Directly Coating Hydrogel on Filter Paper for Effective Oil–Water Separation in Highly Acidic, Alkaline, and Salty Environment

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The separation of oil–water mixtures in highly acidic, alkaline, and salty environment remains a great challenge. Simple, low-cost, efficient, eco-friendly, and easily scale-up processes for the fabrication of novel materials to effective oil–water separation in highly acidic, alkaline, and salty environment, are urgently desired. Here, a facile approach is reported for the fabrication of stable hydrogel-coated filter paper which not only can separate oil–water mixture in highly acidic, alkaline, and salty environment, but also separate surfactant-stabilized emulsion. The hydrogel-coated filter paper is fabricated by smartly crosslinking filter paper with hydrophilic polyvinyl alcohol through a simple aldol condensation reaction with glutaraldehyde as a crosslinker. The resultant multiple crosslinked networks enable the hydrogel-coated filter paper to tolerate high acid, alkali, and salt up to 8 M H_2SO_4, 10 M NaOH, and saturated NaCl. It is shown that the hydrogel-coated filter paper can separate oil–water mixtures in highly acidic, alkaline, and salty environment and oil-in-water emulsion environment, with high separation efficiency (>99%).

1. Introduction

The frequent oil spill accidents and the rapid expansion of manufacturing industries have highlighted the challenge of effective oil–water separation.[1–4] Oil–water separation is essentially an interfacial issue, and thus intelligent strategies based on special wettability have been demonstrated to be effective and advantageous.[5,6] Since the time when the superwettable coating mesh has been developed for the separation of oil and water, it has been witnessed an explosion in the development of various materials based on special wettability to separate oil–water mixtures in the past few years.[7–20] These synthesized oil–water separation membranes are generally categorized into "oil-removing" and "water-removing." However, most of these membranes are usually used for separation of oil slick in gentle environment. It is difficult for them to separate oil–water mixtures in complex environment, such as highly acidic, alkaline, and salty environment or surfactant-stabilized oil–water emulsion environment. On the one hand, because most of these materials are fabricated by modifying with the porous substrate, such as stainless steel, copper grid, polyester fabric, etc., they are exceedingly not resistant to highly acidic or alkaline solutions. On the other hand, subjecting to the "size-sieving" effect,[21] these membranes are usually ruled out for the separation of oil–water emulsion. Meanwhile, the separation of these emulsions usually has the aid of additional procedures, such as pump filter, that are limited, either by energy-intensive or by high operation costs. The more complicated environment in the practical oil–water separation, such as high salt concentration of seawater, high acid or alkali, or emulsified level of industrial oily wastewater, makes the separation encounter more challenges and that pose questions regarding the exploration of novel materials.[22–25] Therefore, there is a critical need for the fabrication of novel stable materials which is expected not only to separate oil–water mixtures in gentle environment, but also to separate ones in complex environment, especially those challenged by highly acidic, alkaline, and salty or stabilized by surfactant.

Herein, taking advantage of an exceedingly simple crosslinking process between cellulose filter paper and polyvinyl alcohol (PVA), we report a superhydrophilic–underwater superoleophobic and ultralow oil-adhesive hydrogel-coated filter paper for the effective oil–water separation in complex environment. We demonstrate that the resulting hydrogel-coated filter paper with multiple crosslinked networks is robust enough to separate oil–water mixtures in highly acidic, alkaline, and salty environment. Meanwhile, the hydrogel-coated filter paper can also realize gravity-driven surfactant-stabilized oil-in-water emulsions separation with high separation efficiency (>99.9%).

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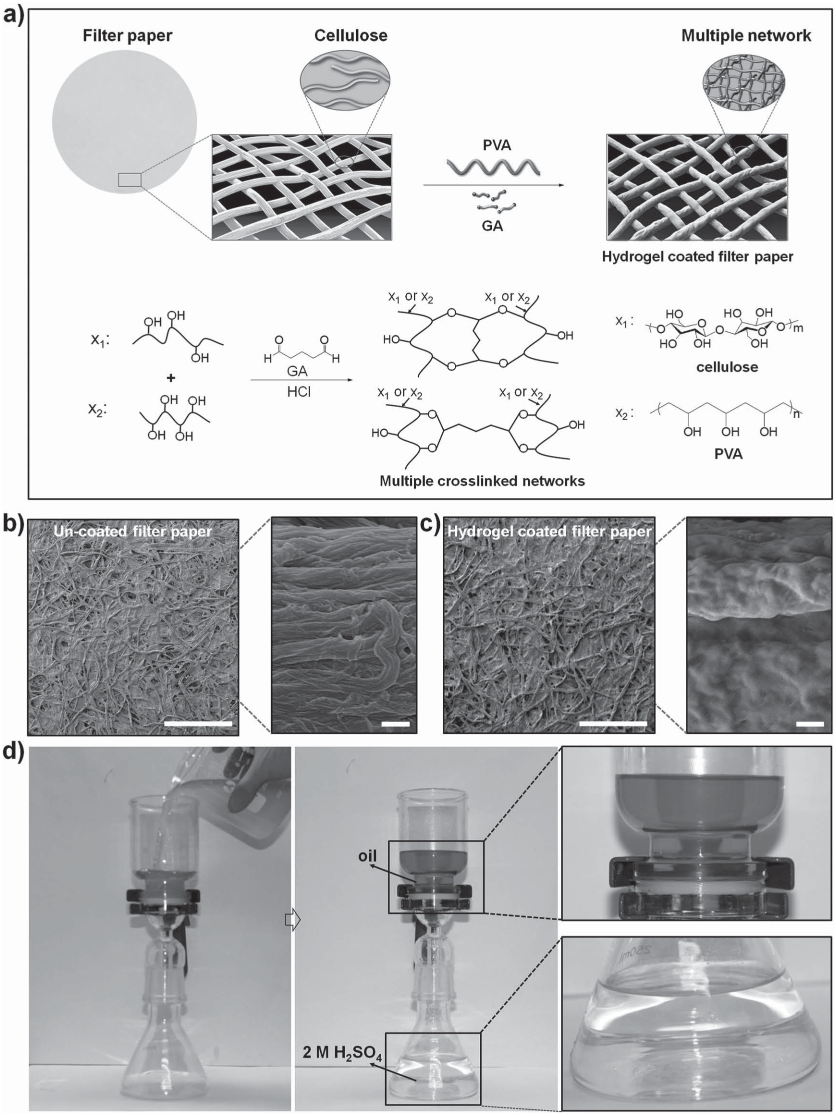

Figure 1. Fabrication of hydrogel-coated filter paper. a) Schematic of the fabrication process of hydrogel-coated filter paper. The process involves a simple aldol condensation reaction between cellulose filter paper and polyvinyl alcohol (PVA), in which glutaraldehyde (GA) serves as a crosslinker and HCl as a catalyst. This process allows the formation of a multiple crosslinked networks. b) SEM images of uncoated filter paper. c) SEM images of hydrogel-coated filter paper. Scale bar, low magnification, 500 µm; high magnification, 500 nm. d) A typical surfactant-free hexane–2 M H_2SO_4 (30:70, v:v) mixtures separation.

2. Results and Discussion

2.1. Direct Covalent Binding Hydrogel on Filter Paper

The hydrogel-coated filter paper is fabricated by a direct crosslinking reaction between cellulose filter paper and hydrophilic PVA, in which glutaraldehyde (GA) serves as a crosslinker and hydrochloric acid (HCl) as a catalyst (**Figure 1**a). The crosslinking reaction is triggered by an aldol condensation between aldehyde groups and hydroxy groups at room temperature. Owing to the native hydroxy groups on the cellulose filter paper and hydrophilic PVA, the reaction would not only makes cellulose molecule and PVA be, respectively, crosslinked with GA, but also allows for the co-crosslinking of the cellulose molecule and PVA.[26–28] Therefore, the process would result in the formation of the ether bond/acetal ring bridged multiple crosslinked networks (**Figure 2**b). This one-step fabrication of hydrogel-coated filter paper is exceedingly simple, low-cost, eco-friendly, and easily scale-up. The fabrication process is solely performed under gentle condition rather than a few specific procedures (such as UV photopolymerization coating mesh,[14] spraying steel mesh,[29] or needle punching membranes.[15] Figure 1b,c show a typical scanning electron microscopy (SEM) image of the uncoated filter paper and hydrogel-coated filter paper, respectively. The results show that PVA is successfully coated on the fiber of filter paper. Moreover, various hydrogel-coated filter paper can be readily prepared by varying the concentration and molecular weight of PVA (Figure S1, Supporting Information). Figure 1d shows a typical surfactant-free oil–2 M H_2SO_4 mixtures can be effectively separated by hydrogel-coated filter paper. The 2 M H_2SO_4 aqueous solutions can easily pass through the hydrogel-coated filter paper while the oil was retained, indicating the effective oil–water separation. Beyond that, the extreme oil–water environment, such as oil–8 M H_2SO_4, oil–10 M NaOH, and oil-saturated NaCl mixtures, can also be effectively separated by hydrogel-coated filter paper.

2.2. The Stability of Hydrogel-Coated Filter Paper in Highly Acidic, Alkaline, and Salty Environment

The resistance of acid, alkali, and salt test demonstrates that the hydrogel-coated filter paper is stable enough to tolerate highly acidic, alkaline, and salty environment. We show the high stability of hydrogel-coated filter paper is attributed to the formation of covalent bond bridged multiple crosslinked networks (Figure 2a). Filter paper is composed of cellulose. In original cellulose, there is a large amount of intra- and intermolecular hydrogen bonds which usually make it difficult to dissolve in common solvents. And thus, some of acid or alkali mediated hydrogen bonds breaking are employed for the dissolution of cellulose.[30,31] In general, the breaking of hydrogen bond would cause the frequency shift of O–H stretching vibration toward higher wave number.[32] Figure 2b shows Fourier transform infrared spectroscopy (FTIR) spectra of uncoated and hydrogel-coated filter paper. The absorption band at 3430 cm^{-1} corresponding to the O–H stretching vibration of cellulose molecule in hydrogel-coated filter paper shifted to a higher wave number in comparison

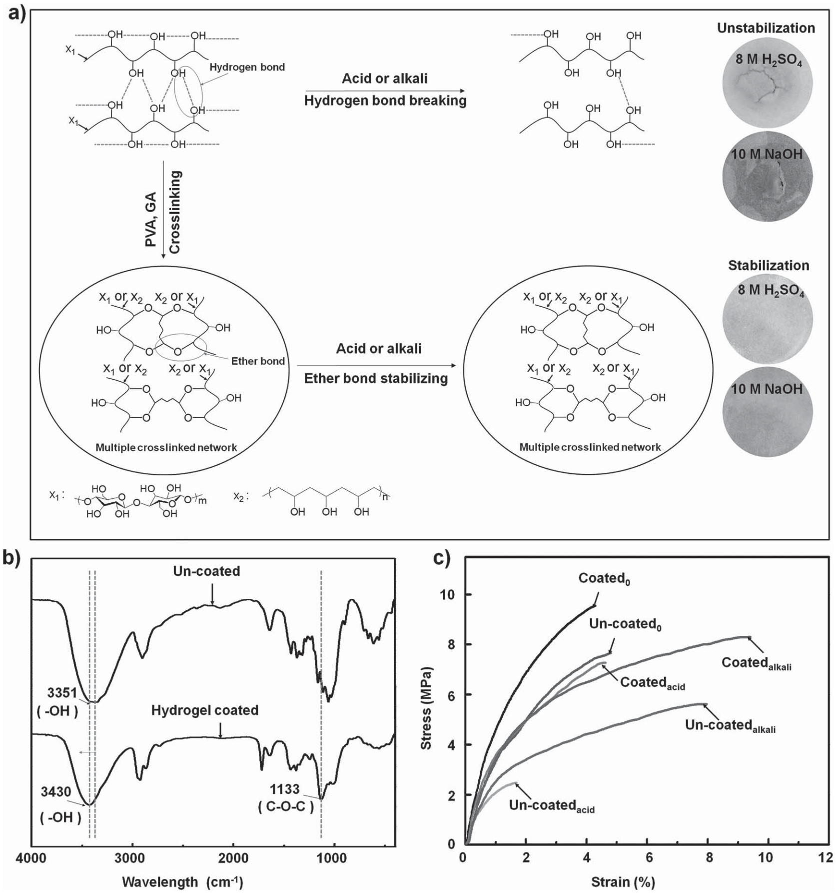

Figure 2. The stability of hydrogel-coated filter paper. a) Schematic of the stability of hydrogel-coated filter paper. Under highly acidic and alkaline environment, the intra- and intermolecule hydrogen bond in cellulose of uncoated filter paper can be broken so that it can significantly decrease the mechanical property. However, the crosslinking reaction between cellulose molecule and PVA forms ether bond bridged multiple crosslinked networks enabling the hydrogel-coated filter paper robust enough to resist high acid and alkali. And thus, the optical photograph shows the uncoated filter paper would be ruptured when the 8 M H_2SO_4 or 10 M NaOH aqueous solution pass through the filter paper, while the hydrogel-coated filter paper still remains intact. b) FTIR characterization of uncoated filter paper and hydrogel-coated filter paper. The absorption band of the O–H stretching vibration of cellulose molecule in hydrogel-coated filter paper obviously shifted to a higher wave number than that of cellulose in the uncoated filter paper, indicating the breaking of hydrogen bond. The new peak at 1135 cm^{-1} corresponds to the C–O–C stretching vibration in hydrogel-coated filter paper, suggesting the formation of ether bond. c) Stress–strain curves of untreated hydrogel-coated filter paper and hydrogel-coated filter paper treated with 8 M H_2SO_4 and 10 M NaOH for 10 min, respectively.

with 3351 cm^{-1} in cellulose of uncoated filter paper. Meanwhile, the band width at 3430 cm^{-1} assigning to O–H in cellulose of hydrogel-coated filter paper became sharper and narrower than that of uncoated filter paper. The strong new peak at 1133 cm^{-1} corresponds to the stretching vibration of C–O–C in hydrogel-coated filter paper. These results demonstrate the breaking of hydrogen bond and the forming of the ether bond in hydrogel-coated filter paper (Figure 2a). At room temperature, the ether bond is relatively stable and thereby it has the capacity to resist acid or alkali in a wide range.[33] Therefore, the direct crosslinking results in the breaking of hydrogen bond network of cellulose replacing with covalent bond bridged multiple crosslinked network, which is very crucial for the hydrogel-coated filter paper to tolerate highly acidic and alkaline environment.

In the highly acidic and alkaline environment, the original hydrogen bond network of cellulose would be quickly broken due to the formation of new hydrogen bond through the dissociative H^+ or OH^- and hydroxy groups of cellulose,[34] leading to the decrease of mechanical property of filter paper (Figure 2a,c). While for the hydrogel-coated filter paper, the covalent bond bridged multiple crosslinked networks would significantly enhance the resistance to high acid and alkali. Compared to the uncoated filter paper, the tolerating highly acidic, alkaline, and salty experiment indicates that the hydrogel-coated filter paper would not destruct when the hexane–8 M H_2SO_4 or hexane–10 M NaOH mixtures pass through it (Figure 2a). SEM images show that the morphology of hydrogel-coated filter paper nearly has no obvious change under these environments. Even immerged into 2 M H_2SO_4, 2 M NaOH, and saturated NaCl aqueous solutions for 30 d, the hydrogel-coated filter paper still remains intact (Figure S2, Supporting Information). The extremely stretchable results show that the hydrogel-coated filter papers are even more remarkable than the uncoated filter paper (Figure 2c). The uncoated filter paper and hydrogel-coated filter paper were immersed in an aqueous of 8 M H_2SO_4 or an aqueous of 10 M NaOH for 10 min and then rinsed with water and dried at room temperature. The hydrogel-coated filter paper can withstand 3 and 1.5 times more stress than the corresponding uncoated filter paper in 8 M H_2SO_4 and 10 M NaOH, respectively. Therefore, the hydrogel-coated filter paper has good stability to resist highly acidic, alkaline, and salty environment.

2.3. Wettability and Oil–Water Separation Capacity of Hydrogel-Coated Filter Paper in Highly Acidic, Alkaline, and Salty Environment

To demonstrate the oil–water separation capacity in highly acidic, alkaline, and salty environment, we first investigated the wettability and underwater oil-adhesion behaviors of hydrogel-coated filter paper. The formed hydrogel-coated filter paper shows superhydrophilicity with a contact angle (CA) of about 0° for 2 M H_2SO_4 in air and underwater superoleophobicity at different acidic, alkaline, and salty environment with CA larger than 150° (**Figure 3**a,c). The underwater (2 M H_2SO_4, 2 M NaOH, and saturated NaCl) superoleophobicity of hydrogel-coated filter paper is applicable to a wide variety of oils, including gasoline, diesel, hexane, petroleum ether, and toluene (Figure S3, Supporting Information). The underwater

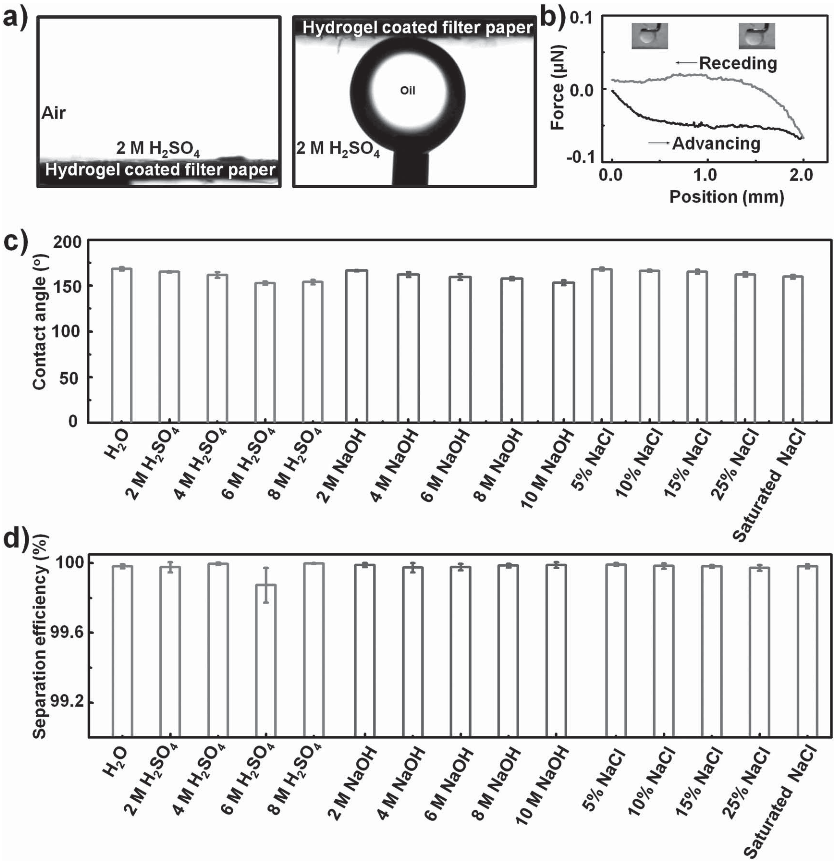

Figure 3. The wettability and oil–water separation capacity of hydrogel-coated filter paper in highly acidic, alkaline, and salty environment. a) Left, a photograph of a 2 M H_2SO_4 droplet on the hydrogel-coated filter paper in air with a contact angle (CA) of about 0°. Right, a photograph of an oil droplet (hexane, 2 μL) on the hydrogel-coated filter paper underwater with an oil CA 170° ± 1.8°. b) Underwater oil-adhesion force of hydrogel-coated filter paper. The as-designed hydrogel-coated filter paper has ultralow oil adhesion property. c) Underwater superoleophobicity of hydrogel-coated filter paper in different acidic, alkaline, and salty environment. d) Separation efficiency of the hydrogel-coated filter paper for the separation of hexane–water mixtures (30:70 v/v) in different acidic, alkaline, and salty environment. The results indicate much high oil–water separation efficiency (>99.9%).

oil-adhesion force was recorded by a high-sensitivity microelectromechanical balance system and the results suggested that the adhesion of oil on the hydrogel-coated filter paper was too low to be detected. No force drop was measured, and the corresponding photographs showed nearly no deformation and loss of the oil droplet (Figure 3b). These results demonstrated that the hydrogel coating can successfully reduce the affinity for oil droplets. When the hydrogel-coated filter paper immerged in water, water molecules can be trapped into the porous structures so that the oil droplets are repelled from the surface of hydrogel-coated filter paper.

Associating with the chemical stability, underwater superoleophobic and ultralow oil-adhesive properties of hydrogel-coated filter paper, we comprehensively carried out a series of oil–water separation experiments in highly acidic, alkaline, and salty environment. Owing to the superhydrophilic and underwater superoleophobic properties of hydrogel-coated filter paper, water permeated easily while the hexane was retained above the hydrogel-coated filter paper. The hexane–water mixtures at different acidic, alkaline, and salty environment were investigated and the results indicated much high separation efficiency (>99.9%), even at 8 M H_2SO_4, 10 M NaOH, and saturated NaCl environments (Figure 3d). The high separation efficiency in these environments implies a high stability of hydrogel-coated filter paper to resist highly acidic, alkaline, and salty environment. Beyond hexane, the separation of toluene–, petroleum ether–, diesel–, and gasoline–water mixtures at 2 M H_2SO_4, 2 M NaOH, and saturated NaCl aqueous solutions also indicated high separation efficiency (Figure S4, Supporting Information).

2.4. The Controlled Fabrication Mediated Oil–Water Separation and Recyclability of Hydrogel-Coated Filter Paper

The effects of various hydrogel-coated filter paper with different particle retention sizes of filter paper, the concentration of PVA and the molecule weight of PVA on separation efficiency were also studied. The hydrophilic property of PVA endows it with high water permeability, contaminants retaining ability, and oil resisting ability.[35] The good chemical, thermal, and mechanical stability of crosslinked PVA[36,37] makes it exhibit good acid, alkali, and salt resistance. Compared to the uncoated filter paper, the hydrogel-coated filter paper exhibited much higher oil resistance capacity, demonstrating PVA hydrogel covalent coating plays a decisive role in the effective oil–water separation (Figure S5, Supporting Information). The separation efficiencies of hydrogel-coated filter paper with different particle retention size are higher than 99.9% (**Figure 4**a–c). There is a slight decrease in the separation efficiency with increasing the particle retention sizes of filter paper (Figure 4a). The separation efficiency is proportional to the concentration and molecular weight of PVA, usually larger than 99.9% (Figure 4b,c). Moreover, the recyclability of hydrogel-coated filter paper demonstrates that the separation efficiency still maintains a relative high level (>99.9%). All the decline rates are less than 0.1% after 20 cycles (Figure 4d). The hydrogel-coated membrane remains clean and almost has no fouling (Figure S6, Supporting Information). In traditional view, filter paper is usually used to retain some solid particles in solutions and it encounters discard after one-time use. Here, this simple crosslinking process endows the hydrogel-coated filter paper with good stability, recyclability, and antifouling.

2.5. Surfactant-Stabilized Oil-in-Water Emulsion Separation

We have also employed our hydrogel-coated filter paper to separate surfactant-stabilized oil-in-water emulsions. The

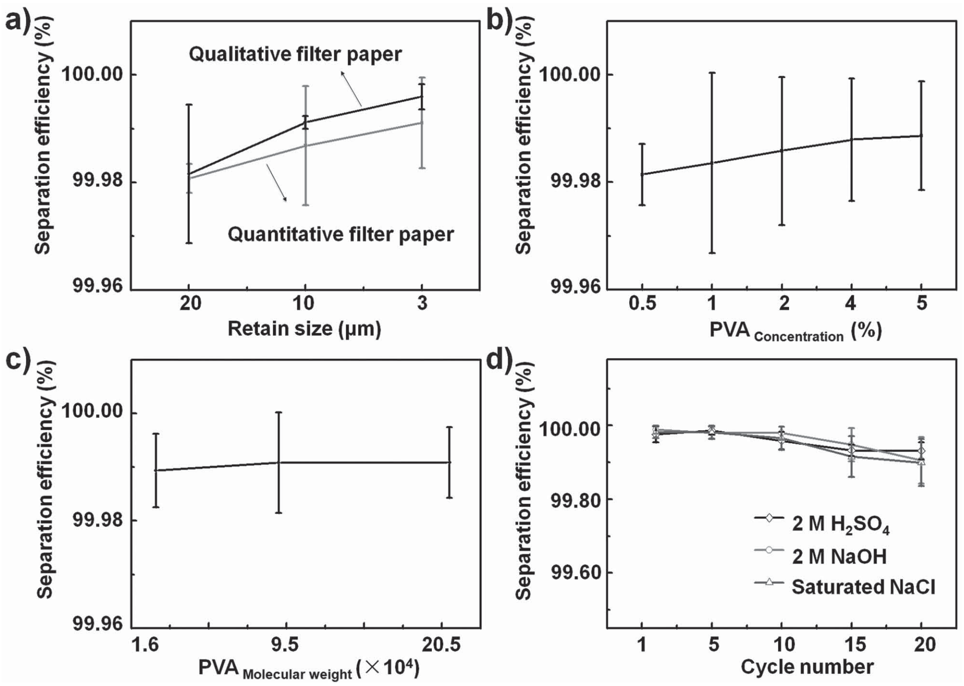

Figure 4. The controlled fabrication mediated oil–water separation and recyclability of hydrogel-coated filter paper. a) The separation efficiency of hexane–water mixtures (30:70 v/v) depends on the particle retention sizes of filter paper. b) The separation efficiency of hexane–water mixtures (30:70 v/v) depends on the concentration of PVA. c) The separation efficiency of hexane–water mixtures (30:70 v/v) depends on the molecular weight of PVA. d) The separation efficiency of hexane–2 M H_2SO_4/2 M NaOH/saturated NaCl aqueous mixtures (30:70 v/v) after 20 cycles. The separation efficiency nearly has no change after 20 cycles.

superhydrophilicity and underwater superoleophobicity, ultralow oil-adhesion, and densely interlaced structures of hydrogel-coated filter paper are very critical for the separation of oil-in-water emulsions. All types of surfactant-stabilized hexane-in-water emulsions, including anionic sodium dodecyl benzene sulfonate (SDBS), cationic cetyltrimethylammonium bromide (CTAB), and nonionic Tween 80 and PVA, are considered. The size distribution of hexane droplets in various surfactant-stabilized aqueous solutions shows that the highest number fraction of droplet diameters are 1–20 μm (**Figure 5**a). There are no demulsification or precipitation observed in the emulsion solutions when placed in ambient conditions for one week and the droplet sizes of the emulsions were still kept in the range of 1–20 μm as determined by optical microscopy, indicating good stability of the emulsions (Figure S7, Supporting Information). The separation apparatus is the same as the separation of surfactant free oil–water mixtures. The emulsion is added to the upper tube and the hydrogel-coated filter paper is pre-wetted by water in advance. The separation is performed solely driven by gravity. When the hexane-in-water emulsion was added into the separation apparatus, water-rich permeate through the hydrogel-coated membrane. Once the hydrogel coatings contacting with the oil droplets, water can be trapped in the rough structures and form an oil–water–solid composite interface.[38] The trapped water molecules will greatly decrease the contact area between oil droplet and solid surface, and thereby repelled the oil droplets from the solid surface. With a large number of water permeating away, the emulsions gradually become viscous, suggesting the occurrence of demulsification process. The process makes the surfactant-stabilized oil droplets aggregate and coalesce as the water passing through the hydrogel-coated filter paper. After separation, no oil droplets were observed in the collected water. The water-rich permeate through the hydrogel-coated filter paper while the hexane-rich are retained above the hydrogel-coated filter paper. The hydrogel-coated filter paper allows all types of surfactant-stabilized oil-in-water emulsions to separate completely, with >99% efficiency (Figure 5c), demonstrating the hydrogel-coated filter paper has the capacity to separate any surfactant-stabilized oil-in-water emulsions. Furthermore, to realize the effective separation of surfactant-stabilized emulsion in a wide range of droplet sizes, we prepare three SDBS-stabilized hexane-in-water emulsions with different droplets sizes which depend on the oil volume fractions. The volume fractions of oil in the mixtures are 10, 20, and 30 vol% hexane, which correspond to the droplet size ranging from 500 nm to 10 μm, 5–15 μm, and 10–25 μm (Figure S8, Supporting Information), respectively. These emulsions can be effectively separated by the hydrogel-coated filter paper. The separation efficiency of these emulsions is more than 99.9%, indicating a much high separation capacity of hydrogel-coated filter paper. The permeate flux as a very important parameter for oil-in-water emulsion separation is also considered. The fluxes of the membrane were calculated by measuring the time of almost completely permeating a certain volume of oil-in-water emulsions. By measuring the permeating time of 50 mL SDBS-stabilized oil-in-water (5:95, v:v) emulsions, the flux was determined. As shown in Figure 5d, the fluxes of 63, 56, and 43 L m^{-2} h^{-1} for SDBS/hexane/H_2O, SDBS/diesel/H_2O, and SDBS/gasoline/H_2O, are obtained, respectively. However, although no visible water is observed to

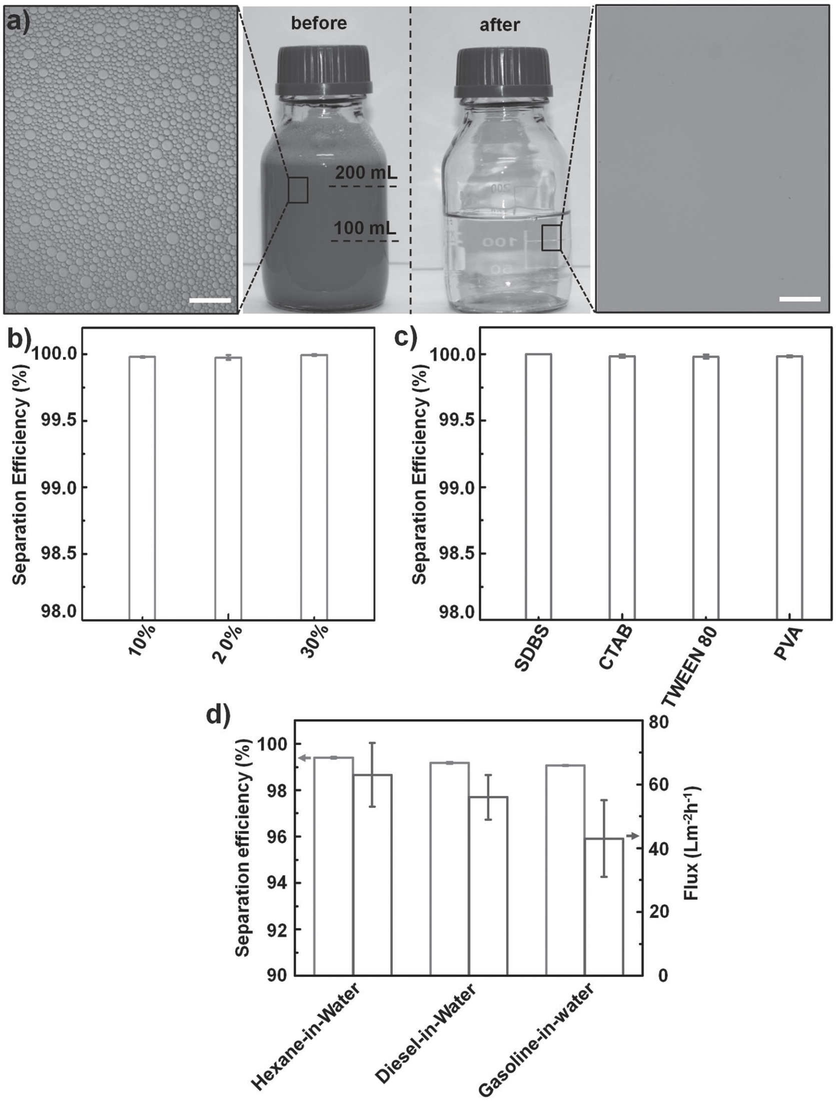

Figure 5. Separation of oil-in-water emulsion. a) The hexane-in-water emulsion before and after separating. Scale bar 100 µm. b) The separation efficiency of SDBS-stabilized hexane-in-water emulsions containing 10, 20, and 30 vol% hexane, respectively. c) The separation efficiency of various types of surfactant-stabilized hexane-in-water emulsions containing 10 vol% hexane. d) The separation capacity and flux of SDBS-stabilized hexane-in-water (5:95, v:v), SDBS-stabilized diesel-in-water (5:95, v:v), and SDBS-stabilized gasoline-in-water (5:95, v:v) emulsion.

permeate from the upper emulsion before calculating the flux, it still has a small amount of residual water in the emulsion because there is not enough pressure to maintain the water filtration under the gravity-driven separation.[38] Therefore, in our study, the retained water in emulsion is investigated by measuring the weight percentage of water. After nearly completely permeating of water, the weight fraction of retained water in the emulsion is about 3%, indicating an average of 97% of water is filtered under this gravity-driven separation.

To demonstrate the efficient separation capacity of hydrogel-coated filter paper, we further studied the intrusion pressure of hydrogel crosslinked filter paper for different oils, including toluene, petroleum ether, diesel, and gasoline. The result shows the intrusion pressure of hydrogel-coated filter paper for different oils is much higher (>5 kPa) than previous reported membrane,[14,15] implying a higher resistance to oil permeation (**Figure 6**). Therefore, the covalent bridged multiple crosslinked networks of hydrogel-coated filter paper provides stable structures which can effectively resist highly acidic, alkaline, and salty environment. The underwater superoleophobicity, ultralow oil adhesion, and high intrusion pressure of hydrogel-coated filter paper ensure effective retainment for oil. Thus, taking advantage of a simple crosslinking process, we achieved the effective oil–water separation in extremely complex environment.

3. Conclusion

In summary, we have developed a simple strategy for fabricating hydrogel-coated filter paper having the capacity to separate oil–water mixtures in highly acidic, alkali, and salty environment and surfactant-stabilized oil-in-water emulsions. The resulting multiple crosslinked networks have proved to be robust to resist highly acidic, alkaline, and salty solutions. Consequently, the hydrogel-coated filter paper can separate various oil–water mixtures in a range of highly acidic, alkaline, and salty environment, with >99.9% efficiency. Meanwhile, the hydrogel-coated filter paper can also achieve effective oil-in-water emulsion separation. The hydrogel-coated filter paper has the capacity to separate all types of emulsions with different droplet sizes.

4. Experimental Section

Materials: PVA with a number-average molecular weight of 16 000 (98% hydrolyzed), 95 000 (95% hydrolyzed), and 205 000 (86.7%–88.7% hydrolyzed) were obtained from J&K, China. GA aqueous solution (50 wt% in water) was obtained from J&K, China. Diesel and gasoline were obtained from China National Petroleum Corporation, China. HCl (37 wt%, extra pure grade), hexane, petroleum ether, and toluene were obtained from Beijing Chemical Works, China. Quantitative filter paper and qualitative filter paper with particle retention size ≈20, 10, and 3 µm were obtained from Hangzhou Whatman-Xinhua Filter Paper Co., Ltd., China. The filter paper was dried under vacuum at 35 °C for 24 h before use.

Fabrication of Hydrogel-Coated Filter Paper: The hydrogel-coated filter paper was fabricated by a direct crosslinking reaction between cellulose filter paper and hydrophilic PVA, in which GA serves as a crosslinker and HCl as a catalyst. In a typical synthesis, 4% w/v PVA solutions were prepared by dissolving PVA powder in deionized water at 95 °C. After cooling to room temperature, the pH of the PVA aqueous solutions was corrected to 2 with HCl. Then the filter paper was immersed into 20 mL of PVA solutions at ambient temperature for 2 h, allowing for the PVA solutions completely wetting the filter paper. Subsequently, the PVA wetted filter paper was taken out and again immersed into 20 mL of

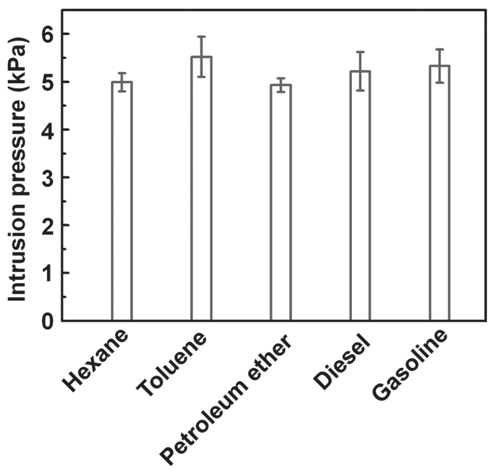

Figure 6. The intrusion pressures of hydrogel-coated filter paper supporting different oils. The intrusion pressure of hydrogel-coated filter paper for different oils is much higher than our previous reported membrane, implying it has the higher resistance to oil permeation during separation process.

GA solutions at ambient temperature for 24 h. Finally, the obtained PVA coated filter paper was washed in water and ethanol several times and then dried under vacuum before use.

Contact Angle, Oil-Adhesion Forces, and Intrusion Pressures Measurement: All measurements of contact angle (in air and under water) were conducted using an OCA20 machine (Data-Physics, Germany) at ambient temperature. The oil droplets (2 μL) were dropped onto the surface of hydrogel crosslinked filter paper, which was immersed in water. The average value of at least three measurements performed at different positions on the same sample was adopted as the CA.

The oil-adhesion forces were measured using a high-sensitivity microelectromechanical balance system (Data-Physics DCAT11, Germany) underwater. An oil droplet (1,2-dichloroethane, 10 μL) was suspended with a metal cap and was programmed to contact with the surface to the hydrogel crosslinked filter paper surface at a constant speed of 0.005 mm s^{-1} and then to leave.

The intrusion pressure of oils flowing through the hydrogel crosslinked filter paper was characterized by measuring the maximum height of oil that the hydrogel-coated filter paper can support. The experimental intrusion pressure (P_{exp}) values were calculated from the following equation

$$P_{\text{exp}} = \rho g h_{\text{max}} \tag{1}$$

where ρ is the density of the oil, g is acceleration of gravity, and h_{max} is the supportive maximum height of oil on hydrogel-coated filter paper. Before measuring, the hydrogel-coated filter paper was wetted by water in advance.

Oil–Water Separation in Highly Acidic, Alkaline, and Salty Environment: The as-prepared hydrogel crosslinked filter paper was fixed between two glass tubes. The volume of upper tube was 300 mL while the below conical flask was 250 mL. The oil–water mixtures were poured onto the as-prepared hydrogel crosslinked filter paper.

Oil-in-Water Emulsions Separation: Hexane-in-water emulsions with different oil content (hexane:water = 5:95, 10:90, 20:80, and 30:70 v:v) were prepared by mixing 0.1% w/v of SDBS of water and hexane using a stir bar (at 1500 rpm) at room temperature for 16 h.

To investigate the separation capacity of hydrogel crosslinked filter paper for different types of surfactant-stabilized oil-in-water emulsions, 0.1% w/v of SDBS of water and hexadecane (hexane:water = 30:70 v:v) were mixed using a stir bar (at 1500 rpm) at room temperature for 16 h.

The obtained hydrogel-coated filter paper was fixed between two glass tubes. The volume of upper tube was 300 mL and the volume of below conical flask was 250 mL. The surfactant-stabilized oil-in-water mixtures with different volume ratio were poured into the as-prepared hydrogel crosslinked filter paper.

The separation efficiency was characterized by measuring the oil concentration of the original oil–water mixtures using an infrared spectrometer oil content analyzer (CY2000, China). CCl_4 was used to extract oils from water. The absorbance at 2930, 2960, and 3030 cm^{-1} were measured. The oil content was obtained by calculating the absorbance and the correction coefficient.

The separation efficiency was calculated by oil rejection coefficient (R(%)) according to

$$R(\%) = \left(1 - \frac{C_p}{C_o}\right) \times 100 \tag{2}$$

where C_p and C_o are the oil concentration of the collected water and the original oil–water mixtures after one time separation, respectively.

Supporting Information

Supporting Information is available from the Wiley Online Library or from the author.

Acknowledgements

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