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REVIEW

Recent Advances in Interfacial Engineering of Bio-Based Polymeric Materials for Oil/Water Separation: From Surface Chemistry to Separation Performance

Jia Li*, Siyuan Liu and Zixuan Guo

School of Materials Science and Chemical Engineering, Harbin University of Science and Technology, Harbin, China

*Corresponding Author: Jia Li. Email: lijia@hrbust.edu.cn

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ABSTRACT: Oil spill accidents and extensive industrial oily wastewater discharge pose severe threats to aquatic ecosystems and public health. Membrane-based separation technology has emerged as a core technology for treating oily wastewater. However, conventional petroleum-derived materials suffer from inherent limitations, including non-renewability, poor biodegradability, and high risks of secondary environmental contamination. Bio-based polymers represent a competitive alternative in oil/water separation due to their renewability, biodegradability, and versatile sources. Interfacial engineering serves as the core strategy for optimizing their high performance and multifunctionality. In this review, the fundamental interfacial chemistry theories of oil/water separation are systematically elaborated first. Recent advances in bio-based polymer systems for this application are then classified and discussed. Mainstream interfacial engineering regulation strategies are subsequently highlighted, such as surface chemical grafting polymerization, nanoparticle hybridization for hierarchical structure construction, biomimetic interface design, and dynamic/smart interfaces. Finally, the critical interfacial determinants governing the oil/water separation performance are systematically elucidated, and future research directions are prospected.

KEYWORDS: Bio-based polymers; oil/water separation; interfacial engineering; wettability; antifouling

1 Introduction

With the rapid expansion of the petrochemical, food processing, and machinery manufacturing industries, massive oily wastewater is discharged, which constitutes a predominant contributor to worldwide water contamination [1,2]. Oily wastewater harms aquatic ecosystems and poses a risk to human health via bioaccumulation in the food chain. To address this threat, a variety of materials have been developed and demonstrated effective performance in oil/water separation [3,4]. Membrane-based separation has gained prominence as a highly prospective approach for addressing oil-contaminated wastewater, distinguished by its high efficiency, operational simplicity, and absence of phase transitions [5,6]. Nevertheless, conventional polymer membranes predominantly originate from non-renewable petroleum resources, presenting post-consumer disposal challenges including recalcitrant environmental persistence and secondary contamination risks, which motivates the exploration of bio-based polymer membranes as sustainable alternatives [7–9].

Bio-based polymeric materials have attracted extensive attention as an ideal alternative to petroleum-based materials. They feature abundant sources, inherent biocompatibility, excellent biodegradability, and rich active groups (e.g., hydroxyl, amino, carboxyl), which provide abundant reactive sites for interface modification [1]. Through technologies such as electrospinning [5,10], three-dimensional (3D) printing [4],

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freeze-drying [11–13], sol-gel [14,15], liquid phase deposition [16], spraying [17], and surface coating [18], bio-based polymers can be processed into membranes [19,20], hydrogels [21], aerogels [13,22], sponges [23] and fibers [24,25], providing a structural basis for efficient mass transfer.

The essence of oil/water separation lies in the regulation of the oil-water-solid three-phase interface. The separation performance of materials is governed primarily by interfacial characteristics (chemistry, wettability, surface charge and micro-roughness), instead of their bulk intrinsic structure [26]. Interfacial engineering, which refers to the precise design of material interfaces at the molecular and nanoscale through chemical or physical means, is considered a promising critical strategy for breaking the performance limitations of bio-based materials and achieving high flux and selectivity, strong antifouling, and durable oil/water separation [27,28].

Starting from the fundamentals of interfacial chemistry, this review systematically elaborates the types of bio-based polymeric materials, interfacial engineering strategies and the determinants.

2 Fundamental Interfacial Chemistry of Oil/Water Separation

2.1 Models of Interfacial Wetting

The selectivity of oil/water separation fundamentally originates from interfacial wettability. The essence of oil/water separation is to selectively allow the permeation of one phase (usually the continuous phase) while retaining the other phase (the dispersed phase).

For an ideal smooth surface, the intrinsic wettability follows Young's equation [29] with the expression as Eq. (1):

$$r_{sv} = r_{sl} + r_{lv} \cos\theta, \tag{1}$$

where r_{sv}, r_{sl} and r_{lv} denote the interfacial tensions of solid-vapor, solid-liquid, and liquid-vapor interfaces, respectively, with θ representing the intrinsic contact angle, as shown in Fig. 1.

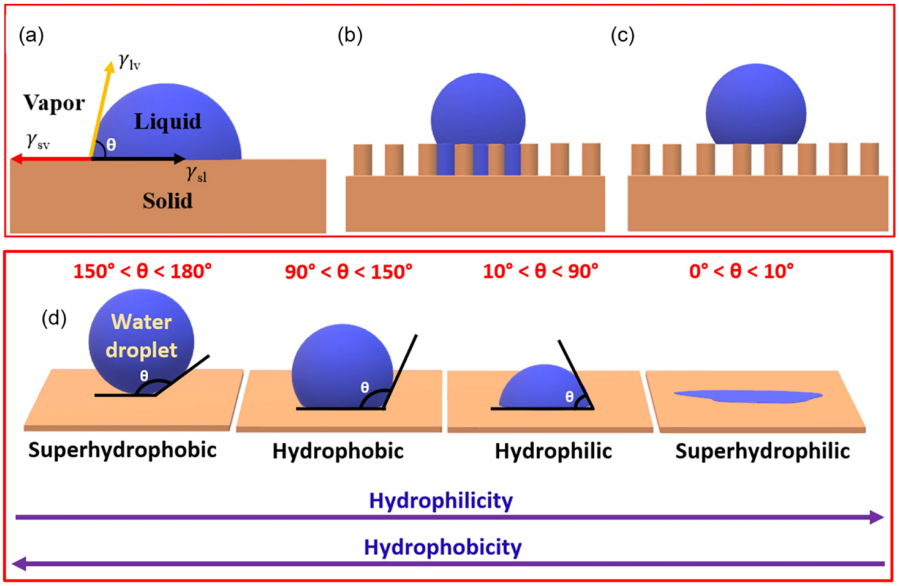

Figure 1: Schematics of wetting models: (**a**) Young model, (**b**) Wenzel model, (**c**) Cassie-Baxter model, (**d**) Diagram illustrating various surface wettability, adapted from [30] licensed under CC BY.

This equation establishes the fundamental relationship between surface chemistry and wettability. Based on water contact angle (WCA) measurements, solid surfaces are classified into four categories: superhydrophobic, hydrophobic, hydrophilic, and superhydrophilic, as illustrated in Fig. 1.

This equation indicates that surface energy dominates intrinsic wettability. Polar groups such as hydroxyl and amino groups rich in bio-based polymers can significantly increase the solid surface energy to achieve hydrophilic modification, while low surface energy groups including long-chain silyl and alkyl groups enable to reduce the surface energy to achieve hydrophobic modification, which is also the core theoretical basis for interfacial chemical grafting regulation.

Bio-based sorbents are mostly rough and porous structures, and their wetting behavior is more consistent with the Wenzel model (Eq. (2)) [31] and Cassie model (Eq. (3)) [32–34]. In the Wenzel model (Fig. 1), surface roughness serves to magnify inherent wetting characteristics: hydrophilic substrates exhibit enhanced hydrophilicity, while hydrophobic substrates demonstrate amplified hydrophobicity.

$$\cos\theta_a = r\cos\theta, \tag{2}$$

where θ_a represents the apparent contact angle of the rough substrates, and r denotes the surface roughness factor (the ratio of the actual surface area to the apparent surface area). This model explains the underlying principle that hierarchical roughness construction via nanoscale hybridization can significantly amplify the intrinsic wettability of bio-based materials and is theoretical support for interfacial morphology regulation. A variety of unique superhydrophobic surfaces widely exist in nature, which provide the original prototype and core inspiration for the construction of artificial superwetting surfaces for oil/water separation (Fig. 2).

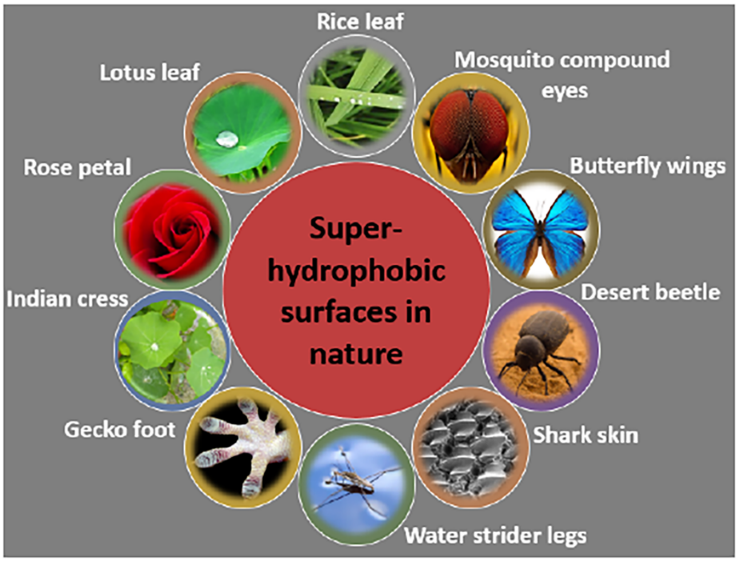

Figure 2: Superhydrophobic surfaces available in nature, reprinted [30] licensed under CC BY.

The Cassie-Baxter model (Fig. 1) describes a composite interface with trapped air as Eq. (3):

$$\cos\theta^* = f_1\cos\theta - f_2, \tag{3}$$

where θ^* denotes the apparent contact angle of the composite interface, f_1 represents the fraction of solid substrate in contact with the liquid, and f_2 indicates the fraction of air in contact with the liquid. The air layer trapped in the rough structure forms a stable hydrophobic barrier, allowing the oil phase to spread

and penetrate rapidly while blocking the water phase. For superhydrophilic/underwater superoleophobic materials, a stable hydration layer will be formed in a rough structure, and a solid-water-oil interface will be established with aqueous phase mediation, thereby blocking direct oil-solid contact and endowing the material with underwater superoleophobicity.

For oil/water separation, two types of surfaces with extreme wettability are usually required. One is the superhydrophilic/underwater superoleophobic surface, which exhibits an extremely high affinity (WCA ≈ 0°) and strong repellency to oil droplets underwater (oil contact angle (OCA) > 150°) [18]. This mechanism relies on establishing a stable hydration boundary at interfaces, which effectively blocks the adhesion and spreading of oil droplets, rendering such surfaces appropriate for oil-in-water emulsion separation. Chitosan (CS) and poly(tannic acid) hydrogel were immersed and coated on cotton fabric and a fabric was obtained with an underwater OCA of 166.84° after oxidative polymerization with a water flux >23,999 $L \cdot m^{-2} \cdot h^{-1}$ and a separation efficiency >99.84% [35]. The other is the superhydrophobic/superoleophilic surface. Microtopographical features efficiently entrap substantial gaseous volumes, establishing continuous air films. These architectures permit oil phase transport while obstructing the aqueous penetration [33,36]. Cardanol-derived benzoxazine monomer was used to prepare a superhydrophobic/superoleophilic membrane on polyester fabric via spraying and thermal curing. For heavy oil/water mixtures, the fabric demonstrated gravity-driven separation property with an oil flux of 6500–9000 $L \cdot m^{-2} \cdot h^{-1}$, a selectivity exceeding 90%, and operational durability [36]. A highly transparent self-cleaning surface was developed from bio-based polyurethane and SiO_2 nanoparticles (NPs) through a simple and eco-friendly approach, exhibiting a sliding angle (SA) <4° and WCA ≈ 158.5°. The coating showed significant robustness after 5 friction cycles, with no obvious decrease in superhydrophobicity [37].

2.2 Interfacial Energy and Antifouling Mechanism

Membrane fouling caused by oil adsorption and coalescence is the main obstacle to long-term operation. Constructing a stable hydration layer is the most effective strategy to resist oil fouling [38]. The hydration layer is a water molecule layer tightly bound to the surface with hydrogen bonds, electrostatic interactions, etc., which can generate a huge steric hindrance and energy barrier to prevent direct contact between oil and the surface [26]. Therefore, the introduction of bio-based polymers rich in high hydrophilic groups such as carboxyl, hydroxyl and amino groups is an effective way to construct a stable hydration layer and achieve underwater superoleophobicity [39].

Dai et al. [38] prepared a stainless-steel mesh coating with guar gum and metaborate hydrogel, which endowed the material with self-cleaning performance and high oil/water separation efficiency by virtue of the excellent hydrophilicity of guar gum. Yao et al. [36] synthesized benzoxazine monomer from cardanol and further prepared a superhydrophobic/superoleophilic membrane on commercial polyester fabric. The fabric featured self-cleaning performance and desirable durability and maintained stability under severe conditions including acid and alkali immersion, high temperature treatment and ultraviolet irradiation. A colorful, superhydrophobic, self-cleaning multifunctional coating for oil/water separation was developed using renewable nanocellulose, which solved the problems of single color and insufficient practicability of conventional superhydrophobic materials [28]. A cost-effective method was adopted to prepare high-performance superhydrophobic coatings using green biomass raw materials. The material exhibited self-cleaning, antibacterial, and anti-ultraviolet properties, accompanied by impressive chemical and mechanical stability. It achieved an oil/water separation efficiency of over 99% and remained stable for 25 cycles [40]. A superhydrophobic and superoleophilic porous sponge was prepared with polydimethylsiloxane (PDMS) and lignin, a waste from the papermaking industry. The material displayed a lotus leaf-like self-cleaning performance, and the pollutants on the surface were taken away by rolling water droplets. The

adsorption capacity for various oils (e.g., chloroform, toluene, olive oil, and ethyl acetate) was 36 to 48 times its mass [33]. A self-supporting bio-based superwetting nanofibrous membrane was prepared using CS, polyvinyl alcohol (PVA) and β-FeOOH NPs as raw materials. With a 99.9% separation efficiency, the FeOOH@CS/PVA-24-24 membrane exhibited excellent separation performance for water/n-heptane emulsion. The material presented significant antifouling performance, owing to its superwetting surface and the photocatalytic activity of particles. After five photocatalytic self-cleaning cycles, both separation efficiency and water flux remained nearly constant, highlighting its strong potential for practical applications [10]. A fully bio-based aerogel was fabricated for fast removal of oils and organic solvents from water with above 96% separation efficiency. The material simultaneously and efficiently adsorbed dyes and heavy metals. In addition, CS contributed antibacterial functionality to the aerogel through its amino groups [41].

2.3 Network Structure and Retention Performance

The network structure of the material (such as pore size and its distribution, porosity, channel orientation) is also a key factor determining the oil/water separation efficiency. For gravity-driven separation of conventional oil/water mixtures, maintaining a suitable porous framework is crucial [42]. At the same time, the high porosity and interconnected channels of the material ensure a high flux [5]. Interfacial engineering allows for precise control of the pore structure of materials through strategies such as tuning the crosslinking density of polymers, incorporating NPs, or utilizing templates [43].

A fully bio-based oil/water separation membrane was prepared through 3D printing technology. Nanographene oxide was used to regulate the printing quality of the bio-ink and the microstructure of the material, finally achieving a separation efficiency >99.5% for n-heptane/water emulsion and a high flux of higher than 38,300 $L{\cdot}m^{-2}{\cdot}h^{-1}$ [4]. Ye et al. [44] prepared a chitin nanofibril (ChNF)/CS aerogel with a highly oriented polygonal structure through directional freezing technology. By adjusting the pore size via compression, emulsions with different particle sizes were effectively separated, yielding a separation efficiency exceeding 99.5%. As a naturally porous material, wood affords numerous open microchannels aligned with its growth direction, making it an ideal skeleton for high-efficiency oil/water separation filters. Cheng et al. [45] fabricated Ag/wood filters by *in situ* deposition of Ag NPs within balsa wood channels. The honeycomb-like porous architecture with large-lumen vessels provided high-speed pathways for water transport (Fig. 3). Benefiting from the intrinsic hydrophilicity of cell wall components, the filter exhibited underwater superoleophobicity with OCA exceeding 150°, achieving selective oil/water separation with water permeating through while oil was retained, at efficiencies of approximately 99% under gravity-driven flow. Lin et al. [46] anchored fluorinated SiO_2 NPs on loofah sponge featuring a WCA of 156° and rapid oil absorption. Yao et al. [36] compounded cardanol-based benzoxazine with SiO_2 NPs. A superhydrophobic and superoleophilic fabric on polyester fabric by spraying and thermal curing was obtained with an oil flux of 6500–9000 $L{\cdot}m^{-2}{\cdot}h^{-1}$.

3 Typical Bio-Based Polymer Systems for Oil/Water Separation

Bio-based polymeric materials refer to polymer materials derived from renewable biomass resources, including natural polymers and their derivatives, as well as polymers synthesized from bio-based monomers. These materials not only feature abundant sources, renewability and biodegradability, but also possess abundant active functional groups, which provide abundant reaction sites for interfacial engineering.

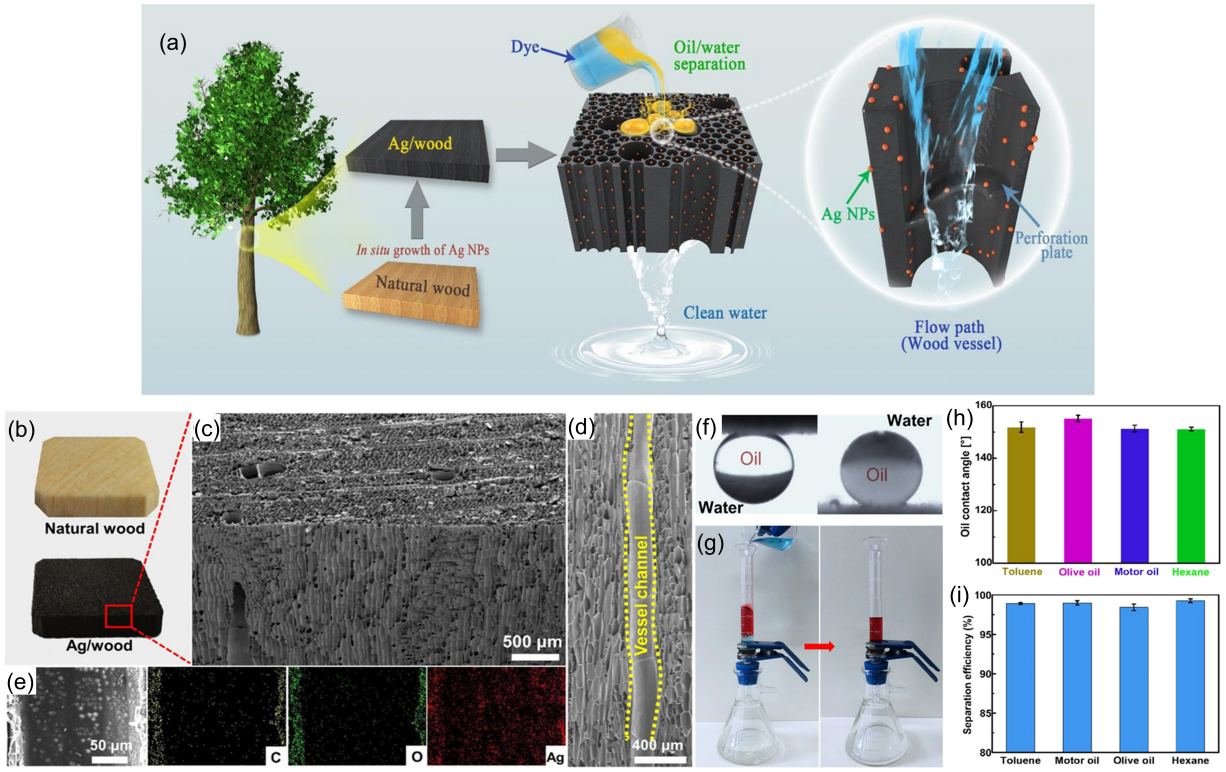

Figure 3: Dual-functional Ag/wood filters. (**a**) Schematic illustration of the fabrication of dual-functional Ag/wood filters for simultaneous oil/water separation and organic pollutant removal. (**b**) Photographs of the natural balsa wood and Ag/wood filter. The wood turned dark after *in situ* growth of Ag NPs inside the wood matrix. (**c**) Scanning electron microscope (SEM) image of Ag/wood filter showing the honeycomb-like porous structure. (**d**) Magnified SEM image showing an elongated vessel channel interconnected via perforation plates, which was highlighted with yellow dashed lines. (**e**) SEM image of the vessel channel and its corresponding elemental maps. (**f**) Photographs of olive oil and dichloromethane droplets sitting on the water-soaked wood filter with high contact angles. (**g**) Photographs of the oil/water separation process in which water flowed through the wood filter, while the oil phase (dyed red) was retained on top. (**h**) Contact angles of Ag/wood filters for various oils demonstrating their underwater superoleophobicity. (**i**) Separation efficiency of the Ag/wood filter for various oils, adapted from [45] licensed under CC BY.

3.1 Cellulose and Its Derivatives

With plentiful hydroxyl groups, cellulose and its derivatives are among the most abundant natural polymers [28]. These materials can be fabricated into various forms [47,48], including fibers, aerogels, and sponges, and chemical modification is generally required.

A bottom-up approach was reported to convert cellulose into hydrogel with nanostructure. The aerogel was modified to improve its elastic recovery [11]. *Metaplexis japonica* seed hair fibers exhibited outstanding superhydrophobic performance with a static WCA of about 151°. For vegetable oil, engine oil and diesel oil, it possessed excellent oleophilicity showing static contact angles of 0°. The adsorption capacities for these oils reached 81.52, 77.62 and 57.22 g/g, respectively, while oil retention rates after 12 h were 79.1%, 75.4% and 72.0%, respectively [24]. Su et al. [26] extracted micro-nanoscale heteromorphic cellulose crystals and cross-linked them with carboxymethyl CS. A cellulose-based coating was obtained, featuring excellent underwater superoleophobicity and resistance to viscous oil. A preparation method of nanocellulose-based superhydrophobic coating was proposed, which drew a variety of colorful patterns. The WCA was 156.6° and remained 150.7° after 70 times of friction [28]. A renewable and degradable superhydrophobic cotton fabric

was fabricated via dip-coating, using cellulose nanocrystals as the rough-structure component and cured epoxidized oil resin as the binder. The resulting fabric selectively absorbed oil from oily water and efficiently separated various oil/water mixtures with an efficiency exceeding 98%, while also exhibiting excellent solvent and chemical resistance [43]. Yang et al. [49] utilized an oxidized starch solution to emulsify cellulose stearoyl ester, yielding a hydrophobic-oleophilic membrane with hierarchical micro/nanostructures and low surface energy (Fig. 4). Its separation efficiency remained over 99% after 10 repeated separation cycles. With a suspension of lignin-containing cellulose nanofibril (LCNF), modified filter paper was prepared in both wet and dry states by coating commercial filter paper (Fig. 5a). The wet filter membrane with the same LCNF coating amount showed higher separation efficiency and flux compared with the dry filter membrane (Fig. 5b,c) [50]. A carbon aerogel based on kapok fiber and regenerated cellulose displayed an exceptional oil sorption capacity ranging from 137.5 to 371.7 $g \cdot g^{-1}$ and retained 89.3% after 10 extrusion cycles [51].

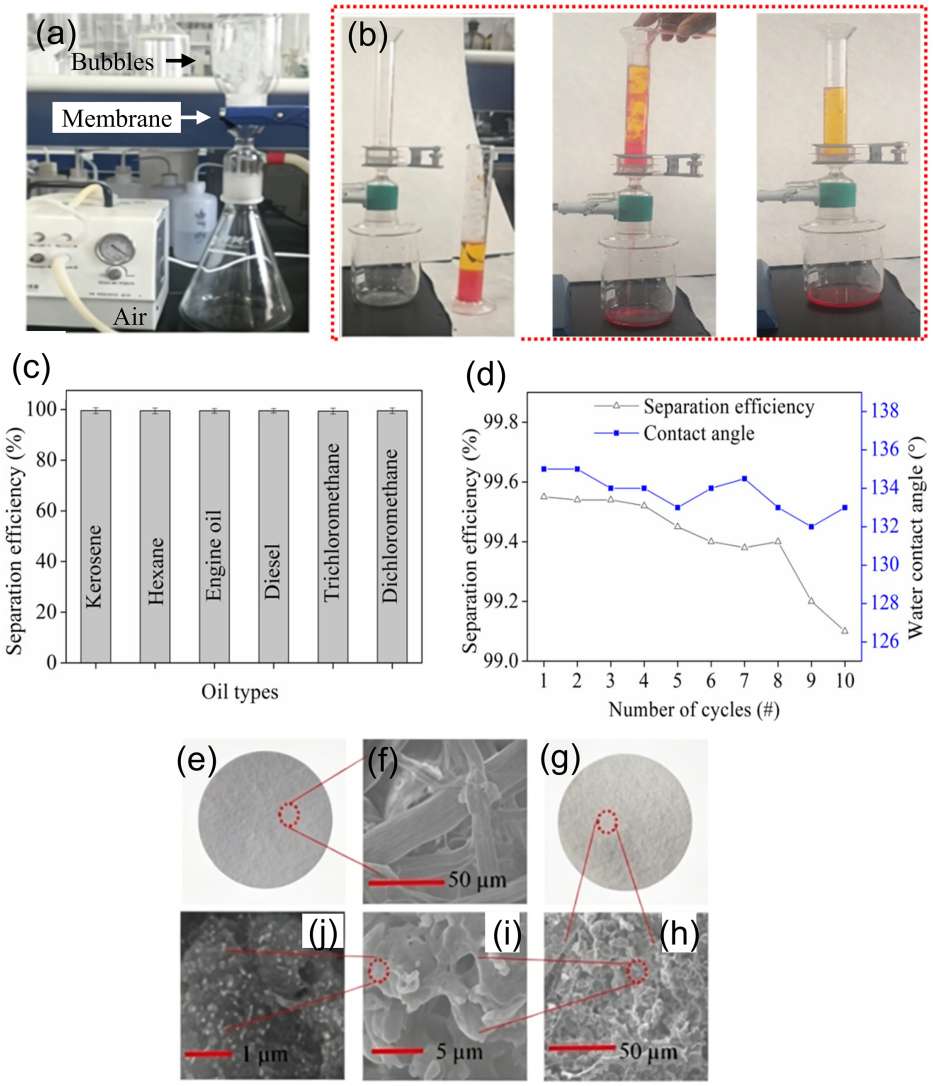

Figure 4: Hydrophobic-oleophilic membrane for oil/water separation. (**a**) Air permeability of the hydrophobic-oleophilic membrane. (**b**) Oil/water separation process. (**c**) Separation efficiency of various oil-water mixture. (**d**) Separation efficiency and contact angle of repeated tests. SEM images of (**e**) the original filter paper, (**f**) the original filter paper at high magnification, (**g**) the modified membrane, (**h**–**j**) highly magnified membrane, adapted from [49] licensed under CC BY.

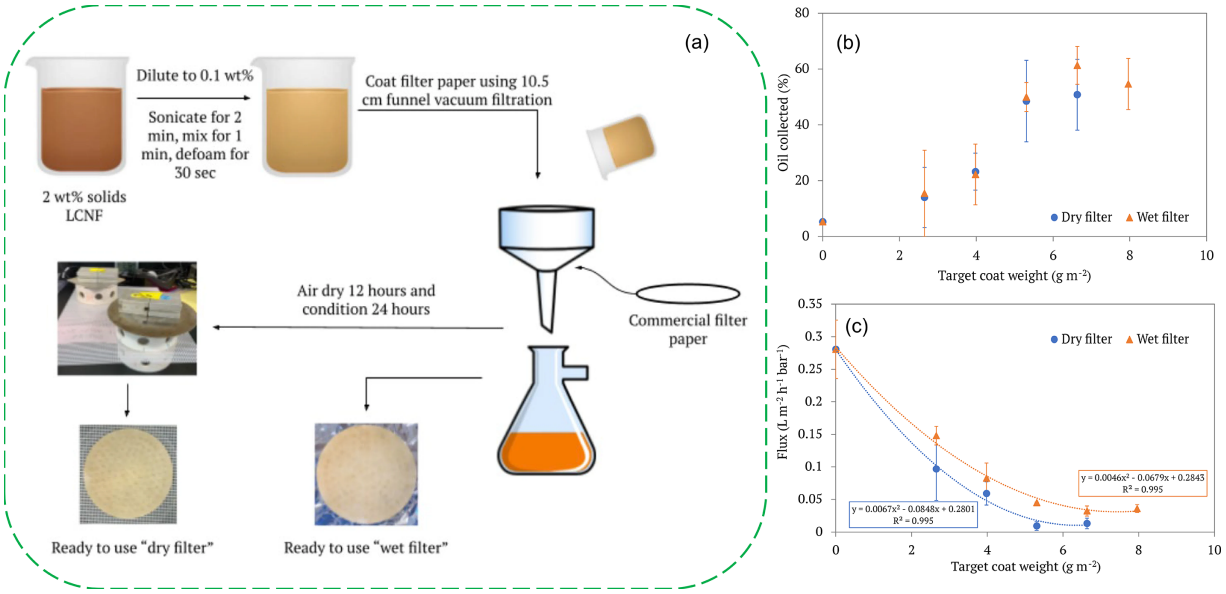

Figure 5: Fabrication and performance of LCNF-modified filter papers. (**a**) Schematic illustration of the coating process for both wet and dry LCNF-modified filter papers. (**b**) The oil separation efficiencies of different LCNF-modified filters by coat weight. (**c**) The flux values of different LCNF-modified filters by coat weight, adapted from [50] licensed under CC BY.

Inspired by the preparation of green tofu cellulose from plant leaves, Zhang et al. [52] extracted cellulose from premna microphylla leaves and reinforced it with sisal cellulose to fabricate a hierarchical porous superhydrophobic carbon-X aerogel with ultra-low density and superior elasticity. The aerogel exhibited excellent superhydrophobicity and a large specific surface area (475.68 $m^2 \cdot g^{-1}$). It demonstrated a remarkable adsorption capacity ranging from 77.7 to 147.3 $g \cdot g^{-1}$ for various oils and organic solvents, a high durability and reusability in 10 oil/water separation cycles. Cheng et al. [53] utilized tunicate cellulose nanocrystals to prepare a superhydrophilic/underwater superoleophobic membrane by vacuum filtration. The underwater contact angle for various oils (isooctane, chloroform, toluene, etc.) was >155°. These studies fully demonstrated the infinite potential of bio-based polymeric materials.

Cellulose materials offer the advantages of wide availability, low cost, and easy modification. Nevertheless, hydrophobic functionalization is commonly required to achieve oil-water separation performance. Nanocellulose (cellulose nanocrystal, cellulose nanofibrils) exhibits a high specific surface area and aspect ratio. It is an ideal unit for constructing hierarchical structures. However, the current preparation process is energy-intensive. Future efforts should focus on developing low-energy fabrication methods for nanocellulose and exploring green modification approaches.

3.2 Chitosan/Chitin

Chitosan/chitin contains abundant amino and hydroxyl groups as active sites for functional modification and has been widely applied in oil/water separation [54], accompanied by intrinsic cationic properties and favorable antibacterial activity [55,56].

An ultra-light and superelastic aerogel was proposed using ChNF and CS. Chemical crosslinking imparted the aerogel with mechanical strength at low density. After 500 compression-release cycles, the deformation recovery rate of the aerogel in air maintained at 88%, showing excellent resilience. For different oils and organic solvents, the absorption capacity of the aerogel was 52–114 g/g [44]. A covalent bond

network was constructed through crosslinking reactions among nanofibers, CS, glutaraldehyde and glycerol. This significantly enhanced compressibility, fatigue resistance and elasticity. It featured structural stability, high porosity and hydrophobicity (WCA = 141°, SA = 7.4°) with a high separation efficiency (99.3%) after 10 oil/water separation cycles [57]. Chitin nanocrystals scaffolds with oriented channels were prepared by liquid nitrogen-assisted directional freezing method. After crosslinking with glutaraldehyde, it displayed good mechanical and water stability, along with excellent resistance to salt, acid, and alkali during oil/water separation [58]. A hydrophilic membrane was prepared and modified with genipin, a natural and nontoxic crosslinking agent, which markedly enhanced the stability of the membrane in aqueous environment. With a separation efficiency of over 98%, the permeation fluxes for crude and cooking oil were 420 and 698 $L \cdot m^{-2} \cdot h^{-1}$, respectively [59].

The advantage of CS lies in the cationic properties and antibacterial activity brought by the amino groups. The poor solubility of CS limits its processing methods. It exhibits poor mechanical properties which should be enhanced by crosslinking. The development of non-toxic or low-toxic bio-based crosslinking agents is an important direction.

3.3 Lignin

Lignin possesses abundant hydroxyl groups [42], and its network structure is favorable for constructing micro-nano rough surfaces [60].

Li et al. [60] reported the preparation of Janus cellulose paper sheets derived from industrial lignin residue and nano-silica. A hierarchical network structure was constructed by simple spraying, and its flux was 654.46 $L \cdot m^{-2} \cdot h^{-1}$ in dichloromethane-water mixture. In addition, due to the excellent interfacial adhesion, the mechanical properties, folding resistance and tear resistance of the resulting material were notably enhanced. After 8 cycles of use, a separation efficiency of over 90% was sustained by the Janus paper sheets. Graphene aerogel was modified with a hydrophobic and porous skeleton using renewable biomass lignin and a compressible, ultra-light and flame-retardant lignin-modified graphene aerogel was prepared. In addition to efficiently absorbing petroleum oils, the lignin-modified graphene aerogel also showed significant uptake of hazardous solvents, including chloroform, carbon tetrachloride and toluene, achieving an absorption capacity reaching up to 350 times its mass, which was better than pure graphene aerogel and at the highest level among the adsorbents reported previously. Even after several compression-release cycles, its adsorption capacity remained at 96%. In addition, the adsorption capacity was further improved by carbonization, up to 522 times its own weight [61]. Polyvinylidene fluoride membrane was blended with renewable CS and lignin, achieving a permeate flux of 1.6862 $L \cdot m^{-2} \cdot h^{-1}$, an oil rejection of 99.26% and a reduced WCA of 17.34° [62].

Lignin is derived from the waste liquid of the papermaking industry, with low cost, huge output and significant sustainability. However, the complex structure and poor batch stability limit its high-value utilization. The establishment of standardized extraction methods to improve its dispersibility and compatibility in different matrices is the focus of current research.

3.4 Alginate

Alginate has abundant carboxyl groups and can form hydrogels via ionic crosslinking under mild conditions.

After soy protein isolate and sodium alginate were combined by sol-gel method, surface functionalization with tetraethyl orthosilicate and methyltrimethoxysilane (MTMS) was carried out. Surface was modified successfully to transform the aerogel into hydrophobicity. The aerogel absorbed many pollutants such as

waste engine oil, heavy oil and chloroform, with an adsorption capacity spanning from 1.34 to 10.3 g/g. After oil adsorption, the aerogel underwent significant degradation within 70 days [15].

The advantage of alginate lies in the mildness and reversibility of ionic crosslinking, which can be used to prepare recyclable and degradable hydrogel materials. However, owing to its relatively inferior mechanical properties, it often requires compounding with other materials for reinforcement. In addition, alginate exhibits excessively strong hydrophilicity, so appropriate hydrophobic modification is required for oil/water separation.

3.5 Natural Porous Biomass

Natural biomass materials such as wood and corn have hierarchical pore structures, allowing their direct use as oil/water sorbents or as substrates for functional modification [63].

Natural porous biomass materials are attractive owing to their low cost and biodegradability. However, their properties are highly sensitive to environmental factors (e.g., source, season, and production area), resulting in limited batch stability.

Wang et al. [14] fabricated a porous bio-based aerogel featuring a 3D network structure by compounding agricultural waste corn stalk with konjac glucomannan. Achieving over 96.26% separation efficiency and a permeation flux exceeding 2381 $L \cdot m^{-2} \cdot h^{-1}$, the aerogel exhibited excellent performance in separating oil-in-water emulsions. Li et al. [64] sprayed a mixture of waste potato residue powders and water-based polyurethane on a stainless-steel mesh. After pre-wetting with water, the mesh allowed efficient gravity-driven removal of water from light oil/water mixtures, with separation efficiencies exceeding 96.5% for various light- and heavy-oil mixtures. For kerosene-water mixture, it maintained over 98.0% separation efficiency and stable reusability even after 40 cycles. As a naturally porous material, wood affords many open microchannels aligned with its growth direction, which makes it an ideal skeleton for the development of high-efficiency water treatment filter materials. Cheng et al. [45] developed a bifunctional filter by *in-situ* deposition of Ag NPs in the channels of balsa wood, enabling oil/water separation and removal of organic dyes from water in one step. The Ag/wood filter selectively separated water from oil with an efficiency of approximately 99% and enabled the efficient removal of both soluble pollutants and insoluble oils in a single step, providing a promising solution for water purification. As an abundant agricultural by-product, corn residues possess potential for functional reuse. Kim et al. [65] reused untreated raw corn cob and corn husk for oil/water separation and energy generation. Without any chemical treatment, both materials exhibited strong hydrophobicity to water (WCA > 120°) and strong oleophilicity to different oils (OCA ≈ 0°) to effectively remove floating oil and sunken oil in water. For 5 representative oils, the oil absorption capacity of corn cob varied between 62 and 234 wt%, and that of corn husk ranged from 127 to 593 wt%. Cork membrane was treated with simple ethanol and H_2O_2 without any coating. The reversible switch between pre-wetting induced superhydrophilic/underwater superoleophobic (OCA > 160°) and superoleophilic/under-oil superhydrophobic (WCA > 150°) states was realized [66]. This provided a scalable strategy for the reuse of biomass under conditions of limited resources. A superhydrophobic, recyclable and renewable wood-based sponge from natural balsa wood was prepared. The sponge exhibited a porosity of 98.68%, an ultra-low density of 35.4 $mg \cdot cm^{-3}$ and a contact angle of 159.2°. After 50 absorption cycles, its contact angle and absorption capacity for CCl_4 were 150.8° and 52 g/g, respectively [16]. Starting from the pith core of the Shola plant, a natural adsorbent was obtained with an oil absorption capacity of 166 g/g by using its inherent 3D porous structure [67].

3.6 Other Bio-Based Polymers

Polysaccharides have attracted great attention because of their low toxicity, cost effectiveness and good biocompatibility [3]. Tannic acid, which provides phenolic hydroxyl groups and exhibits strong adhesion, can serve as both a crosslinking agent and a functional coating [39]. Protein polymers such as gelatin and collagen possess good film-forming properties and biocompatibility [59]. Polylactic acid (PLA) [68,69] and poly(propylene 2,5-furandicarboxylate) (PTF) [5], as bio-based synthetic polymers, are biodegradable and can be electrospun into nanofibrous membranes. Stable hydrophobic networks can be formed by vegetable oil-based polymers, such as epoxidized soybean oil [70], castor oil [37] and cardanol [36]. These networks can further enhance the hydrophobicity and stability of the material surface, thus providing reliable support for efficient oil-water separation.

Various main-chain benzoxazines derived from biological resources were synthesized from renewable raw materials and loaded on a melamine sponge scaffold through a simple dipping-squeezing-curing process. The modified sponge showed a WCA of 137.8°–144.5° and chloroform adsorption capacities of 117.0–140.64 g/g. After 15 adsorption-extrusion cycles, the retention rate of its adsorption capacity was 67.47% to 76.29%. Notably, its excellent adsorption selectivity was reflected in the fact that only trace amounts of water (ppm level) were contained in the oil phase after separation [71]. 28 different sorbents were prepared using plant-derived *D*-limonene and β-myrcene as renewable units [72]. It was observed that the ratio of *D*-limonene and β-myrcene was a critical parameter to control the pore morphology and physicochemical properties. Through carbonization and PDMS modification, a material was developed from discarded fermented steamed buns for the recovery of high-viscosity oils. Derived from wheat, the discarded fermented steamed buns possessed renewable and environmentally friendly characteristics. Studies indicated that the material exhibited a WCA of 145.39°. Furthermore, it maintained a separation efficiency of 97.69% over 10 cycles of oil/water separation occurring under gravity [73].

As illustrated in Table 1, bio-based polymers can be fabricated into multiple forms such as aerogels and fibers, each exhibiting distinct advantages for separation. The diversity in material forms also reflects the adaptability of interfacial engineering strategies to different structural requirements and operational conditions.

Table 1: Representative bio-based polymers for oil/water separation and their performance.

Bio-Based Polymer	Material Form	Wettability Type	Key Performance	Reference
Tung oil	Sponge	Hydrophobic (pH = 7) and hydrophilic (pH = 13)	Reversible pH-responsive wettability, superelasticity and good thermal/chemical stability.	[23]
Cellulose	Surface modified fiber	Hydrophobic	After 120 adsorption/desorption cycles, capacity retention = 81%, WCA >145°.	[25]
Cellulose	Hydrogel	Hydrophilic	The separation efficiencies of different viscosities of oil and water mixtures >99.5%.	[21]
Cellulose	Nanofibril	Hydrophobic	The oil collection of wet and dry LCNF-modified filters = 61% and 51% (unmodified: 5%).	[50]

(Continued)

Table 1 (continued)

Bio-Based Polymer	Material Form	Wettability Type	Key Performance	Reference
Cellulose	Microparticle	Hydrophobic	Self-cleaning and WCA = 156.6°. After 70 abrasion cycles, WCA = 150.7°.	[28]
Cellulose	Nanocrystals	Hydrophilic	Water flux = 1036–1734 $L \cdot m^{-2} \cdot h^{-1} \cdot bar^{-1}$. The rejections for toluene, chloroform, isooctane and n-hexane >99.99%.	[53]
Cellulose	Aerogel	Superhydrophobic	Oil absorption = 20–65 g/g. Flux = 18,000–28,000 $L \cdot m^{-2} \cdot h^{-1}$.	[57]
Chitosan	Membrane	Superhydrophilic/ underwater oleophobic	WCA = 9.5 ± 3.5°. The underwater OCA >140°.	[10]
Chitosan	Fabric	Superhydrophilic/ underwater superoleophobic	A separation efficiency >99%. A water flux >15,000 $L \cdot m^{-2} \cdot h^{-1}$.	[18]
Lignin	Aerogel membrane	Superhydrophobic	WCA = 161°.	[8]
Lignin	Sponge	Superhydrophobic	WCA = 160°.	[33]
Lignin	Emulsion	Hydrophobic	Mechanical stability and storage stability, thermal stability and hydrophobicity.	[42]
Alginate	Aerogel	Superhydrophilic/ underwater superoleophobic	Light-driven self-cleaning ability, underwater superoleophobicity with an OCA >150° for various oils.	[13]
Konjac glucomannan	Fabric membrane	Superhydrophilic/ underwater superoleophobic	A separation rate = 99.9%.	[3]
Tannic acid	Hydrogel	Superhydrophilic/ underwater superoleophobic	Salt tolerance and wide pH stability.	[39]
CS/Gelatin	Membrane	Hydrophilic	Selective separation, separation efficiency >98%, recyclability and anti-fouling performance.	[59]

Each bio-based polymer system possesses complementary strengths and inherent weaknesses that must be balanced for targeted oil/water separation applications. Cellulose and its derivatives offer exceptional abundance, low cost, and versatile chemistry modification, but their intrinsic hydrophilicity necessitates hydrophobic conversion, and nanocellulose isolation remains energy intensive. Chitosan provides unique cationic charge and antibacterial activity valuable for anti-fouling membranes, yet its poor solubility in neutral media and weak mechanical strength require extensive crosslinking or polymer blending. Lignin is highly sustainable and economical when valorized from pulping waste, and its hydrophobicity facilitates

rough surface construction. However, structural heterogeneity and batch inconsistency challenge reproducible engineering. Alginate enables mild, reversible ionic gelation for eco-friendly hydrogel processing, but excessive hydrophilicity and mechanical fragility restrict standalone use without hydrophobic modification. Natural porous biomass offers ready-made hierarchical pores with minimal processing, though geographical and seasonal variability cause poor standardization and mechanical durability.

4 Interfacial Engineering Strategies for Bio-Based Polymers

The intrinsic properties of bio-based polymers provide the basis for their application, whereas precise regulation of the surface via interfacial engineering is indispensable to achieve efficient oil/water separation. The core goal of interfacial engineering is to optimize wettability, charge properties and antifouling performance of materials by precisely modulating the chemical components, physical morphology and surface microstructure of the materials.

4.1 Surface Chemical Grafting and Polymerization

Surface grafting is one of the most straightforward and effective strategies to regulate surface energy. Through esterification, etherification, amidation and other reactions, hydrophilic molecules can be grafted onto the surface for hydrophilic modification.

Crosslinked CS/cellulose hydrogels were prepared using a dicationic epoxy resin with abundant quaternary ammonium groups. The material exhibited excellent dye adsorption and oil/water separation performance over a wide pH range [21]. Flexible and oleophilic polycaprolactone (PCL) diol segments were incorporated into PTF, thus greatly increasing the copolymer's flexibility and resistance to water. PTF-b-PCL nanofibrous membranes were prepared by electrospinning technology and exhibited excellent mechanical properties. The membranes showed a flux as high as 7774 $L \cdot m^{-2} \cdot h^{-1}$ and a separation efficiency greater than 98.5% [5]. By means of dip coating, the degradation products of waste polyethylene terephthalate were coated onto a stainless-steel mesh, which increased the surface WCA to 123°. An oil/water separation flux of 9825 $L \cdot m^{-2} \cdot h^{-1}$, a separation efficiency of 98.99% and favorable recycling performance were obtained [6]. The surface energy of the material can be significantly reduced to achieve hydrophobic modification by grafting long-chain alkyl groups (e.g., stearic acid [74], octadecylamine [70]), fluoroalkyl groups [42] or silane coupling agents carrying hydrophobic functional groups [46,75–77]. MTMS [28], octadecyltrimethoxysilane (ODS) [40], methyltrichlorosilane (MTS) [75], dodecyltrimethoxysilane (DTMS) [76] are commonly used as hydrophobic modifiers. Fig. 6 illustrated the hydrophobization of regenerated cellulose (RC) via chemical vapor deposition (CVD) using MTS, a silane coupling agent carrying hydrophobic -CH_3 groups. MTS vapor reacted with surface -OH groups of RC through a gas-solid reaction, yielding transparent films with progressively denser siloxane aggregates on the surface as the deposition time increased from 10 to 60 min. FT-IR confirmed successful grafting via characteristic peaks of Si-CH_3 (1272 cm^{-1}) and Si-O-Si (782 cm^{-1}), while the WCA rose monotonically from 58.9° to 83.2°, demonstrating effective surface energy reduction by hydrophobic silane coupling agents [75].

Meng et al. [76] reported a two-step strategy for fabricating superhydrophobic balsa wood sponges, wherein alkali treatment removed hemicellulose and lignin to generate a flexible porous cellulose skeleton, and subsequent DTMS impregnation formed a hydrophobic polysiloxane coating, affording a WCA of 150.5° and robust oil-water separation recyclability. The adsorption capacity reached 12–31 g/g, and showed no significant change after 10 cycles with a WCA of 143.2°. In another study, Renewable balsa wood was employed to synthesize MTMS-modified wood aerogel with a WCA of 151.8°. The adsorption capacity for dichloromethane attained 25.1 times its own weight [77]. Photo-induced polymerization is also an efficient method. A photo-induced "thiol-ene" click reaction was used to spray-deposit castor oil-based thiolated

oligomer (thiol-CO) and modified SiO_2 NPs on various substrates. Characterized by a WCA of approximately 160.5° and a SA of nearly 1°, a superhydrophobic self-cleaning coating was yielded without significant attenuation of performance after 25 cycles [78].

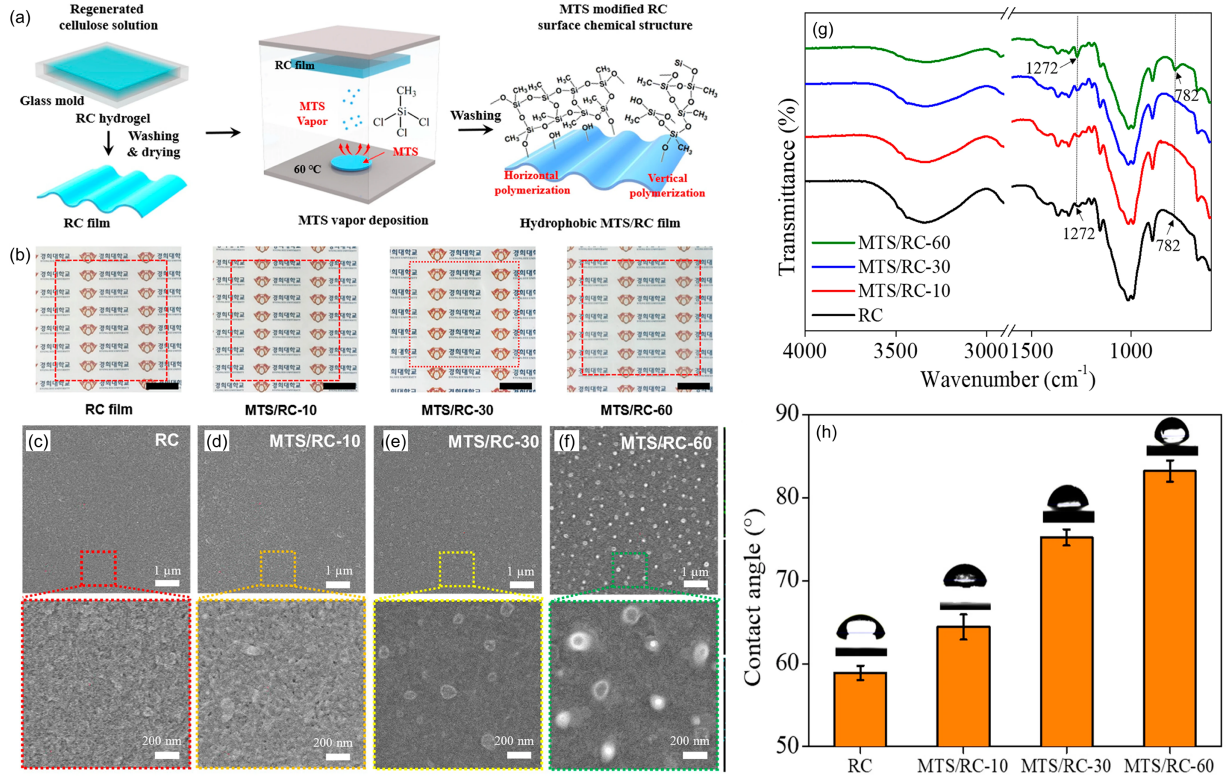

Figure 6: Fabrication process, morphology, chemical structure and wettability of hydrophobic silylated cellulose films. (**a**) Fabrication process of hydrophobic films via CVD. Firstly, the cellulose solution was poured into a glass mold and coagulated into RC films. Next, the dried RC film was placed in a desiccator, and MTS was vapor-deposited on the surface of the RC film for 10, 30, or 60 min. (**b**) Photographs of the pristine RC film and the modified films with different deposition times. Inset scale bar: 2 cm. Thickness of the films: 70 μm. (**c**–**f**) Top-view field emission scanning electron microscopy (FE-SEM) images of the pristine RC film and the modified films as a function of the MTS deposition time (10, 30, and 60 min). (**g**) Fourier transform infrared (FT-IR) spectra of the pristine RC film and the MTS/RC films with different MTS deposition times. (**h**) WCA of the pristine RC film and modified films, adapted from [75] licensed under CC BY.

Overall, surface grafting polymerization has the advantages of precise modification, stable interfacial bonding and significant functionalization effect. It is the crucial method for the regulation of interfacial chemical composition. However, most grafting procedures require organic solvents and suffer from complicated operations and harsh reaction parameters, restricting large-scale continuous industrial production. In addition, some grafting reactions will destroy the intrinsic biodegradability of bio-based polymers.

4.2 Construction of Hierarchical Rough Structures via Nanoparticle Hybridization

Introducing NPs into polymer materials is a common method to construct hierarchical micro-nanoscale rough structures. In recent years, there are two categories of commonly used NPs. One is inorganic NPs, such as SiO_2 [79,80], ZnO [81], TiO_2 [13], CuO [82], Ag [83,84], CuS [48], Fe_3O_4 [85] and metal-organic frameworks (MOFs) [86,87]. According to the classical Wenzel model, surface roughness serves to amplify

the intrinsic wettability of materials. Specifically, the construction of hierarchical rough structures not only enhances the intrinsic wettability of bio-based materials but also imparts multifunctional properties such as photocatalysis, antibacterial activity, and photothermal conversion.

After nanofibrous membranes were prepared by electrospinning using CS and PVA, β-FeOOH NPs were *in-situ* grown on the material surface. Through precise control of the growth time of β-FeOOH NPs and the crosslinking time of CS and PVA, the membrane's nanopore size and rough surface structure were modulated. FeOOH@CS/PVA-4-12 exhibited superwettability, and FeOOH@CS/PVA-24-24 showed excellent separation capability for water/n-heptane emulsions with a separation efficiency up to 99.9% [10]. Cationic starch and tannic acid were used, together with ODS-modified nano-SiO_2, to construct a superhydrophobic and highly durable coating. Benefiting from the hierarchical micro-nanoscale rough structure and long-chain alkyl groups with low surface energy, the coating demonstrated superhydrophobic behavior with WCA above 170° [40]. Inspired by solar-driven viscosity reduction, a photothermal 3D PDMS/CuS/carbon fibers/reduced graphene sponge was efficiently fabricated through a combined process of self-assembly, carbonization, freeze-drying and hydrophobization. When subjected to 0.3-sun simulated irradiation at room temperature, the surface temperature spontaneously reached 75.9°C, which consequently decreased the *in-situ* viscosity of crude oil and substantially enhanced its diffusion coefficient and adsorption rate. As a result, the material attained an oil sorption capacity of 66.8 times its own weight, indicating excellent absorption efficiency [48]. Bone ash particles were prepared using bovine bone biomass, which were loaded on a cotton fabric substrate. Epoxy resin and stearic acid were used for impregnation modification to prepare bone ash/cotton cellulose composite membranes. The bone ash particles constructed a rough micro-nanoscale architecture at the fiber surface, while the stearic acid provided low surface energy. The two factors synergistically imparted excellent superhydrophobicity and superoleophilicity to the membrane. The separation efficiencies were all >99%, and the light oil flux was 53,635.2 $L \cdot m^{-2} \cdot h^{-1}$ [74].

The second category is bio-based NPs, such as chitin nanocrystals [58], which are renewable nanomaterials that can provide both roughness and ensure the fully bio-based properties of the material. Bio-based composites were fabricated with cardanol/eugenol-based benzoxazine as the matrix and a combination of natural fibrous materials (coir felt, kapok fabric, jute felt, and rice husk) and bio-silica as reinforcing components. Characterization revealed that the composite reached a maximum WCA of 147° and a minimum dielectric constant of 2.0. The coated cotton fabrics exhibited WCAs of 159° and 157°, which corresponded to oil/water separation efficiencies of 96% and 95%, respectively [80]. Cellulose-derived nano-CuO@carbon aerogel composites (CNCAs) were fabricated via a process involving freeze-drying, carbonization and subsequent impregnation. The results showed that the CNCAs effectively separated oil from water-in-oil emulsions stabilized by surfactants with a separation efficiency of 92.3% and a WCA of 158° [82]. A magnetically controllable and efficient oil-absorbing bio-based aerogel with Fe_3O_4 NPs was prepared through freeze-drying and hydrophobic modification with CS and itaconic acid as the skeleton and the crosslinking agent, respectively. The aerogel exhibited both superparamagnetism and superoleophilicity with a density of 0.0655 g/cm^3, a porosity of 92% and a WCA of 147.1°. Floating oil and submerged oil were adsorbed under the control of a magnetic field. The upper limit of adsorption capacity was determined to be 22.38 g/g. The adsorption efficiency reached 98.5% while the adsorption performance remained above 85% after 10 cycles [85]. Dip-coating was first performed using a dispersion of carbon nanotubes (CNTs) wrapped with poly(sodium 4-styrenesulfonate), then proceeding to *in-situ* deposition of polydopamine (PDA). Then a superhydrophilic and underwater superoleophobic cotton fabric was obtained. An underwater OCA of 160° was exhibited with a water flux of 10,834 $L \cdot m^{-2} \cdot h^{-1}$ and a separation efficiency over 99.8% [88]. An antibacterial composite paper with sandwich-structure was prepared utilizing pulp fibers functionalized with Ag NPs and reinforced with cellulose nanofibers (CNFs). Demonstrating superhydrophilicity, the

composite paper attained stable cycling for 20 times and a 96% separation efficiency for a range of oil-in-water emulsions. CNFs provided nanoscale pore size for the paper and intercepted nearly 100% of bacteria through physical barrier effect. At the same time, Ag NPs endowed the strong antibacterial activity against *Escherichia coli* [89]. Inspired by the hierarchical architecture of coral reefs, a multifunctional paper-based material with superhydrophobicity, superoleophilicity, electromagnetic shielding and self-cleaning functions was fabricated with the renewable cellulose filter paper as the substrate via electroless copper plating, followed by silver nitrate etching and 1-hexadecanethiol modification [90]. The cellulose fibers within the filter paper featured a 3D interwoven architecture and were fully used as the skeleton. The biomimetic micro-nanoscale structure was constructed by means of the displacement reaction occurring between Ag^+ and the coated copper (Fig. 7).

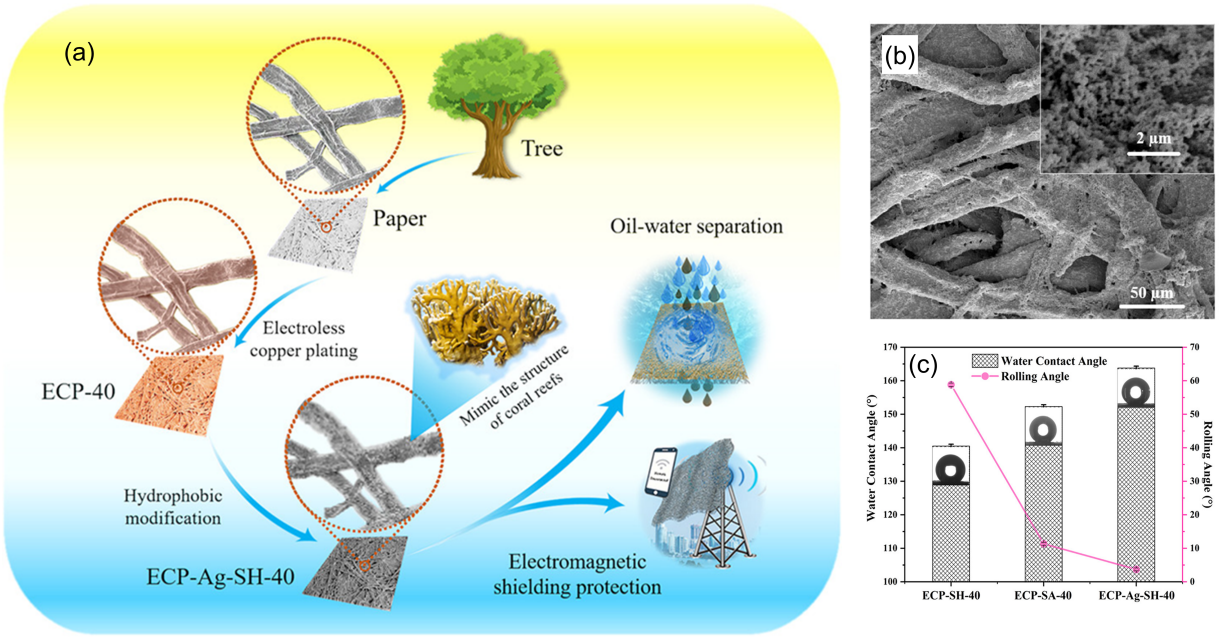

Figure 7: Fabrication, morphology, and wettability of coral reef-like superhydrophobic paper. (**a**) Illustration of the preparation procedure for the superhydrophobic paper by conventional electroless copper plating and surface modification. (**b**) SEM image showing the coral reef-like hierarchical micro/nanostructures. (**c**) WCA and rolling angle of the modified papers, demonstrating the excellent superhydrophobicity, reprinted with permission from [90]. Copyright ©2025, John Wiley and Sons.

In addition, the *in-situ* growth method is also highly favored. Ni-Co layered hydroxide nanosheets on the surface of cotton fibers were obtained by an *in-situ* hydrothermal growth method and then grafted with stearic acid to prepare cotton-based composite which exhibited a WCA of 161°. The oil absorption capacity exceeded 10 times its own weight. The introduction of layered double hydroxides constructed a rough structure with hierarchical micro-nanoscale features and the long alkyl chain of stearic acid provided low surface energy. The two synergistically provided the material with excellent superhydrophobicity [91]. Oligo-chitosan and aminopropyltriethoxysilane were used to *in-situ* grow a hierarchical Janus rough structure on the amino-pulp surface. It revealed water resistance, self-cleaning, anti-fouling and good color stability [92].

The main constraint of NPs hybridization is attributed to the predominantly physical nature of the interfacial interactions between NPs and bio-based matrix. Such weak interfacial bonding renders the materials susceptible to NPs detachment during prolonged cyclic operation, consequently leading to progressive performance degradation. Moreover, the incorporation of inorganic NPs inevitably compromises, to some

extent, the inherent biodegradability of the bio-based matrix. Therefore, it is imperative to improve the interfacial interaction between NPs and the matrix through rational surface modification strategies.

4.3 Biomimetic Interface Design

Many organisms in nature feature unique interfacial properties, which offer valuable guidance for the design of bio-inspired materials.

Inspired by the strong adhesion of marine mussels, a superhydrophilic and underwater superoleophobic fabric was fabricated via *in-situ* deposition of PDA particles onto cotton fabric and then dip-coating CS. Featuring >99% separation efficiency and a water flux of >15,000 $L{\cdot}m^{-2}{\cdot}h^{-1}$, the fabric was resistant to wear, ultrasound, acid, alkali and salt erosion [18]. CS and PDA hydrogel coated CF (CS&PDA/CF) were constructed via surface coating combined with oxidative polymerization of CS and dopamine mixtures. As a result of the formation of a rough surface structure composed of a hydrophilic composite hydrogel, the material displayed superhydrophilicity and underwater superoleophobicity and showed favorable oil/water separation performance for several mixtures of oil and water, with a separation efficiency higher than 99.5%. In addition, it showed excellent tolerance to a range of severe conditions, including concentrated salt solution, ultrasonic treatment, and boiling water, owing to the strong mussel-inspired adhesion that stabilized the structure and morphology [54]. A PDA-functionalized silica (SiO_2) interlayer was fabricated on a stainless-steel mesh substrate through the simultaneous hydrolysis of tetraethoxysilane and oxidative polymerization of dopamine. Dip-coating technology was used to deposit the outermost composite hydrogel layer, which consisted of PVA-tannic acid and TiO_2@PDA. This structured hydrogel coating displayed multiple functions, such as superoleophobicity, broad pH stability, and salt tolerance, which were essential properties for achieving reliable separation performance. In combination with 800-mesh and 1200-mesh stainless steel mesh, it promoted the successful demulsification toward oil-in-water emulsions [39]. Ramalingam and Das [93] prepared stearic acid-coated biomaterial functionalized graphene oxide aerogel, achieving Janus asymmetric wettability with a WCA of 40.35–129.4°. This aerogel realized high oil uptake up to 227.53 g/g. The anti-oil fouling characteristics of fish scales in water were imitated. A carrageenan hydrogel was coated on stainless steel mesh, obtaining an underwater OCA of no less than 158°, a flux of 1380.75 $L{\cdot}m^{-2}{\cdot}h^{-1}$. It exhibited a separation efficiency exceeding 97.9% for various oils and unchanged performance after 20 cycles [94]. Inspired by superhydrophobic structures found in biological systems, a new type of superhydrophobic and superoleophilic porous material was prepared for efficient oil/water separation by a simple spraying method using renewable biomass raw materials: beeswax, lignin and cotton. Beeswax provided low surface energy, endowing the composite porous material with simultaneous superhydrophobicity and superoleophilicity. The sample containing lignin still maintained superhydrophobicity after heating at 120°C for 60 min [95]. Mimicking the hierarchical architecture of coral reefs, a superhydrophobic surface featuring micro-nanoscale composite structures was engineered on cellulose filter paper. The material exhibited outstanding superhydrophobicity (WCA = 163.8°), superoleophilicity (OCA = 0°), and the separation efficiency for various oils such as gasoline, diesel, toluene and chloroform still maintained above 89% after 12 cycles [90]. Wu et al. [96] constructed a biomimetic nacre-like microstructure, which effectively avoided the structural collapse during atmospheric drying, and endowed the aerogel with excellent compressive strength. The aerogel absorbed low-viscosity engine oil quickly (<2 s) and high-viscosity crude oil with the assistance of light (full adsorption within 150 s). Yu et al. [28] developed a colorful nanocellulose-based superhydrophobic coating (WCA = 156.6°) by mimicking lotus leaf structures. The coated paper exhibited efficient oil/water separation performance and excellent self-cleaning ability and retained superhydrophobicity after 70 abrasion cycles (Fig. 8).

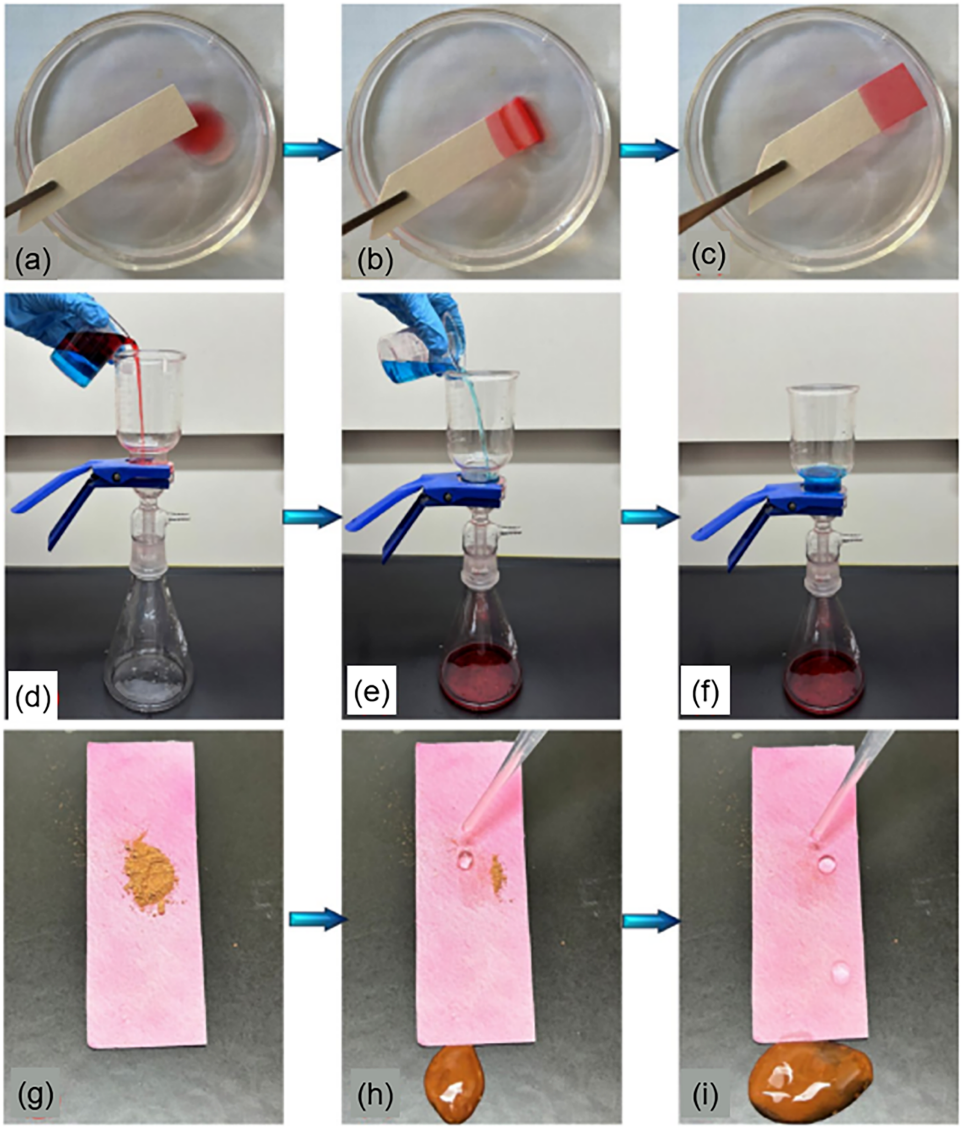

Figure 8: Oil/water separation and self cleaning performance of the colorful nanocellulose based superhydrophobic coating. (**a**–**c**) Adsorption process of n hexane (dyed red) from water. (**d**–**f**) Gravity driven separation of stratified n hexane/water mixture. (**g**–**i**) Self cleaning test with soil as contaminant, reprinted with permission from [28]. Copyright ©2025, John Wiley and Sons.

4.4 Dynamic and Smart Interfaces

Smart responsive interfaces are designed by grafting environment-sensitive groups such as pH, light, heat, magnetism, and salt on the surface of bio-based materials and realize reversible wettability switching for on-demand separation and intelligent response. This strategy breaks through the application limitations of conventional single wettability materials, realizing *in-situ* regeneration of materials and desorption of pollutants through external stimuli. Smart interfaces represent a cutting-edge direction for intelligent wastewater treatment.

Reversible transitions of surface charge and conformation are achieved in pH-responsive materials by the incorporation of ionizable groups on the surface, thus enabling tunable wettability and pore dimensions. Choi et al. [97] fabricated a pH-responsive chitosan-coated polyethersulfone membrane via dip-coating

and glutaraldehyde cross-linking (Fig. 9a). As shown in Fig. 9b, the wettability switching was driven by the reversible protonation of chitosan amino groups. This charge reversal near pH 6 was confirmed by zeta potential analysis (Fig. 9c), correlating with the sharp decrease in underwater OCA at pH 12 (Fig. 9d). Consequently, the membrane achieved >97% oil rejection under acidic conditions, declining to ~84% at pH 12 (Fig. 9e) [97].

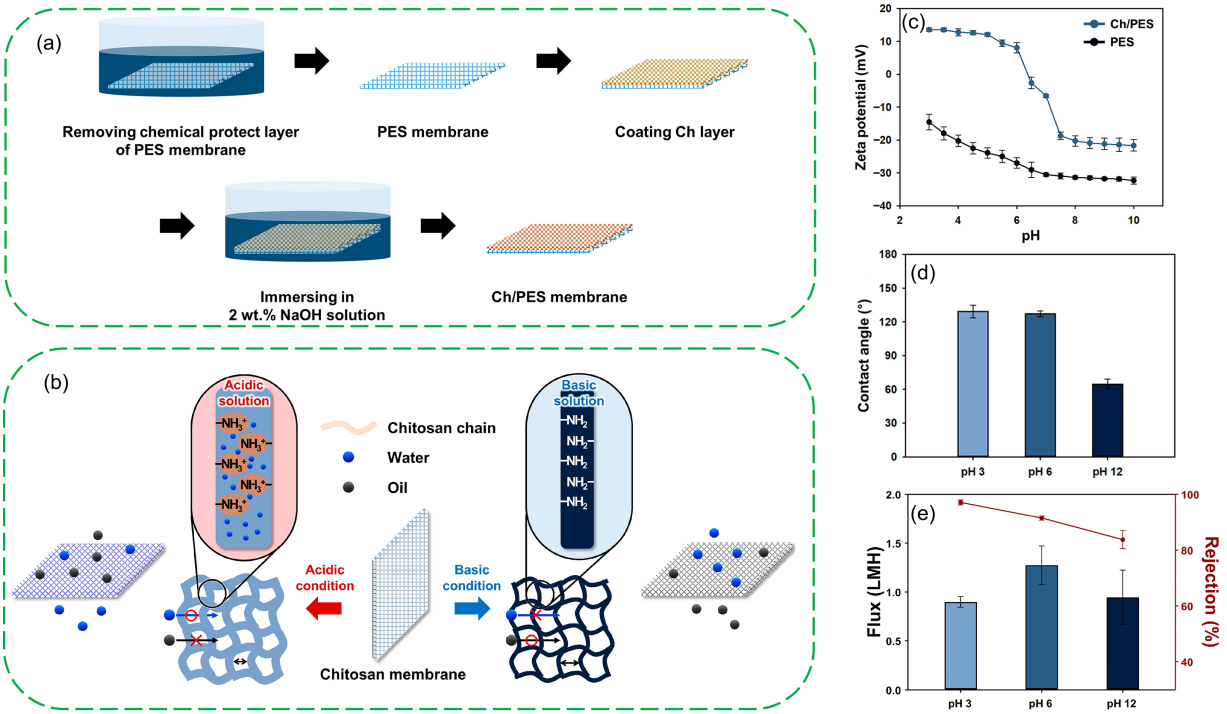

Figure 9: pH-responsive chitosan-coated polyethersulfone membrane for oil–water separation. (**a**) Schematic illustration of the fabrication process for the pH-responsive chitosan-coated polyethersulfone membrane. (**b**) Schematic diagram of the pH-responsive swelling behavior of the chitosan-coated polyethersulfone membrane. (**c**) Zeta potential of polyethersulfone and chitosan-coated polyethersulfone membranes measured across a range of pH values. (**d**) Underwater oil contact angle of chitosan-coated polyethersulfone membranes at varying pH levels. (**e**) Flux measurements and oil rejection efficiencies of chitosan-coated polyethersulfone membranes during emulsified-oil separation tests at varying pH levels, adapted from [97] licensed under CC BY.

Mimicking the trabecular bone structure, a sponge-like cellular network derived from tung oil was prepared via hypersalinity-confined thermopolymerization. The obtained material, with a separation efficiency exceeding 99.4%, possessed pH-responsive wettability and superelasticity [23]. After photothermal materials such as CuS [48], CNTs [98], MXene [96] and Fe_3O_4 [99] were embedded, the heat could be generated when exposed to light to reduce the viscosity of high-viscosity crude oil, which facilitated rapid adsorption and separation. Using CNTs and PDMS, a modified sponge was prepared and exhibited a WCA of 142° and a maximum oil/water separation efficiency of 98%, demonstrating its hydrophobicity, self-cleaning performance, and ability to absorb crude oil and organic solvents. After multiple intermittent on-off light cycles at an irradiation of 1.0 kW/m^2, the surface temperature of the sponge was maintained above 60°C, which proved its excellent photothermal stability [98]. A PDMS/CuS/carbon fibers/reduced graphene sponge was fabricated, which reached a surface temperature of 75.9°C under 0.3-sun irradiation and exhibited an oil absorption capacity of 66.8 g/g [48]. A hydrophobic magnetic CS-based aerogel was obtained through dip-coating using itaconic acid, CS and Fe_3O_4 NPs. For various organic solvents, the adsorption capacity

was in the range of 17.7–43.8 g/g. The average adsorption rate ranged from 3.3 to 74.5 g/(g·s). The separation efficiency and WCA for oil/water mixtures were 98.0%–99.6% and 147.9°, respectively [99]. Self-healing materials utilize dynamic covalent bonds or reversible noncovalent interactions to enable the damaged material surface to recover its original wettability and separation performance. Zhao et al. [100] synthesized functionalized polysiloxane based on β-pinene, with a WCA of about 170° and a SA of nearly 0°. It displayed superhydrophobicity, anti-fouling properties along with resistance to scratching and abrasion. The damaged superhydrophobicity was quickly repaired by flame and still maintained excellent hydrophobicity after 10 cycles. Ashrafi et al. [101] studied bacterial nanocellulose fibers, whose 3D network structure firmly anchored abundant water molecules through hydrogen bonding and physical adsorption. After the membrane material was physically scratched, squeezed or partially damaged, the bound water inside the network flowed spontaneously to the damaged area driven by capillary force and surface energy, and quickly filled the defects and cracks. Therefore, the hydration layer quickly recovered to a continuous and complete state, realizing rapid self-healing at room temperature without external stimuli.

5 Interfacial Determinants of Oil/Water Separation

5.1 Surface Wettability

Interfacial chemical composition governs the intrinsic wettability of materials, which in turn dictates the selectivity in oil-water separation. In the separation of oil-in-water emulsions, materials that are superhydrophilic and underwater superoleophobic tend to adsorb water preferentially, facilitating the formation of a robust hydration layer. This hydration layer enables high-flux water permeation under low applied pressures while effectively repelling oil droplets [102]. Ho et al. [102] fabricated dendrite-type silica nanowires on stainless steel meshes via chemical vapor deposition, using silicon wafers as the silicon source. The nanowires uniformly covered the mesh wires and reduced the pore size from approximately 25 μm to below 10 μm, creating a superhydrophilic surface with a WCA of 0° and underwater superoleophobicity with an OCA of 166° (Fig. 10). Benefiting from the sieving effect and enhanced wettability, the nanowire mesh achieved 100% separation efficiency for both layered mixtures and emulsions, whereas the bare mesh only reached 12.9% for emulsions. Gelatin and tannic acid were coated onto a stainless-steel mesh. With a water flux of 1.74×10^5 $L \cdot m^{-2} \cdot h^{-1}$ and a separation efficiency >99.0%, no obvious attenuation was observed after 90 cycles. Regeneration proceeded merely through rinsing with deionized water [103]. When separating oil/water mixtures, superhydrophobic/superoleophilic materials facilitated rapid oil spreading and penetration, while effectively repelling water droplets. Separation selectivity increased with the content of low-surface-energy groups, such as long alkyl chains and silyl groups. The rough structure allowed for superhydrophobicity (WCA > 150°) and superoleophilicity. A superhydrophobic ramie fabric was fabricated, which featured high oil-water separation efficiency along with excellent heat resistance, mechanical and chemical stability even under extreme conditions. The separation efficiency exceeded 96.29% for six oil/water mixtures and was still higher than 95.36% after 30 cycles [104]. A superhydrophobic composite based on rice starch and shellac was coated on hydroxypropyl methylcellulose film by spray deposition method. Rice starch was added to build hierarchical surface roughness, with shellac acting as a natural low-surface-energy binder and interface compatibilizer. The results showed the formation of a uniform coating, enhanced interfacial hydrogen bonding and finely tuned micro-nanoscale structure. It exhibited a WCA of 153.1° and an SA of 2.7°, resulting from the synergistic balance between starch concentration and ball milling time [105]. Cavalcante and Szekely [106] leveraged the natural hydrophobin proteins of *Pleurotus ostreatus mycelium* to fabricate a self-growing Janus membrane without any additives. The membrane exhibited hydrophobicity (WCA = 140°) on the top surface and superhydrophilicity (WCA = 9°) at the interface with the PBI membrane contactor, achieving an ultrahigh oil-water selectivity of 622.5 (Fig. 11).

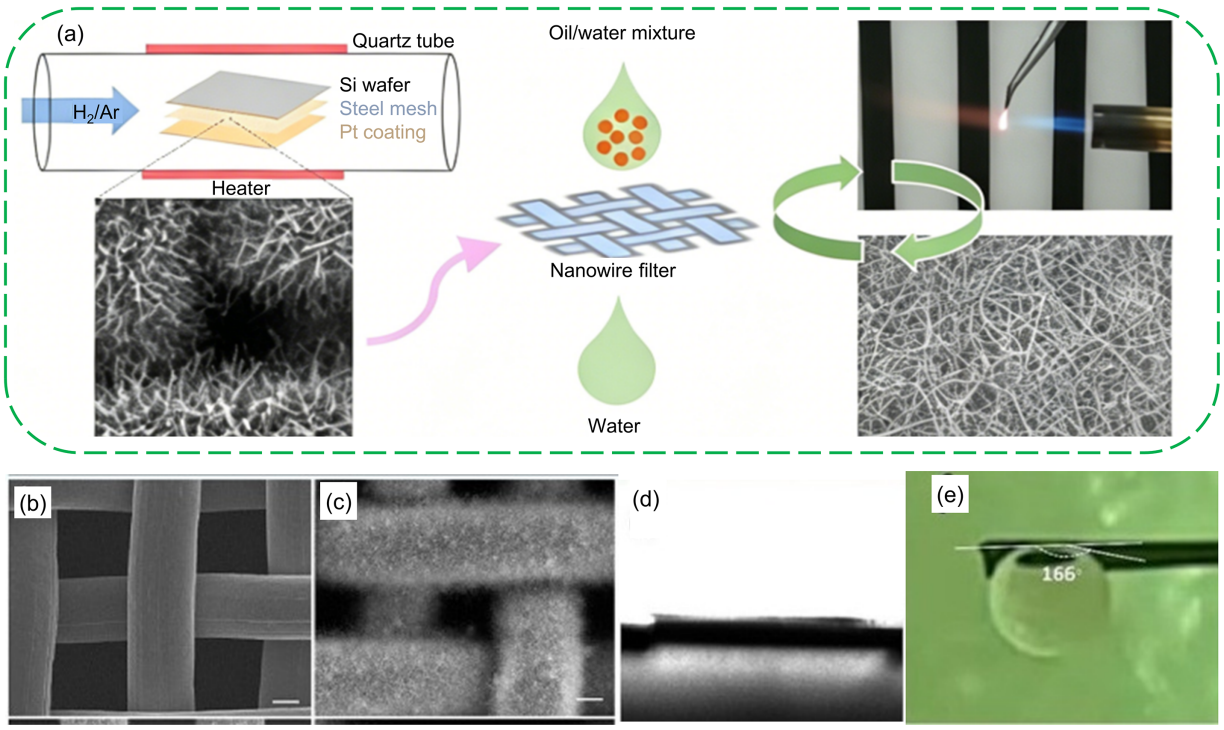

Figure 10: Fabrication, microscopic morphology and wettability performance of silica nanowire. (**a**) Fabrication process of silica nanowires grown on a steel mesh. SEM images of the (**b**) Bare stainless steel mesh and (**c**) Dendrite-type nanowires grown on the stainless steel mesh (scale bars: 10 μm). (**d**) Contact angle image of a water droplet spreading on the nanowire mesh to demonstrate its superhydrophilicity. (**e**) Photograph of a hexane droplet contacting the nanowire mesh immersed in water to demonstrate its underwater superoleophobicity, adapted from [102] licensed under CC BY.

5.2 Surface Charge

Surface charge and charging effect are essential for dealing with charged pollutants. For oil/water mixtures containing ionic surfactants or dyes, the zeta potential of the surface facilitates the efficient retention or adsorption of specific pollutants through electrostatic attraction or repulsion. Oil droplets are usually negatively charged, so the negatively charged hydrophilic interface has better antifouling performance. Kang et al. [107] prepared a nanofibrous membrane, which had a separation efficiency exceeding 99.5% for oil-in-water emulsions of various oils, for instance, toluene, diesel oil and petroleum ether. Following the synthesis of a di-cationic epoxy resin N,N′-diglycidyl-N,N,N′,N′-tetramethyl-1,3-propane diammonium dichloride as the crosslinking agent, CS/cellulose hydrogel was prepared. Due to the numerous quaternary ammonium groups, the material exhibited superhydrophilicity and underwater superoleophobicity, enabling efficient separation of various oil/water mixtures (efficiency >99.5%) with excellent recyclability, mechanical durability, and chemical resistance [21].

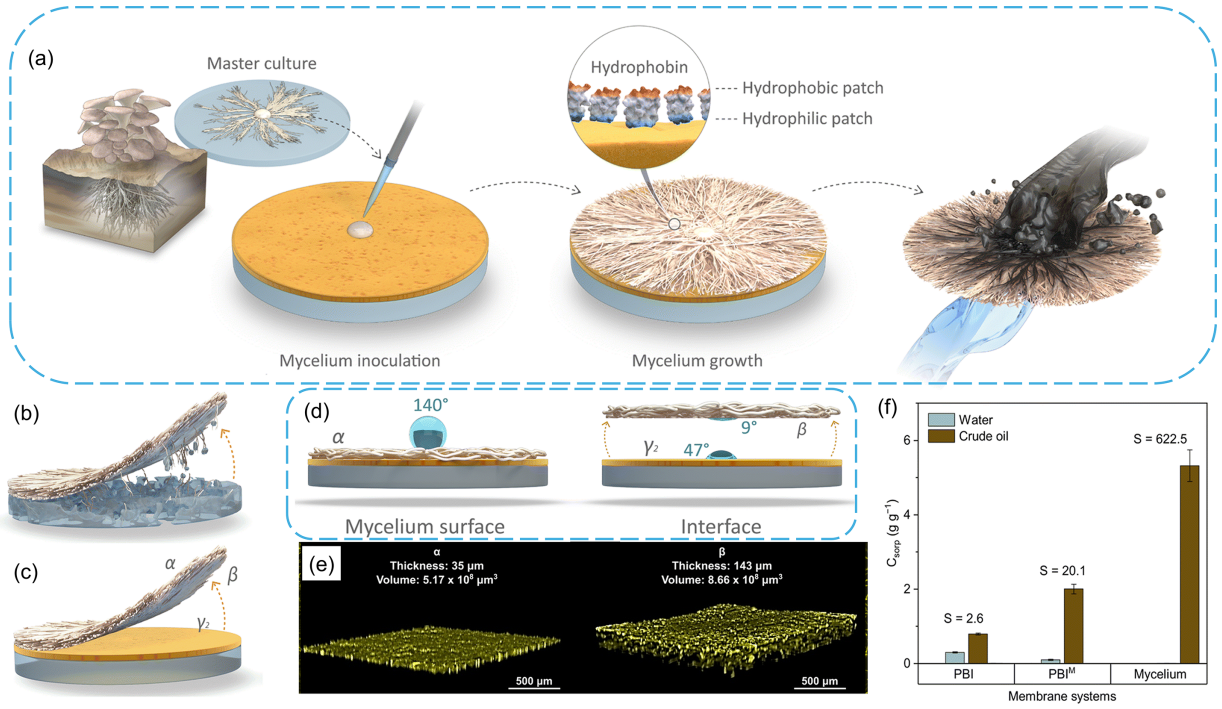

Figure 11: Superamphiphilic mycelium membrane derived from *Pleurotus ostreatus* for oil-water separation. (**a**) Schematic illustration of the fabrication process of *Pleurotus ostreatus mycelium*. The *mycelium* master culture was isolated and then inoculated on top of the membrane contactor, which allowed the *mycelium* membranes to grow. Through surface engineering via precise hydrophobin alignment, a superamphiphilic biomaterial suitable for oil-water separation was created. Detachment of *mycelium* membrane from with (**b**) and without (**c**) a nano-porous membrane contactor. (**d**) Surface definitions and schematic illustration of the WCA of each investigated surface. (**e**) 3D hyphae distribution of the *mycelium* membrane. (**f**) Crude oil and water sorption after 1 min treatment, adapted from [106] licensed under CC BY.

5.3 Stability of Network Structure

The practical application of materials is based on their mechanical and chemical stability. Strategies such as crosslinking [108,109], rigid NPs [27], and mussel-inspired strong adhesion [39] can significantly enhance the structural integrity of materials in harsh environments. Aminated lignin (AL) was loaded onto cotton fibers and subsequently crosslinked with green ethyl cellulose (EC) to synthesize hydrophobic/oleophilic AL@Cotton-EC. Considering the low-surface-energy feature of MTMS, researchers further constructed a superhydrophobic AL@Cotton-EC/MTMS separation membrane. The cotton fiber surfaces were wrapped with EC, and an interpenetrating porous structure featuring excellent wettability was fabricated. The AL@Cotton-EC/MTMS achieved an oil/water separation efficiency of 99.33% and good cycle stability. Furthermore, AL@Cotton-EC/MTMS was found to stay superhydrophobic even under simulated outdoor acidic, alkaline, and salt environments [108]. Polyethylene oxide (PEO) was employed as a pore-forming agent to synthesize stereocomplex PLA aerogels via freeze-drying and leaching of the pore-forming agent, which exhibited excellent oil/water separation and thermal insulation performances. It was found that the stereocomplex PLA aerogel exhibited a hierarchical micro/nano porous structure with complete stereocomplex crystallization. With the increase of PEO content, the pore size, porosity and hydrophobicity of the aerogel increased, and the density decreased [69]. Wu et al. [96] prepared MXene-based aerogels using CNFs. Owing to the plentiful functional groups of CNFs, the chemical crosslinking of MXene-based aerogels was promoted, leading to a significant improvement in water resistance, oxidation stability and

hydrophobicity. The ultra-thin one-dimensional characteristics of CNFs minimized the interlayer gap of MXene and created numerous heterogeneous interfaces, so that the aerogel exhibited outstanding electrical conductivity and electromagnetic interference shielding performance. Liu et al. [110] prepared hydrophobic plant fiber sponges using hexamethyldisilazane. The sponge also possessed high porosity, outstanding mechanical robustness and superior hydrophobicity with a WCA of 145°, presenting excellent oil absorption capability (15–40 g/g) and high selectivity. Assisted by a miniature diaphragm pump, it enabled continuous oil/water separation. A moderately rough structure can simultaneously improve separation efficiency and flux. For the superhydrophilic/underwater superoleophobic system, the rough structure can amplify the intrinsic hydrophilicity, strengthen the stability of the hydration layer, improve the oil droplet retention efficiency, and reduce the water phase penetration resistance to realize the simultaneous improvement of flux and efficiency. However, excessive roughness will lead to pore blockage, resulting in flux attenuation instead. When the pore size of the material is smaller than that of the emulsion droplets, effective retention can be achieved through size sieving, and the separation efficiency is enhanced with the reduction of pore size. However, too small pore size will significantly increase the fluid resistance, leading to a decrease in flux, so it is necessary to balance the two by adjusting the porosity. Benefiting from directional freezing-induced oriented channel architecture, bio-based MXene/carboxymethyl chitosan (CCS) composite aerogels presented remarkable structural stability [111]. The interconnected and aligned channel network not only reinforced mechanical robustness against compression but also maintained intact conductive pathways and porous structure under cyclic deformation (Fig. 12). Meanwhile, strong interfacial interactions between MXene nanosheets and CCS chains effectively prevented structural collapse and component detachment, thereby guaranteeing long-term structural stability in service. Such hierarchical and oriented network design offered a universal strategy for enhancing the structural stability of bio-based aerogels toward durable oil/water separation and multifunctional applications.

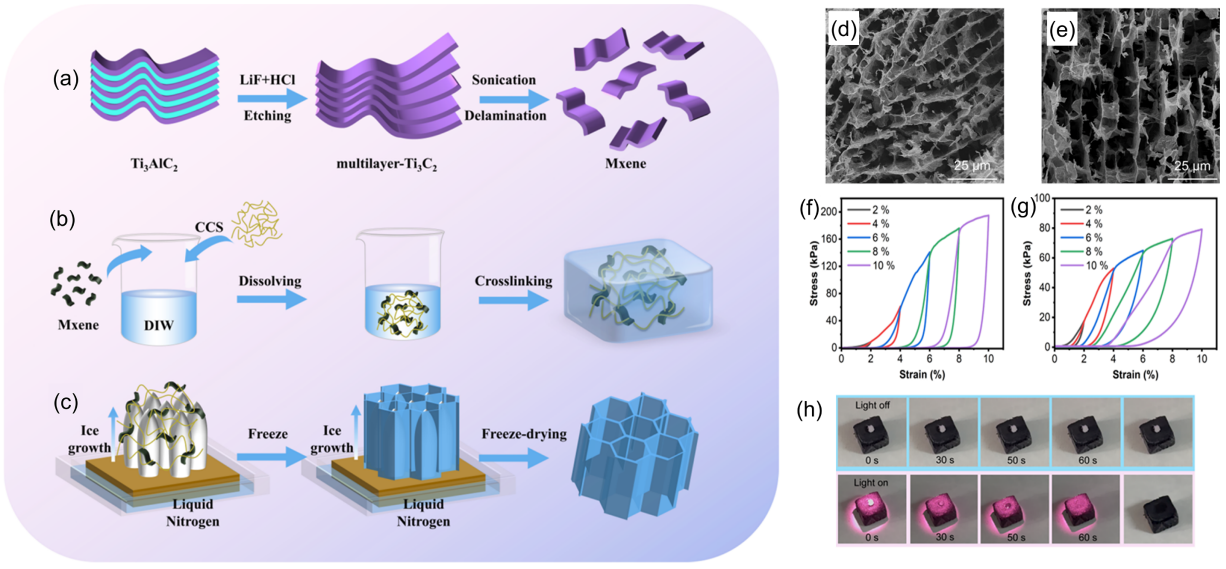

Figure 12: Fabrication and performance of MXene/CCS composite aerogel. (**a**) Preparation process of MXene nanosheets. (**b**) Dissolution gel of MXene/CCS. (**c**) Directional freeze-drying process of MXene/CCS. SEM images of the aerogel. (**d**) Along the vertical ice crystal growth direction and (**e**) along the ice crystal growth direction. Compression stress-strain curves of the composite aerogel. (**f**) Along the ice crystal growth direction and (**g**) perpendicular to the ice crystal growth direction. (**h**) Adsorption capability of the MXene/CCS composite aerogel for high-viscosity oil before and after laser irradiation. Adapted with permission from [111]. Copyright ©2026 American Chemical Society.

6 Summary and Outlook

Bio-based polymers show remarkable potential for oil/water separation owing to their sustainability and structural designability. Interfacial engineering is the core for achieving their high performance. Through strategies such as surface chemical grafting, NPs hybridization, biomimetic design, and smart interface construction, researchers have precisely regulated the surface wettability, charge, roughness, and network structure, thereby realizing high-flux, high-selectivity separation with durable antifouling performance.

Despite significant progress, scalable manufacturing represents an urgent challenge that must be overcome. Many interfacial engineering strategies are still in the laboratory stage, and continuous fabrication processes remain economically uncompetitive. Therefore, developing cost-effective, industrially scalable, and green-chemistry-compliant manufacturing technologies represents a crucial future direction. Fabrication is not the only outstanding challenge. Systematic data on material durability under harsh conditions, such as high salinity, mechanical abrasion, and biofouling, remain scarce, and the re-generation mechanisms of fouled membranes during continuous operation are still unresolved. Future work could explore stimuli-responsive interfaces that enable on-demand separation via pH, temper-ature, or light triggers, while integrating heavy-metal and dye adsorption would allow a single membrane to treat complex wastewater streams. These strategies suggest potential applications in portable purifiers, offshore spill cleanup, and protective textiles, provided that fouling resistance and recyclability can be substantially improved over repeated cycles. Finally, the biodegradability and end-of-life fate of these materials require rigorous evaluation to prevent secondary pollution, thereby substantiating their sustainability claims.

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Abbreviations

3D	Three Dimensional
WCA	Water Contact Angle
OCA	Oil Contact Angle
SA	Sliding Angle
CS	Chitosan
PDMS	Polydimethylsiloxane
PVA	Polyvinyl Alcohol
ChNF	Chitin Nanofibril
MTMS	Methyltrimethoxysilane
PLA	Polylactic Acid
PTF	Poly(propylene 2,5-furandicarboxylate)
PCL	Polycaprolactone
MTS	Methyltrichlorosilane
DTMS	Dodecyltrimethoxysilane

ODS	Octadecyltrimethoxysilane
CNT	Carbon Nanotube
PDA	Polydopamine
CNFs	Cellulose Nanofibers
LCNF	Lignin-Containing Cellulose Nanofibril
LCNFs	Lignin-Containing Cellulose Nanofibrils
NPs	Nanoparticles
MOF	Metal-Organic Framework
PEO	Polyethylene Oxide
AL	Aminated Lignin
EC	Ethyl Cellulose
SEM	Scanning Electron Microscope
FE-SEM	Field Emission Scanning Electron Microscope
FT-IR	Fourier Transform Infrared Spectroscopy
CVD	Chemical Vapor Deposition
CCS	Carboxymethyl Chitosan
RC	Regenerated Cellulose

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