 20 μL of N-hexane forming a droplet diffused into the surface of PP fiber material immediately as the droplet dropped onto the surface due to the PP fiber material's capillary action. A droplet having 10 μL of water stood on the PP fiber surface, the contact angle of the water droplets was about 156° (Figure 4(D)). Generally, the superhydrophobic property is the result of the rough structure and low surface energy (Xu et al., 2011). For the PP fiber material, the roughness of surface plays a more important role in the process of obtaining superhydrophobic property. This is because when a water droplet was suspended on the rough structure, it allowed air trapping between the rough structures on a surface underneath the droplet (Rioboo et al., 2008; Zhang et al., 2009). These phenomena proved the superhydrophobic and superoleophilicity of functionalized PP fiber material, which suggested that it could be used as an oil sorption material.

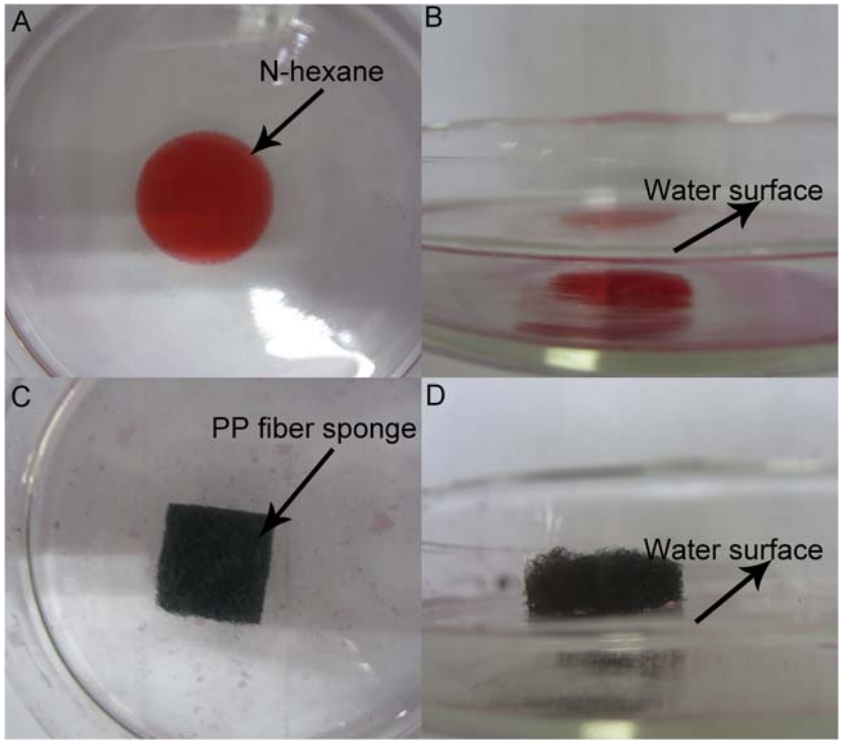

Figure 5. Photographs of the oil uptake process. (A) A red oil droplet was dropped onto the middle of the water surface. (B) The side view of (A). (C) The oily droplet was absorbed as soon as it made contact with the PP fiber material. (D) The side view of (C).

In an attempt to investigate the absorbability of the as-prepared PP fiber material, the following experiment was carried out. A 100 μL droplet of N-hexane colored with oil red was dropped onto the middle of the water surface (Figure 5(A) and (B)). Then, a piece of PP fiber material with dimensions of $1.5 \times 1.5 \times 0.6\,cm^3$ (length, width, and thickness) was held to approach the droplet. Due to its superoleophilicity and superhydrophobicity, the oily droplet was absorbed by the PP fiber material in a few minutes, and it was still floating in the water (Figure 5(C) and (D)).

The stability of surface is important for oil/water separation system. As shown in Figure 6(A), a droplet of 20 μL of water presenting a ball-like shape on the superhydrophobic surface was nearly unchanged with the water's temperature increasing from room temperature to 85°C indicating that surface has good stability. Moreover, as shown in Figure 6(B), this PP fiber material has high absorption for normal alkane, aromatic, and common silicone oil at room temperature due to the reticulated network of the PP fiber material's pores by capillary action which indicates that the PP fiber material could be widely used in different kinds of oil/water system.

In addition that the PP fiber material could be reused with high absorbability is an important issue for the oil spill cleanup (Figure 6(C)). The absorption can be expressed by weight gain. A piece of PP fiber material with dimensions of $1.5 \times 1.5 \times 0.6\,cm^3$ (length, width, and thickness) was immersed into the N-hexane and pentane solvents for about 5 min, respectively. Then, the PP fiber material was took out and weighted. Finally, the PP fiber material was dried carefully at 60°C and was made ready for next

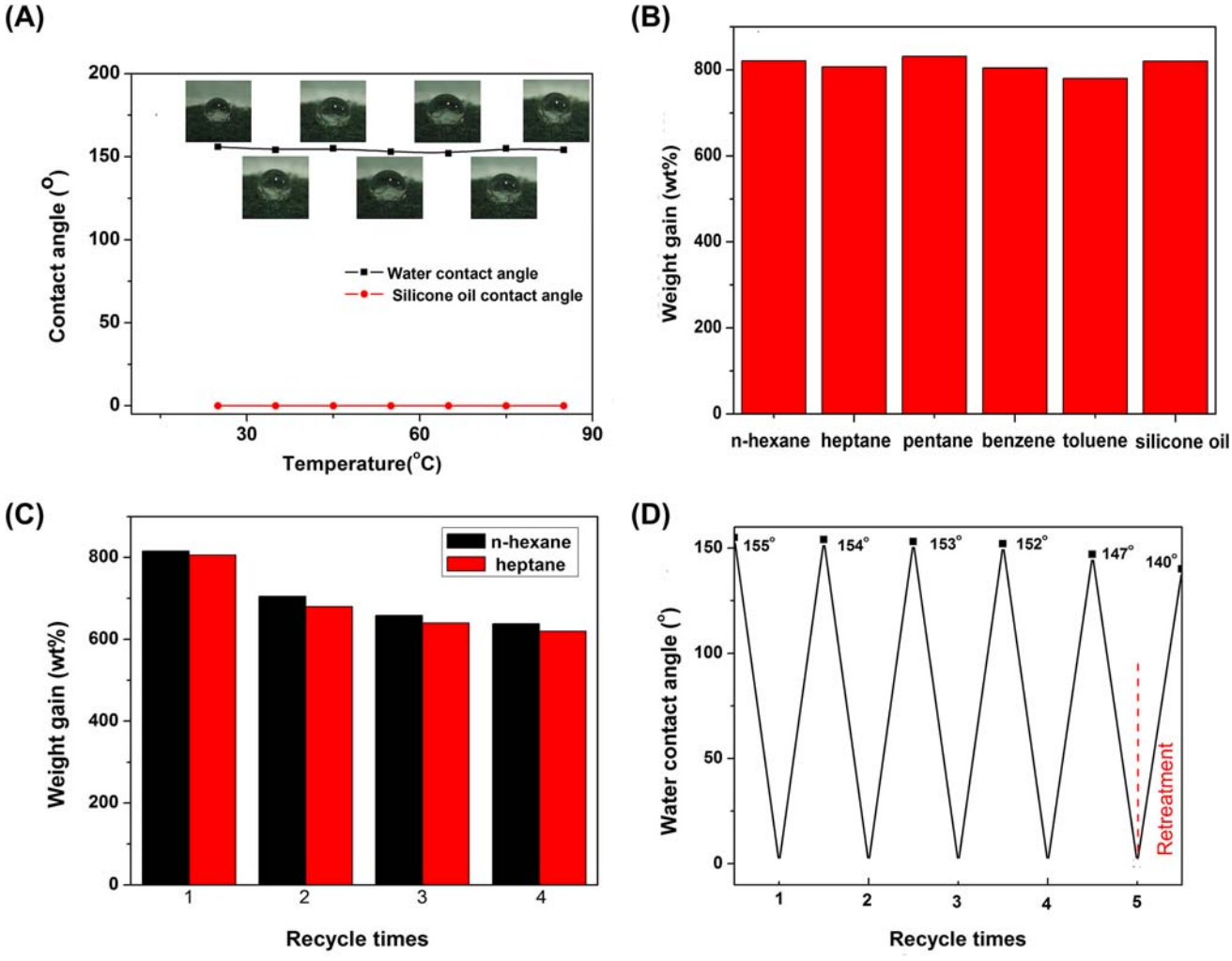

Figure 6. (A) Contact angle of water droplet and oil droplet standing on the fibers at different temperatures. (B) Absorption capacities for a selection of organic solvents and oils in terms of its weight gain. (C) Demonstration of the recyclability of the PP fiber. The absorption capacity of the PP fiber material after multiple cycles was normalized by weight gain. (D) Reversible wettability of the functionalized PP fiber material for water droplets.

testing. This process was cycled four times. Firstly, the PP fiber material's absorption for N-hexane and pentane were 816 and 806%, respectively. After four times, the PP fiber material' absorption for N-hexane and pentane decreased to 638 and 620%, respectively. The PP fiber material still keeps its high absorption rate and exhibits the possibility for recycle use.

Moreover, the reversible wettability of the functionalized PP fiber material for water droplets is an important issue for repeated use. In this experiment, a piece of PP fiber material with dimensions of $1.5 \times 1.5 \times 0.6\,cm^3$ (length, width, and thickness) was immersed into the N-hexane solvents for about 5 min. Then, the PP fiber material was taken out. Finally, the resulting material was dried carefully at 60 °C and tested for its wettability, this process was recycled five times. However, as shown in Figure 6(D), the functionalized PP fiber material loses superhydrophobicity when it is used for oil-absorbing more than five times, but if the PP fiber is retreated by trichloro (octyl)silane chemical vapor phase deposition, the superhydrophobicity will recover. It may be caused by the lost of hydrogen grafted trichloro(octyl)silane during the process of organic solvents absorption and volatilization.

Conclusion

In summary, we have developed an efficient multifunctional PP fiber by immersing method and trichloro(octyl)silane chemical vapor phase deposition technique for oil spill cleanup. It has the advantages of high efficiency and reproducibility, which is suitable for many types of organic solvents or oils. The fabrication technique is simple and easy to be scaled up, while the employed materials are inexpensive and some of them can be recycled. We believe this water/oil separation system has great prospect in processing oil pollution.

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