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# A Review of Research on Materials for the Separation of Oil/water Mixtures

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## ABSTRACT

Water pollution caused by oil spills at sea and industrial and daily wastewater discharges are causing serious damage to the ecological environment, not only in terms of economic losses but also in terms of human health and survival, a problem that needs to be addressed urgently. Oil/water separation is a global challenge, and while these problems are frequent, various oil/water separation strategies have been extensively investigated in recent years. The efficiency of the materials prepared is a key factor, as are the environmental friendliness and low cost of the methods and raw materials used in the experiments. This work reviews methods and materials applied in oil/water separation in recent years, including natural textile materials, metal meshes, synthetic membranes, particulate adsorbent materials, foams, sponges, aerogels, smart controllable special wettable separation materials.

## INTRODUCTION

With rapid industrialization, oil spills in the food (Zhong et al. 2003), metal manufacturing (Rubio et al. 2002), petroleum (Pendashteh et al. 2011), pharmaceutical (Yu et al. 2017), weaving (Cheryan & Rajagopalan 1998) and other industries have led to mixed oil and water pollution of water resources (Wang et al. 2015). In addition to offshore oil spills, all areas of human life are subject to varying degrees of oil and water pollution. In addition to offshore oil spills, all areas of human life are subject to varying degrees of oil and water pollution. Oily wastewater has many compounds that are difficult to decompose, which causes serious damage to the ecological environment and endangers human health. It causes serious damage to the ecological environment and endangers the economy and health of human beings, and water pollution treatment is a global concern (Borges et al. 2016, Kang et al. 2020). Many studies have been carried out in recent years on efficient oil/water separation materials, with adsorption and filtration being the main means of treating oil/water mixtures (Dunderdale et al. 2015).

In addition to separation efficiency, environmental and cost issues and good recycling are key challenges for oil/water separation materials (Wang et al. 2009). This work describes the various methods and materials used in recent years for oil/water separation, including natural textile fibers (Lahiri et al. 2019), synthetic membranes (Lin et al. 2017), adsorbent particles or powders (Wang et al. 2018b), aerogels (Zhou et al. 2018a), sponges, and foams (Feng & Yao 2018, Zhou et al. 2018b), filtering metal meshes and intelligently controlled wettability separation specialties. The various modification methods and oil/water separation materials have advantages, disadvantages, and limitations. Adsorbent substances are commonly used for their simplicity, high absorption rate, recyclability, and environmental friendliness (Lü et al. 2016). Commercial sponges can be used as substrates in the oil/water separation field due to their low price, high specific surface area, low density, and high porosity. Still, they can't be used directly for oil/water separation (Ke et al. 2014). High lipophilicity can be achieved by roughly structuring the sponge surface or modifying it with low surface energy materials (Chen et al. 2008). Aerogels have the advantage of ultra-low density and high specific surface area for oil absorption and are commonly used to treat oil slick materials (Reynolds et al. 2001). Natural cellulose-based materials are widely developed due to their comprehensive source, good biocompatibility, and low cost. Fabric surfaces generally modify them to reduce free energy and construct rough structures (Gui et al. 2013, Zhu et al. 2009). Finally, the outlook for the application of these materials is presented.

## OIL/WATER SEPARATION MATERIALS

### Textile Materials

Natural textile materials can be used as good substrates for oil/water separation due to their low cost, high yield, stability, non-toxic, and environmentally friendly nature when handled properly (Mao et al. 2023, Xue et al. 2014). It is well known that the hydroxyl groups on the surface of natural fiber fabrics make them adsorb both oil and water, and they will not be used directly in the field of oil/water separation. The oleophilic and hydrophobic or hydrophilic properties are generally achieved by modifying the fabric's surface with chemicals to reduce the surface free energy and create a rough surface structure (Zhao et al. 2021). Various synthetic fibers are also attracting increasing interest (Gao et al. 2019b, Peng et al. 2020). These fabrics have good oil absorption properties and oil retention capacity under external forces and have promising applications in oil spill remediation (Lin et al. 2012).

Su et al. (2018) reported a magnetic field manipulation method for preparing bifunctional superhydrophobic textiles with asymmetric rolling/fixed states. Iron oxide (F-$Fe_3O_4$) nanoparticles were modified by fluorosilanes and fabricated along the magnetic direction in the presence of polydimethylsiloxane (PDMS), forming surfaces with different levels and roughness on both sides of the fabric. The prepared textiles have superhydrophobicity, magnetic responsiveness, good chemical stability, adjustable surface morphology, and controllable adhesion. A schematic diagram of the fabrication of a bifunctional superhydrophobic fabric is shown in Fig. 1. Han et al. (2018) prepared a superhydrophobic cotton fabric by depositing polydimethylsiloxane (PDMS) coated $SiO_2$ nanoparticles on the surface of the cotton fabric using a hot gas deposition. It was found that the samples could only repel surfactant solutions and pure water when the PDMS/$SiO_2$ content deposited on the surface was further increased and the nanoparticles completely covered the surface gaps. The separation efficiency was stable in solutions with poor chemical conditions. Zhou et al. (2017) introduced phytate metal complexes on the fabric's surface to generate a rough hierarchical structure, followed by PDMS modification. A superhydrophobic cotton fabric was prepared. It has excellent resistance to UV radiation, high temperature, organic solvent impregnation, and good mechanical wear properties. The superhydrophobic/superhydrophilic fabric successfully separated oil/water

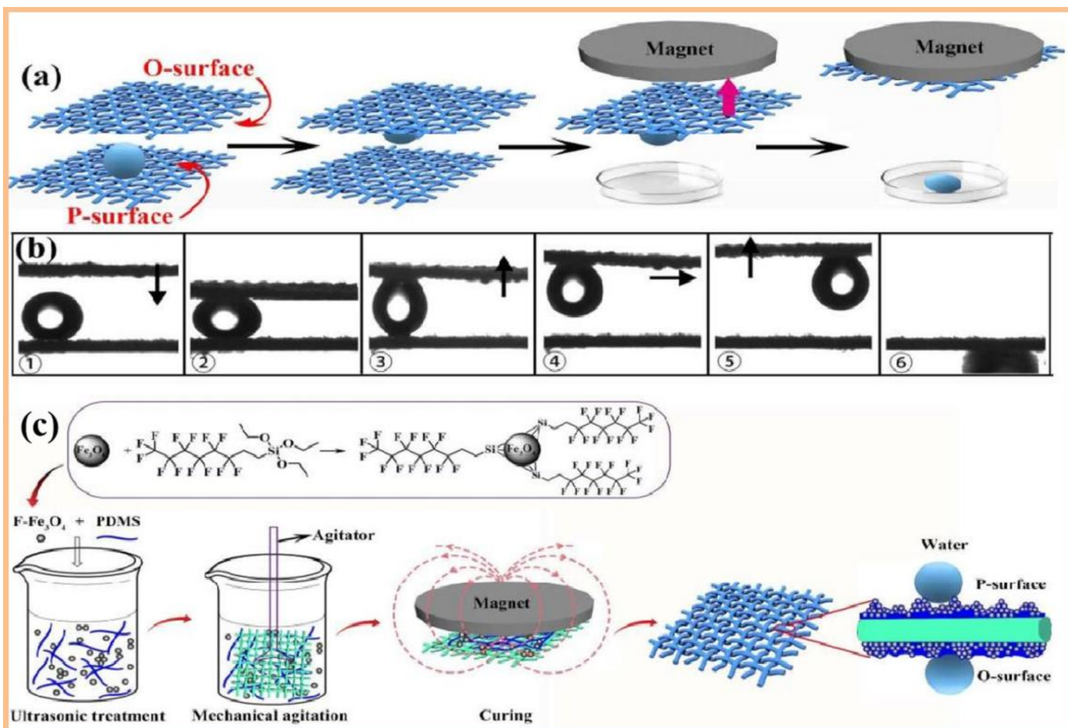


Fig. 1: (a) Schematic process and (b) detailed process of water droplet transportation, (c) schematic illustration for fabricating dual-functional superhydrophobic textile with roll-down/pinned states.

mixtures with a separation efficiency of 99.5 %. These super-resistant fabrics with phytate-metal complexes and polydimethylsiloxane (PDMS) are environmentally friendly, cost-effective, sustainable, and easily scalable.

The sol-gel method is the hydrolytic polymerization or hydrolytic condensation of raw materials in solution into porous spatially structured solids in a certain way (Antonelli & Ying 1995). Currently, nanoparticles such as silica are mainly used to build superhydrophobic surfaces on the surface of substrates, and this method is simple, effective, and product-controlled. Xie et al. (2019) used tetraethyl orthosilicate and hexadecyltrimethoxysilane, and cetyltrimethoxysilane to chemically modify cellulose membranes to prepare sustainable superhydrophobic cellulose membrane, which is a simple and efficient method that is environmentally friendly and durable, with an oil/water separation efficiency of up to 98 % even under harsh conditions. A schematic diagram of the separation mechanism, the separation efficiency of various oil/water mixtures, and the SEM diagram are shown in Fig. 2.

Wang et al. (2016) prepared silylated cellulose using the sol-gel reaction of biodegradable microcrystalline cellulose and hexadecyltrimethoxysilane. The separation efficiency reached 99.93%. The modified silylated cellulose is a porous structure material, and its special hydrophobic properties give it good separation performance. Fig. 3 (a) shows the experimental silylated synthesis route.

Previous superhydrophobic cotton fabrics were not robust enough, so Cheng et al. (2019) prepared durable superhydrophobic cotton fabrics by a one-step solvothermal method. They were resistant to abrasion and sonication and maintained high efficiency in hydrophobic separation after treatment with high-temperature liquid nitrogen. Unlike other methods that use toxic fluorinated reagents to reduce surface-free energy, the team used divinylbenzene to form surface deposits on the cotton fabric under thermal polymerization of the solution, which is non-toxic and

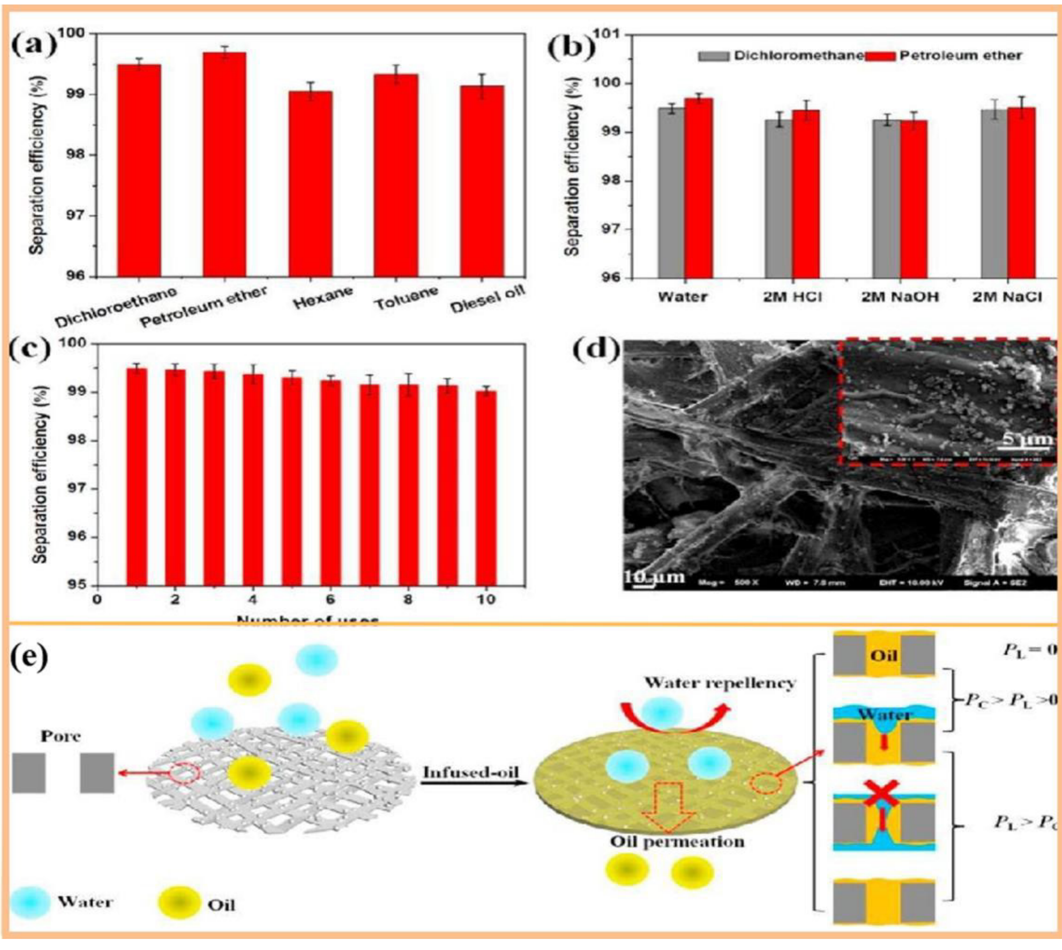


Fig. 2: (a) Separation efficiency for various oil/water mixtures, (b) and oils/corrosive liquids (2 M of acid, alkali, and salt solution), (c) separation efficiency for recycled separation, (d) SEM images of SOCM after 10 recycles, (e) schematic diagram of separation mechanism of SOCM.

reduces production costs, with separation efficiencies of up to 98 %. Fig. 3 (b) shows a schematic diagram of the one-step solvent and thermal polymerization method for preparing PDVB@CF.

Hu et al. (2020) synthesized an asymmetric wettability and Janus composite fabric by combining surface hydrophobic, oxidative, and chemical bath deposition treatments. The asymmetric wettability means that the composite fabric is partly silanized to obtain the hydrophobic side and partly oxidized at the interface to obtain the hydrophilic side. Based on this, the material can transport water in one direction and is more permeable than untreated fabrics. The method also combines gravity-driven effects to improve oil/water separation, which can be applied to other substrates and has potential applications. Fig. 3 (c) shows a schematic representation of the preparation process of the composite fabric. The process includes surface hydrophobization, oxidation treatment, and chemical bath deposition. Zhang et al. (2016) prepared a superhydrophobic and superoleophilic fabric by forming a certain roughness on the surface of the polyester fabric through the etching effect of alkali on the fibers and then dipping it in a mixture of intrinsic microporous polymer (PIM-1) and fluorinated alkyl silane (PTES) to reduce the surface free energy.

The main route to self-cleaning oil/water-separating cotton fabrics is using $TiO_2$ and its nanocomposites, which can achieve self-cleaning by decomposing pollutants due to their photocatalytic activity and hydrophilicity after light induction. The materials are inexpensive, environmentally friendly, and efficient (Montazer et al. 2011, Tung & Daoud 2013). Shaheen et al. (2019) used a sol-gel method to combine and condense titanium dioxide nanosol and silica nanosol on the surface of cotton fabrics. Unlike the traditional immobilized particle method, they used in situ deposition to improve the durability of the modified fabrics and combined with the photocatalytic effect of $TiO_2$ to produce superhydrophobic fabrics with certain self-cleaning ability, anti-bacterial and UV protection, and also have the prospect of mass production. Fig. 3 (d) shows a schematic diagram of the in-situ preparation of nanosol on cotton fabric and its surface modification. Yang et al. (2018a) deposited silica nanoparticles encapsulated by polydimethylsiloxane on the surface of the non-woven fabric by impregnation vapor phase deposition to obtain hydrophobic cotton samples that remained stable under harsh chemical conditions such as acidic, alkaline, and NaCl solutions.

Layer-by-layer is a top-down method for preparing environmentally friendly, inexpensive, multifunctional solid coatings. It allows control of the thickness and shape of the coating compared to other methods (Li et al. 2014). The driving forces (Borges & Mano 2014) for layer-by-layer assembly include electrostatic interactions (Decher & Hong 1991), hydrogen bonding (Kim et al. 2008), and coordination (Wang et al. 2012). Yang et al. (2017) prepared superhydrophobic paper by assembling and depositing cationic starch and sodium alginate on the paper surface and silylating the surface with non-fluorosilane and trichloromethylsilane. They showed excellent resistance to moisture, self-cleaning ability, and higher tensile strength compared to the original paper.

Joung & Buie (2015) used electrostatic self-assembly techniques to deposit silica nanoparticles in a homogeneous dispersion on the surface of polyester fabrics to obtain a superhydrophobic surface by this rapid modification. In their previous work (Xu et al. 2013), superhydrophobic surfaces were also prepared using electrophoretic deposition (EPD) combined with polydimethylsiloxane (PDMS) modified $SiO_2$ nanoparticles (PDMS-$SiO_2$), which is not only fast, but can be controlled by electric field and time during the experimental reaction to control the thickness and shape of the coating, largely improving the durability of the modified fabric, and the combination of hybrid techniques defeats the combination of hybrid techniques eliminates the problem of not being able to control the deposition thickness through electrophoretic deposition methods.

Cellulose-based film materials can be used as candidate substrates for achieving superhydrophobic and superoleophilic surfaces by impregnation, spin coating, or spraying. Kollarigowda et al. (2017) prepared an oil/water separation material using cellulose membranes (CM) as a substrate through a simple chemical modification. Environmentally friendly anti-pollution membranes for oil/water separation, litter particle filtration, and thiol odor barrier were prepared by modifying the superhydrophilic nature of CM with silane and lauryl monomer block copolymers via reversible addition-transition transfer (RAFT) polymerization. The CM surface was modified from its original superhydrophilic nature to superhydrophobic, establishing efficient oil/water separation and thiol filtration. The film is environmentally friendly, biodegradable, and can effectively adsorb particles in complex water-oil particulate systems. Yong et al. (2018) reported the preparation of underwater oil sheets by a simple mechanical drilling process after forming microporous arrays on the surface of wood sheets. Composed of hydrophilic cellulose nanofibres, lignin, and hemicellulose, the wood was hydrophilic, and its inherent porous structure gave the surface excellent underwater superoleophobic properties. The synthetic flakes were successfully applied to separate oil/water mixtures and showed good separation capabilities. This separation method was green and cost-effective.

### Metal Mesh

Metal mesh materials have a large surface area and porous structure with a controllable structure. They can also selectively adsorb oil/water mixtures under the influence of gravity, so gravity drive is a factor to be considered in preparing metal mesh separation materials. However, such meshes are sensitive to acid, alkali, or salt solutions and are only suitable under mild conditions. Usually, by treating the metal mesh with high surface energy and surface roughness underwater with hydrophobicity, water penetrates and forms a water film which forms a repellent effect on the oil in the mixture, thus achieving oil/water separation (Kollarigowda et al. 2017, Xu et al. 2013). Commonly prepared by etching a modified metal surface and then by coating the particles (Yong et al. 2018).

Such special wettable materials can be effective for oil/water separation but are also susceptible to oil contamination or blockage, as their lipophilicity affects the separation efficiency. Dai et al. (2020) prepared polytetrafluoroethylene (PTFE) nanofibre coatings by molecular self-assembly. Thermally aged PTFE nanoparticles could undergo molecular rearrangement to obtain ultrathin PTFE nanofibre coatings with high homogeneity and excellent chemical stability by using electrostatic interactions as the driving force for self-assembly. The fiber is superhydrophobic and superlipophilic, effectively separates various oil/water mixtures, has good reusability, and has proven highly resistant to strong acids, bases, and salt solutions. A schematic diagram of the PTFE nanofibre coating preparation process is shown in Fig. 4 (a).

The anchoring ability and accessibility of plant polyphenols inspire them. Chen et al. (2018) prepared an underwater super-oil repellent coating based on metal sheets using TA modified by dip-coating method to obtain a TA-coated copper surface with excellent underwater super-oil repellent properties. The anchoring effect of TA molecules with the metal surface resulted in a tantalum-plated copper surface with excellent underwater superhydrophobicity and ultra-low adhesion, easy processing, and low cost. The design of Du et al. (2018) was inspired by the superhydrophobicity of the lotus leaf surface. The stainless-steel mesh was etched with hydrofluoric acid, coated with silver nanoparticles, and modified with stearic acid (STA) to construct a superhydrophobic oleophilic structure with micro/nanostructures. The treated stainless-steel mesh is superhydrophobic and lipophilic, showing up to 98 % separation efficiency in oil/water separation experiments and maintaining a high separation efficiency after multiple cycles. In addition, the mesh shows good chemical resistance under harsh mixed solution conditions and good wear resistance in hot water. A schematic diagram of the fabrication of a superhydrophobic/superoleophilic stainless-steel mesh is shown in Fig. 4 (b). Inspired by superhydrophobic stainless-steel filters, Zeng et al. (2017) produced a superhydrophobic/superoleophilic stainless-steel

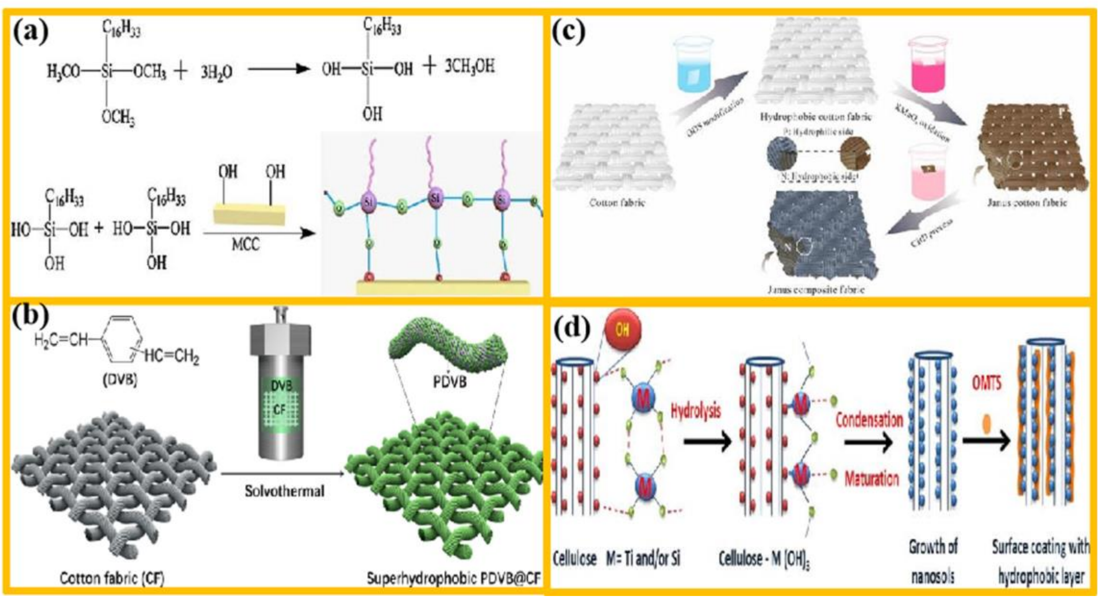


Fig. 3: (a) Synthetic route for silanization of MCC, (b) schematic illustration of the fabrication of PDVB@CF via a one-step solvothermal polymerization, (c) schematic illustration of the preparation process of a Janus composite fabric. The process includes surface hydrophobization, oxidation treatment, and chemical bath deposition, (d) diagram illustration of the in situ preparation of nano sols onto cotton fabrics, and their surface modification with OMTS.

mesh (SBS-SSM) filter. The hydrophilic silica particles were assembled on the stainless-steel mesh by electrostatic self-assembly. Then a fluoropolymer/$SiO_2$ nanoparticle solution was coated on the substrate, allowing the stainless-steel mesh surface to mimic beetle water back and obtain oil/water separation, which is simple and environmentally friendly compared to other superhydrophobic/superoleophilic surface preparation methods. The principle diagram of the SBS-SSM preparation is shown in Fig. 4 (c). Wang et al. (2018a) prepared TA/PVP coated stainless steel mesh using natural water-soluble polyphenols (TA) and highly biocompatible polymers (PVP). The materials are superhydrophilic and submerged superhydrophobic and can effectively separate oil/water mixtures. TA is a biocompatible, low toxicity, and superwet material. In addition, the TA/PVP coated stainless steel mesh exhibits good anti-fouling properties and is made into a funnel shape. Underwater oil collection units with TA/PVP coated stainless steel mesh in combination with underwater oil/water separation systems provide continuous collection and removal of oil contaminants from the underwater environment.

Zhou et al. (2021) used a fast, low-cost electrodeposition method to attach a metal-organic framework consisting of $Co^{2+}$ ions and 2-methylimidazole to the copper mesh, which can be prepared in less than ten minutes. The difference in the action of $Co^{2+}$ with specific groups (carboxyl or sulfhydryl) produces adsorption properties on mixed oil and water solutions with a long-lasting and stable separation capacity. After ten cycles, it still has 98 % separation efficiency.

Cao and Liu (2021) first ultrasonically pretreated the copper mesh and then prepared an underwater superoleophobic copper mesh by layer-by-layer self-assembly of inorganic sodium silicate and alumina powder (SSA) on copper (Cu) mesh. The modified copper mesh achieved an oil/water separation efficiency of 95 % even after heat treatment in a 700°C solution and maintained an efficiency of over 96 % after thirty cycles. Moreover, this experimental preparation required a short time and low raw material costs.

Gao et al. (2021) prepared a stainless steel mesh film with switchable wettability by in situ generation of ZnO nanowire arrays of ZnO-NAs on the surface of the stainless steel mesh, modified with hydrophobic properties. The preparation process also utilizes electro-wetting-induced oil/water separation, using ZnO-NAs to coat the SSM and a silane coupling agent as a bridging agent to improve the adhesion between the film and the metal mesh. Applying a low electric field controls this intelligent switchable wettability, and the response time is very fast. This method is extremely effective in treating wastewater and has good practicality. The modified material is not only durable but also has some antifouling ability.

Zhang et al. (2021) constructed by etching and in situ growth of a Prussian blue analog on a nickel grid with a coarse micro- and nano-scale grid structure. The composite grid exhibits strong superhydrophilicity and submerged

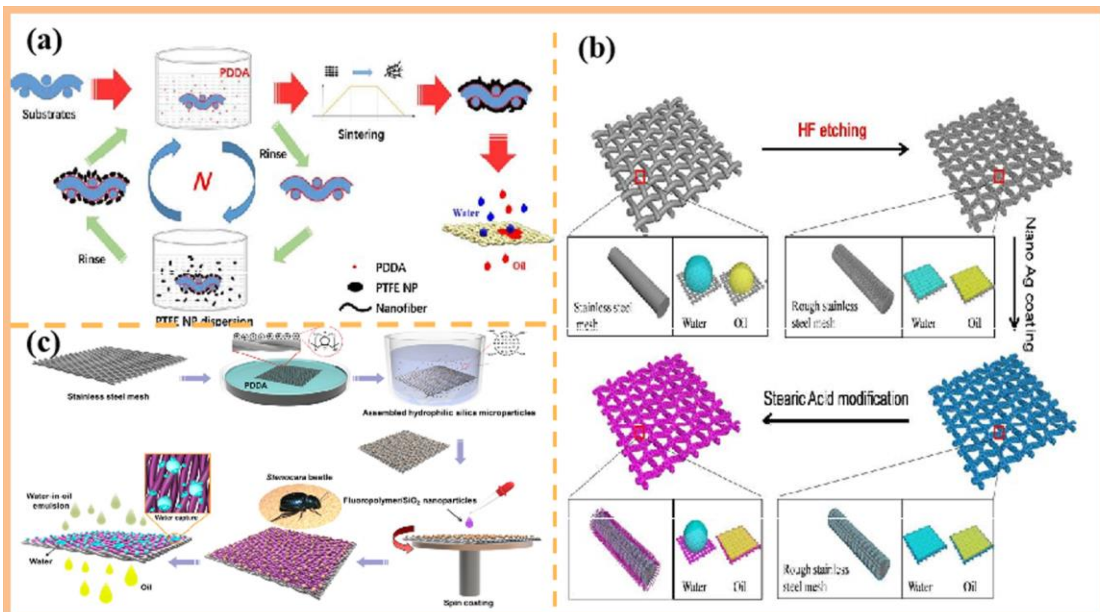


Fig. 4: (a) Schematic diagrams of the preparation process of PTFE nanofibrous coatings, (b) schematic illustration of the fabrication of the superhydrophobic/superoleophilic stainless-steel mesh, (c) schematic illustration of the fabrication of the superhydrophobic/superoleophilic stainless-steel mesh (SBS-SSM) filter.

superoleophobicity, and its stable separation efficiency and resistance to acids and bases make it a promising material for wastewater separation.

It is well known that separation materials are contaminated by crude oil as the surface free energy of contact with crude oil decreases in the dry state. Hence, materials must be pre-hydrated before oil/water separation (Peng et al. 2018, Yang et al. 2019). Filter membrane materials are commonly used in wastewater treatment, but their lack of pre-hydration is a major challenge (Zhao et al. 2019). Wang et al. (2021) used chitosan (CS)-cellulose nanocrystals (CNCs) in this way, the modified metal mesh has better hydration ability in both air and wastewater, reducing the risk of oil contamination while providing excellent oil/water separation performance. The raw material of this experiment, chitosan, is inexpensive and environmentally compatible, and the material prepared from cellulose is economical and environmentally friendly (Ma et al. 2021).

## Synthetic Membrane Materials

Membrane separation technology is widely used in wastewater treatment due to its low energy (Lee & Park 2013), simple operation, and lack of secondary contaminants (Shi et al. 2019). However, conventional separation materials tend to have low efficiency, stability, and selectivity (Matsubayashi et al. 2017), limiting their application in oil/water separation (Wang et al. 2022).

Shi et al. (2018) used mussel adhesion protein to stimulate dopamine-modified poly (lactic acid) (PLA) nonwovens to obtain polydopamine (PDA)/PLA nonwovens. As a hygroscopic and oil separation material, it has high adsorption performance and selectivity and excellent photocatalytic degradation ability for various soluble organic pollutants under UV irradiation. The controlled wetting performance of the nonwoven was achieved by controlling the relative ratio of butyl titanate ($Ti(OBu)_4$) to heptadecanoic acid (HFA) during the preparation process. Wu et al. (2018) coated dopamine on polyvinylidene fluoride (PVDF) membranes and then modified the superhydrophobic PVDF membranes by adding glutathione (GSH) through a simple reaction, reducing the serous membrane contamination caused by oil/water separation. The membrane was modified by adding glutathione (GSH) to the superhydrophobic PVDF membrane in a simple reaction, reducing the serous membrane contamination caused by oil/water separation and improving the reusability of the material. The PVDF membrane with high pure water permeability, good anti-fouling properties, and reusability, making it suitable for long-term efficient membrane separation. Fig. 5 shows FE-SEM images of PVDF (a, d), PVDF@PDA (b, e), and PVDF@PDA-GSH (c, f) surfaces. The PVDF@PDA-GSH membranes were prepared schematically, as shown in Fig. 5 (g).

Gupta and Kandasubramanian (2017) polymerized non-ionic surfactant-soaked nano tetrafluoroethylene

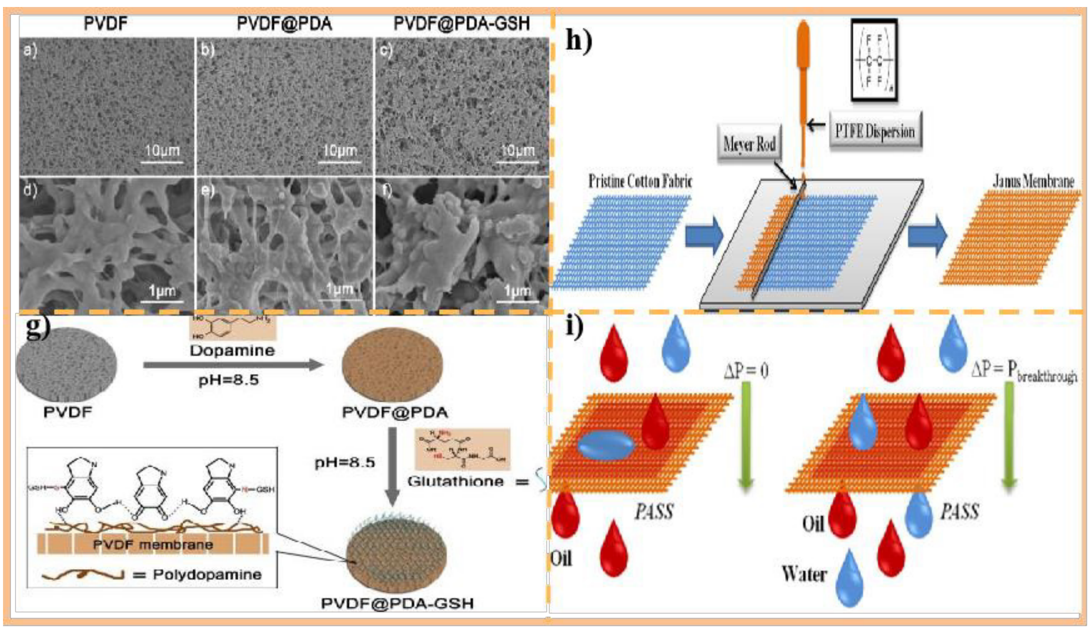


Fig. 5: (a-f) FE-SEM images of the surface of (a, d) PVDF, (b, e) PVDF@PDA, and (c, f) PVDF@PDA-GSH. The time of PDA and GSH treatment was 4 and 2 h, respectively, (g) schematic illustration for the fabrication of PVDF@PDA-GSH membrane, (h) schematic of the fabrication of Janus membrane, (i) Janus membrane as a directional fluid diode.

(PTFE) dispersions into modified superhydrophobic/superhydrophilic oil Janus films using the Meyer rod coating technique. This Janus film has excellent non-homogeneous wettability and anti-icing properties. We can still perform very good oil/water separation in some extreme environments and also have some flame retardancy and recyclability. An et al. (2018) prepared charged Janus membranes (CNTs) and hydrophobic microfiltration membranes using vacuum filtration. Hydrophilicity regulates the thickness of the carbon nanotube coating by controlling the number of carbon nanotubes deposited on the substrate membrane. This is because Janus films use the charge shielding effect to emulsify and separate heavy and light oils in emulsions. This team solved the problem that the normal Janus film preparation method couldn't control the thickness of the hydrophilic emulsion breaking layer. Fig. 5 (h) shows a schematic diagram of the fabrication of the double mask, and Fig. 5 (i) shows a schematic diagram of the double mask as a directional fluid diode.

## Granular or Powdered Materials

Granular or powdered oil absorbent materials have excellent oil absorption properties (Zhang et al. 2020) and have been the star material in oil and water separation, which is particularly effective in the face of large areas of contamination, which has also received attention. Common metal oxides include $TiO_2$, $SiO_2$, $Fe_3O_4$, ZnO, $Al_2O_3$, and others, generally used as hydrophobic agents to modify other substrates (Eskandari et al. 2021). However, they must often be modified with hydrophobic polymers to obtain better oil absorption or improve hydrophobicity. High cost, low efficiency, and environmental pollution, the performance of the material is prone to rapid degradation due to surface corrosion damage by the external environment (Ishchenko et al. 2021, Tabrizian & Amoozadeh 2016), the mechanical durability of the surface structure, chemical resistance remains a challenge, which makes the large-scale use of granular oil absorption materials limited.

Qu et al. (2018) produced a superwetted material based on kaolinite nanoparticles, exhibiting good oil removal properties in the air and underwater. The addition of perfluorooctanoic acid to form long chains under alkaline conditions greatly reduced the surface energy of the kaolin, giving the modified kaolin material good oil removal properties, and further improved the durability and chemical resistance of the modified kaolin material by binding tightly to the substrate in the presence of bis(3-trimethoxysilylpropyl)amine. Unlike previous materials, this material is suitable for almost any substrate and exhibits good hydrophilicity in air and water. Compared to conventional functional materials, the coating is stain-resistant, energy efficient, and easily recyclable. Fig. 6 (a) shows the wetting behavior and oil/water separation of modified superhydrophilic/superoleophobic kaolin materials. Wang et al. (2018b) prepared superhydrophobic particles by modifying $TiO_2$, infiltrated the modified particle suspension into the material, and prepared 3D materials with superhydrophobic and superhydrophilic properties by simple impregnation and coating. By adding a certain

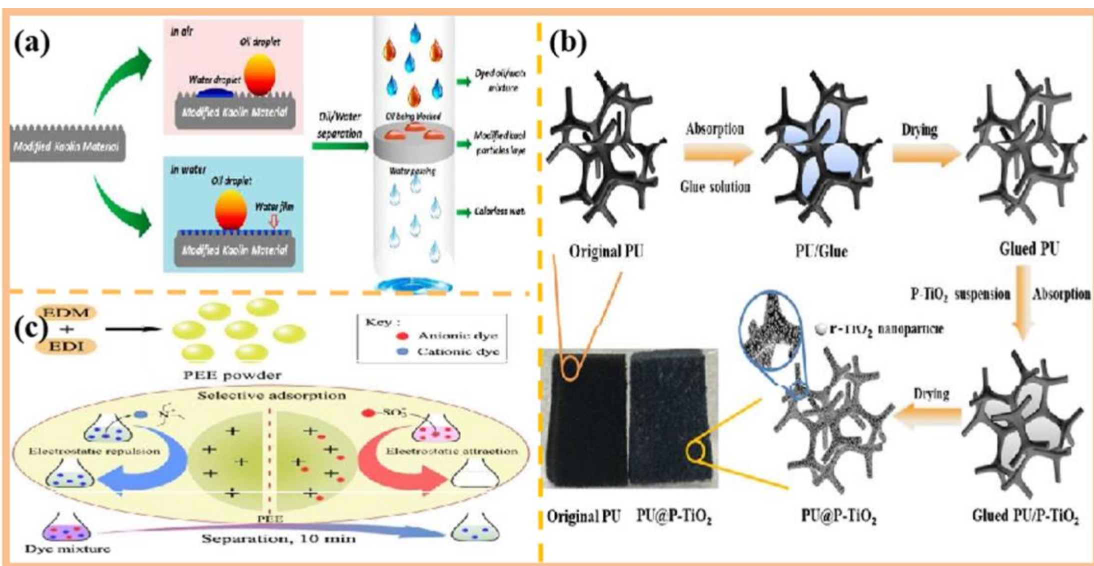


Fig. 6: (a) Schematics of the wetting behavior and oil/water separation of the modified superhydrophilic/superoleophobic kaolin materials, (b) schematic illustration of the fabrication process for PU@P-$TiO_2$ composite, (c) schematic of selective adsorption and separation process of dyes.

amount of $TiO_2$ nanoparticles to the hydrolyzed PTES solution, the hydroxyl groups on the $TiO_2$ surface condensed with water, and hydrophobic groups grafted onto the $TiO_2$ nanoparticle surface. And due to the glue's stability and the material's environmentally friendly nature, the oil removal function is efficient and long-lasting by means of good reuse and recyclability. A schematic diagram of the preparation process of PU@P-$TiO_2$ composites is shown in Fig. 6 (b).

Li et al. (2017) prepared quaternary ammonium salt hydrogels by a simple one-step copolymerization method as an effective adsorbent for removing dyes and insoluble oils and fats from water. It is an organic cationic adsorbent with large $R_4N^+$ groups and has a much higher selective adsorption efficiency for anionic dyes than other commonly used adsorbents. The poor adsorption capacity of cationic dyes can be used for efficient and rapid adsorption on hybrid solutions. In addition, the adsorbent is reusable, efficient, and durable, making it a simple, inexpensive, and truly efficient adsorbent. The principle diagram of the dye-selective adsorption separation process is shown in Fig. 6 (c).

**Aerogels**

Aerogels are wet gels filled with air pores, usually obtained from dried organic or hydrogels (García-González et al. 2012, Pierre & Pajonk 2002). Using the properties of the porous structure, lightweight materials such as carbon nanotubes, graphene, silica, and alumina can be prepared, which are characterized by a large specific surface area and low density and have a wide range of applications (Liebner et al. 2008, Mohanan et al. 2005). However, aerogels are expensive, complex, and non-biodegradable, limiting their applications. Zhang et al. (2018) inspired cancellous bone. They successfully prepared many HAP nanopores with three-dimensional interconnection, porous mesh structure, special structure with high porosity and interconnection, and better adsorption of oil and organic solvents by using HAP nanowires freeze-drying method without using organic solvents. Compared to organic aerogels, HAP nanowire aerogels have good, environmentally friendly, and low-cost biocompatibility. The hydrophobic HAP nanowire aerogels have good PM 2.5 filtration efficiency, low airflow resistance, and high adsorption capacity for different oil and organic solvents. In addition, the hydrophobic HAP nanowire aerogels have excellent elastic properties, high reusability, and good recycling capacity(Hou et al. 2020, Liu et al. 2014). Fig. 7 shows (a) a schematic diagram of the cancellous bone microstructure, (b) digital images and SEM micrographs of cancellous bone, and (c) digital images and SEM micrographs of the prepared HAP nanowire aerogel.

Gao et al. (2018) used a mussel adhesive as an inspiration and a polydopamine interlayer as a medium. The polydopamine (PDA) coating prepared from the mussel adhesive was uniformly spun on nano-fibrillated cellulose (NFC) scaffold, linking the hydrophilic NFC with the desired hydrophobic octadecyl amine (ODA) molecules to give nano-fibrillated NFC surface hydrophobicity. With a high contact angle, the new superhydrophobic NFC aerogel allows the NFC scaffold to come into complete contact with the modifier, generating a structurally homogeneous aerogel that maintains its original porous structure intact and can absorb various oils and solvents in aqueous solutions. In nature, mussels have strong adhesion to almost all types of substrates through the secretion of adhesion proteins. The composite aerogel has excellent superhydrophobicity, ultra-low density, and high porosity. It has good oil/water absorption selectivity and can adsorb various organic solvents.

Yang et al. (2018b) successfully synthesized peptidoglycan aerogels using short peptides and polysaccharides by molecular self-assembly and freeze-drying. The microporous peptidoglycan aerogel was prepared by freeze-drying a hydrogel co-assembled with a simple dipeptide and the polysaccharide konjac dextran (KGM), which had good separation properties for various oils and fats. The peptide consists of a base methoxy attached to the C-terminus of a diphenylalanine peptide (Fmoc, FF), which self-assembles in water into a hydrogel composed of one-dimensional nanofibres. KGM is a functional polysaccharide with good hydrophilicity and gelation properties. The microstructure of the aerogel can be controlled by varying the mass ratio of Fmoc, FF, and KGM. KGM solution is rapidly freeze-dried in liquid nitrogen to form an aerogel with relatively weak mechanical strength compared to other inorganic aerogels. Still, the synthesized composite aerogel has good biocompatibility with peptide materials in oil/water separation.

Zhou et al. (2018a) successfully constructed superhydrophobic aerogels using silylated cellulose nanofibres and silica nanoparticles by a simple freeze-drying method. The prepared aerogel has high porosity, low density, good hydrophobicity, and a large water contact angle. It can be used for the filtration separation of surfactant-stabilized oil-in-water emulsions. The aerogel has a porous gradient structure with high roughness and low surface energy. The hydrophobic modifier reduces the surface energy of cellulose nanofibres and silica nanoparticles. The modified aerogel's density and specific surface area are increased by adding silica nanoparticles. By adding silica nanoparticles to improve the surface roughness and porosity of the material, the rough laminar structure and the lower surface can make the

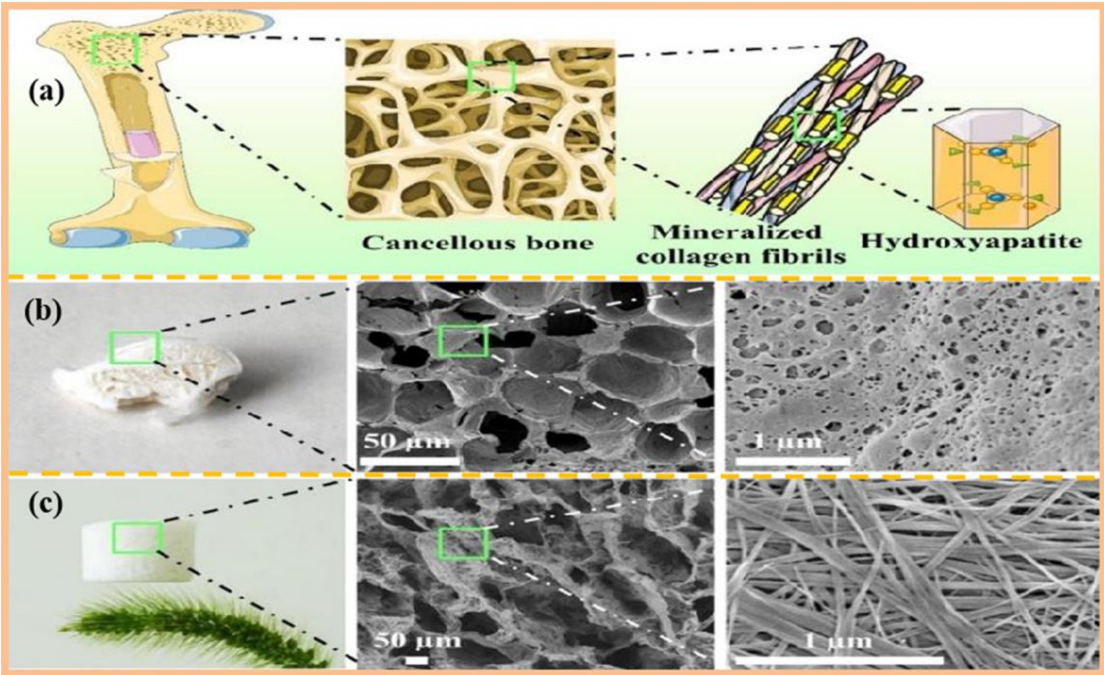


Fig. 7: (a) Schematic illustration of the microstructure of the cancellous bone, (b) digital image and SEM micrographs of the cancellous bone, (c) digital image and SEM micrographs of the as-prepared HAP nanowire aerogel.

aerogel superhydrophobic and highly efficient in separating oil/water emulsions without external pressure. It also has good anti-fouling properties and recyclability with long service life.

## Sponge Materials

Common sponge materials include polyurethane and melamine sponges (Liu et al. 2017, Lou et al. 2004). Due to their low price, high specific surface area, low density, and high porosity, they can be used as oil/water separation substrates. Still, they can't be used directly for oil/water separation. High lipophilicity can be achieved by constructing the sponge surface structure more roughly or modifying it with low surface energy materials (Matsubayashi et al. 2017). Peng et al. (2018) reported superhydrophobic polydimethylsiloxane (PDMS) via UV-assisted thiol and prepared superhydrophobic and lipophilic melamine sponges from sponge materials by membrane functionalization. Due to the special strip microstructure and the hydrophobicity of the silicone, the new material has a high contact angle. First, the polymercapto cross-linker polymethyl vinyl silicone (PMVS) is soaked onto the melamine sponge, and UV-assisted curing is carried out. The resulting material has self-cleaning properties and different adsorption capacities for different oils and solvents. The prepared sponges had a convenient pore and regular strip structure with superhydrophobic and lipophilic properties.

Feng & Yao (2018) reported that hydrophilic melamine sponges (MS) can be prepared into sponges with hydrophobic properties through modification by several methods: 1. Direct charring of MS can yield hydrophobic foam carbon, and the charred MS obtained has different adsorption properties at different temperatures. 2. Introduce hydrophobic functional group modification on the surface of MS, wrap the functional group monomer on the surface of MS, and then gel or polymerize to obtain the MS with a hydrophobic surface. 3. Treatment in HCl solution to obtain protonated mass spectra. The protonated mass spectra have excellent oil/water separation performance, usage stability, and anti-pollution performance. 4. MS or carbonic acid MS was used as a carrier, and the sponge surface was coated with graphene oxide (GO), metal-organic backbone (MOF), or other nanomaterials. Feng & Yoa (2018) coated the MS by furfuryl alcohol immersion and acid solution polymerization to modify the original MS. Furfuraldehyde reacted and polymerized on the surface of the MS, making the modified MS hydrophobic. Fig. 8 shows a schematic diagram of the preparation process (a) and SEM images at different magnifications (b-d) of the melamine filament-coated $NiCo_2S_4$ composite. Schematic diagram of the synthesis of alginate sponge and its application in copper (II) adsorption (e).

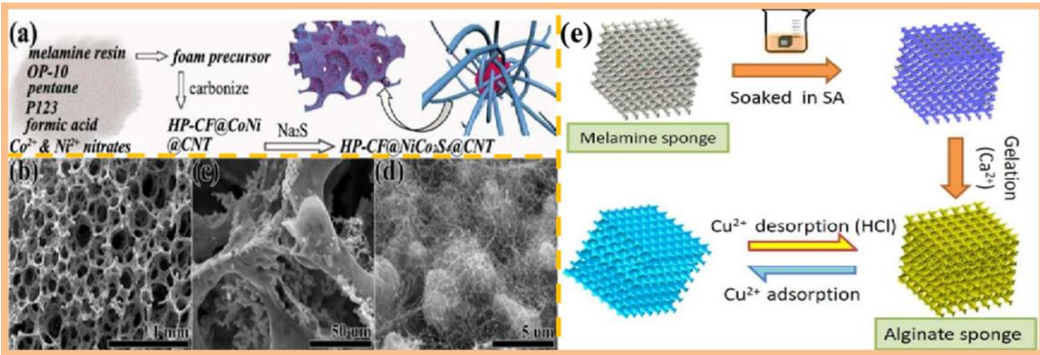


Fig. 8: Illustration of the fabrication process (a) and SEM images with different magnifications of melamine wires wrapped $NiCo_2S_4$ composites (b-d), (e) schematic illustration of the synthesis of alginate-MS and its application for Cu(II) adsorption.

## Foam Materials

Unlike sponge materials, foam materials are inherently hydrophobic. They can be used directly for oil/water separation (Pan et al. 2015), and the roughness of the surface structure after construction can further obtain superhydrophobicity (Yu et al. 2015). Metal foam materials are also common, with very good flexibility and high mechanical strength, which can be called good oil/water separation materials after proper modification. Zhou et al. (2018b) dipped copper foam (CFs) into an aqueous hydrochloric acid solution, roughened the surface by solution etching, and then put them into TD nanorods synthesized by heating in an aqueous solution. After coating the TD nanorods, the CFs were superhydrophilic and superlipophilic. Modified TD-CF has a WCA value of approximately 0°, exhibits superhydrophobicity at a WCA value of 160°, and superhydrophobicity at an OCA value of 159°. In addition, TD is an excellent photocatalyst, and the water absorbed by the TD-CF material will return to its original state after evaporation. The absorbed oil decomposes into non-toxic substances such as carbon dioxide and water under UV irradiation, indicating that the material is efficient and durable with good recovery rates. The metal skeleton provides a high-strength material to cope with the external stresses encountered and resists wear and tear during use. The nanorods form a stable composite three-phase interface for oil/water separation through laminar pores and millions of conical micropores, eliminating the need for complex chemical modification processes to achieve high/low surface energy (Fujishima et al. 2008, Wang & Zhang 2012)87-89. Fig. 9 shows the general preparation route and use of TD-CFs for efficient continuous oil/water separation processes.

## Smart Controllable Special Wettable Separation Materials

In recent years, stimuli-responsive polymers have received much attention and development due to their great potential for research and application in manufacturing smart controllable materials (Zhang et al. 2020, Zheng et al. 2020). Oil/water mixtures are usually highly acidic and alkaline, which tends to corrode the separation material, thus affecting the stability of the separation efficiency. It is also easy to cause oil contamination to clog the material voids, resulting in a reduction in separation efficiency or even the outright failure of the separator. Compared to other oil/water separation materials, intelligent and controllable separation materials reduce consumption and speed up the separation process(Ren et al. 2020) . However, only a few studies have reported on smart controllable separation materials.

Cao et al. (2014) prepared temperature and pH-responsive oil/water separation materials using stainless steel as a substrate for free radical polymerization triggered under UV light. The modified meshes have superhydrophilic and submerged superoleophobic properties with excellent separation efficiency. The method is novel and effective and opens up a new field of oil/water separation by selectively collecting oil or water from wastewater under temperature or pH regulation using dimethylaminoethyl methacrylate (DMAEMA), a raw material with excellent biocompatibility. A schematic diagram of the preparation of the hydrogel-coated mesh and the relative wettability of the mesh when in contact with oil is shown in Fig. 10 (a).

Cheng et al. (2015) reported the formation of smart controllable separation membranes by assembling responsive thiol molecules on the surface of copper mesh by an assembly technique. The modified material was extremely oleophobic under alkaline conditions and superhydrophobic under non-alkaline conditions and suggested that nanostructure and proper pore size on the substrate were keys to controllable separation.

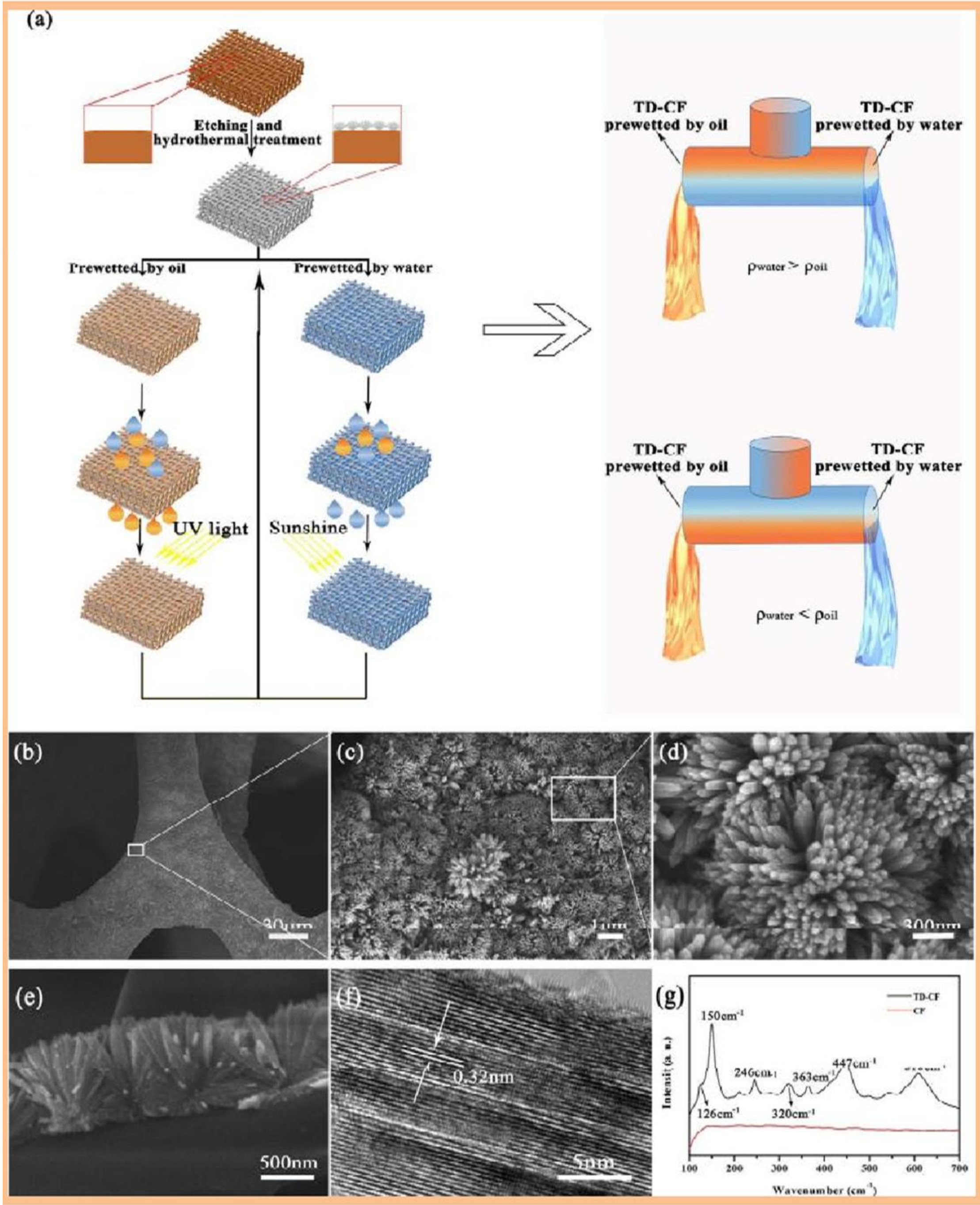


Fig. 9: (a) Illustration of the overall preparation route and usage of TD-CFs for high-efficiency and continuous oil-water separation process, (b-d) FESEM images on TD-CFs surface morphology with different magnification, (e) a cross-section FESEM image of TD coating, (f) HRTEM image of TD nanorods, (g) Raman spectra of TD-CF and commercial CF.

Xu et al. ( 2020) prepared the first visible light-responsive photocatalytic and pH-responsive super-wet melamine sponge (MS) with a porous elastic structure, providing a larger specific surface area and photocatalytic sites with a greater ability to adsorb contaminants, showing high oil absorption and photocatalytic effects. The excellent and stable separation efficiency is expected to be a multifunctional wastewater treatment material.

Fu et al. (2017) reported a superhydrophobic antibacterial fabric prepared by cross-linking reaction of nanoparticles with adipic diisocyanate using cotton fabric as the base material. The modified cotton fabric had underwater lipophilic and hydrophobic properties and was more effective in separating oil and water in acidic emulsions. The cotton fabric substrate is low in environmental protection, has a long service life, and has a certain self-cleaning ability after oil and water contamination.

Li et al. (2019) first prepared cellulose nanofibers (CNFs) aerogels by surface-initiated atomic technology, grafting poly(N, N-dimethylamine-diethyl methacrylate), the modified aerogel surface hydrophobic can be converted to hydrophilic by the concentration of carbon dioxide, when carbon dioxide is dissolved by other gases materials transformed into hydrophobic crude oil. The intelligent switchable separation properties effectively prevent the separation material from being contaminated with oil. The cellulose feedstock makes the aerogel very environmentally friendly and shows good separation efficiency even when dealing with surfactant-stabilized emulsion-modified materials, which has great potential for development. The $CO_2$-responsive cellulose nanofibre aerogel reaction mechanism is shown in Fig. 10 (b).

Gao et al. (2019a) prepared wettability switchable biomimetic $TiO_2$-titanium meshes (BTTMs) using a one-step dip coating method with polyvinylidene fluoride and modified $TiO_2$ suspensions. The modified metal mesh can be wettability switched intelligently by the action of surface hydroxyl groups and can self-healing and self-cleaning in addition to the effective separation of wastewater, which improves the material's recyclability and reduces energy loss. These properties increase the recyclability of the material and reduce energy losses. The preparation method is inexpensive and simple and has many applications. A schematic representation of the preparation is shown in Fig. 10 (c).

## CONCLUSIONS

This work has reviewed the preparation and application of oil/water separation materials in recent years, granular or powdered materials, metal meshes, natural textile or synthetic film materials, aerogels, sponges, foams, and intelligent, controllable humidification separation materials. Special attention is paid to cellulose-based superlipophilic/superhydrophobic, superhydrophilic/superoleophobic, and smart, responsive oil/water separation materials. Cellulose is a widespread and inexpensive biomass material that exhibits unique oil absorption properties due to its inherent porous structure and is applied in oil/water separation. To improve oil/water selectivity, oil/water separation materials are prepared by modified graft copolymerization, surface

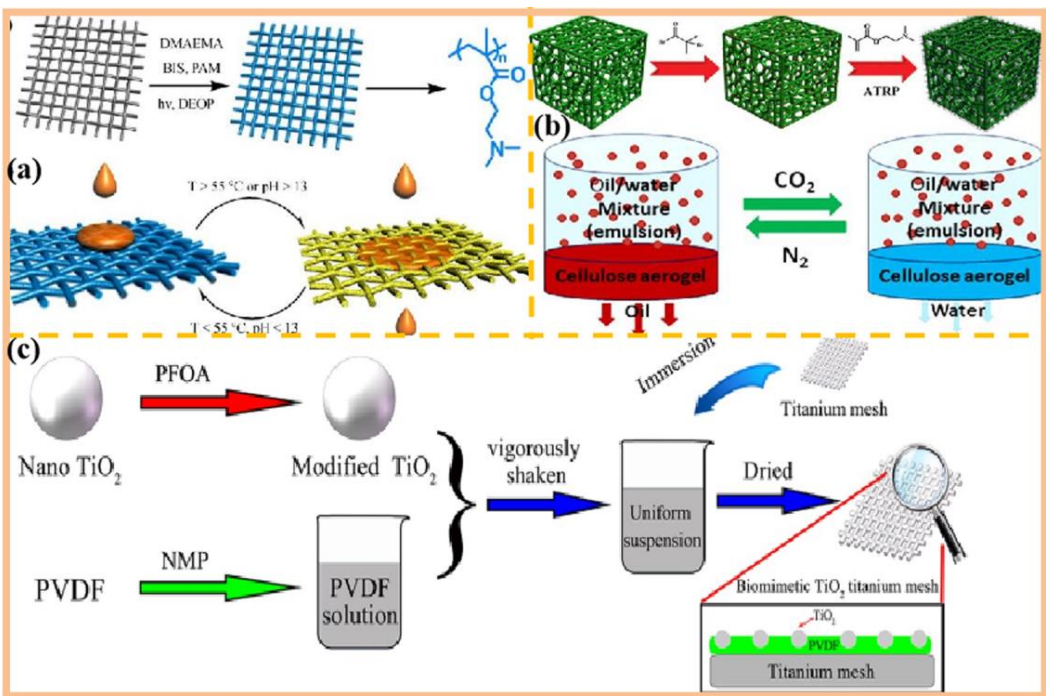


Fig. 10: (a) Schematic description of the preparation of PDMAEMA hydrogel coated mesh and the opposite wettability of the mesh when contacted with oil, (b) schematic illustration of the preparation of CNF-g-PDMAEMA Aerogel, (c) schematic illustration of the preparation process.

coating, particle adsorption, and construction of material surfaces or internal structures.

The durability and recyclability of the materials are still to be solved, and attention needs to be paid to the problem of separation materials being easily blocked by oil molecules. Without improving the anti-fouling properties of the materials, this will seriously affect the separation efficiency. There are also some key technical defects, such as the complexity of the synthesis process, the high cost, whether the environment is friendly, etc. Because of the complexity of the process, many successfully prepared materials are still limited to the laboratory research range; it is difficult to mass production applications. Therefore, low-cost, simple to prepare, self-cleaning, and durable oil and water separation materials should be vigorously developed.

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