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Efficient separation of oil-water emulsions: Competent design of superwetting materials for practical applications

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ABSTRACT

Globally, the issue of oil-water emulsion is continuously tickling in the minds of researchers. The oil-water emulsion is hazardous to marine ecological safety and human health around the globe, indicating that we urgently need to develop efficient strategies to tackle emulsion issues. Materials with porous morphology and selective superwettability towards certain liquids are essential in separating oil-water emulsions. Recently, there has been increased demand for smart oil-absorbent and water-absorbent superwetting porous materials. Over the past decade, several innovative advancements and promising industrial developments to separate oil-water emulsions have been spotted. This review article overviews the latest advancements in the design of superwetting materials for oil-water emulsion separation and thoroughly discusses oil-absorbent and water-absorbent superwetting porous materials, demulsification, and superamphiphilic materials. In each section, the design strategies, base materials and oil-water emulsion separation performance of superwetting porous materials have been systematically discussed. Then, common challenges of superwetting materials in practical application are discussed. Finally, conclusions and future perspectives concerning oil-water emulsion separation based on superwetting porous materials are summarized. This review article can allow helping hand to early career scientists to elegantly understand the oil-water emulsion separation issues.

1. Introduction

Oil-water emulsions have become a major global issue due to their detrimental and long-lasting effects on the environment, animals, and plants. Offshore oil spills can produce oil-water emulsion, which can have a negative impact on the fresh water [1–7]. The five largest and disastrous oil spills in history include Gulf War - Persian Gulf (1991), Deepwater Horizon - Mexican Gulf (2010), Ixtoc -1 - Mexican Gulf (1980), Atlantic Empress - Caribbean Sea (1979), and ABT Summer - Offshore Angola (1991) [8]. Additionally, several industrial processes release large volumes of oil containing wastewater into the fresh water annually. In a day, petro-refinery industries generate over 250 million barrels of oily wastewater, out of which around 40% is discarded into the environment [9]. Due to inadequate distribution of oil stores and consumption areas, over 71% of oceans are infected with oil, causing immense damage to marine and human life [10]. Oil creates a layer on the water surface that prevents oxygen from reaching aquatic plants and animals. Due to the oil layer on the sea surface, the hypothermia effect caused to the sea birds, fishes, seals, and sea otters, which can be fatal. Fish and other sea life can suffer from poisoning, internal injury, ulcers, damage to red blood cells, kidneys, liver, and the immune system after ingesting oil. Whales can die due to clogged blowholes from oil, and humans can suffer various diseases such as lipid pneumonia, lung diseases, folliculitis, and skin diseases due to exposure to oily water [11–14]. The primary source of generation of oil-water emulsion and its impact on the ecosystem is compiled in Fig. 1.

In fact, emulsion separation is a worldwide challenge [15,16]. Conventional methods have been developed to tackle this issue, however find numerous limitations. The heat separation method, for instance, involves heating oil to reduce its viscosity, which releases abundant hydrocarbon components in the environment, causing secondary pollution such as air pollution [17]. On the other hand, gravity separation takes much longer time and relies on the density difference between oil and water [18]. Coalescence separation involves the

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collision of tiny droplets to form larger ones, which can then be separated [19]. Centrifugation is an industrial-scale process that separates oil-water emulsion using centrifugal force, but its efficiency depends on the specific gravity of components. The coagulation method involves destabilization of emulsion through aggregation and flocculation, but non-biodegradable coagulants used in this process create secondary pollutants. Generally, these conventional methods are inefficient, expensive, and prone to secondary pollution [20–23]. Therefore, cutting-edge techniques are urgently required for easy and sustainable oil-water emulsion separation, using environmentally friendly materials.

The wetting phenomena occurring on many natural surfaces brought the idea of superwetting. A water drop rests on the lotus leaf like a tiny ball due to rough surface morphology and the presence of low surface energy wax crystalloids on the lotus leaf [24,25]. Furthermore, the surface and interfacial studies of fish scales exhibit superhydrophilic (SHL) and underwater superoleophobic (SOP) characteristics [26]. This outstanding wetting characteristic is due to its rough surface structure and outer fish skin secretes a slippery mucus layer consisting of hydroxyl, amine, and carbonyl groups. This principle can be utilized to develop materials that can either absorb oil or water or form a layer that prevents other liquids from passing through the pores [27–30].

As promising alternatives, superwetting materials have many benefits, such as being affordable, easy to make, efficient, and environmentally friendly [31–35]. These materials have demonstrated exceptional performance in various applications, including self-cleaning [36], anti-icing [37], anti-fouling [38], self-healing [39], anti-corrosion [40], and oil-water separation [41–43]. Porous superwetting surfaces were found beneficial for efficient oil-water emulsions.

However, large pore-size materials are unable to separate emulsions effectively. The pore size can be controlled using the size-sieving effect by adjusting the pore size smaller than emulsion droplets for efficient separation [44]. In academic and industrial research, different types of materials, including meshes, polymer membranes, and foams have been used to create superhydrophobic/superoleophilic (SHP/SOL) and superhydrophilic/underwater superoleophobic (SHL/SOP) porous materials. A SHP/SOL porous material can be used to separate water-in-oil (W/O) emulsion, whereas SHL/underwater SOP materials can be adopted to separate oil-in-water (O/W) emulsion. Many attempts have been made to fabricate porous superwetting materials for oil-water emulsion separation [45–48].

Several review articles on oil-water emulsion separation are found in the literature discussing a variety of membranes, polyimide materials, fibrous materials, and natural superwetting materials [49–55]. However, their applications from small laboratory scale to large industrial level have not been summarized well. This review aims to fill this gap by explaining the mechanism of emulsion separation by superwetting materials. We provide an overview of recent advancements in the fabrication of superwetting materials for emulsion separation, consequently adding subsections on oil-absorbent and water-absorbent superwetting porous materials, demulsification, and superamphiphilic materials. In each section, the design strategies, base materials, and oil-water emulsion separation performance of superwetting porous materials have been thoroughly discussed. Moreover, this review highlights the exceptional properties of porous superwetting materials, including their extended wettability, efficiency, reusability, mechanical strength, and scalability in manufacturing, making them ideal for emulsion separation. Consequently, common challenges of superwetting materials in practical applications are briefly discussed. The conclusions and future perspectives concerning oil-water emulsion separation based on superwetting porous materials are briefed at the end.

2. Mechanisms of oil-water emulsions and their separation

2.1. Basics of oil-water emulsions

An emulsion is a fine mixture of water and oil that contains small droplets of the opposite liquid dispersed throughout. There are two main types of emulsions: oil-in-water (O/W) and water-in-oil (W/O) [56]. In an O/W emulsion, oil droplets are dispersed in a continuous water phase (schematically shown in Fig. 2A, top). In contrast, the continuous and dispersed phases are switched in a W/O emulsion (schematically shown in Fig. 2A, bottom). Stabilized microemulsion droplets are formed when surfactants, biopolymers, or fine particles are present in the emulsion [55,57]. Surfactants have hydrophilic and hydrophobic groups that tend towards the water and oil phases, respectively, which reduce the interfacial tension. Water-soluble surfactants are present in O/W emulsions, while oil-soluble surfactants are present in W/O emulsions.

2.2. Oil-water emulsion separation mechanism

For oil-water emulsion separation, the superwetting porous materials can be designed by controlling pore size along with surface morphology and surface energy. The superwetting porous materials can completely wet with one phase of oil-water emulsion and pass through the pores to achieve oil-water emulsion separation. Pore size-sieve plays a significant role in the design of superwetting porous materials for emulsion separation. According to the size-sieving effect, oil-water

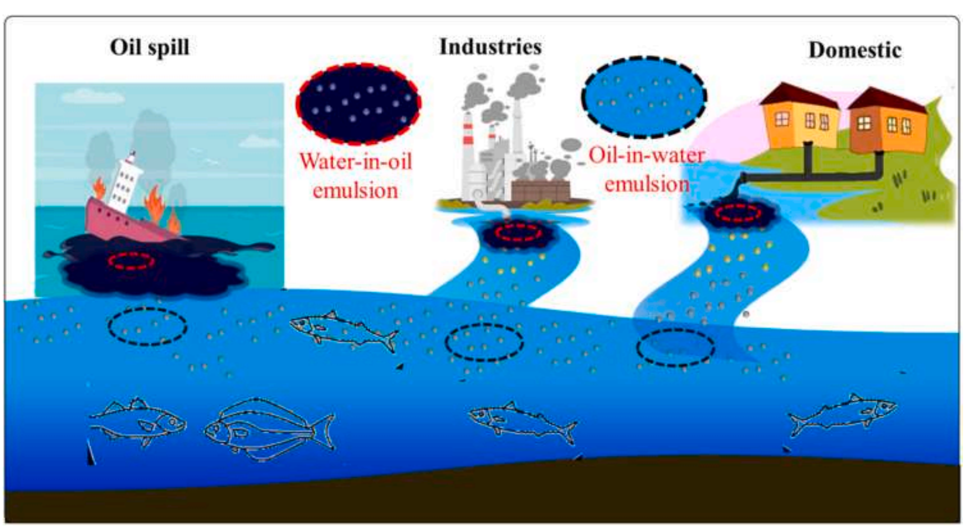

Fig. 1. Suffering of aquatic life due to oil spill and industrial and domestic oily wastewater dumping in water bodies.

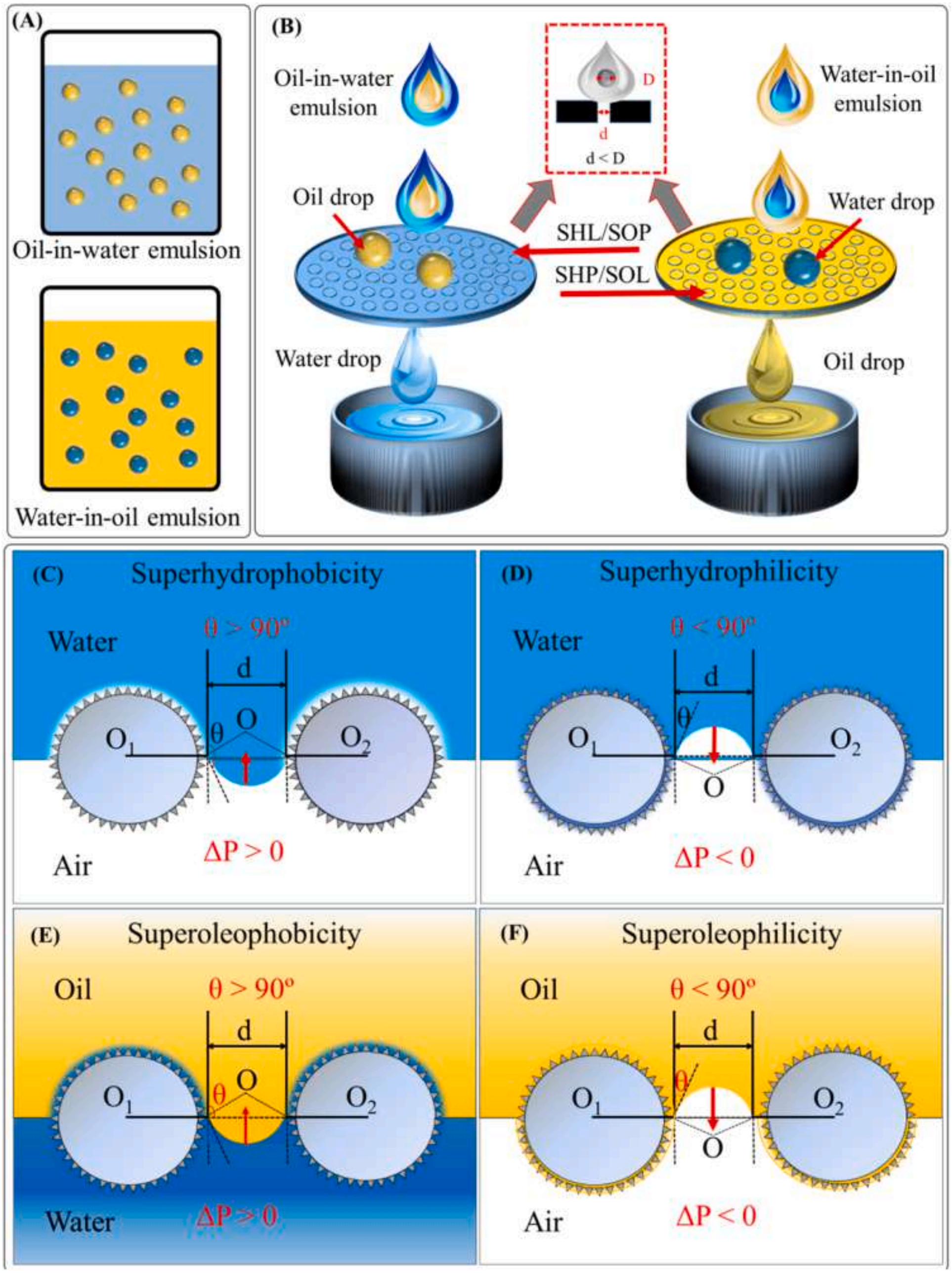

Fig. 2. (A) Schematics of O/W and W/O emulsion. (B) Schematic representation of the mechanism of separation of oil-water emulsion by superwetting materials with a small pore size based on size-sieving. (C) Water cannot wet a porous surface when CA > 90° in air. (D) Water can wet a porous surface when WCA < 90° in air. (E) Oil cannot permeate superoleophobic porous surfaces when prewetted by water. (F) Oil can wet a porous surface when OCA < 90° in air.

emulsions can be separated by the small pore sized porous materials where the pores are much smaller than the emulsion droplets (schematically shown in Fig. 2B) [58,59]. Another physical characteristic, the intrusion pressure (ΔP), needs to be considered in the emulsion separation process. Intrusion pressure is the maximum pressure that porous materials can withstand without liquid passing through their pores. It is determined by the wettability of the material. Liquid intrusion pressure can be determined using the following equation:

$$\Delta P = -(l\,\gamma\,\cos\theta)/A \quad (1)$$

where γ is surface tension, l and A is a pore's circumference and cross-sectional area. θ is the liquid contact angle on the flat surface. The superhydrophobic porous material possesses a water contact angle (θ_w) > 90°, enabling it to repel water effectively (Fig. 2C). This is due to the presence of a thin air film trapped within the porous structure of the material, which acts as a barrier against water intrusion. In contrast, water can enter the superhydrophilic porous material that has $\theta_w < 90°$ (Fig. 2D). When water prewets a porous surface; it replaces trapped air. The water-prewetted porous surface readily forms a capillary bridge of water between two cylinders of pores and is opposed to the intrusion of oil into the porous structure. As a result, water-prewetted porous materials become superoleophobic with oil contact angle $\theta_o > 90°$, indicating external pressure is needed to penetrate oil into pores ($\Delta p > 0$) (Fig. 2E). In the case of superhydrophilic or superoleophilic porous materials, which have a contact angle of less than 90°, only gravity can cause water or oil to penetrate easily. Therefore, these materials have an intrusion pressure of $\Delta P < 0$ (Fig. 2 D and F).

Further, the thickness along with the pore size of superwetting porous materials can influence the permeation flux of oil-water emulsions. The Hagen–Poiseuille relation can be used to calculate the permeation flux (F) of the membrane, which depends on the porosity (ε), pore radius (r_p), thickness of the material (L), intrusion pressure (ΔP), and viscosity of the liquid (μ) [60]; is given by following equation,

$$F=\Delta P \left(\varepsilon r_p^2 / 8\mu L\right) \tag{2}$$

The reduction in the pore size of porous materials decreases separation flux. Moreover, emulsion droplets clogged in small pores, resulting decline in flux. To overcome this issue, the oil-water emulsion can be separated based on demulsification. Superwetting materials can absorb emulsion droplets during demulsification. Emulsion droplets on a superwetting surface coalesce into bigger droplets due to the coalescence effect. The coalesced droplets form a layered oil/water mixture that can be separated. This separation process is not affected by the pore size of superwettable materials and does not require increased flux to be effective [61,62]. Therefore, superwettable materials can lead to the highest possible oil-water emulsion separation.

3. Recent progress in the design of superwetting materials for oil-water emulsion separation

Extensive research is being conducted on porous superwetting materials, which are proving highly effective for separating oil-water emulsions. There are two common types of superwetting materials: superhydrophilic and superoleophilic. These materials are designed to exhibit extreme affinity for either water or oil, respectively. Adjusting pore size concerning emulsion droplets and making them robust for industrial applications is challenging. The different types of porous materials, including metal mesh/foam, polymer membranes, polymer sponges, cellulose materials, cotton fabric and filter paper, have been used to prepare superwetting surfaces for emulsion separation (Fig. 3A) [63–66]. The superwetting membranes are popular due to their high porosity, adjustable pore size, thin architecture, and affordability. Different techniques, including spray [67,68], spin [69], dip [70,71], brush coating [72,73], electrospinning, electrospray, electrochemical [74–76] and phase separation [77], have been used to fabricate superwetting porous materials. The common fabrication methods of superwetting porous materials are schematically represented in Fig. 3 (B-G).

Along with wettability and porous morphology, physical and chemical stability are essential factors for superwetting porous materials to be used in oil-water emulsion separation applications. The various durability testing methods, including abrasion test, sand impact test, tape-peeling test, UV exposure, deep corrosive liquid stability, heating, antifouling, and reusability have been mostly used. Fig. 4 (A-H) represents a schematic illustration of durability testing methods for superwetting porous materials. The recent progresses in the design of superwetting materials for oil-water emulsion separation are discussed in the following subsections.

3.1. Oil absorbent superwetting porous materials

Huge amounts of oil-water emulsions are continuously formed as a result of oil spills; the viscosity, temperature, and density differences between the oil and water play significant roles in the formation of emulsions [78]. In addition, W/O emulsions are developed due to water injection during oil extraction, water cooling during petroleum cracking, or long-distance transportation by underground pipelines or oil tankers. The micro emulsion droplets are captured by superoleophilic porous materials, wet the surface completely and pass through the pores. However, water may strike on the surface, resulting in effective W/O emulsion separation [79].

For the first time, a lotus leaf-like surface that strongly repels water and quickly absorbs oil was fabricated by coating low surface energy polytetrafluoroethylene (PTFE) on stainless steel mesh [80]. The mesh separated oil and water mixture successfully, however, it was incapable in separating the oil-water emulsion because of the large pore size. Considering this drawback, researchers have designed superwetting materials with microporous morphology, especially for oil-water emulsion separation [81,82]. Recently, Lin assembled a cobweb-like multi--sized pore-based network on stainless steel mesh (SSM) by a vacuum suction method for W/O emulsion separation [83]. Dispersion of carbon nanofibers (CNFs) and single-walled carbon nanotubes (SWCNTs) was filtered through chemically etched SSM (ESSM) under pressure. The CNFs-SWCNTs network embedded mesh was dipped into polydimethylsiloxane (PDMS) for superhydrophobicity. Fig. 5A depicts the schematic of an embedded network structure of CNFs–SWCNTs–PDMS on ESSM. The dual scaled pore diameters of 500 nm and 200 nm were formed through cross-linking between CNFs and SWCNTs, respectively, which could separate 100 nm sized W/O emulsion with separation flux of 430 $L{\bullet}m^{-2}{\bullet}h^{-1}$. Besides, Stenocara beetle's back structure gives a new idea to design superwetting materials, in which the water droplets get captured from water-in-oil emulsion and improve separation efficiency. The beetle's back structure consists of a unique structure of hydrophilic bumps and superhydrophobic background. Hydrophilic bumps capture atmospheric water molecules and then coalesce and roll down the superhydrophobic area under gravity. Thus, Zeng et al. have simulated the superwetting Stenocara beetle's back structure for collecting micro water droplets dispersed in oil [84]. The schematic of preparation of superhydrophilic bumps−superhydrophobic/superoleophilic stainless steel mesh (SBS-SSM) by electrostatic self-assembly and spin-coating methods is shown in Fig. 5B. The superoleophilicity and superhydrophobicity were achieved by depositing hydrophilic silica particles on SSM having pore size of 10 μm followed by spin-coating of a nanocomposite solution of fluoropolymer/SiO_2 nanoparticles. Hydrophilic bumps (due to hydrophilic silica particles) could capture tiny water droplets from emulsion that are smaller than the pore size of the stainless steel mesh. Due to superoleophilicity, oil permeates through the pores of SHP/SOL surface and gets separated (99.95%).

Immoderately small and large sized pores of materials are

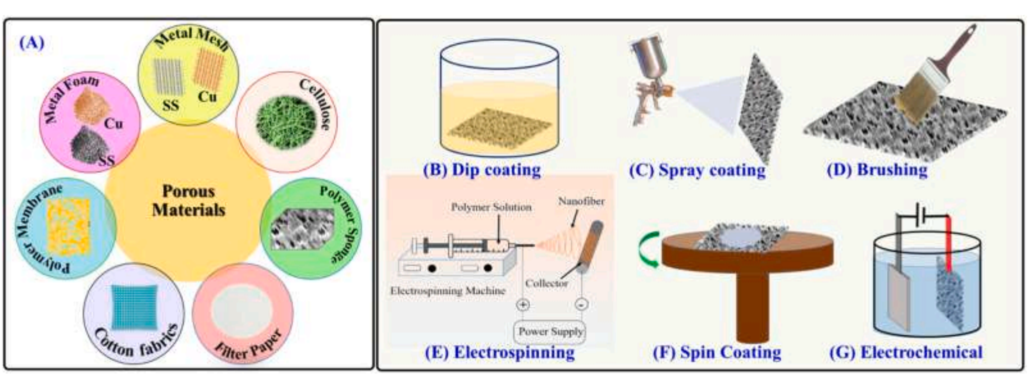

Fig. 3. (A) Schematic of various porous materials. (B-G) Common fabrication methods of superwetting porous materials.

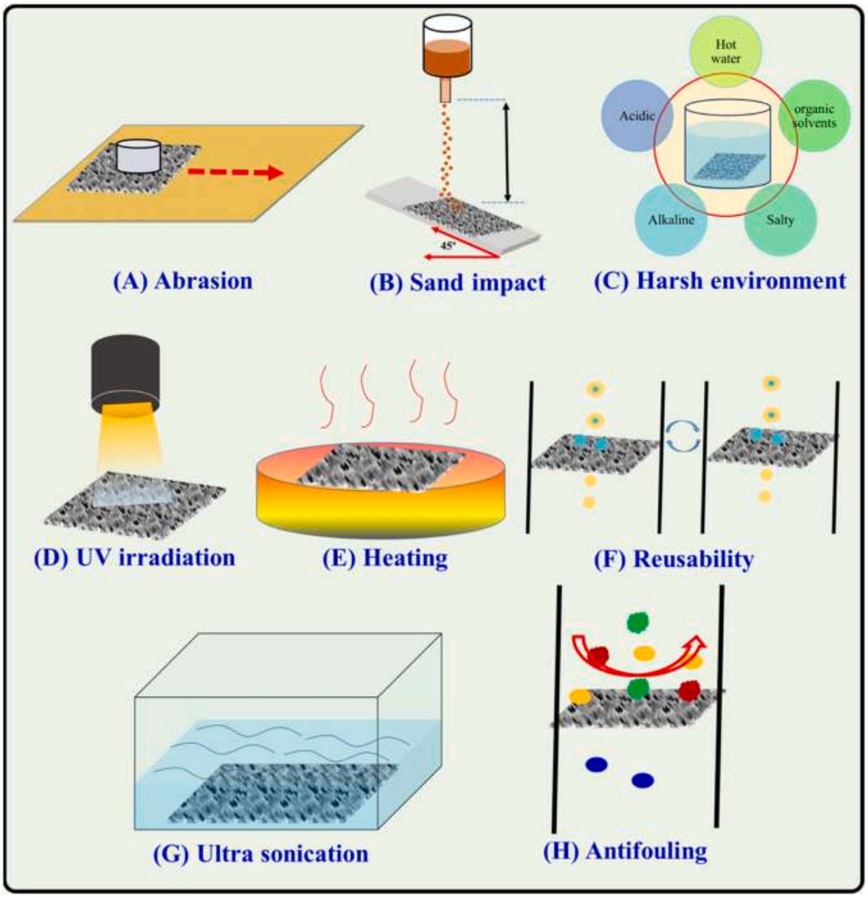

Fig. 4. (A-H) Common durability testing methods of superwetting porous materials.

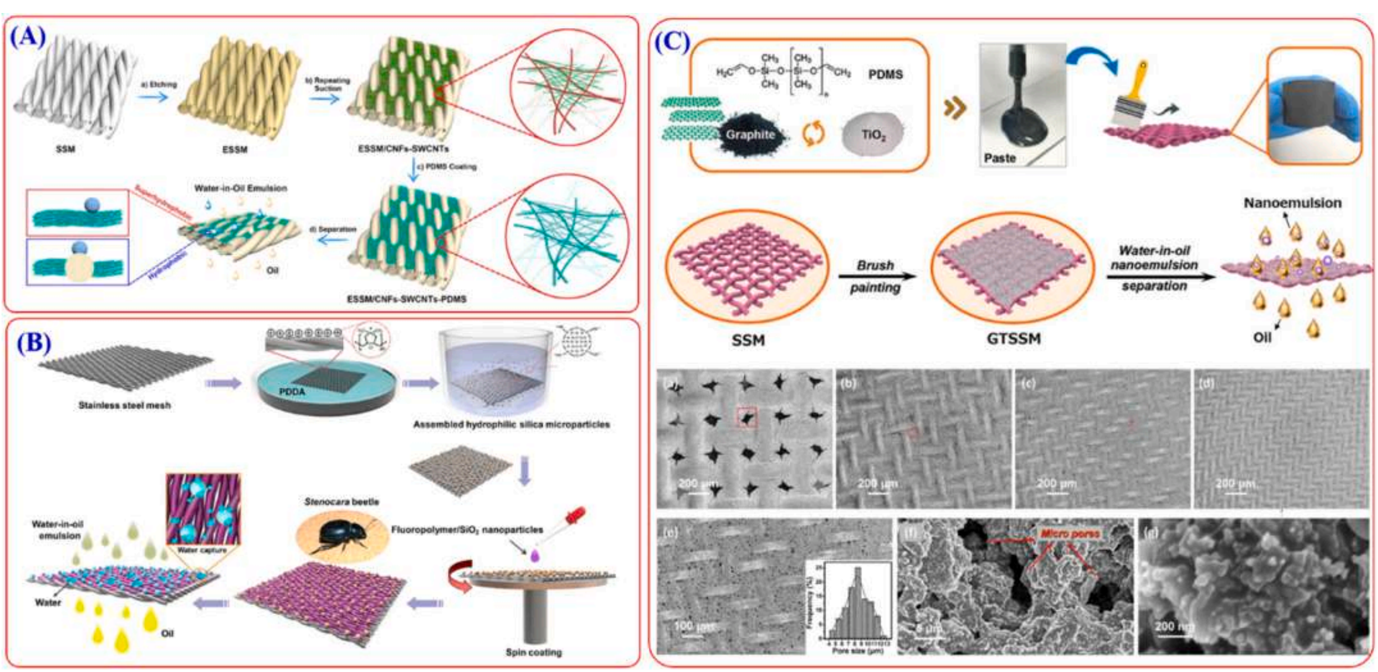

Fig. 5. (A) Schematic of embedded network structure of CNFs–SWCNTs–PDMS on mesh [83]. (Copyright 2017, American Chemical Society. Reproduced with permission from Ref. [83]). (B) Schematic of preparation of SBS-SSM by electrostatic self-assembling and spin-coating methods [84]. (Copyright 2017, American Chemical Society. Reproduced with permission from Ref. [84]). (C) Schematic of preparation of superhydrophobic coating on mesh and SEM images of coated mesh [85]. (Copyright 2019, American Chemical Society. Reproduced with permission from Ref. [85]).

inappropriate for emulsion separation. Du et al. have analyzed the emulsion separation performance against different pore sized SSM [85]. The superhydrophobic paste (graphite, TiO_2 nanoparticles, and polydimethylsiloxane (PDMS) was applied on SSM of different mesh numbers (80, 200, 300 and 400) having different pore diameters as shown in Fig. 5C. The micro- scale graphite and nano-scale TiO_2 were responsible to attain hierarchical micro-/nano- structures on mesh surface and PDMS reduces the overall surface energy, which can eventually enhance the superhydrophobic property of the coating. The 8-9 μm pore size obtained for the paste applied on 300-mesh, could separate W/O emulsion droplets (smaller than 8 nm) of gasoline, isooctane, xylene, dichloromethane, and n-hexane with separation efficiency of 99.9%. In Scanning electron microscopy analysis of coatings (Fig. 5C), it was observed that many rough and irregular protuberances conglomerated on the coating, which enhances oil absorption and water drops strike on oil wetted mesh surface. Along with superwettability and pore size control, mechanical robustness is considered for long time separation of oil-water emulsion. For the fabrication of mechanically robust superhydrophobic surface, Li et al. have used aluminum phosphate (AP) as an inorganic adhesive in the suspension of octadecyltrimethoxysilane-modified attapulgite and spray coated on SSM (Fig. 6 A**)** [86]. The roughness attained due to attapulgite nanorods and low surface energy by virtue of octadecyltrimethoxys executes SHP/SOL SSM. The Si-O bond on octadecyltrimethoxysilane breaks and Si atom makes new bonding with O atom of attapulgite, which eventually produces long-chain of hydrophobic alkyls on the attapulgite surface. Furthermore, there are several hydrogen bonds between the functional groups of the aluminum phosphate binder and the oxygen atoms on the attapulgite, which will aid to improve adhesion. Therefore, the prepared coating has exhibited more robust wetting properties (WCA>150°) against 200 sandpaper abrasion cycles and sand impact tests (Fig. 6 B and D**)**. Even after 200 sandpaper abrasion cycles, the mesh had showed outstanding emulsion separation (99.4%) (Fig. 6 C**)**. The hydroxyl groups and reactive double bonds (P=O) in AP create cross-linked structures, improving the attractive force and cohesion between nanoparticles and the substrate [87].

Further, polymer-based materials have been extensively utilized for emulsion separation due to their flexibility, controlling pore structure, and low corrosion possibility. The polymeric porous materials are mostly synthesized by electrospinning and phase inversion [88,89]. In the electrospinning technique, nanofibers attain porous structured membrane. In phase inversion, the porous morphology was developed by non-solvent induced phase separation (NIPS), solvent evaporation-induced phase separation (SEIPS) and thermally induced phase separation (TIPS). Poly (vinylidene fluoride) (PVDF) [90], poly (vinyl alcohol) (PVA) [91], polyurethane (PU) [92], poly(vinyl chloride) (PVC) [93] and poly(arylene ether nitrile) (PEN) [94] are mostly used for fabrication of porous polymeric materials. The pristine polymeric porous materials are hydrophobic in nature, but unsuitable for high wettability and oil-water emulsion separation. The addition of inorganic nanoparticles in polymer matrix substantially improves the wettability, emulsion separation ability and mechanical strength. For example, the addition of SiO_2 nanoparticles and micro-scale fluorinated ethylene propylene particles into the polysulfone/fluorinated ethylene propylene membrane resulted in a hierarchical roughness of 0.712 μm, which revealed a WCA of 153.3° and rolling angle of 6.1° [95]. This membrane effectively separated water-in-kerosene and water-in-diesel emulsions with high efficiency. Xiong et al. have attained PET membrane by electrospinning technique and immersed into ZnO seed solution followed by grafting of hydrophobic functional groups by dipping into sodium laurate solution (Fig. 7A) [96]. The SEM micrograph of PET membrane reveals that the PET membrane consists of smooth PET nanofibers which form porous network structure. After in situ growth of ZnO, nanofiber surface become rougher due to the coverage of ZnO nanopillars (Fig. 7B). The ZnO nanopillars, self-entanglement and network-like pores provide flow channel during emulsion separation. The as fabricated membrane exhibited excellent chemical stability in corrosive liquid and showed separation efficiency of 99.6% even after 10 separation cycles (Fig. 7C).

Compared with the two-dimensional superwetting materials, the three-dimensional structure yield space for the storage of oil droplets and its selective superwetting facilitates rapid and effective separation. For continuous and longtime emulsion separation, Xu et al. have designed a novel tubular-like three-dimensional nanofiber membrane using polyvinyl chloride (PVC) and hydrophobic SiO_2 nanoparticles (Fig. 7D) [97]. The tubular-like three-dimensional PVC nanofiber membrane was prepared using three-dimensional microspheres and two-dimensional nanofibers via electrospinning process. The

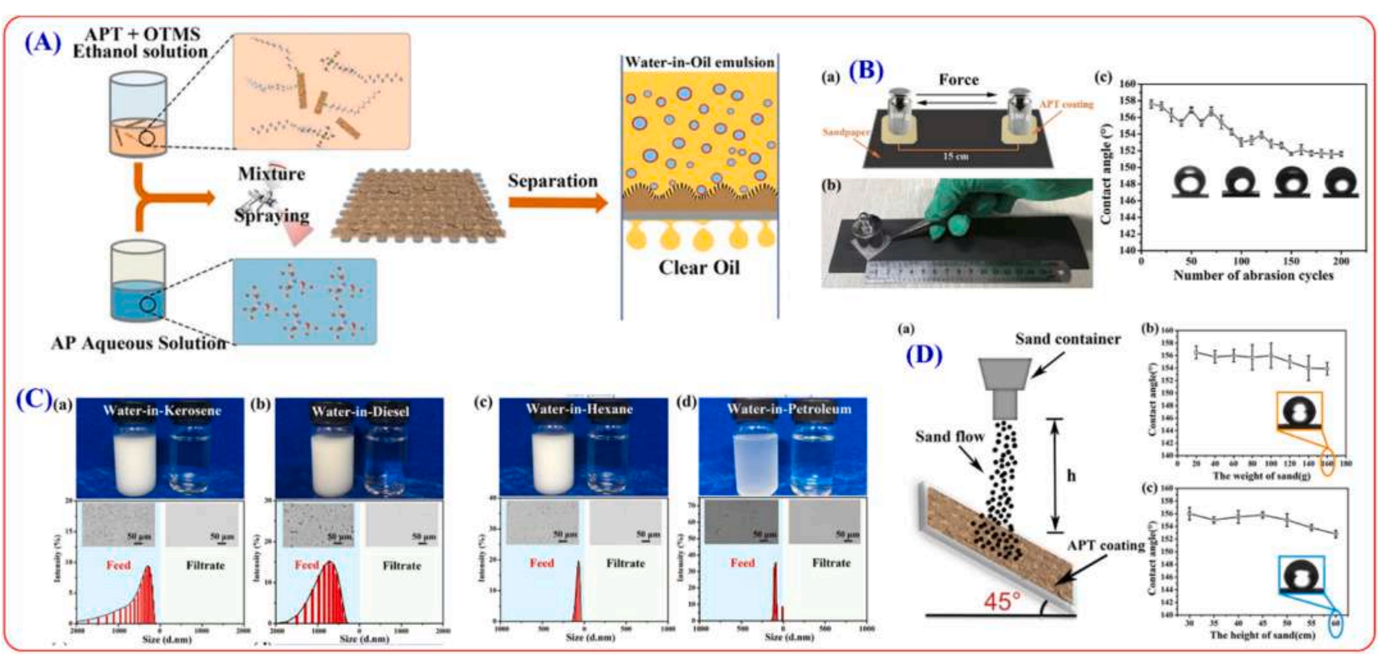

Fig. 6. The experiment process of coating (A), the evolution of mechanical durability (B and D) and the emulsion separation performance (C) of coated mesh [86]. (Copyright 2019, Elsevier. Reproduced with permission from Ref. [86]).

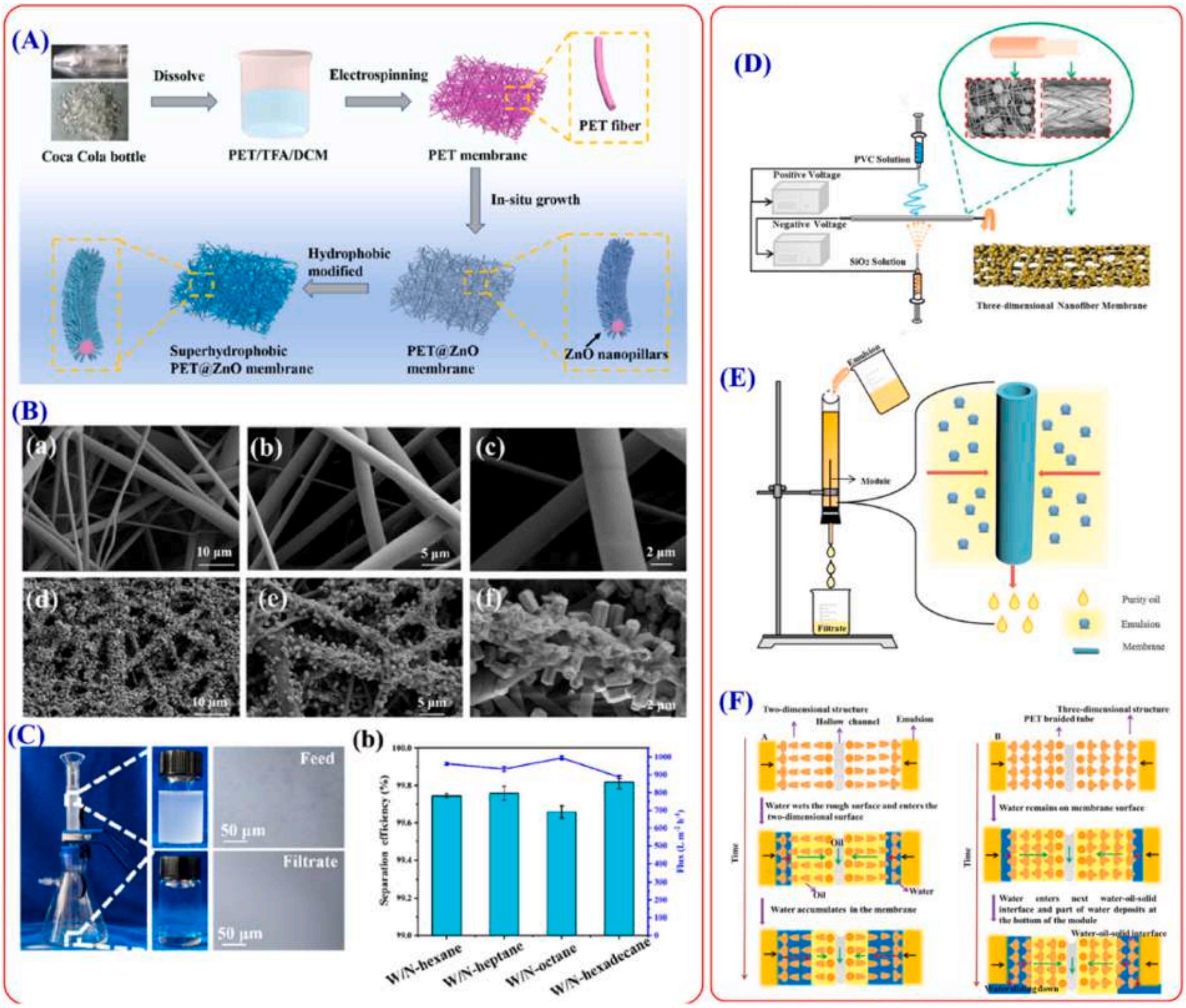

Fig. 7. Schematic of fabrication process of PET membrane (A), SEM images (B) and W/O emulsion separation performance (C) of PET membrane [96]. (Copyright 2022, American Chemical Society. Reproduced with permission from Ref. [96]). Schematic of experimental process of fabrication of PVC/SiO_2 membrane (D), emulsion separation device (E) and wetting behavior of two dimensional and three dimensional membrane (F) [97]. (Copyright 2021, Elsevier. Reproduced with permission from Ref. [97]).

hydrophobic SiO_2 nanoparticles were electrosprayed. The as-prepared membrane revealed lipophilicity and under oil superhydrophobicity. Fig. 7E shows the schematic of W/O emulsion separation device. Compared with two dimensional nanofiber membrane, three dimensional nanofiber membranes has fast oil wetting process due to continuous water-oil-solid interface and reduce possibilities of water wetting. After longtime emulsion separation process, the direction of water droplets flow changed from horizontal to vertical, as shown schematically in Fig. 7F.

A nepenthes pitcher plant-inspired slippery liquid-infused porous surfaces (SLIPS) have been designed for emulsion separation [98,99]. Excellent water repellency can be achieved by infusing lubricating oil into the rough micro/nanostructured surface, which enables water drop to slide off. Zhao et al. have developed a slippery liquid-infused membrane for W/O emulsion separation [100]. The hydrophobic trimethoxy (1H,1H,2H,2H-heptadecafluorodecyl) silane (FOTS) functionalized polyethylene terephthalate (PET) membrane with pore size of 4 μm and thickness of 270 μm was infused by oil to obtain a slippery liquid-infused membrane. Such membrane repels water and showed more than 98.6% separation efficiency for low viscous oil-water mixture and more than 99.4% for high viscosity oil-water mixture.

Nevertheless, the emulsion separation research has paid attention to aerogels [101–103], bio-waste materials [104], covalent organic framework (COF) [105] and metal-organic framework (MOF) membrane [106] in oil-water emulsion separation. Aerogels' unique wettability and 3-D porous structure enables emulsion droplet coalescence and the formation of larger droplets. Apart from this, the framework materials have highly tunable structures and functionalities towards wettability. Table 1 depicts W/O emulsion separation performance of recently developed oil absorbent superwetting porous materials.

3.2. Water absorbent superwetting porous materials

As discussed in the previous section, the reverse wetting property of porous materials towards water and oil makes them superhydrophilic (SHL) and superoleophobic (SOP), which can separate O/W emulsions. The unique hierarchical surface structure and a thin mucus layer on fish skin exhibit SHL/ underwater SOP [116]. To date, many efforts have been made on the design of SHL/underwater SOP porous materials for O/W emulsion separation. Using an in-situ chemical etching method, He et al. have developed a fish scale-like wettability on stainless steel mesh [117]. The vertically grown CuC_2O_4 nanosheet layer on mesh exhibited high water absorbing ability to firm the water layer into a rough surface and consequently showed superoleophobicity with OCA greater than 150°. A phenomenon of water capturing ability of desert beetle from atmosphere can be adopted to separate O/W emulsion separation by

Table 1
W/O emulsion separation performance of oil absorbent superwetting porous materials.

Materials	Method	flux L•m^{-2}•h^{-1}	Separation efficiency (%)	Number of cycles	References
Metal felt, glue and SiO_2	Dip	4000	99.7	30	[107]
SSM, Cu NPs, aluminum phosphate and Octadecanethiol	Spray	75	99.975	10	[108]
SSM, copper stearate/ Fe_3O_4 Nps	Dip	503	98	10	[109]
SSM, copper (II) chloride, sodium hydroxide, and polyurethane, and n-dodecanethiol	Dip	11.5	99.0	10	[110]
PVDF Membrane	Electro-spinning	88167	99	-	[111]
Carbon Membrane nanotube/poly(vinylidene fluoride-cohexafluoro trimethoxy (1H, 1H, 2H, 2H-heptadecafluorodecyl) silane propylene)	Electro-spinning	632.5	99.9	20	[112]
PET Membrane	Electro-spinning	1100	99.99	40	[113]
PVDF, PMMA, PAN	Electro-spinning	2408	92	5	[114]
SSM, CNFs-PDMS	Vacuum-based filtration	984	-	-	[115]
Copper foam, N-dodecyl mercaptan,	Electro-deposition	6560	99.89	30	[61]`

collecting oil droplets from an emulsion. Inspired by the desert beetle back structure, Li et al. have constructed an inverse desert beetle back-like structured membrane for the collection of oil from the emulsion [69]. This membrane consists of oleophilic ZIF-8 bumps and superoleophobic polyacrylonitrile (PAN), that exhibit a similar structure and opposite wettability that found on desert beetles. The bumps on the material can capture small oil droplets. The process of capturing and demulsifying oil droplets on the surface of ZIF-8 is attributed to both electrostatic attraction and hydrophobic interaction. The positive charge on the surface of ZIF-8 can attract negatively charged oil

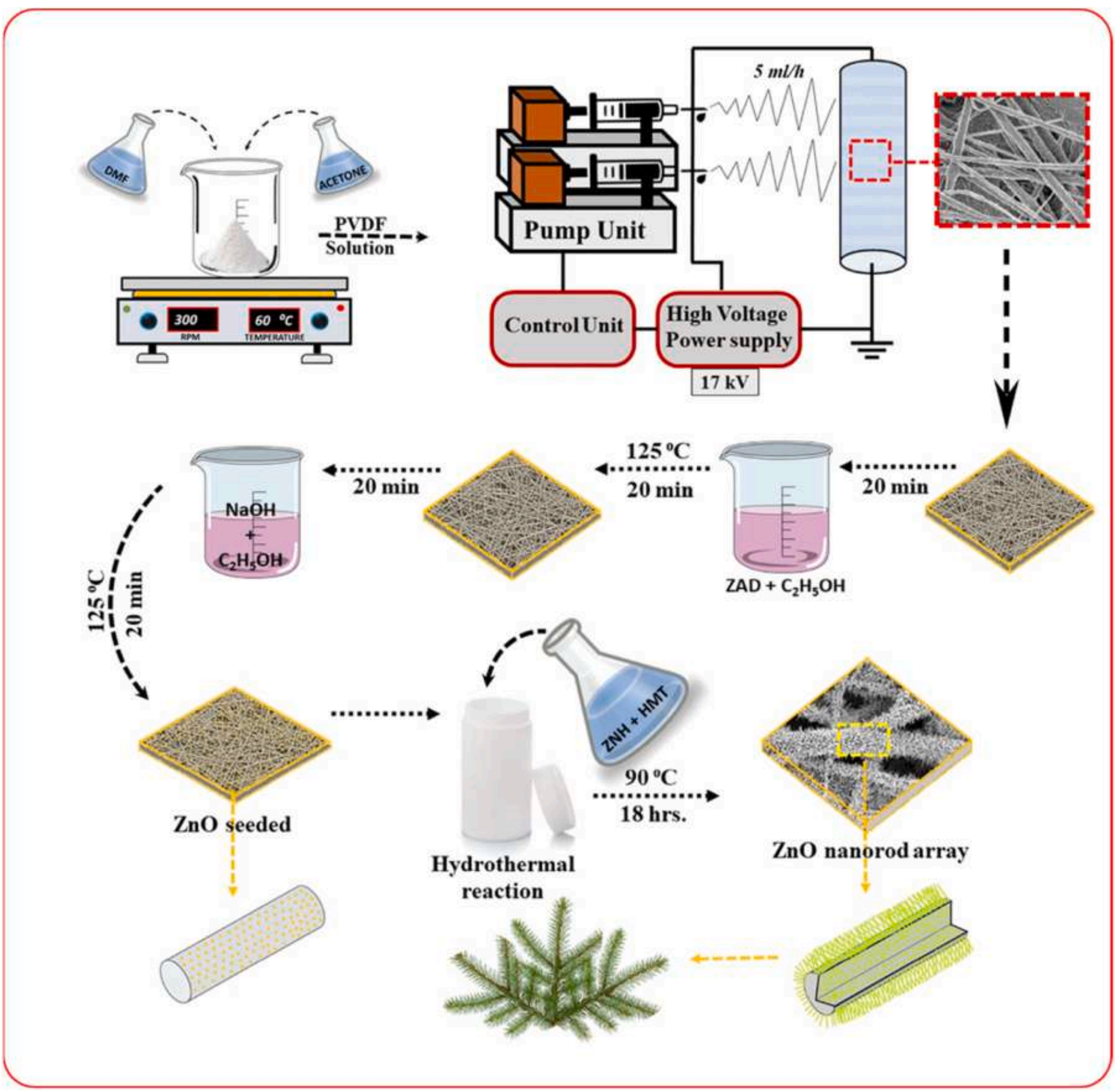

Fig. 8. Experiment fabrication process of nanofibrous PVDF@ZnO nanorod membrane fabrication [118]. (Copyright 2022, Elsevier. Reproduced with permission from Ref. [118]).

droplets, while the hydrophobic nature of ZIF-8 interacts with the hydrocarbon chains of the oil droplets, which leads to their adsorption. The captured oil droplets form a thin layer on the particle surface; at the same time, further oil droplets get adsorbed and aggregated. With time, size of the oil droplets grows and eventually detach from ZIF-8 particle and these large oil droplets bounce off the superhydrophilic PAN surface. ZIF-8/PAN membrane had showed 99.59% of O/W emulsion separation efficiency.

The electrospinning technique can be adopted to attain randomly oriented nanofibers of polymers that have a high surface-to-volume ratio and form ultrahigh porosity, which leads to a high permeation flux to liquids. The addition of hydrophilic nanomaterials in the membrane makes it best for separating O/W emulsions. Ahmed et al. have hydrothermally decorated ZnO nanorods on PVDF nanofibrous membrane (Fig. 8) [118]. Due to the high surface energy and rough micro-/nano structure of PVDF@ZnO nanorods, water droplets spread on the membrane and form an infused water layer on the surface; consequently, it repels oil. Due to the photocatalytic activity of ZnO, the membrane could easily decompose methyl orange and ciprofloxacin, which are major pollutants in organic dyes and pharmaceutical wastewater. The photocatalytic degradation of organic pollutants (stearic acid) by the membrane was due to the photocatalytic nature of ZnO existing in PVDF membrane. The WCA was measured to confirm photocatalytic degradation and self-cleaning capability. Such membrane exhibited high water flux and separation efficiency of 99% for O/W emulsion and exhibited good chemical stability in acidic, alkaline, hot, and cold surroundings.

In another work, Qing et al. have grown SiO_2 nanoparticles on the PVA nanofibers [119]. The hydrophilic PVA nanofibrous membrane was attained using PVA by electrospinning technique (Fig. 9A). During this process, the average diameter of nanofibers increased (from 460 nm to 770 nm); meanwhile, the pore size slightly decreased (532 ± 3 nm to 433 ± 5 nm). Such membrane efficiently separated kerosene-in-water emulsion having a droplet size of 1.36 μm. Membrane shows high water permeation flux, while n-heptane, kerosene, toluene, and chloroform are strongly repelled (Fig. 9B). Based on practical applications like wastewater treatment, authors have studied the effect of oil densities on emulsion separation performance. They concluded that the oils have lower densities than water and showed higher permeation flux and good oil rejection than higher-density oils. The oil density is one of the critical factors during the emulsion separation process; the separation of lighter and higher-density oil from water is hypothetically shown in Fig. 9C.

Compared with membrane materials, three dimensional superwetting porous materials are most promising in O/W emulsion separation [120]. Due to high porosity, these materials could collect more oil droplets from emulsion and exhibited high separation efficiency. Recently, Li et al. have prepared phenolic-resin-based sponge composed of entangled nanofibers using phenol, 2-aminobenzimidazole, and CTAB via a simple hydrothermal method [121]. He et al. have prepared a magnetic SHP sponge using Fe_3O_4 particles and stearic acid through immersion [122]. The magnetic SHP sponge drove magnetically to collect oil from the O/W emulsion, avoiding direct contact with toxic and hazardous solvents. The as-prepared sponge showed more than 90% of separation efficiency, with good chemical resistance (soaking for 7 days) and good mechanical stability against 80 times cyclic compression by 200 g of weight. Table 2 shows the list of recently developed water absorbent superwetting porous materials for O/W emulsion separation.

3.3. Demulsification

As discussed in previous sections, superwetting technologies for separating oil-water emulsions have several issues, such as fouling, pore blocking, and inadequate mechanical strength. Therefore, these techniques slightly fail to produce satisfactory results. However, demulsification is a promising method that overcomes the drawbacks of previous methods. Unlike chemical and heating methods, demulsification based on superwetting is fast, inexpensive, and eco-friendly. This method uses the coalescence effect to merge emulsion droplets into larger ones before separating them. Materials with selective wettability capture and coalesce emulsion droplets, forming larger droplets that eventually detach from the surface. Water droplets coalesce readily on a SHL surface, while

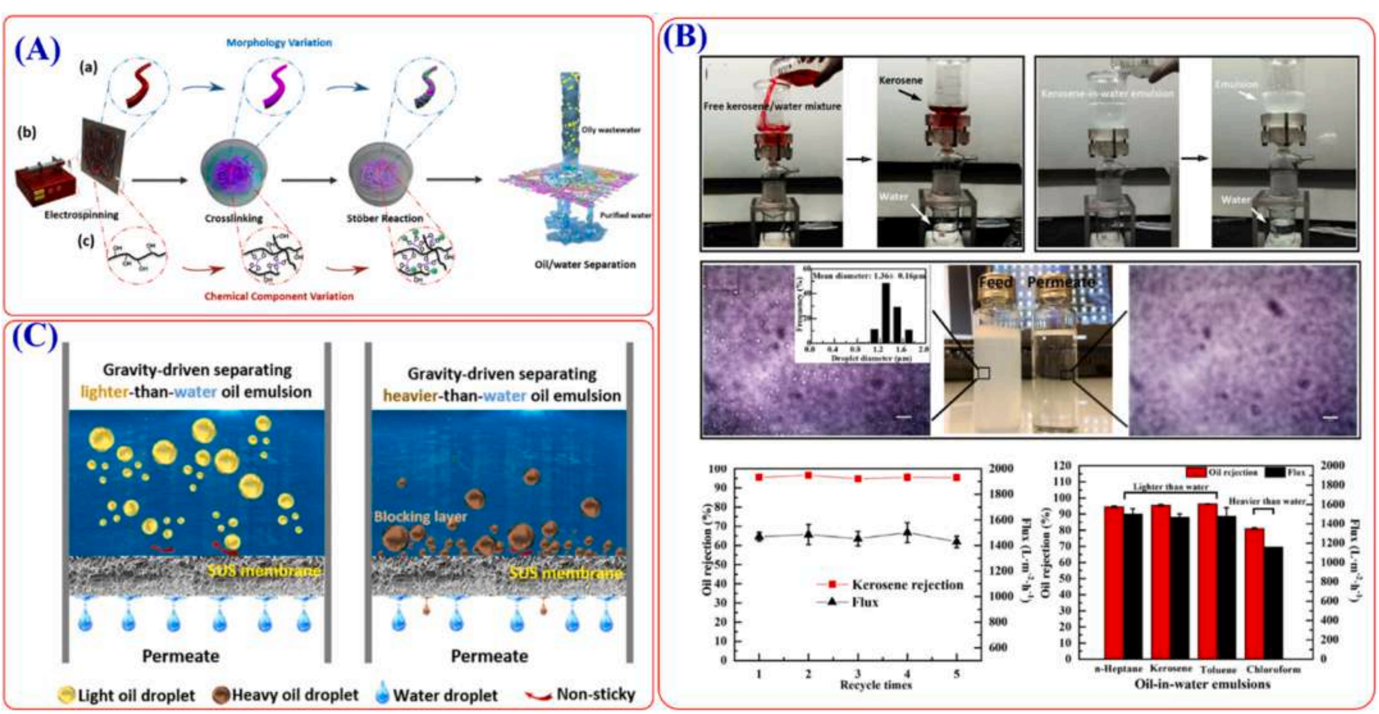

Fig. 9. (A) Schematic representation of experimental procedure and variation of morphology and chemical components during preparation. (B) Oil/water emulsion separation performance of SiO_2@PVA membrane. (C) Hypothetical illustration of emulsion separation based on different densities of oil in the emulsion [119]. (Copyright 2020, Elsevier. Reproduced with permission from Ref. [119]).

Table 2
O/W emulsion separation performance of water absorbent superwetting porous materials.

Materials	Method	flux $L\bullet m^{-2}\bullet h^{-1}$	Separation efficiency (%)	Number of cycles	References
SSM, polyurethane@graphitic-carbon nitride@perflourooctanoic acid	Dip coating	50.5	99		[123]
Cellulose acetate membrane, chitosan and TiO_2	vacuum-assisted filtration technique	6002.5	97	10	[124]
SSM, TiO_2 nanowires	Casting	5859.54	99.1	30	[125]
Glass fibrous membrane, poly (diallyldimethylammonium chloride), TiO_2	Dip	2329	94.75		[126]
Wood membrane, sodium hydroxide, sodium sulfite and hydrogen peroxide	Chemical treatment	460.8	99.43	10	[127]
3D cellulose sponge, Row cotton, Zinc chloride, sodium sulfate	Freeze drying	290	99.3	10	[128]

oil droplets coalesce on a SOL surface [129]. Emulsion droplets move toward the surface of the coalescing medium due to hydrodynamic, electric, or other forces and coalesce to form a layered oil/water mixture that can be easily separated. Recently, Wang et al. have developed a PVDF membrane with a dual-scale hyperporous structure that can continuously separate oil-water emulsions by demulsification (Fig. 10A) [130]. The membrane has two layers with different pore sizes: the exterior layer has pores ranging from 0.5 to 2.5 μm, while the interior layer has pores ranging from 0.1 to 0.3 μm. The membrane revealed a WCA of 59 ± 1° and an OCA of 174 ± 1°. It showed a high permeability of 1078 ± 50 $L\bullet m^{-2}\bullet h^{-1}.bar^{-1}$ and can separate toluene-in-water and D5-in-water emulsions with 99% efficiency under 1.0 bar pressure.

Cao et al. have developed a SHP nanofiber membrane (NFM) made of polytetrafluoroethylene (PTFE) using electrospinning and electrospraying techniques [131]. The membrane was capable of separating O/W and W/O emulsions through demulsification. The PTFE and polyacrylonitrile (PAN) solution was spun out simultaneously by an electrospinning machine to create the PTFE NFM. Subsequently, the PTFE solution was sprayed onto the NFM using electrospray, forming microspheres of PTFE, which get attached firmly on the NFM. This NFM was finally sintered at 380 °C for 10 minutes (Fig. 10B). The developed NFM showed a flux of 1867.93 $L\bullet m^{-2}\bullet h^{-1}$ and a separation rate of 98.46% for W/O emulsions and a flux of 122 $L\bullet m^{-2}\bullet h^{-1}$ and a separation rate of 98.40% for O/W emulsions in multiple cycles. In the demulsification process, oil droplets quickly spread and completely wet the fiber surface, while the water phase flowed through the NFM continuously under pressure. As continuous pressure drove the attached tiny oil droplets to aggregate into larger oil droplets, the aggregated oil droplets would fall off the attaching site, pass through the NFM and seep out from the bottom. However, new oil droplets would attach to the locations where the previous ones were attached, and oil passes through NFM under pressure. The aggregated oil droplets and permeated water form a stable layered oil/water mixture. The demulsification performance is illustrated in Fig. 10B. The comparative survey of the performance of demulsification based on superwetting materials is summarized in Table 3.

3.4. Superamphiphilic porous materials

Porous materials which are superamphiphilic in nature can absorb both oil and water, making them useful for separating W/O and O/W emulsions. These materials can change their wettability between

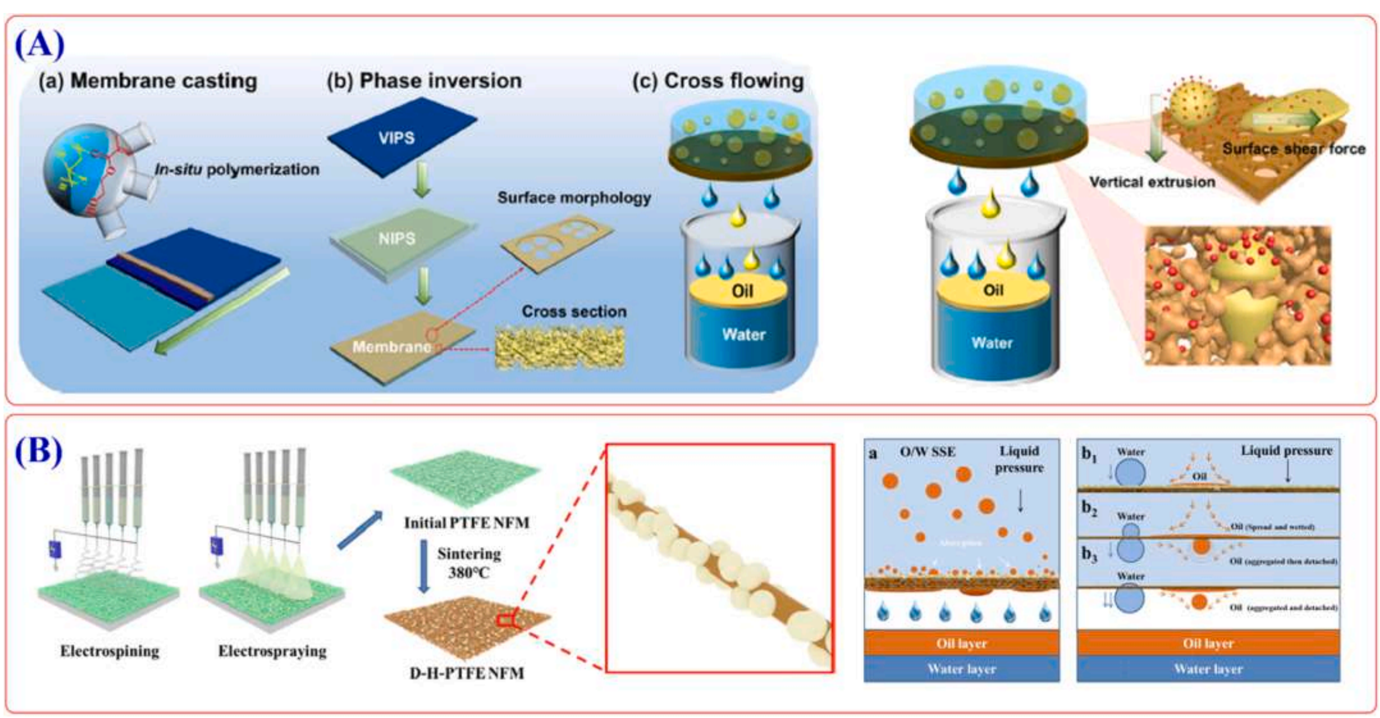

Fig. 10. (A) Schematic representation of the membrane fabrication and membrane demulsification process of PVDF membrane [130]. (Copyright 2021, American Chemical Society. Reproduced with permission from Ref. [130]). (B) Schematic illustration of preparing PTFE electrospun NFM and converting O/W emulsion to stratified oil/water mixture through demulsifying tiny oil droplets under pressure [131]. (Copyright 2022, Elsevier. Reproduced with permission from Ref. [131]).

Table 3
Water-in-oil emulsion separation performance based on demulsification.

Materials	Method	flux L•m^{-2}•h^{-1}	Separation efficiency (%)	Number of cycles	References
SSM, poly-(N,N-dimethylaminoethyl methacrylate) and poly (divinylbenzene)	Immersion	1200	99.32	10	[132]
Copper mesh, TiO_2	Immersion	750	99.97	20	[46]
PVA/SiO_2	Electro-spinning	2398	-	10	[133]
Glass wool, glass fiber and cellulose fiber, thermoplastic polyurethane resin		11,000	99.3		[134]
Cotton fabric, TiO_2, Silver nitrate, octadecyl trichlorosilane,	spray	924.16	98.99	60	[135]
Melamine sponges, asphaltenes and polyethyleneimine	Dip	15945	98.7	10	[136]

superhydrophobic and superhydrophilic states due to external factors like pH value, temperature, electric field, and light illumination. The selective oil-water emulsion separation was achieved based on these materials.

Using a simple, eco-friendly, and cost-effective electrochemical method, Lei et al. have created a superamphiphilic SSM [137]. The process involved electroplating of the mesh with $CuSO_4$ and H_2SO_4 solution and then anodic oxidation with NaOH solution. These reactions could change the surface wettability of the mesh to superamphiphilic in nature. The superamphiphilic mesh had over 99% separation efficiency for O/W and W/O emulsions. Additionally, it showed good mechanical resistance against sand impact tests and anti-fouling against reusable experiments. In another work, amphiphilic bamboo powder and PVDF composite foam was successfully used to create underwater superoleophobicity and underoil SHP [138]. The foam was prewetted with water and can repel oil droplets in an O/W emulsion. The unstable oil droplets coalesce with each other and form larger oil droplets on the foam surface. The coalesced larger oil droplets then float towards the water surface from the low adhesion foam surface and form an oil layer. Table 1 gives a comparative survey on the performance of oil-water emulsion separation by recently developed superamphiphilic porous materials. Table 4

4. Challenges in practical applications

The previous sections have covered various methods for separating oil-water emulsions using superwetting porous materials. Though these methods are effective, the short lifespan of these materials limits their use in industrial applications. There are still challenges in real-time applications of superwetting materials for oil-water emulsion separation, such as recyclability, durability, and resistance to fouling. These issues and potential solutions are discussed in detail.

4.1. Recyclability

Superwetting materials can be evaluated for their long-term usability through cyclic separation tests. These materials have a high affinity for liquids, which quickly get adsorbed. In the continuous wetting process, superwetting surfaces are diminishing. As a result, the separation efficiency and permeation flux decrease. In most of the studies, superwetting porous materials are washed/re-modified after every emulsion separation cycle to maintain permeation flux, which is a hectic process.

For example, Ye et al. have modified multiwalled carbon nanotube film (MWCNT) by zeolitic imidazolate framework (ZIF)-8 and Co-polydimethylsiloxane (Co-PDMS) to obtain SHP/SOL membrane (Fig. 11 A-B) [147]. In situ, growth of ZIF-8 formed a micro-/-nanolayered structure on the MWCNT and Co-PDMS coating, which provides 0.62 μm of average pore size. Four types of W/O emulsions were prepared using chloroform, toluene, xylene and n-hexane with span-80 as an emulsifier. In the recyclability test, an equal amount of emulsion was used for the separation test and the membrane was washed by ethanol during the cyclic interval. Even after ten separation cycles, the separation efficiency (>99.9%) and flux (177.6 L•m^{-2}•h^{-1}) remained unaltered. Zhang et al. have fabricated a tubular composite membrane by electrospraying PVDF/graphene (GE) composite into an electrospun PVDF nanofibrous network. The combination of PVDF micro/nanospheres and GE nanosheets with the PVDF nanofibrous network produces a 3D rough composite membrane [148]. A laboratory scale setup was designed to separate W/O emulsions using a vacuum suction process (Fig. 11 D). The separation efficiency of the membrane was up to 99.8%, and it sustained its separation efficiency up to 10 cycles for four types of emulsions, namely, water-in-kerosene (W-K), water-in-toluene (W-T), water-in-chloroform (W-C), and water-in-diesel (W-D). After each cycle, the membrane was washed with ethyl alcohol and dried in the oven for reuse. In conclusion, superwetting porous materials are found promising for oil-water emulsion separation, but need improvement in recyclability for practical applications.

4.2. Mechanical durability

Mechanical durability is one of the important factors of superwetting materials that limit its application in oil-water emulsion separation. The superwetting ability of porous materials is influenced by their surface microstructure. The mechanical contact or continuous oil-water

Table 4
Water-oil emulsion separation performance based on superamphiphilic porous materials.

Materials	Method	flux L•m^{-2}•h^{-1}	Separation efficiency (%)	Number of cycles	References
SSM, zeolitic imidazolate framework-8/PVA	Immersion	457.6	98.6	10	[139]
Polyurethane/fluorine-modified silica	Electro-spinning	428	99	10	[140]
Candle soot, waterborne polyurethane (PU)	leaching	75	99.9	7	[141]
Poly(N,N-dimethylamino-2-ethyl methacrylate)		-	99.6	-	[142]
Graphene oxide (GO), PVDF	vapor-induced phase separation	-	98	-	[143]
Cellulose diacetate, Sodium hydroxide	Electro-spinning /immersion	38,000	96.9	10	[144]
Polydopamine (PDA)/Cu/Zn, SiO_2 and $Zn(OH)_2$ nanoparticles	Electro-deposition/dip	115.97	99.91	10	[145]
Nitrocellulose membranes, Dopamine hydrochloride, Tris(hydroxymethyl) aminomethane and polyethylenimine	immersion	1400	99.9	10	[146]

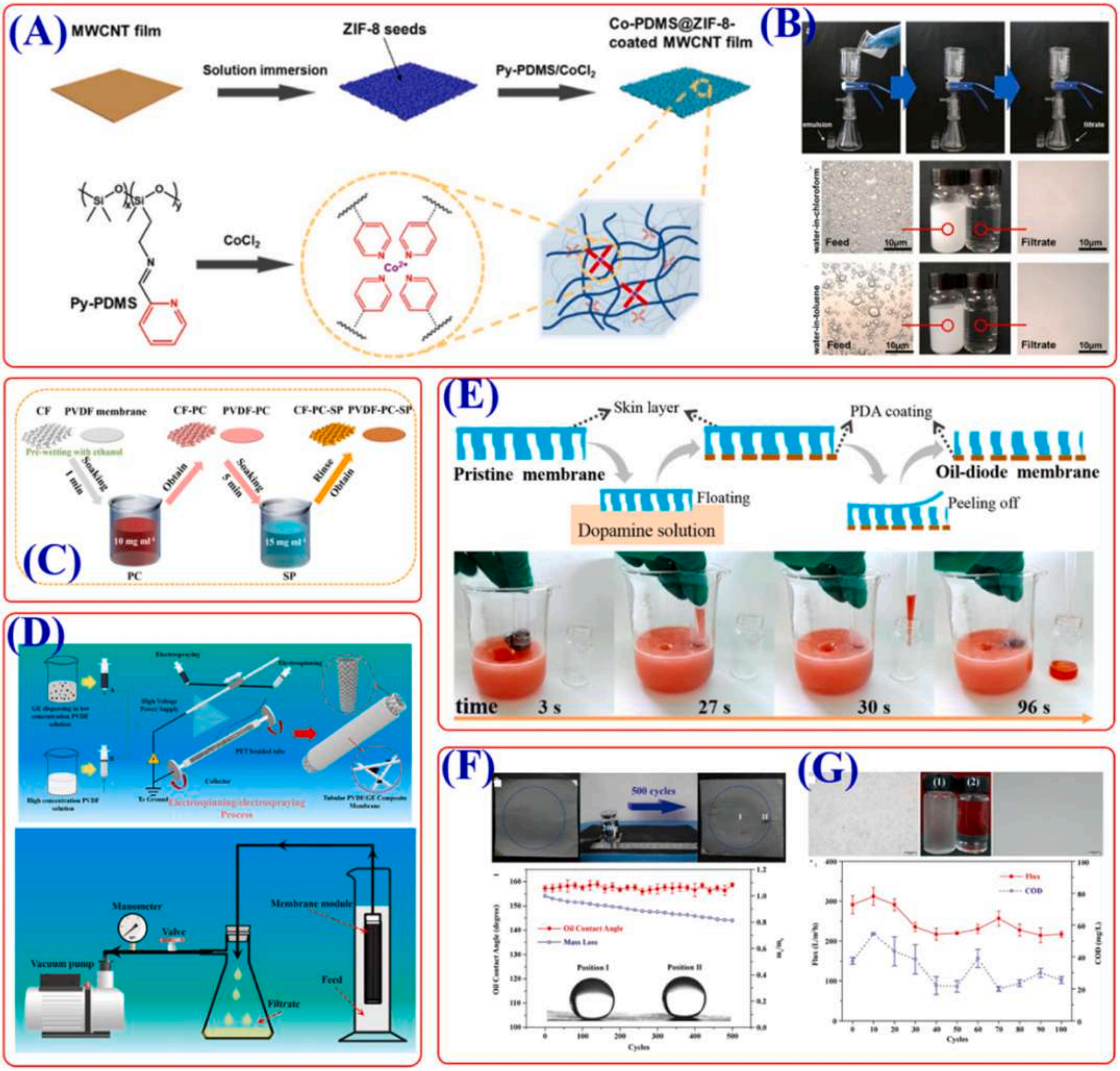

Fig. 11. (A-B) Schematic of the Fabrication of the ZIF-8- and Co-polydimethylsiloxane (PDMS)-Modified MWCNT Films and photographs of the water-in-toluene emulsion separation process [147]. (Copyright 2019, American Chemical Society. Reproduced with permission from Ref. [147]). (C) The fabrication process of the SHL CF-PC-SP and PVDF-PC-SP [149]. (Copyright 2022, Elsevier. Reproduced with permission from Ref. [149]). (D) Schematic illustrating the fabrication process of the tubular PVDF/GE composite membrane and continuous W/O emulsions separation setup [148]. (Copyright 2021, Elsevier. Reproduced with permission from Ref. [148]). (E) Schematic fabrication illustration of oil-diode Janus membrane (JM-O) via the floating coating and peeling off strategy and the snapshots about trapping oil from SDS stabilized toluene-in-water emulsion by a homemade collector capped by the JM-O (the hydrophilic layer faced outward) at one end [150]. (Copyright 2018, Elsevier. Reproduced with permission from Ref. [150]). (F-G) Wetting behavior and separation performance of mesh after abrasion test [151]. (Copyright 2017, American Chemical Society. Reproduced with permission from Ref. [151]).

emulsion separation can damage the micro/nanoscopic hierarchical rough architecture, causing the surface to become smoother and lose its superwetting state. Consequently, it fails in oil-water emulsion separation. The mechanical robustness was determined by various mechanical tests such as abrasion, bending, and ultrasonication [152,153]. Therefore, highly robust superwetting porous materials are on demand for practical applications. To address this problem, Liu et al. have produced superwetting coating on a stainless steel mesh by spraying a titanium dioxide (TiO_2) suspension in combination with aluminum phosphate (AP) as an adhesive [151]. The mechanical durability of the stainless steel mesh was assessed through a sandpaper abrasion test. The mesh was dragged on sandpaper (grit no. 320) under 200 g weight by an external force for 10 cm and then returned to complete one cycle. Even after 500 sandpaper abrasion cycles, the mesh maintained its superwettability. The as-prepared TiO_2-AP coated mesh was used to separate three types of O/W emulsions prepared by mixing oil (hexane, petroleum ether, and isooctane) in water. This mesh sustains a separation efficiency of more than 99.98% even after 100 abrasion cycles (Fig. 11

F-G). Additionally, the mesh showed excellent photocatalytic self-cleaning and anti-fouling characteristics against oleic acid and oleic oil, and remained stable up to 100 abrasion cycles. This study demonstrates that combining inorganic adhesives with functional nanomaterials can extend the service life of superwetting materials for practical applications.

4.3. Chemical stability

Surface energy, along with the surface structure of porous materials, plays a significant role in determining the affinity of the surface of a material to oil and water. However, in practical applications, harsh chemical environments such as acidic, alkaline, salty, and organic solvents can destroy specific functional groups that alter the wetting characteristics of superwetting materials. Therefore, developing chemically stable superwetting materials is crucial to maintain their long-term wetting characteristics.

He et al. have fabricated a chemically stable SHP paper for separating W/O emulsions [154]. The polydopamine (PDA) layer was applied on the paper by dipping in PDA solution, followed by dipping in TiO_2 nanoparticles suspension. Finally, the TiO_2/PDA paper was modified using n-hexane solution of 1H,1H,2H,2H-perfluorodecyltrichlorosilane (FDTS) to achieve superhydrophobicity. The SHP paper was kept in different solvents, acidic and alkaline solutions for different times to determine its chemical stability. The SHP paper has maintained water contact angles (WCA) of over 158°, even after being immersed in 3.5 wt % NaCl solution, n-hexane, ethanol, and DMF for 60 minutes. When immersed in HCl and NaOH for 120 minutes, the WCAs remained greater than 151°, indicating the high chemical stability of the SHP paper. Besides, SHP TiO_2/PDA paper showed thermal stability, mechanical durability and high emulsion separation efficiency.

4.4. Fouling resistant

The problem with porous material is that the pore sizes are too small, causing liquid droplets to get stuck and obstruct permeation, leading to fouling. Surfactant-stabilized emulsions can also contain contaminants contributing to fouling [155–160]. Therefore, anti-fouling surfaces must be designed.

Zhang et al. have fabricated anti-oil fouling SHL PVDF membrane for oil-water emulsion separation [149]. The PVDF membrane was first dipped into a proanthocyanidin buffer solution prepared in tris-hydrochloride and then dipped in an aqueous sodium periodate solution. (Fig. 11C). The SHL PVDF membrane showed a maximum flux and efficiency of 108 $L{\bullet}m^{-2}{\bullet}h^{-1}$ and 99.7% under gravity-driven O/W emulsion and under external pressure, it showed 681 $L{\bullet}m^{-2}{\bullet}h^{-1}$ and 99.5%. The flux and efficiency remained the same even after five cycles of separation. The authors also studied the oil-fouling-resistant properties and found that the prepared SHL fabric and membrane had excellent anti-oil-fouling properties.

4.5. Universal strategy

The gravity-driven emulsion separation method is a cost-effective and easy approach that does not require external energy. It is also highly efficient in separation compared with other methods that rely on external pressure. However, it has some limitations when used in large-scale applications or harsh conditions. Hence, the design of superwetting materials needs to be improved significantly to achieve better results.

Yang et al. have developed a Janus oil-unidirectional membrane to separate O/W emulsion [150]. A Janus membrane has a thin hydrophilic layer and a thick hydrophobic layer over hydrophobic PET/PTFE composite porous membrane. The hydrophilic layer was prepared by floating the membrane on dopamine hydrochloride and tris(hydroxymethyl) aminomethane solution, while the other side was glued with adhesive tape. A homemade collector was constructed with a transparent tube capped by a Janus membrane with a hydrophilic layer towards the emulsion for oil collection. Before collection, the hydrophilic layer was pre-wetted with water, and the hydrophobic layer was wetted with oil. The collector was dipped into the emulsion for oil collection. The fabrication of the Janus membrane and collection of oil is shown in Fig. 11 E.

Sheng et al. have coated the melamine formaldehyde (MF) sponge by iron (III) chloride solution and then modified using PDMS through an immersion technique [161]. The sponge was then loaded into a stainless steel cylinder, where it was compressed to achieve an average pore size of about 45 μm. On a large-scale experiment, the sponge could separate 2 L of 1000 ppm O/W emulsion composed of refined mineral oils with hydrocarbons from C10 to C34 and additives. The separation efficiency reached to 98.8% in 10 minutes and up to 99.8% in 30 minutes. The O/W emulsion separator and separation result is shown in Fig. 12 (A-B). This ultrahigh efficiency makes it ideal for decanting applications, where recovered water on a temporary storage device may be decanted if the total petroleum hydrocarbons are below a specific amount, for example, 15 ppm. The sponge also exhibited long-term robustness, remained stable for up to 10 cycles, and separated 40 L of emulsion.

Tuteja et al. have used the dip coating technique to create SHL and underwater SOP porous membranes for continuous oil-water emulsion separation [162]. The porous substrates, including stainless steel, polyester fabric, and filter paper, were first coated with hydrophilic poly (ethylene glycol) diacrylate (PEGDA) and then modified with low surface energy material 1H, 1H, 2H, 2H - Heptadecafluorodecyl polyhedral oligomeric silsequioxane (F-POSS). The PEGDA-coated substrate exhibited HL/OL; while coated with (F - POSS), the surface becomes SHL/SOP. They used a gravity-assisted capillary force-driven separation (CFDS) device consisting of a membrane sandwiched between two vertical glass tubes to separate O/W emulsion and W/O emulsion. The membrane showed continuous separation of both types of emulsion for over 24 hours with self-cleaning and fouling-resistance ability. This work was registered under United States Patent in 2019.

5. Conclusions and future perspectives

The challenge of separating oil-water emulsions is a global concern. The use of superwetting porous material provides a promising solution to address this challenge. The superwettability and pores of materials are crucial in separating oil-water emulsions. This review presented the latest advancement in fabricating superwetting porous materials that can effectively separate oil-water emulsions with ease. The SHP/SOL and SHL/underwater SOP surfaces are used to separate W/O and O/W emulsions, respectively. Several factors, such as separation effectiveness, flux rate, antifouling, reusability, physical stability and chemical stability have proven that superwetting porous material is an excellent candidate for emulsion separation. However, there is limited literature on the practical application of separating oil-water emulsions using superwetting porous materials. The practical products are expected to be cost-effective, optimum separation efficiency, switchable wettability, multifunctional features, multiple reusabilities, and easy fabrication processes for smart superwetting porous materials and operation.

The future perspectives of superwetting materials for oil-water emulsion separation are outlined and graphically represented (Fig. 13),

1) The superwetting porous materials at the lab scale showed excellent oil-water emulsion separation ability. For real-life use, it is necessary to produce them in an environmentally friendly and scalable manner. For large-scale application of superwetting porous materials for oil-water emulsion separation, it's essential to simplify the synthesis process and reduce preparation costs.
2) Gravity-driven oil-water emulsion separation is fascinating field that expands the application of superwetting porous materials. Additionally, chemically stable, anti-fouling and robust superwetting

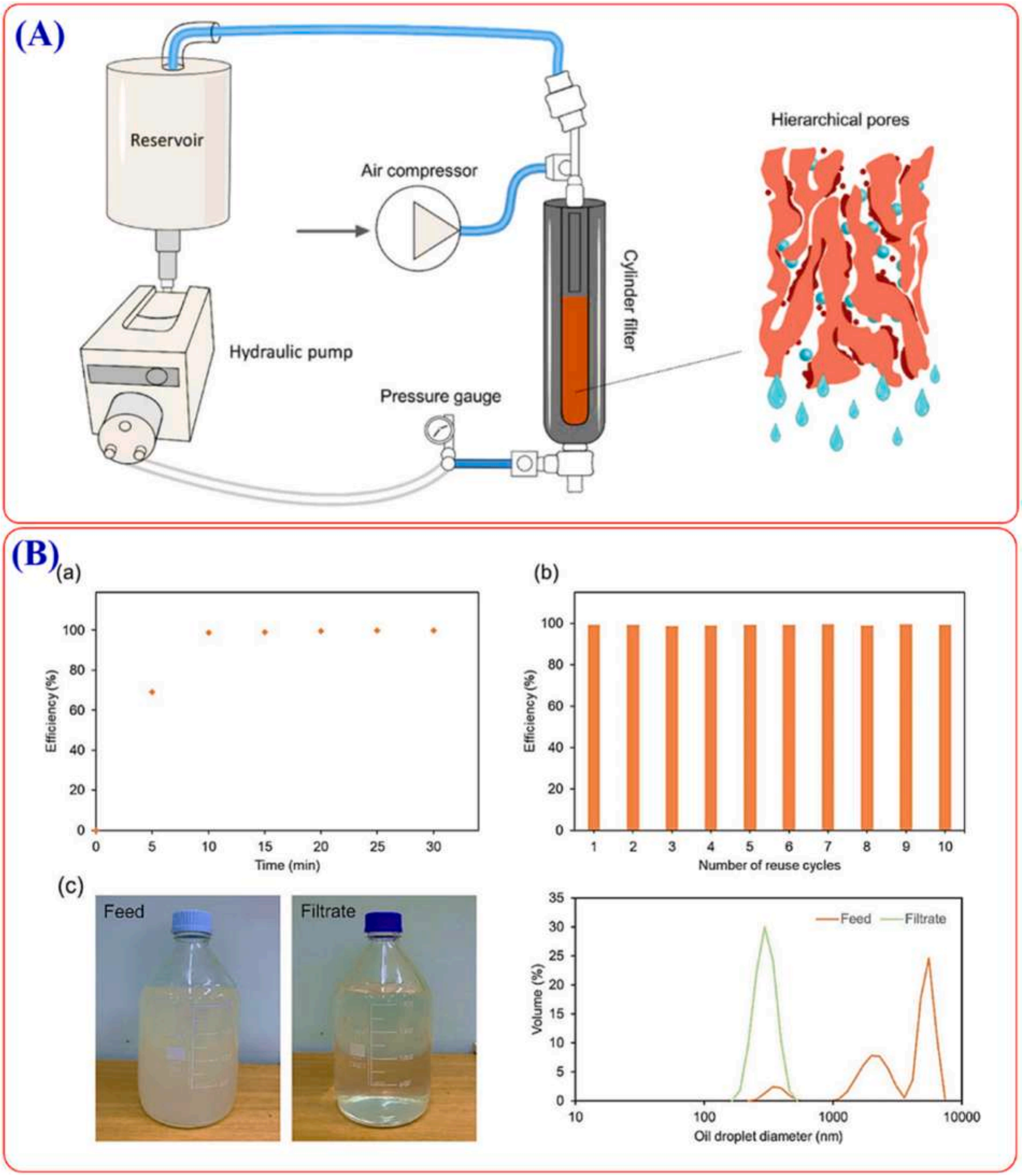

Fig. 12. (A) Schematic of an O/W emulsion separator with an HCP sorbent filter. (B) O/W separation efficiency of MF-HCP-PDMS as a function of (a) time and (b) number of reuse cycles. (c) Picture of before and after emulsion separation and the size distribution of emulsions in the feed (average 3.6 μm) and filtrate (average 0.35 μm) [161]. (Copyright 2022, American Chemical Society. Reproduced with permission from Ref. [161]).

porous materials should be extended their application in oil-water emulsion separation.

3) The phenomenon of superwetting occurs at the liquid-solid interface, and surface structure can be easily damaged by repeated separation, fouling, surface contamination (microorganisms, inorganic salts, and dyes), and physical and chemical contact. To address these challenges, a multifunctional surface that is self-cleaning, self-repairing, and photocatalytic must be designed.
4) Until now, most superwetting materials have effectively separated low-viscosity oil-water emulsions. They often fail to separate highly viscous oil and oily wastewater. It is crucial to enhance demulsification technologies by combining selective pore size and wettability of porous materials to separate high-viscosity oil-water emulsions effectively.

Declaration of Competing Interest

The authors declare that they have no known competing financial interests or personal relationships that could have appeared to influence the work reported in this paper.

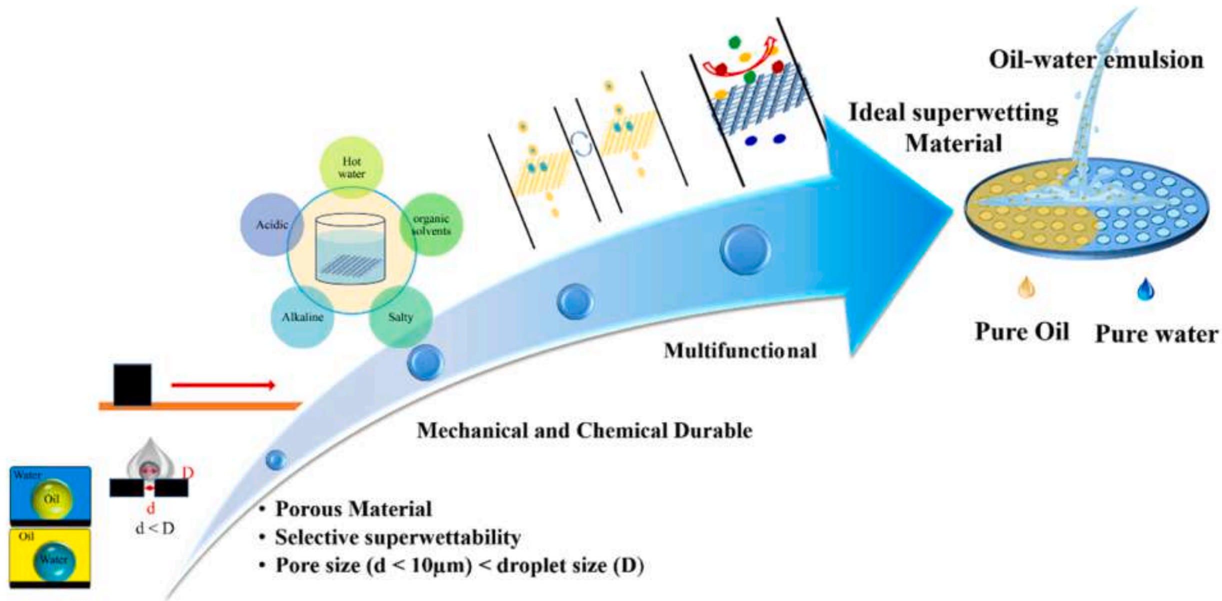

Fig. 13. Future outlook of porous superwetting materials for smart oil-water emulsion separation.

Data Availability

No data was used for the research described in the article.

Acknowledgments

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