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REVIEW

Advanced Electrospun Nanofibrous Materials for Efficient Oil/Water Separation

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Abstract

The frequent occurrence of crude oil leakage accidents and the massive discharge of industrial oily wastewater not only caused huge damage and pollution to the ecosystem but also wasted a lot of precious resources. Therefore, it is urgent to solve the worldwide problem of oil/water separation. As a leader in advanced fiber materials, nanofibrous materials prepared by electrospinning have the advantages of high permeability, high separation efficiency, large specific surface area, adjustable wettability, simple preparation process, and low cost, making it attracted more attention of researchers in oil/water separation. This article mainly reviews the recent progress of various electrospun nanofibrous materials for oil/water separation field. The preparation and synthesis of nanofibrous adsorbents, nanofibrous membranes, and nanofibrous aerogels in recent years based on different applications, design principles, and separation approaches are systematically summarized. Finally, this review discusses the challenges and future development directions in oil/water separation.

Keywords Oil/water separation · Electrospinning · Nanofibrous adsorbents · Membranes · Aerogels

Abbreviations

3D	Three-dimensional
ACNTs	Acid treated carbon nanotubes
BAF-a	Bifunctional benzoxazine
CFMHF	Carbon fiber membrane
DA	Dopamine
DMF	N, N-dimethylformamide
F-PS	Foam-expanded polystyrene
FIBER	Isotropically-bonded elastic reconstructed
GSH	Thiolated graphene
MA	Myristic acid
NFAs	Nanofiber-based aerogels
OCA	Oil contact angle
P2VP	Poly(2-vinylpyridine)
P4VP	Poly(4-vinylpyridine)
PAA	Polyacrylic acid
PAN	Polyacrylonitrile
PDA	Polydopamine
PFTS	1H, 1H, 2H, 2H-perfluorooctyltriethoxysilane
PI	Polyimide
PMMA-*b*-PNIPAAm	Poly(methyl methacrylate)-block-poly(N-isopropylacrylamide)
PP	Polypropylene
PS	Polystyrene
PSBR	Polystyrene polybutadiene rubber
PU	Polyurethane
PVA	Polyvinyl alcohol
PVC	Polyvinyl chloride
PVDF	Poly (vinylidene fluoride)
SiNFs	Silicone nanofilaments
TCMS	Trichloromethylsilane
THF	Tetrahydrofuran
$Ti(OBu)_4$	Tetrabutyl orthotitanate

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UECS	Ultra-light electrospun cellulose sponge
WCA	Water contact angle
ZIF-8	Zeolitic imidazolate framework-8

Introduction

During the development and transportation of oil, the frequent occurrence of oil spills causes a lot of energy loss and ecological pollution [1–3]. Additionally, industrial wastewater from many industries such as papermaking, textile processing, and food also cause serious pollution. Therefore, researchers pay more attention to the treatment of oily wastewater and environmental protection issues [4–6]. Statistics show that a large amount of oily wastewater are discharged into rivers, lakes, and the ocean [7–9]. After oil flows into the sea, a series of complex changes occur, such as diffusion, evaporation, dissolution, emulsification, microbial oxidation, photochemical oxidation, and sedimentation. Although there are differences in size and sequence of these changes, most processes are carried out interactively [10, 11]. As a result, oil spills have a catastrophic impact on the environment and humans, and it increases the difficulty of treating oily wastewater. The current oil spill treatment methods (such as skimming and ultrasonic separation) have the disadvantages of high energy consumption, low separation efficiency, and poor recycling performance, which cannot meet the demands of complex environment [12, 13]. Therefore, developing efficient oil/water separation methods is urgent.

As a simple and efficient spinning technology, electrospun fibrous materials from various polymer solutions has been widely used in many fields [14–16]. Compared with other preparation methods of fibrous materials, electrospinning has the advantages of simple and efficient preparation process, low cost, and controllable fibrous structures [17–19]. In the spinning process, the metal needle is used as the spinneret and conductive device. When the voltage is applied, the charge accumulates on the surface of the solution, and the repulsive force is generated when the solution reaches the same polarity. When the charge repulsion force is bigger than the surface tension of the solution, a Taylor cone is generated, and the solution jet is ejected. Then, the solvent evaporates while the jet travels towards the collector, and fibers are deposited on the receiving plate [20–22]. The fibers obtained by electrospinning usually have large specific surface areas and the structure and chemical components of fibers are easy to control. At present, electrospun nanofibers have been used and developed into functional materials in the fields of filtration [23, 24], biomedicine [25, 26], electricity [27–29], photocatalysis [30, 31], and so on.

With the in-depth research, the special wettable nanofibrous materials can make oil or water freely penetrate through the materials because of their different wettability [32]. Therefore, it plays a great role in the field of the separation of oily wastewater. Using materials with the selectivity of oil/water for separation is a highly promising separation method [33–35]. Materials with different selectivity for water and oil (such as hydrophobicity and lipophilicity) can be obtained by adjusting the structure and components of the solid surface for oil/water separation. Our group has made some achievements in special wettable fibrous membranes. We have prepared fibrous membranes by melt blow spinning, solution blow spinning, and electrospinning method, and modified the membranes by dip coating, hydrothermal method, etc. Then, the hydrophobic/lipophilic membranes achieved with excellent performance in oil/water separation flux and separation efficiency [4, 5, 36]. Recently, electrospinning has been widely used as an efficient fiber preparation technology, which can effectively prepare multifunctional nanofibrous materials with controllable components and structures [37, 38]. In addition, the properties of the material can be improved through the composite of multiple components, such as porous structure and high surface area ratio [38–40]. Thus, electrospinning provides more methods for preparing special wettable materials.

In this review, we summarize the latest developments in the design, preparation, and application of nanofibrous materials by electrospinning in oil/water separation. Based on the different uses of electrospun nanofibrous materials, this review is divided into three main research directions: nanofibrous adsorbents (including polymer adsorbents, composite adsorbents, and biomass adsorbents) for the cleanup of the oil spill, nanofibrous membranes (including hydrophobic-lipophilic membranes, hydrophilic-oleophobic membranes, and switchable wettability membranes) for the separation of oil/water mixture, and nanofibrous aerogel for the separation oil/water emulsion (Fig. 1). We discuss the recent related

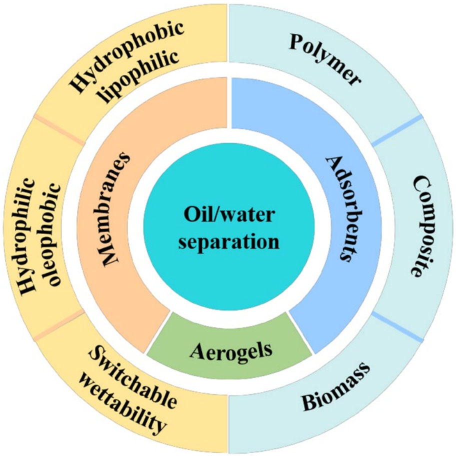

Fig. 1 Electrospun nanofibrous materials for oil/water separation

work, focusing on the optimal design and properties of these materials. Last of all, the challenges and development prospects of electrospun nanofibrous separation materials are discussed.

Theoretical Basis

Wetting Phenomenon and Models

The wettability is an important characteristic of the oil/water separation functional materials, and the state of wettability can be expressed by the contact angle. The free energy and the rough structure are two main factors affect the wettability of the surface, so the surface with special wettability can be constructed by changing the two factors [41]. For smooth surfaces, the free energy and roughness of the material surface can determine its wettability. As an important parameter to measure the wettability of a solid surface, the contact angle can be expressed by the Young's equation (Fig. 2a) [42].

$$\cos\theta = \frac{\gamma_{SV} - \gamma_{SL}}{\gamma_{LV}} \tag{1}$$

Among then, θ represents the contact angle under the Young's model, and γ_{SV}, γ_{SL}, and γ_{LV} are the surface tensions of the solid–vapor, solid–liquid, and liquid–vapor interfaces, respectively. Young's model describes the wettability of an ideal solid surface. However, many actual solid surfaces are rough and defective. To clarify the influence of roughness on the contact angle between the surface and droplet of the material, the Wenzel model was proposed. In fact, the liquid can fully contact with the solid rough surface and penetrate into the grooves, and it can be expressed by the Wenzel equation (Fig. 2b) [43]:

$$\cos\theta_W = r\cos\theta \tag{2}$$

Among them, r represents the roughness factor (≥ 1), which is the ratio of the actual area to the apparent area, and θ_W represents the apparent contact angle. The analysis shows that the roughness will enhance the hydrophilic or hydrophobic of the surface. When the roughness of the solid surface increases, the surface ($\theta_W < 90°$) becomes more hydrophilic. Conversely, the hydrophobic surface ($\theta_W > 90°$) becomes more hydrophobic with the increase of the roughness.

In the Wenzel model, the groove is filled by the droplet completely. However, when there are some small protrusions on the surface and air enters the grooves, the Wenzel model cannot explain it well. Therefore, Cassie–Baxter proposed a new theoretical model to demonstrate the phenomenon, and it can be explained by the following equation (Fig. 2c) [44]:

$$\cos\theta_C = f_{SL}\cos\theta_{SL} + f_{LV}\cos\theta_{LV} \tag{3}$$

where θ_{SL} represents the contact angle of the solid–liquid phase, θ_{LV} is the contact angle of the liquid–vapor phase, f_{SL} and f_{LV} are the surface fraction of the solid–liquid phase and liquid–vapor phase, respectively. Obviously, θ_C is increased relative to the smooth surface of the same material composition due to gas filling.

When the surface of the material is rough, a little liquid added to the droplet will make the contact angle increase, which is called the advancing contact angle. If a small amount of liquid is taken out, the droplet becomes flatter without moving the perimeter, and the contact angle becomes smaller which is called the receding contact angle. In addition, the advancing angle and receding angle exist at the same time while the surface is oblique. Furthermore, the actual wetting condition is very complicated

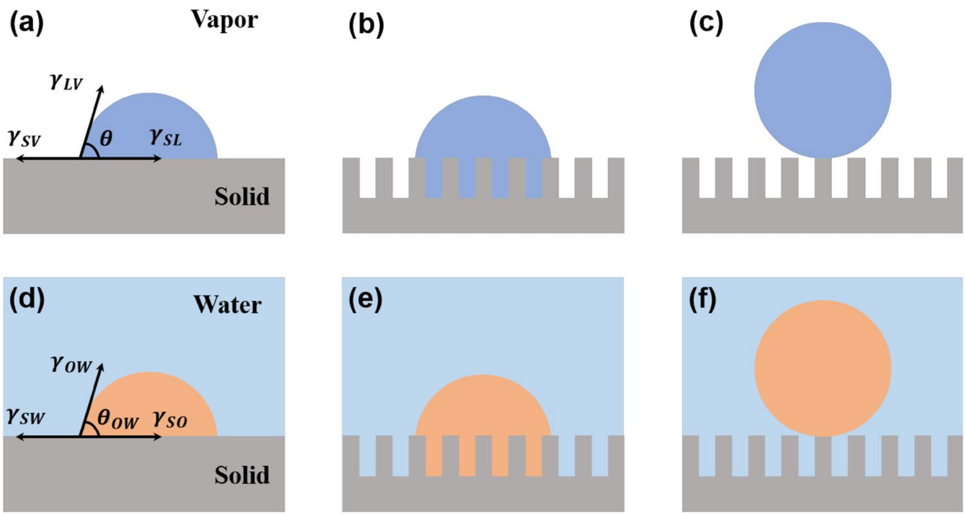

Fig. 2 The wetting models of a liquid droplet on the solid surface in the air: **a** Young's model, **b** Wenzel's model, and **c** the Cassie–Baxter model. The wetting models of a liquid droplet on the solid surface under water: **d** Young's model, **e** Wenzel's model, and **f** the Cassie–Baxter model

and there are many factors can influence it. Therefore, the above-mentioned models are not sufficient to explain all the wetting states.

While oil droplets are deposited on underwater surfaces, a triple interfacial phase among solid, water, and oil occurs. The following Young's equation can explain the behavior (Fig. 2d) [45]:

$$\cos\theta_{OW} = \frac{\gamma_{SW} - \gamma_{SO}}{\gamma_{OW}} = \frac{\gamma_{OA}\cos\theta_O - \gamma_{WA}\cos\theta_W}{\gamma_{OW}} \tag{4}$$

where γ_{SW}, γ_{SO}, and γ_{OW} are the surface tension of solid-water, solid-oil, and oil–water, respectively. θ_O and θ_W are the contact angle of oil and water in air, respectively. The Wenzel and Cassie–Baxter equation can also express the underwater oil contact angle (Fig. 2e, f) [46]:

$$\cos\theta_{OW(W)} = r\cos\theta_{OW} \tag{5}$$

$$\cos\theta_{OW(C)} = f\cos\theta_{OW} + f - 1 \tag{6}$$

where $\theta_{OW(W)}$ and $\theta_{OW(C)}$ are the Wenzel's and Cassie–Baxter's underwater contact angle, respectively. r is the roughness of the surface, and f represents the fraction of the surface contacted with oil.

Mechanism of Oil/Water Separation

If the material is used for oil/water separation, the oil and water need expressing different wettability on the solid surface. When there is a pressure difference on both sides of the membrane, the material become wet due to a specific phase in the oil/water mixture, thus achieving the selective moving of oil or water. Moreover, pore size and intrusion pressure are two important factors to consider in oil/water separation. Intrusion pressure (ΔP_C) represents the maximum static pressure that the material can resist, as demonstrated in Young–Laplace formula [47]:

$$\Delta P_C = -\frac{2\gamma_L\cos\theta}{R} \tag{7}$$

where γ_L is the surface tension of the liquid, θ represents the contact angle of liquid from inside, and R is the radius of the pore. When $\theta < 90°$, the liquid can penetrate to the pores of the material. However, a certain pressure is needed to allow the droplets to penetrate the material when $\theta > 90°$ [48]. To separate water and oil selectively, it must be ensured that the intrusion pressure of one phase of is positive, and the other phase of oil/water mixture has a high intrusion pressure. Moreover, the wettability of the solid material can be controlled by adjusting the surface tension of the materials.

Oil/water mixtures are normally separated by the materials with selective wettability. The interface of solid–water–oil three-phase occurs when a special wettable material is used for oil/water separation. For hydrophobic-lipophilic materials, the oil droplets can penetrate to the solid surface quickly, and squeeze the air out of the grooves. The oil are penetrate the grooves and repel the water phase, making the material remain hydrophobicity [49]. In addition, the excellent adsorption capacity for oil is also attributed to the capillary force generated by the micro-nano structure of the surface, which can make the oil/water mixture effectively separated. In addition, when the ΔP of the lipophilic surface is negative, the surface can be quickly wetted by the oil. The hydrophilic surface in the air usually exhibits oleophobicity underwater because of the surface tension of oil is lower than water. Therefore, a superhydrophilic and underwater oleophobic surface can allow water to pass through, then a water layer formed and repel oil to removing water. Moreover, superhydrophilic and underwater superoleophobic materials express better oil/water separation performance because they are not easily contaminated by oil. Meanwhile, the separation of emulsions can also be achieved by sieving or demulsification of the above two separation materials. When sieving is used to separate emulsions, the aperture of the superwetting film needs to be smaller than or proportional to the size of the emulsion droplet. The surfaces with superhydrophobic/superoleophilic properties are commonly used to separate water-in-oil emulsions, while water-removing separation materials are commonly used to treat oil-in-water emulsions. In general, the separation efficiency of the emulsion through screening will be lower than that of immiscible oil/water mixtures due to the smaller pore size and reduced flow rate.

Nanofibrous Adsorbents for the Cleanup of Oil Spill

Oil spill adsorption materials stand out by their simplicity and high efficiency and have developed into a key research object in the field of oil spill emergency treatment technology [50–52]. At present, most of the oil adsorption materials in the market are melt-blown polypropylene (PP) nonwovens. However, due to the relatively thick diameter of fibers and the low porosity of the material, it has the disadvantages of low oil absorption and poor oil/water selective wettability [53]. Therefore, the design and preparation of floating oil adsorption materials with a high oil absorption ratio and high oil/water selective wettability have important practical and economic significance. Many factors are affecting the adsorption rate and dynamic equilibrium of adsorption materials, mainly including surface energy, porosity, specific surface area, viscosity, and surface energy of oils [54, 55]. In the first place, to realize oil spill adsorption, the oil spill adsorption material should be hydrophobic-lipophilic

[56]. Secondly, porosity not only affects the oil adsorption efficiency of oil spill adsorption materials but also affects oil absorption [57]. In addition, the specific surface area of oil spill adsorption material directly determines the contact area between oil spill adsorption material and oil, and has a great impact on oil absorption rate and adsorption capacity. Therefore, an excellent oil spill adsorption material should have reasonable porosity and specific area structure while having appropriate surface energy [58, 59]. Recently, electrospun nanofibrous adsorbents exhibit excellent oil absorption capacity because of the high specific surface area and high porosity [60, 61]. By adjusting the physical and chemical structure of electrospun fibrous materials, preparing oil absorbent materials with a high oil absorption capacity, and great oil/water selective wettability are expected to play an important part in oil spill treatment. Compared with PP nonwovens, electrospun nanofibrous adsorbents show better adsorption capacity, but they are still limited. A large amount of nanofibrous material is consumed in cleaning up the oil spill, causing serious economic problems, and recovery of oil from adsorbents is also a challenge.

Polymer Adsorbents

At present, preparing high molecular polymers into nanofiber adsorbents by directly electrospinning has become an effective method for absorbing oil spills [62, 63]. An important parameter for oil spill cleaning is the oil/water selectivity of the adsorbents. The hydrophobic-lipophilic porous fibrous material prepared by the electrospinning exhibits excellent oil absorption performance, and the oil absorption capacity can be several times that of PP nonwoven fabric. Furthermore, the structure and adsorption performance of the fiber can be controlled by adjusting the composition and concentration of the spinning solution.

Polystyrene (PS), as a polymer with good spinnability and fiber-forming properties, is widely used in the preparation of superhydrophobic materials [64]. PS fibers with different micro-nano structures can be prepared in one step by adjusting the properties of the PS solution and spinning parameters [65]. Isik et al. adjusted the morphology and porosity of PS fibers to obtain porous foam-expanded polystyrene (f-PS) fibers with the best adsorption capacity (Fig. 3a, b) [66]. The morphology of the fibers can be regulated by adjusting the ratio of N, N-dimethylformamide (DMF): tetrahydrofuran (THF) and the concentration of the solution. The experimental results show that the fiber membrane obtained by electrospinning PS dissolved in a mixture of DMF: THF = 1:3 has the highest porosity. By observing the state of water droplets and oil droplets on the top of fibrous membrane, it can be concluded that the f-PS fiber membrane exhibits good hydrophobicity with a water contact angle (WCA) of

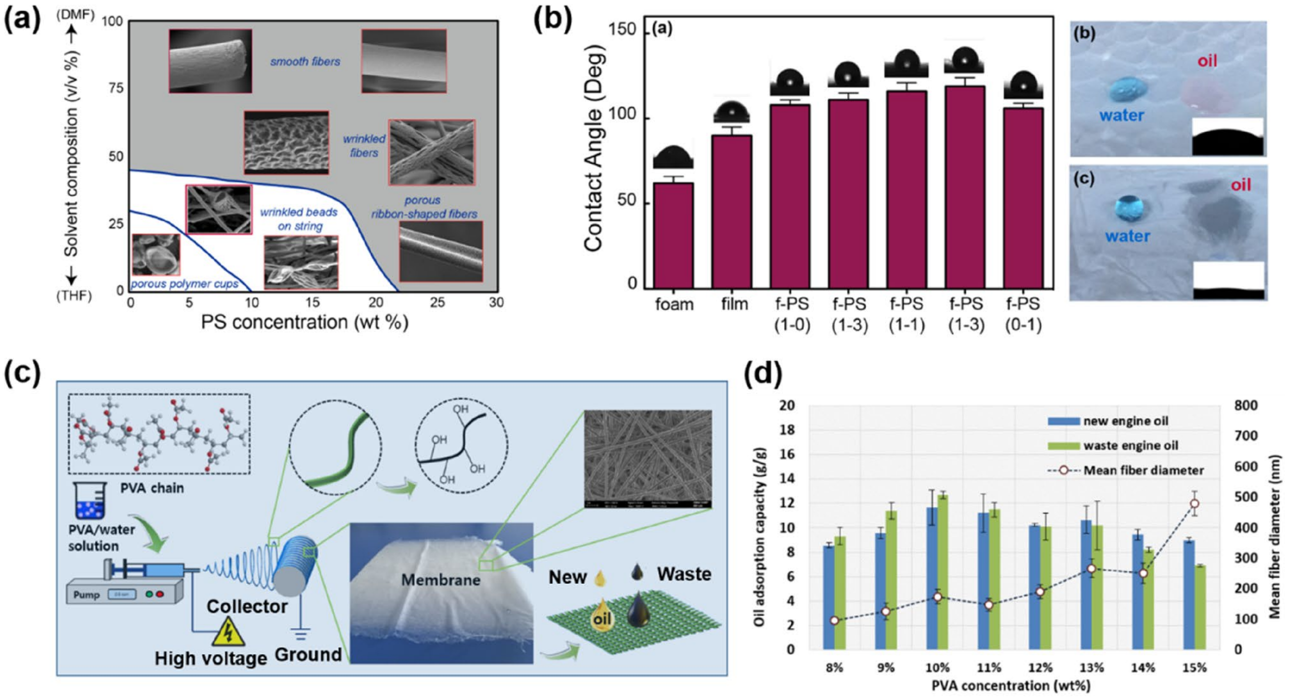

Fig. 3 **a** Effects of DMF:THF ratio and f-PS solution concentration on fiber morphology. **b** Water contact angle of f-PS under different DMF:THF ratio and the digital image of water and oil droplets on the top of f-PS fibrous sorbent. Reproduced with permission of Ref. [66], Copyright of ©2018 Elsevier. **c** Preparation process of PVA fibrous membrane. **d** Column of the relationship between PVA solution concentration, fiber diameter and adsorption capacity. Reproduced with permission of Ref. [67], Copyright of ©2021 MDPI

120° and lipophilicity with oil contact angle (OCA) of 10°, and has a good adsorption capacity of 124 g g^{-1}. Moreover, PVA has the advantages of degradability, low cost, good lipophilicity, etc. The PVA fibrous membrane with excellent adsorption capacity can be produced by electrospinning by adjusting the concentration of the PVA solution (Fig. 3c, d) [67]. In addition, there are many high molecular polymers prepared by direct electrospinning into fibrous materials for oil spill treatment which all show good adsorption properties, such as polyimide (PI) [68], polyvinyl chloride (PVC) [69], and poly (vinylidene fluoride) (PVDF) [70].

Composite Adsorbents

Although the fibrous membrane obtained by direct electrospinning is simple, the performance of the obtained membrane cannot meet the more complex requirements [71]. For example, the reusability of PS fibrous adsorbent is poor, and the strength of the fiber decrease after oil adsorption. Therefore, combining a single-component polymer material with other organic or inorganic materials to form a composite material by electrospinning can selectively enhance the hydrophobicity, mechanical strength, and chemical resistance of the composite material [72, 73]. Not only multi-component composite can be carried out in solution configuration, but also single-component fibers can be post-processed to composite with other materials, such as dip coating, spray coating, in-situ growth, etc. The compounding makes the material more applicable and has a wider range of applications. When selecting the combined components, adding polyurethane (PU) [74], PVC [75], and PVDF [76] as reinforcing agents can improve the mechanical properties and durability of single-component fibers. In addition, some inorganic materials (such as SiO_2 and Fe_3O_4) can be added to increase the strength, hydrophobicity, and recyclability of the fiber [77, 78].

Directly electrospinning after preparing two or more components into a mixed solution is a simple and effective preparation method. Akanbi et al. prepared PS and PU hybrid nanofibrous sorbent by electrospinning, which enhanced the mechanical strength and adsorption properties of the fibers (Fig. 4a) [74]. The adsorption capacity used to adsorb motor oil and sunflower oil are 144.52 g g^{-1} and 110.89 g g^{-1}, respectively. In addition, polymer materials can also be combined with inorganic nanoparticles. Ding et al. obtained a superhydrophobic composite fiber by electrospinning a mixed solution of PS and SiO_2 nanoparticles with WCA of 153° (Fig. 4b, c) [79]. It shows excellent adsorption performance for a variety of oils with an adsorption capacity of 122.7 g g^{-1}, which is much higher than commercial PP fiber adsorbents (Fig. 4d). Furthermore, it is also possible to combine organic and inorganic substances by other methods. Gao et al. prepared SiO_2/PVDF composite fibrous membranes by a combination of electrospinning and electrostatic spraying (Fig. 4e) [80]. The addition of SiO_2 microspheres increases the rough structure of the fibers, and increase the hydrophobicity of the membrane surface. Moreover, the composite adsorbent can successfully separate oil in corrosive environments, including acid, alkali, and salt solutions (Fig. 4f).

Biomass Adsorbents

Biomass materials is the most abundant renewable resource on the earth and has the advantage of excellent environmental friendliness [81, 82]. However, a large amount of biomass materials not been effectively used. In order to make full use of biomass resources, researchers are committed to developing biomass adsorbents with special wettability, which can selectively adsorb oil pollutants [83]. Because of the chemical stability, good biodegradability, and biocompatibility of cellulose, it is considered to be a good choice for the preparation of oil/water separation materials [84–86].

Cellulose is the most abundant renewable organic polymer resource in nature, and it has been widely used in nanomedicine, medicine, energy, environment, biology, and agriculture. Further effective use of cellulose resources and expansion of its applications are the hotspots of domestic and foreign research. Xu et al. prepared an ultra-light electrospun cellulose sponge (UECS) by electrospinning and freeze-drying (Fig. 5a) [56]. The three-dimensional (3D) cellulose sponge exhibits good hydrophobicity (WCA = 141.2°) and compressibility (Fig. 5b, c). Therefore, this sponge has excellent adsorption capacity for various oils and organic solvents, and the adsorption capacity can reach 232 times of its own weight.

There are some common cellulose materials in our daily life, including cotton, straw, and wood, etc. Recently, Yang et al. used natural loofah to prepare a 3D ultra-light electrospun fibrous sponge [87]. First, polyacrylonitrile (PAN) nanofibers were prepared by electrospinning, and the chopped electrospun PAN nanofibers and natural loofah were bonded through polyvinyl alcohol (PVA) to obtain a uniform suspension. Then, a hydrophobic 3D nanofiber sponge was obtained by freeze-drying. During the preparation process, the content of the loofah was adjusted to construct a spider web structure with enhanced adsorption capacity (Fig. 5d). As shown in Fig. 5e, f, the obtained PAN/loofah ultra-light sponge can effectively adsorb various oils.

In summary, electrospun nanofibrous absorbents greatly improves the efficiency of cleaning up oil spills. Compared with other materials such as metal materials [88] and powder materials [89], it has the advantages of adjustable structure, recyclable using, and high adsorption performance, which provides a more effective method for oil spill cleanup.

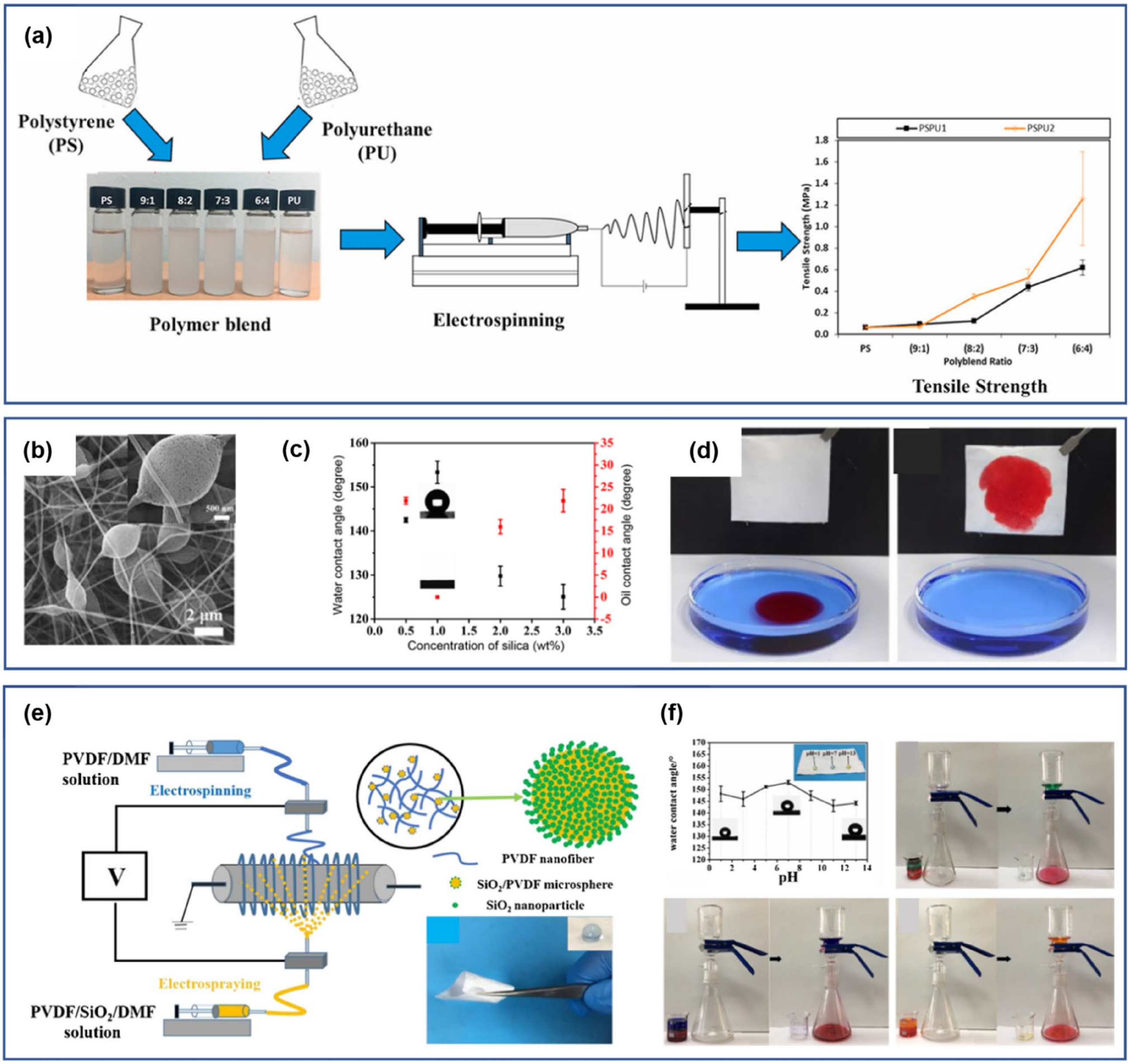

Fig. 4 **a** Preparation of PS/PU composite fibers and the diagram of tensile strength test. Reproduced with permission of Ref. [74], Copyright of ©2021 Elsevier. **b** SEM images of PS/SiO_2 composite nanofibers. **c** The influence of SiO_2 content on the water contact angle of PS/SiO_2 composite fibrous sorbent. **d** Photograph of PS/SiO_2 composite fiber membrane adsorbing soybean oil. Reproduced with permission of Ref. [79], Copyright of ©2019 The Korean Fiber Society. **e** Preparation diagram of SiO_2/PVDF composite fibers. **f** Water contact angle of SiO_2/PVDF membrane under different pH, and the separation of chloroform/various corrosive solutions. Reproduced with permission of Ref. [80], Copyright of ©2018 Korean Society of Industrial Engineering Chemistry

Nanofibrous Membranes for the Separation of Oil/Water

Currently, there are three types of the oil/water separation methods: chemical method, biological method, and physical method [12, 13, 90]. The advantages and disadvantages of different separation methods are shown in Table 1. Due to the advantages of simple operation, low energy consumption, wide applicability, and high separation efficiency, membrane separation is considered as the most promising method for treating oil/water mixture or emulsion among these methods [91–93]. The main factors affecting the permeation flux of the oil/water separation efficiency of fibrous membranes are the wettability, porosity, and thickness of the fibrous membrane. By controlling the geometric structure and chemical composition of the membrane surface, fibrous membranes with different infiltration properties ca n be selectively prepared [94–96]. Generally, these separation

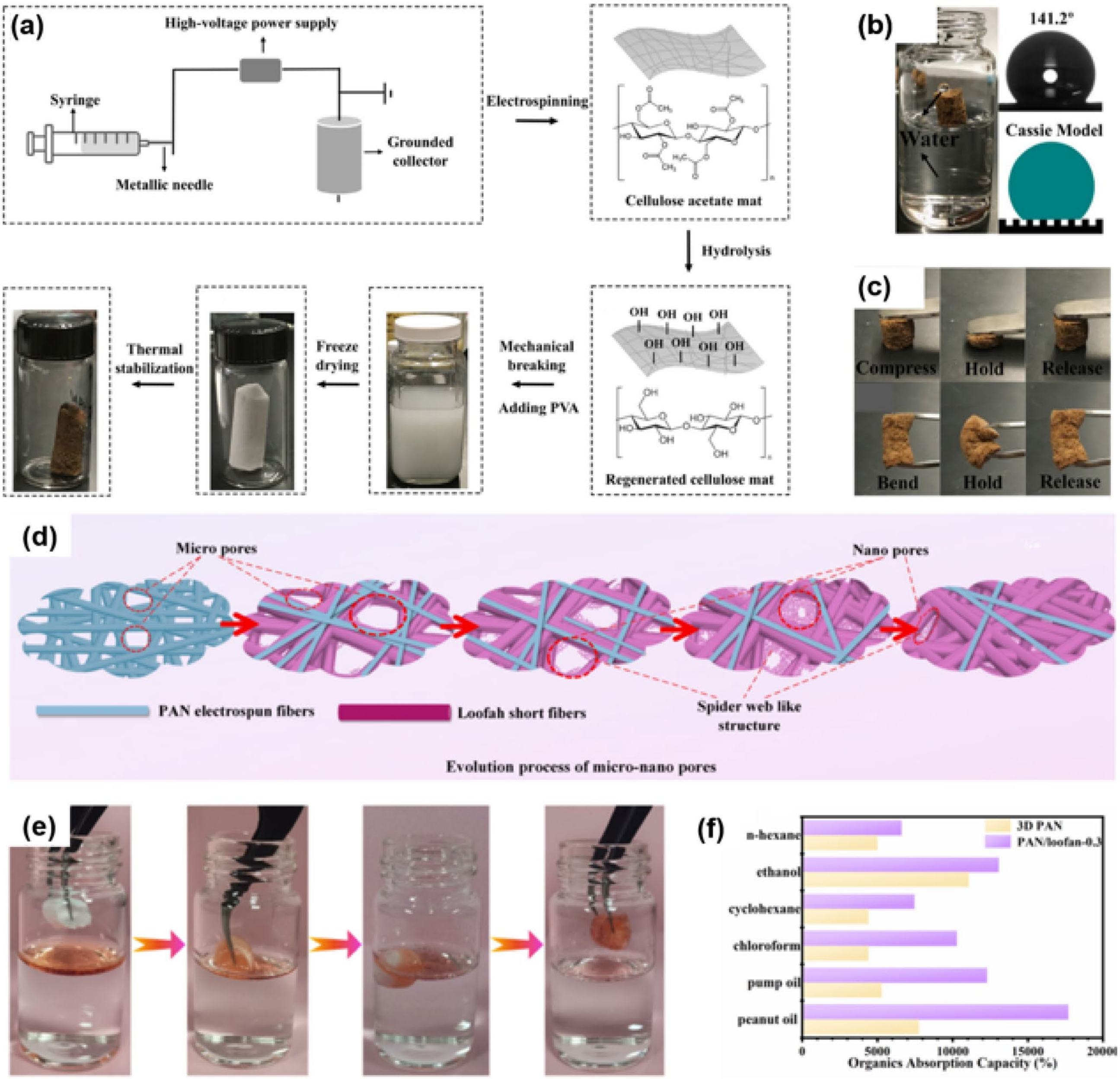

Fig. 5 **a** Scheme of the preparation steps of UECS. **b** The WCA image of UECS. **c** Photographs of the elasticity of UECS under different forms of deformation. Reproduced with permission of Ref. [56], Copyright of ©2018 Elsevier. **d** The formation mechanism of PAN//loofah spider-web structure. **e** Photos of oil absorption process of PAN//loofah sponge. **f** The histogram of adsorption capacity of PAN/loofah sponge for different oils. Reproduced with permission of Ref. [87], Copyright of ©2021 Wiley

membranes are divided into three types: hydrophobic-lipophilic membranes, hydrophilic-oleophobic membranes, and intelligent switchable wettability membranes.

Hydrophobic-Lipophilic Membranes

Although the oil spill absorbent material has a high oil absorption capacity and can achieve rapid in-situ adsorption of large-scale oil on the sea. However, it is difficult to effectively remove the small amount of water contained in the oil. At present, among the commonly used oil removal methods, membrane separation plays an important part in oil/water separation research because of its advantages of simple operation, low energy consumption, and no secondary pollution [101–103]. To achieve the separation of a mixture system with more oil and less water, the membrane material is required to have good hydrophobic-lipophilic properties [104, 105]. The preparation of a hydrophobic

Table 1 Comparison of different oil/water separation methods

Methods	Advantages	Disadvantages	References
Chemical condensation	Simple operation High separation efficiency	High usage of chemicals, easy to produce scum	[12]
Biological oxidation	High separation efficiency Wide application range	Complex device, large land occupation, long processing period	[97]
In-situ combustion	Effectively inhibit the spread of crude oil	Large energy consumption, waste of resources, secondary pollution	[98]
Flotation separation	Large processing capacity High separation efficiency	Expensive equipment, difficult to apply on a large scale	[99]
Membrane separation	Simple operation Low energy consumption Strong applicability High separation efficiency	Easily blocked, high cost	[100]

surface must meet the following two conditions, one is to have a low surface free energy, and another one is to have a rough surface structure [36, 106]. Currently, the preparation of superhydrophobic materials can be divided into two main types: one is to construct a micro-nano multi-level rough structure of the material with low surface energy; the other is to modify the surface with low surface energy substance after constructing a micro-nano multi-level rough structure of the material [107]. Superhydrophobic fibrous membranes with macro-porous structures have good flux in separating oil/water mixtures or emulsions, but may suffer from serious fouling problems. In contrast, superhydrophobic membranes with hierarchical surface structures were shown to have better antifouling ability. According to reports, various superhydrophobic fiber membranes have been effectively prepared through these two approaches.

With the rise of researches on bionic biomaterials, the special wettability of organisms in nature has been continuously explored [108]. For example, the self-cleaning properties of lotus leaves (Fig. 6a) [109], the moisture resistance of water strider legs (Fig. 6b) [110, 111], and the hydrophobicity of shark skin (Fig. 6c) [112, 113]. Inspired by these creatures, the researchers imitated the rough structure of these creatures to build the surface of superhydrophobic materials. Our group has also made some membranes with biomimetic structures to construct superhydrophobic surfaces, such as the protruding structure of bionic lotus flowers and pine needle-like nanorod in the fibers [5, 36]. These special biomimetic structures provide new ideas for the preparation of superhydrophobic surfaces. In order to increase the rough structure of the fiber, Ma et al. loaded zeolitic imidazolate framework-8 (ZIF-8)@thiolated graphene (GSH) on PI nanofibers by a hydrothermal method [114]. The constructed nano-level rough layered structure made ZIF-8@GSH/PI composite fiber membranes exhibits superhydrophobicity (WCA ≈ 153.25°) (Fig. 7a). Furthermore, the highest oil separation flux of 5625 L m^{-2} h^{-1} was obtained for dichloromethane/water mixture, and the fluxes of trichloromethane, dichloroethane, chlorobenzene, tetrachloromethane/water

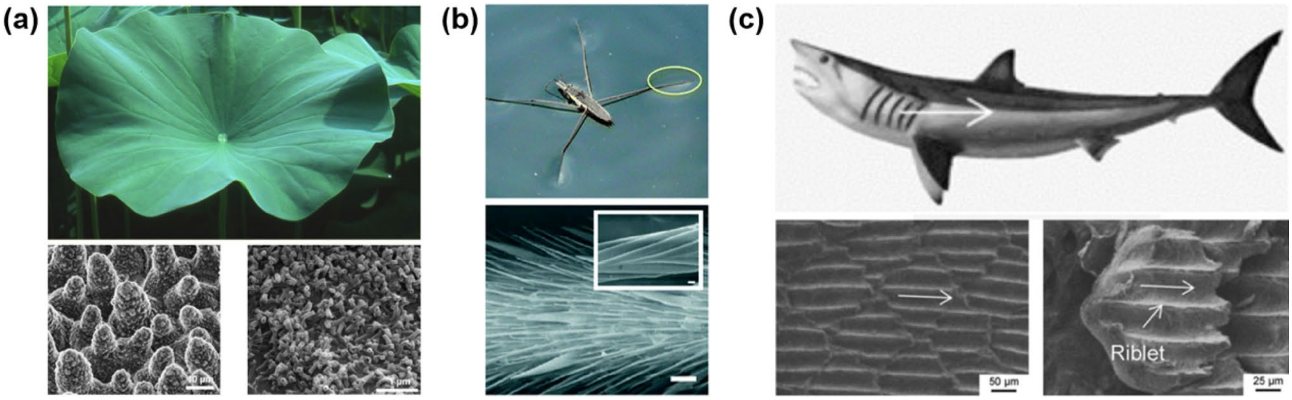

Fig. 6 Digital images and SEM images of **a** lotus leaf. Reproduced with permission of Ref. [109], Copyright of ©2011 Beilstein-Institut Zur Forderung der Chemischen Wissenschaften. **b** water strider legs, Reproduced with permission of Ref. [110], Copyright of ©2010 American Chemical Society. Reproduced with permission of Ref. [111], Copyright of ©2004 Springer Nature. and (c) shark skin. Reproduced with permission of Ref. [113], Copyright of ©2013 Wiley–VCH Verlag

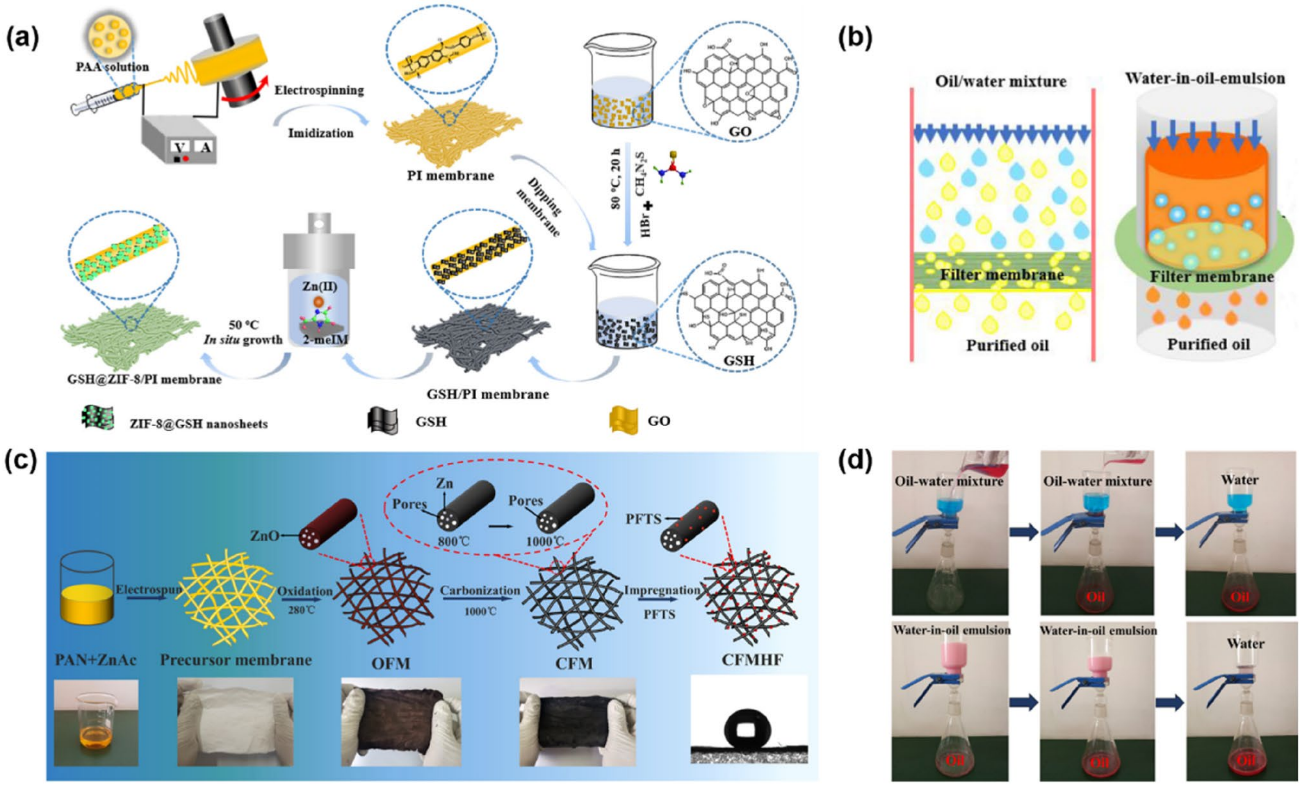

Fig. 7 **a** Preparation diagram of ZIF-8@GHS/PI nanofibrous membrane. **b** Preparation of the oil/water mixture and emulsion separation by ZIF-8@GHS/PI nanofibrous membrane. Reproduced with permission of Ref. [114], Copyright of ©2020 American Chemical Society. **c** Preparation process of the preparation process of CFMHF. d Images of the oil/water mixture and emulsion separations by CFMHF. Reproduced with permission of Ref. [37], Copyright of ©2021 Elsevier

mixtures are 5043, 4613, 4835, and 4196 L m^{-2} h^{-1}, respectively. The separation efficiency was all more than 99%. Moreover, the separation efficiency for stable water-in-kerosene emulsions is also as high as 99.96% (Fig. 7b).

In addition, the hydrophobicity of the material can be enhanced by adjusting the substance of low surface energy on the fiber. Sun et al. prepared PAN/ZnAc composite fiber membranes via electrospinning and then carbonized them by calcination to achieve porous carbon fiber membrane [37]. Finally, the prepared carbon fiber membrane was pretreated with HCl and then immersed in a 1H, 1H, 2H, 2H-Perfluorooctyltriethoxysilane (PFTS) solution for fluorination treatment to obtain carbon fiber membrane (CMFHF). The adhesion of fluorine-containing substances greatly reduces the surface energy of the fiber and increases its hydrophobicity (Fig. 7c). As shown in Fig. 7d, the CMFHF can successfully separate oil/water mixtures and emulsions with excellent separation flux and efficiency. In addition, CMFHF also exhibits good corrosion resistance, which solves the problem that fiber membranes are difficult to apply in harsh environments.

Hydrophilic-Oleophobic Membranes

Hydrophilic oleophobic separation membrane can filter oil components in water, which is a common and effective method for oily wastewater treatment. Since the surface tension of water is usually higher than that of oil ($\gamma_{oil} < 35.0$ mN m^{-1}, $\gamma_{water} = 72.8$ mN m^{-1}), it is theoretically more difficult to prepare oleophobic membranes [48]. At present, researchers can prepare superhydrophilic-underwater oleophobic electrospun fibrous membrane through hydrophilic modification of the fiber surface and construction of micro/nano multi-level rough structures [115, 116]. The hydrophilic-oleophobic membrane is not easy to be polluted due to its repellency to oil, and will not cause clogging and scaling problems which makes the fibrous membrane have better recyclability.

Recently, Qing et al. prepared PVA nanofibrous membrane by electrospinning and then grown SiO_2 nanoparticles on PVA nanofibers in-situ (Fig. 8a, b) [117]. Stable SiO_2 nanoparticles form a nano multistage rough structure on PVA fibers, which makes the composite membrane show

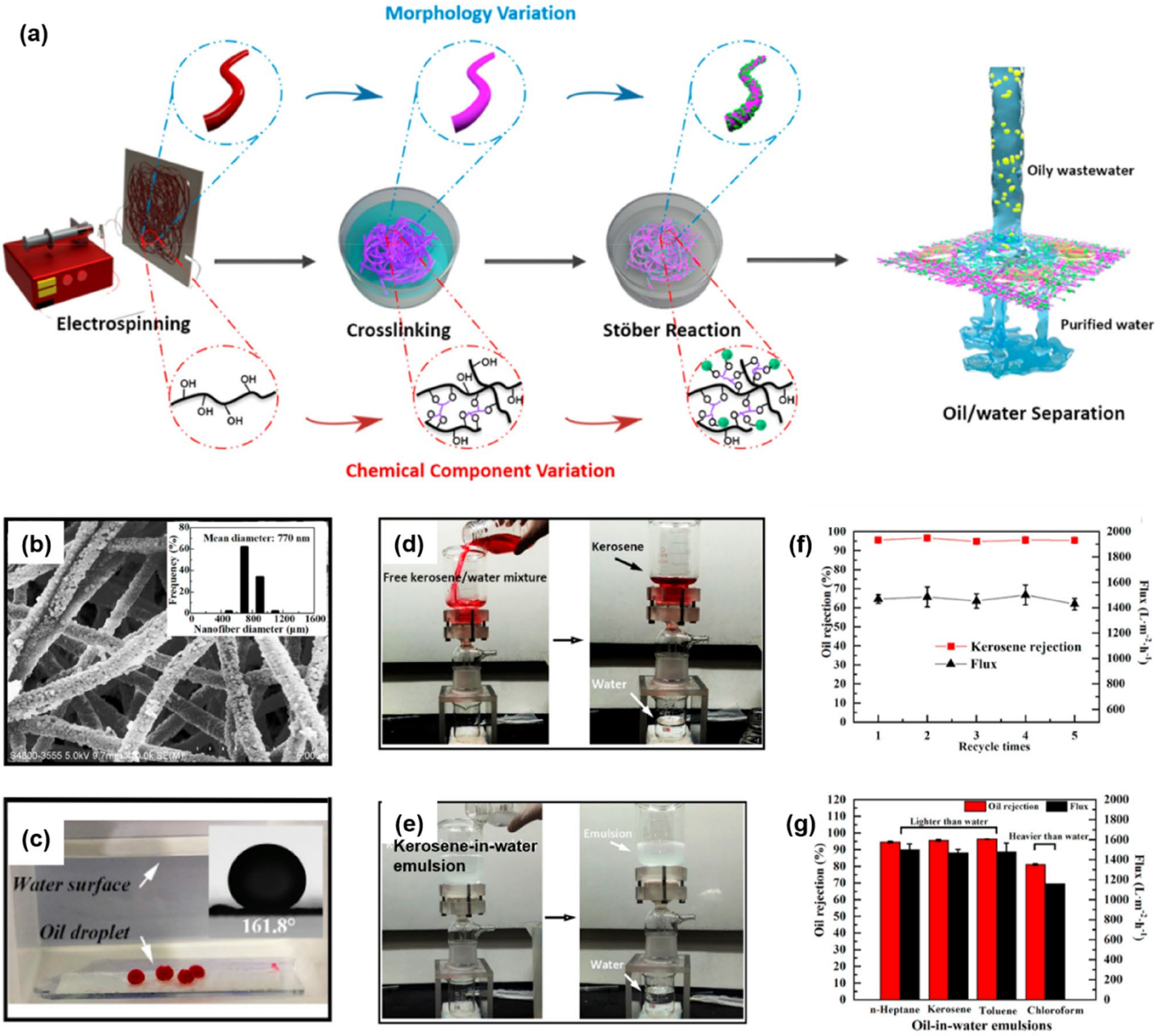

Fig. 8 **a** Preparation diagram of the SUS SiO2@PVA membrane. **b** SEM image of SiO_2@PVA nanofibers. **c** Underwater oil contact angle of SiO_2@PVA nanofibrous membrane. The separation process of **d** oil/water mixture and **e** oil/water emulsion by SiO_2@PVA nanofibrous membrane. **f** The separation flux and efficiency of kerosene-in-water emulsion of SiO_2@PVA nanofibrous membrane. **g** The separation flux and efficiency of different oil-in-water emulsions of SiO_2@PVA nanofibrous membrane. Reproduced with permission of Ref. [117], Copyright of ©2020 Elsevier

superhydrophilicity and underwater oleophobicity with the underwater oil contact angle of 161.8° (Fig. 8c). Moreover, the n-heptane, kerosene, and toluene/water mixtures can be separated by the SiO_2@PVA nanofibers membrane successfully (Fig. 8d, e) with the excellent separation efficiency (> 95%) and separation flux (~ 1500 L m^{-2} h^{-1}) (Fig. 8f, g).

In addition to the method of constructing micro/nano multilayer rough structure, hydrophilic-underwater oleophobicity fibrous materials can also be prepared by low-temperature plasma treatment and surface coating of hydrophilic materials. Huang et al. obtained a flexible nanofibrous composite membrane of superhydrophilicity and core–shell structure [118]. Acid-treated carbon nanotubes (ACNTs) were decorated on electrospun PU nanofibers, and then dopamine (DA) was self-polymerized onto the surface of ACNTs@PU fibers to form polydopamine (PDA) with core–shell structure PDA/ACNTs@PU nanofibers (Fig. 9a). The coating of PDA increased the hydrophilicity of the fiber and showed underwater superoleophobicity for different oils (Fig. 9b, c). In addition, the combination of multiple interfacial hydrogen bonds between ACNT, PDA, and PU nanofibers improves the mechanical properties of the

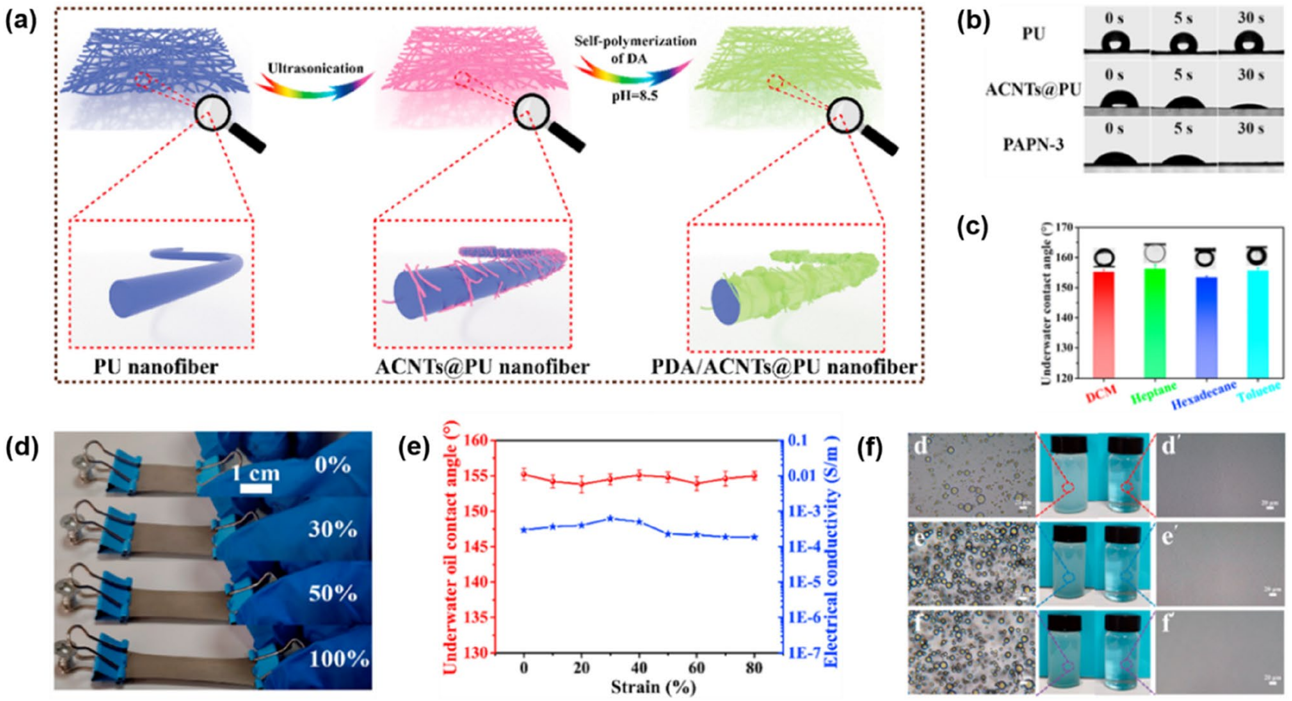

Fig. 9 **a** Preparation diagram of PDA/ACNTs@PU nanofibers. **b** The change of WCA of PU fibers after gradual modification. **c** The underwater oil contact angle of PDA/ACNTs@PU fibrous membrane to different oils. **d** Photos of PDA/ACNTs@PU fibrous membranes under different tensile strengths. **e** The electrical conductivity and underwater oil contact angle of PDA/ACNTs@PU fibrous membranes under different tensile strengths. **f** Optical microscope images before and after the separation of oil/water emulsion. Reproduced with permission of ref. [118], Copyright of ©2020 Elsevier

fibers (Fig. 9d). ACNTs@PU nanofiber membranes exhibit excellent underwater superoleophobicity and conductivity (Fig. 9e). Moreover, the obtained PDA/ACNTs @PU fibrous membrane can successfully separate oil-in-water emulsion by pressure-driven (0.05 MPa). The water separation flux of heptane-in-water, toluene-in-water, and cyclohexane-in-water emulsion are 4108, 5293 and 7240 L m^{-2} h^{-1}, respectively. Meanwhile, all the separation efficiency are higher than 99.7% (Fig. 9f).

Switchable Wettability Membranes

A smart surface is defined as a surface that is sensitive to external stimuli and can produce a special response. Smart materials with switchable wettability have stimulated the interest of many researchers, especially in oil/water separation [119, 120]. The intelligent membrane for separation has the characteristics of stimulus–response and wettability. It can realize the reversible transformation of hydrophilicity and hydrophobicity under certain external stimuli, including temperature, light, pH, and electric field [121–124]. Therefore, it shows great advantages in high-efficiency on-demand oil/water separation applications.

Thermally responsive membranes have received widespread attention due to their simple and easy-to-obtain characteristics. Li et al. prepared a temperature-sensitive poly(methyl methacrylate)-block-poly(N-isopropylacrylamide) PMMA-*b*-PNIPAAm smart membrane by electrospinning [125]. The membrane exhibits temperature-controllable wettability due to the addition of the thermally responsive component PNIPAAm (Fig. 10a). In order to study the change process of the thermal response of the fibrous membrane, it can be observed that the fibrous membrane exhibits hydrophilicity and underwater oleophobicity at the temperature below lower critical solution temperature (LCST). Subsequently, the membrane was heated in-situ to a temperature higher than the LCST, the membrane becomes hydrophobic and lipophilic. The water reserved in the membrane is gradually replaced by oil, and the oil penetrates through the membrane (Fig. 10b, c). In addition, compared with membranes cast by polymer solutions, electrospun fibrous membranes have controllable oil/water wettability and are more suitable for treating industrial oily wastewater. When the temperature decreases from 50 to 15 °C, the underwater oil contact angle of the electrospun fibrous membrane increases from 37° to 153°, which is higher than the variation range of the cast membrane (55°-142°). In addition, the fibrous membrane exhibits excellent water flux (9400 L m^{-2} h^{-1}) and oil flux of petroleum ether (4200 L m^{-2} h^{-1}) with the high separation efficiency (> 98.5%).

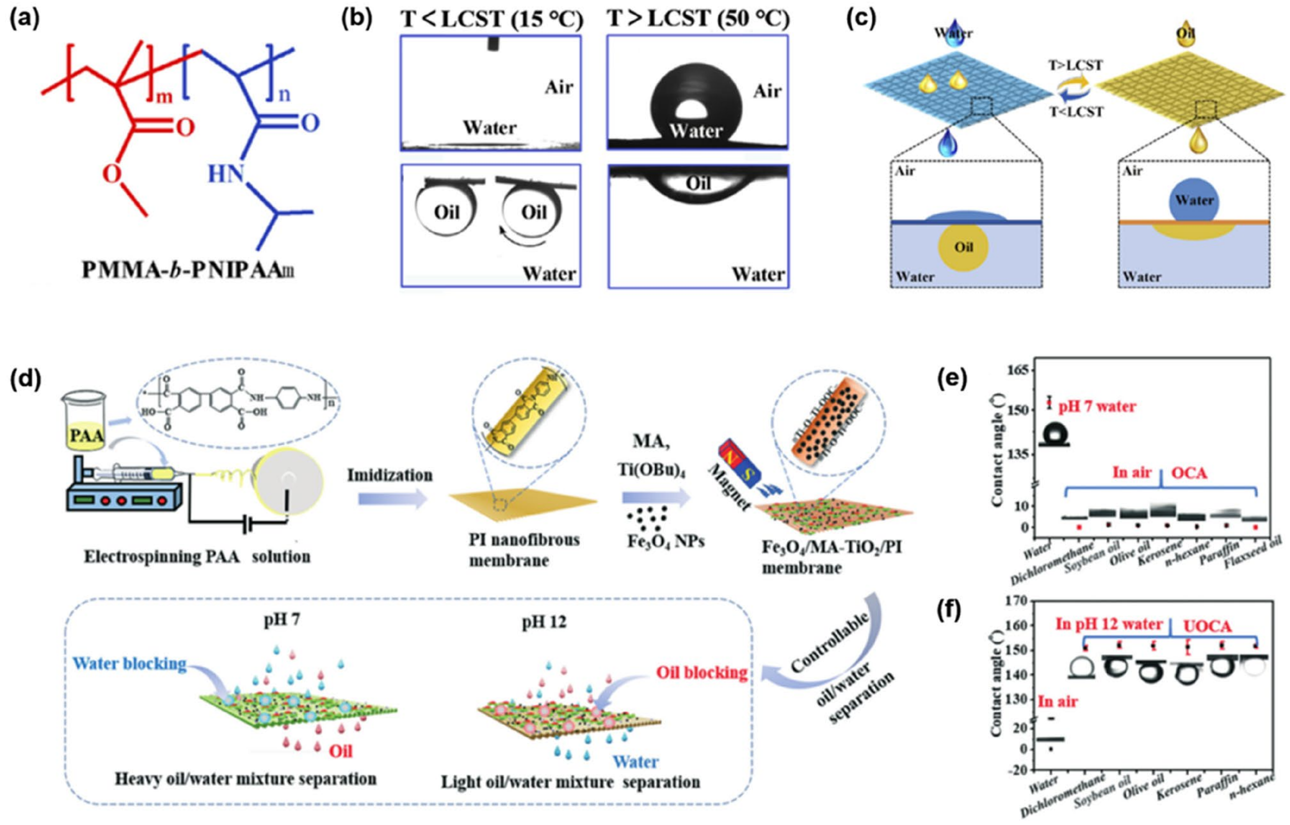

Fig. 10 **a** PMMA-*b*-PNIPAAm macromolecule. **b** Temperature response switchable wettability PMMA-*b*-PNIPAAm membrane. **c** Water passes through the membrane when the temperature is below LCST, and oil passes through the membrane when the temperature is above LCST. Reproduced with permission of Ref. [125], Copyright of ©2016 Elsevier. **d** Preparation diagram of Fe_3O_4/MA-TiO_2/PI and the separation performance under different pH. **e** OCA of Fe_3O_4/MA-TiO_2/PI membrane in the air. **f** The underwater OCA of different oils (in the water of pH = 12). Reproduced with permission of Ref. [126], Copyright of ©2019 Royal Society of Chemistry

Polymers containing poly(4-vinylpyridine) (P4VP), poly(2-vinylpyridine) (P2VP), and polyacrylic acid (PAA) usually exhibit pH-sensitive switchable wettability due to their conformation and charge are affected by pH. Recently, Ma et al. used PI (containing PAA component) as a substrate for the first time to prepare a pH-responsive flexible and magnetic smart nanofiber membrane [126]. Then the fibrous membrane was immersed into a pre-gel solution of tetrabutyl orthotitanate ($Ti(OBu)_4$), myristic acid (MA), and Fe_3O_4 to obtain a magnetic pH-responsive Fe_3O_4/MA–TiO_2/PI smart fibrous membrane (Fig. 10d). In the air, Fe_3O_4/MA–TiO_2/PI exhibits superhydrophobicity (WCA = 150°, pH = 7) and lipophilicity (Fig. 10e). However, when the membrane was immersed in water with a pH of 12, the membrane becomes superoleophobic (Fig. 10f). In addition, the magnetic properties of the membrane make it easier to recover after oil/water separation. Therefore, this environmentally friendly and low-energy smart membrane provides a better choice for oil/water separation.

Nanofibrous Aerogels for Emulsion Oil/Water Separation

Emulsion pollutants are widely present in the fields of petroleum and natural gas, energy and chemical engineering, environmental science, etc., and stable emulsions are not conducive to industrial production and resource recycling [127]. On the one hand, the water content of the oil increases the load, corrosion, and fouling of the storage tanks of the transportation pipeline. It also affects the quality of the oil and the operation of the machine. On the other hand, the discharge of oily sewage would cause pollution to the environment [128, 129]. Therefore, it is also necessary to separate oil/water emulsions. Because of the increasingly complex development of the petroleum industry, the existing form and structure of emulsion are becoming more and more complex, and there are many kinds of emulsifiers, which leads to the increasing difficulty of emulsion demulsification [130]. Thus, it is urgent to find a demulsification method with strong applicability. As a new type of 3D porous material, aerogels have attracted wide attention in many fields [131–134]. Aerogels are expected to further improve the separation flux due to the high tortuosity, high porosity, and large specific surface area. Compared with the pressure-driven separation membrane, the gravity-driven separation membrane reduces the energy consumption. Meanwhile, the fibrous aerogel has higher separation flux and separation efficiency than the reported two-dimensional membranes that can be gravity-driven for the separation of water-in-oil emulsions [135, 136].

Si et al. proposed a novel method for preparing super-elastic and superhydrophobic nanofibrous aerogels by electrospinning and freeze-drying [137]. The preparation process is shown in Fig. 11a. Firstly, PAN nanofibers, SiO_2 nanofibers, SiO_2 nanoparticles, and bifunctional benzoxazine (BAF-a) prepared by electrospinning were uniformly mixed to form a homogeneous nanofiber dispersion. Then the dispersion was freeze-dried to make nanofiber aerogel, and the aerogel was thermally cross-linked to obtain a fibrous, isotropically-bonded elastic reconstructed (FIBER) aerogel (Fig. 11b). Among them, the rigidity of SiO_2 nanofibers plays a supporting role in the aerogel. BAF-a can adhere SiO_2 nanoparticles to the surface of the fiber and endow the aerogel with low surface energy. Moreover, SiO_2 nanoparticles construct micro-nano particles in the aerogel. The hierarchical structure makes the prepared aerogel have excellent hydrophobicity, lipophilicity, and elasticity (Fig. 11c-e). Furthermore, the

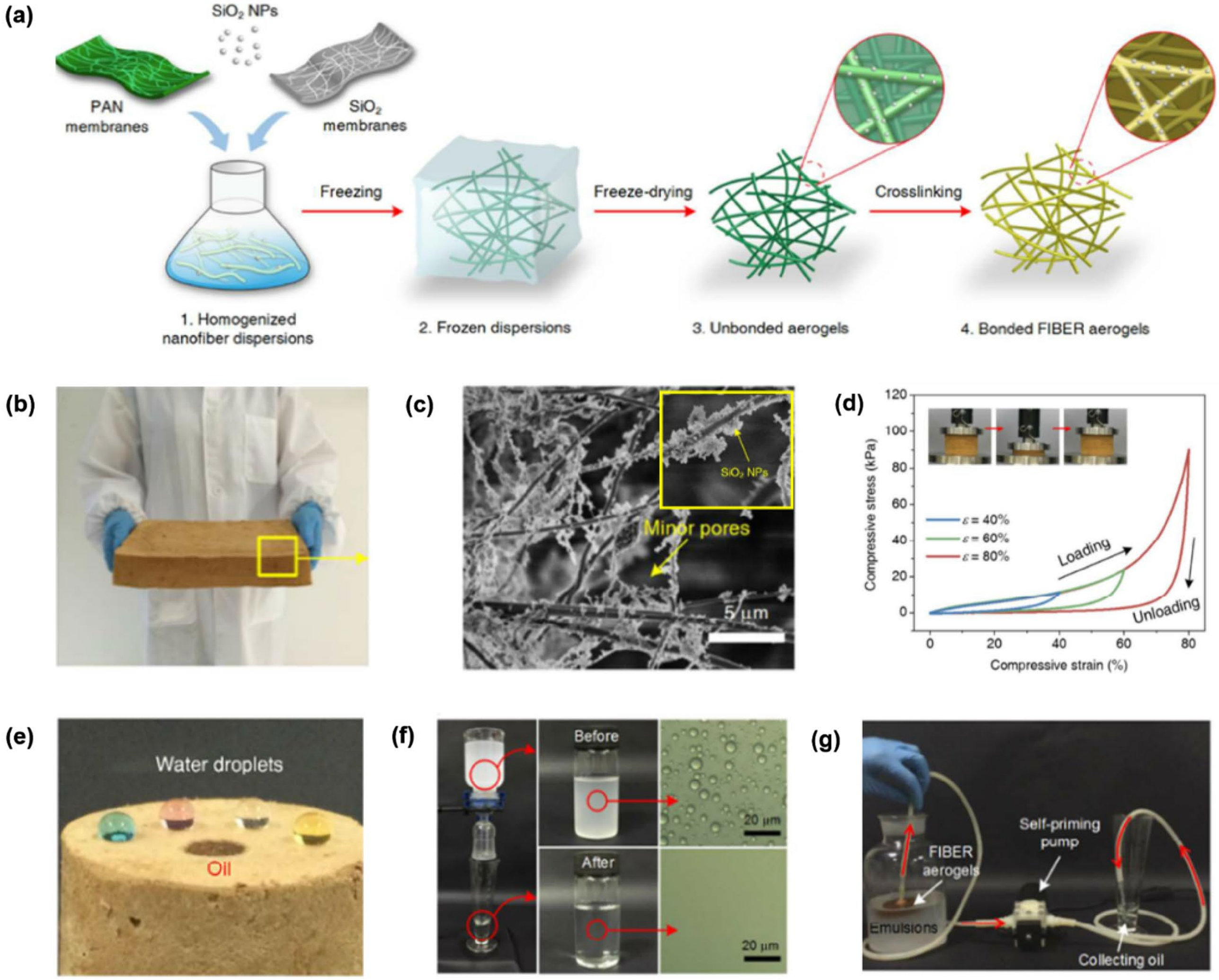

Fig. 11 **a** Preparation diagram of FIBER aerogel. **b** Image of a FIBER aerogel (2.5 L). **c** SEM images of FIBER aerogel. **d** Compressive stress versus strain curves for FIBER aerogels along the loading direction. Insets: Photos of the FIBER aerogels under a compressing and releasing cycle. **e** Water and oil droplets on the surface of FIBER aerogel. **f** Digital and optical images of water-in-oil emulsion by gravity-driven separation using FIBER aerogels. **g** Oil recovery apparatus for collecting oil from water-in-oil emulsions. Reproduced with permission of Ref. [137], Copyright of ©2015 American Chemical Society

superhydrophobic and superlipophilic properties of FIBER aerogels make it possible to quickly separate emulsions by gravity-driven with the excellent flux (8140 ± 220 L m^{-2} h^{-1}) and the separation efficiency of 99.995% (Fig. 11f). In addition, the FIBER aerogels used with a peristaltic pump can continuously collect oil from water-in-oil emulsions more quickly and efficiently (Fig. 11g).

Recently, Shen et al. prepared nanofiber-based aerogels (NFAs) with a separation efficiency of up to 100% due to the good mechanical properties and chemical resistance of PI [138]. As shown in Fig. 12a, PI nanofibers were firstly prepared by electrospinning and made into a homogeneous nanofiber dispersion. After freeze-drying, the PI-NFAs were fumigated in DMF steam to form cross-linked intersections. Afterward, the treated PI-NFAs were immersed in a heptane solution containing trichloromethylsilane (TCMS) and water for different times to coat the surface of the fiber with silicone nanowires to improve the hydrophobicity of the aerogel. Finally, it was freeze-dried again to obtain cross-linked PI-NFAs. The decoration of silicone nanofilaments (SiNFs) makes the SiNFs/PI-NFAs exhibit superhydrophobicity, and the dyed water droplets on the top of a SiNFs/PI-NFAs reflects the low adhesion of the aerogel (Fig. 12b–d). Furthermore, it can be observed from the microscope picture that the SiNFs/PI-NFAs can successfully separate all water-in-oil emulsion by gravity-driven with high flux (Fig. 12e).

In general, different micrometer-sized water droplets can be effectively separated even if the size of the water droplets is smaller than the pore size of the fibrous aerogel. It can be explained that the separation process of fibrous aerogels is based on coalescence separation rather than sieving surface filtration. This process mainly intercepts emulsified droplets through droplet coalescence in many tortuous microchannels. Therefore, fibrous aerogels are expected to achieve higher separation fluxes at low driving pressures.

Conclusion and Perspective

In this paper, the mechanisms and applications of electrospun nanofibrous materials for oil/water separation are reviewed. Different design principles and separation

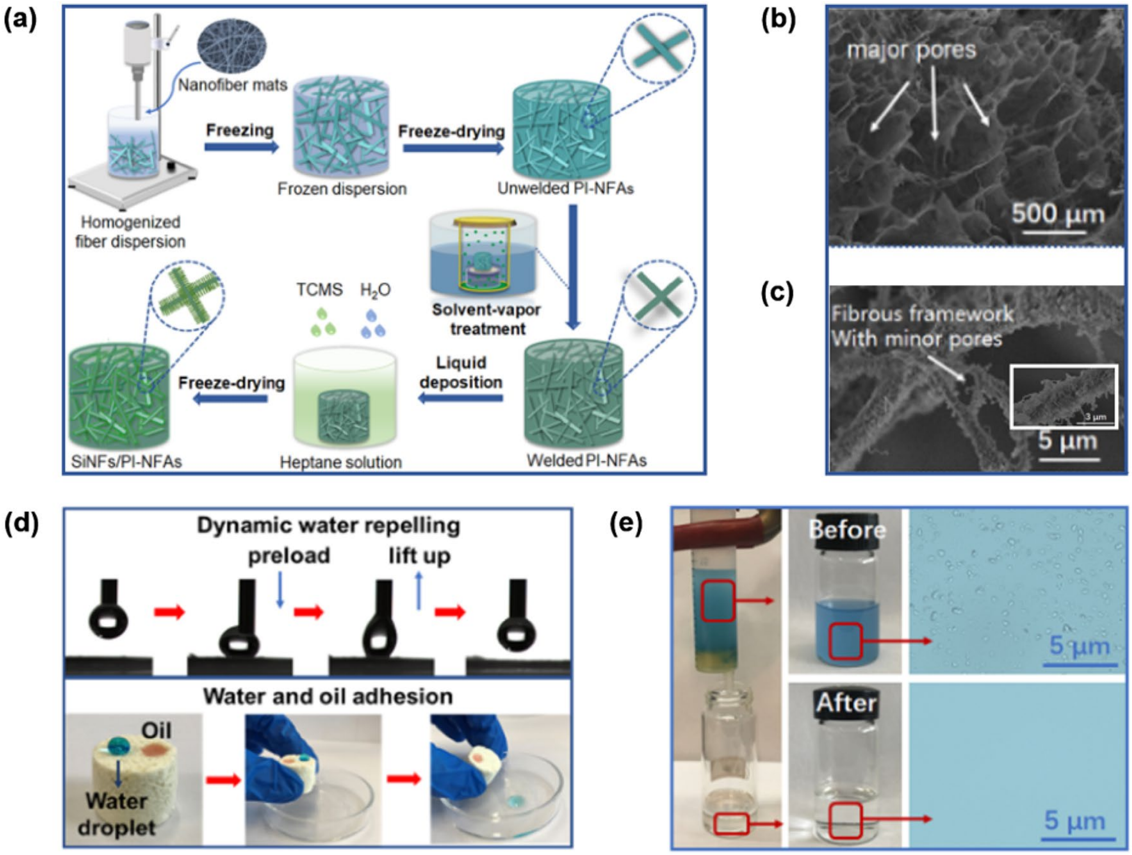

Fig. 12 **a** Preparation diagram of the SiNFs/PI-NFAs. **b**, **c** SEM images of SiNFs/PI-NFAs at different magnifications. d The adhesion of water droplets on the surface of SiNFs/PI-NFAs, and the dynamic measurement photos of water droplets and oil droplets on the surface. **e** The microscopy images of emulsion before and after separation by SiNFs/PI-NFAs. Reproduced with permission of Ref. [138], Copyright of ©2021 American Chemical Society

Table 2 Comparison of various nanofibrous materials for oil/water separation via electrospinning

Type	Materials	WCA(°)/OCA (°)	Oil adsorption performance (g g^{-1})	Separation flux (L m^{-2} h^{-1})	Separation efficiency (%)	References
Nanofibrous sorbents	PS	120/10	95–124	–	99	[66]
	PVA	–	11.7–12.7			[67]
	PI	–	57.4–76.3	–	–	[68]
	PVC	130/0	16–38	–	–	[69]
	PVDF	155 ± 4.9/0	93–149	–	–	[70]
	PS/PU	135/0	110.9–144.5	–	–	[74]
	PVC/PS	–	37–149	–	–	[75]
	SiO_2/PS	153/0	122.7	–	–	[79]
	PAN/loofah	130/0	122.7–176.9	–	–	[87]
Nanofibrous membranes	ZIF-8@GSH	153.25/0	–	4196–5625	> 99	[114]
	CFMHF	155.9/0	–	2800–3590	> 98	[37]
	SiO_2@PVA	0/161.8	–	1500	> 95	[117]
	PDA/ACNTs@PU	0/> 150	–	4195–7240	99.9	[118]
	PMMA-*b*-PNIPAAm	0/153 (T < 15℃) 130/37 (T > 50℃)	–	9400 (water) 4200 (oil)	99	[125]
	Fe_3O_4/MA–TiO_2/PI	150/5 (pH = 7) 5/150 (pH = 12)	–	6038 ± 100 (water) 4281 ± 100 (oil)	99	[126]
Nanofibrous aerogels	FIBER aerogel	138/0	–	8140 ± 220	99.99	[137]
	SiNFs/PI-NFAs	151.7/0	–	75,000–120,000	99.49	[138]

approaches of nanofibrous adsorbents (polymer adsorbents, composite adsorbents, and biomass adsorbents), nanofibrous membranes (hydrophobic-lipophilic membranes, hydrophilic-oleophobic membranes, and switchable wettability membranes), and nanofibrous aerogels are systematically summarized. In addition, their oil/water separation performance are systematically compared, as shown in Table 2. Although electrospinning nanofibrous materials have made great progress in oil/water separation, and various nanofibrous materials have been prepared for oil/water separation treatment in different environments, but it still facing many challenges. First, it is necessary to construct a suitable multi-level rough structure on the surface of nanofibers to achieve the nanofibrous materials of special wettability. However, the prepared functional layer is easily damaged under the influence of external factors, which fundamentally shorten the service life of the separation material. In addition, compared with commercial oil/water separation materials, the electrospun nanofibrous materials currently still have poor mechanical properties, which limits their practical applications. Therefore, the stability of composite materials needs to be considered. Secondly, the preparation process of some current electrospun nanofiber materials is complicated and the cost is relatively high. Therefore, it is of great significance to study electrospun nanofibrous materials with simple preparation process and low cost. Moreover, further research is necessary to advance the preparation of electrospun nanofibrous functional materials for industrialization. In addition, electrospun fibrous membranes should be used in more complex environments, such as the handling of trace oils in the aerospace field. Finally, the current report mainly focuses on the preparation of a variety of separation materials with different wettability. Since the process of oil/water separation with nanofiber materials is a multiphase flow separation process, it involves microfluid mechanics, interface chemistry and engineering science, etc. However, few related theoretical studies can reveal the basic principles of this process in-depth and systematically. Therefore, the preparation and performance of electrospun nanofibrous materials in oilwater separation are still being explored, and it is believed that greater breakthroughs can be made in the next few decades.

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Declarations

Conflict of interest On behalf of all authors, the corresponding author states that there is no conflict of interest.

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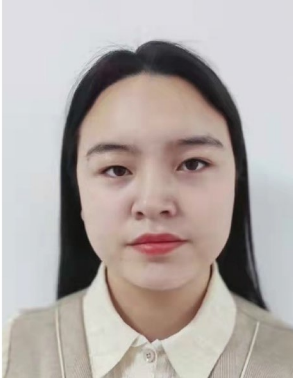

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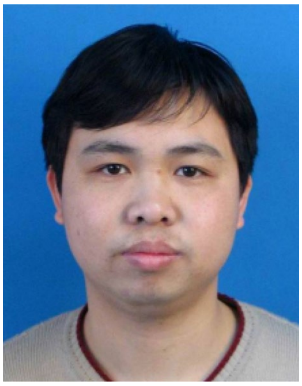

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