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The discharge of waste organic solvents, various oil-water mixtures, and frequent oil pollution infiltration into water threaten ecological and environmental safety. Many researchers have developed separation materials, including filter materials, adsorption materials and smart materials with switch wettability. Among them, natural cotton fabrics have been widely studied as the matrix of separation materials because of their three-dimensional surface structure, porosity, recyclability low cost, and biodegradability. As an oil-water separation material, the matrix surface needs to have micro-nano structures. In this work, the technology of rough texture on the surface of cotton fabric for oil/water separation is reviewed. In this way, this work aligns with the United Nations' Sustainable Development Goals, especially Goal 6, Clean Water and Sanitation.

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Overview of rough surface construction technology for cotton fabrics used in oil/water separation

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Abstract

The waste organic solvents, the discharge of all kinds of oil/water mixture and the frequent infiltration of oil pollution into the water have brought unbearable threats to the ecological environment. The separation and recovery of oil/water mixture are investigated by more and more scholars. Many researchers have developed separation materials with excellent separation efficiency and high separation flux, including filter materials, adsorption materials and smart materials with switch wettability. Among them, the natural cotton fabric as a separation material substrate, because of its three-dimensional surface structure, porous, excellent fiber adsorption

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capacity and recycling, as well as the advantages of low-cost, biodegradable has been widely studied by scholars. As an oil/water separation material, it is necessary for the substrate surface to have micro-nano structure. In general, the researchers use various methods to modify the surface of the fabric with various kinds of micro-nano particles, which form a certain roughness on the surface of the fabric, for example by dip-coating, spray-coating, and grafting reactions, and then further modify it, to obtain separation materials for various purposes. In this work, the technology of rough texture on the surface of cotton fabric for oil/water separation is reviewed.

Keywords: Cotton fabric; oil/water separation; textile materials; micro-nano rough surface; application; fabric modification

1. Introduction

In recent decades, the oil industry has been experiencing rapid growth, significantly benefiting various aspects of people's lives. [1] However, the frequent leakage of oil-carrying vessels at sea has resulted in immeasurable economic losses and inflicted severe damage to the water environment and aquatic organisms. [2] Moreover, even the oily wastewater generated from domestic sewage systems has detrimental effects on the water environment and the entire planet. [3-6] Therefore, to safeguard the environment and promote public health, an increasing number of researchers have devoted their efforts towards developing effective treatments for oil/water mixtures. [7-9] To date, numerous oil/water separation materials with excellent properties

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have been developed, including sponges, [10] foams, [11] metal mesh, [12] synthetic membranes, [13] natural organic materials [14] and so on. [15, 16]

According to the superwetting theory of the lotus leaf, materials need to have a certain level of roughness and hydrophobicity on their surfaces. [17] In combination with theory, researchers have utilized various techniques, such as deposition, [18] dip drying, [19] spray painting, [20] layer-by-layer assembly, [21] in-situ growth, [22] external etching [23] and other strategies [24] to create the required rough structures on the surfaces of materials. By reducing the surface energy with hydrophobic modifiers, these materials can achieve excellent superhydrophobic and superlipophilic properties, which enable them to effectively separate oil from water. [25] This kind of material can achieve the effect of oil/water separation by removing oil. However, one issue with oil-absorbing materials is that the holes on their surfaces can be easily blocked by oil, which greatly reduces their separation effectiveness. [26] Hydrophilic underwater superoleophobic materials provide a good solution to this problem, as they can improve the material's anti-fouling ability and durability. [27]

However, due to the fact that the surface free energy of water droplets is higher than that of oil, it is difficult for a material to exhibit both hydrophilic and oleophobic properties simultaneously. [28] Currently, common methods used to achieve both properties include modifying the surface of the material through the addition of rough and uneven structures, which can increase the

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surface area and improve surface energy. These modifications can ultimately improve the hydrophilic and oleophobic properties of the material. [29] The hydrophilic and oleophobic properties of fluorocarbon surfactants have been utilized for chemical modification of materials. [30] Intelligent controllable oil/water separation materials are being studied extensively due to their unique structures that allow for switchable wettability under specific conditions. These materials are designed to respond to various stimuli such as pH, light, heat, electricity, and gas conversion, making them highly intelligent and efficient in separating oil and water. [31] The distinctive characteristic of switchable wettability renders this material superior to conventional separation techniques in terms of separation efficiency, durability, and pollution resistance. As such, it is highly anticipated in the continuous treatment of oil/water separation. [32, 33]

Compared to conventional separation materials, naturally grown biomass materials possess certain advantages. [34] First, the utilization of natural materials facilitates green environmental protection, as such materials exhibit excellent degradation properties and therefore don't pose a significant burden on the environment. Second, the low cost of biomass materials is highly conducive to their widespread adoption in separation applications. [35] The utilization of natural fiber woven fabrics offers a plethora of advantages. These fabrics possess outstanding hygroscopicity and capillary effect, which are inherent properties of the fibers. The

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three-dimensional and multidimensional structure of the fabric is also favorable for the adhesion of other modified materials. Additionally, the overall aperture of the fabric is highly controllable. [36] Moreover, with the increasing functionalization and diversification of cotton fabrics, there is a wider and wider range of environments in which fabrics can be applied in the field of oil/water separation. [37-39]

This article aims to introduce the fundamental theory of surface wettability and analyze three practical applications of oil/water separation fabric, namely hydrophobic oil-wet fabric, hydrophilic/underwater super oil-wet fabric, and switching wettability fabric. As shown in Fig. 1, the commonly used preparation methods, including dip drying, deposition, sol-gel, and spray methods, are analyzed and discussed. Finally, the oil/water separation fabric is summarized and future prospects are provided.

Fig. 1 is to be inserted here

2. Surface wettability theory

Water droplets in contact with solid surfaces form different ranges of contact angles (WCA), the size of the contact angle represents the material's wettability to water, 10° < WCA < 90° and 90° < WCA < 150° represent hydrophilic and hydrophobic materials, respectively. [40] People pay more attention to the superhydrophilicity and superhydrophobicity of water droplets on the surface 0° < WCA < 10° and 150° < WCA < 180° of the material.

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The most basic theoretical model of contact angle is the Young's equation, which describes the equilibrium relationship of surface tension between solid, liquid and gas phases (Fig. 2). It is proposed on the basis of rationalization and is applicable to solids without surface friction and the conditions of uniform distribution of tension between the three phases, Young's equation is as follows: [41]

$$\gamma_{lv} \cos\theta = \gamma_{sv} - \gamma_{sl} \quad (1)$$

Where, θ is the Young contact angle, γ_{lv}, γ_{sv}, γ_{sl} represent the interfacial tension between liquid and gas, solid and gas, liquid and solid, respectively.

Fig. 2 is to be inserted here

However, in reality, the roughness and tension of solid surface will lead to contact angle lag. [42] Wenzel equation and Cassie-Baxter equation are used to correct the Young contact angle. [43]

The Wenzel equation takes into account the effects of the rough structure of the solid surface and the uneven interfacial tension, and assumes that the liquid is completely filled in the microscopic raised structure of the solid surface, and there is no air between the liquid and the solid. Wenzel equation describes the contact angle θ_w: [44]

$$\mathrm{Cos}\theta_w = r \cos\theta \quad (2)$$

In Fig. 3a, θ is the contact angle of the smooth plane, θ_w is the contact angle of the corrected rough plane, and r is the roughness of the solid surface. The introduction of r can only increase or decrease, but can't change the

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affinity and hydrophobicity.

As shown in Fig. 3b, when there is air between the droplet and the contact uneven solid, and it is not completely paved and unfolded, it is assumed that the area ratio of liquid and gas on the solid surface is f and f1 (f + f1 = 1), θ and θ_1 are the solid-liquid contact angle and gas-liquid contact angle respectively, where $\theta_1 = 180°$. Then the contact angle θ_c described by the Cassie-Baxter equation: [45]

$$\mathrm{Cos}\theta_c = f\cos\theta + f1\cos\theta_1 = f\cos\theta + f - 1 \quad (3)$$

Fig. 3 is to be inserted here

It can be seen that the contact angle θ_c increases with the decrease of the liquid contact proportion.

The above contact angle model has been widely used in many fields. On the basis of these theories, [46] hydrophilic and hydrophobic materials can be prepared by changing the surface energy of materials and constructing micro-nano structures on the surface of materials. [47]

3. Superhydrophobic and oleophilic fabrics

In recent years, researchers have combined various micro and nano ions on the surface of soft fabrics to form rough three-dimensional structures. They then use effective chemical treatment agents to reduce surface energy and obtain modified fabrics with superhydrophobicity and superoleophilicity. [48] In addition, these modified fabrics also have excellent properties such as self-cleaning, anti-fouling, and antibacterial.

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The effectiveness of the material depends more on the firmness of the fabric surface coating and the harsh environment of the sewage water body, [49] so the durability is an important indicator for measuring the effectiveness of fabric preparation, and it is closely related to the method used to create roughness on the fabric surface. Common methods include dip drying, surface deposition, spraying, sol-gel, in-situ growth, and free radical polymerization. Table 1 lists the materials, methods, and efficiency of a typical hydrophobic oil-wet fabric.

Table 1 is to be inserted here

3.1. Dip coating

Dip coating is a commonly used technique for preparing modified materials due to its ease of operation and low difficulty. Typically, the bonding material is dissolved in a solution, and the fabric is fully immersed in the solution. Subsequently, the method of drying or curing to form a rough and firm coating on the surface of the fabric. [50]

Compared to other types of nanoparticles, silica nanoparticles have stable chemical properties, high optical transparency, lower toxicity, and environmentally friendly. As a result, they are widely used in the application field of improving self-cleaning performance. [51] Furthermore, its controllable size and large specific surface area enable it to be effectively integrated onto various fabric surfaces, resulting in a nanoscale structural effect that enhances the material's hydrophobic abilities.

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Lin et al. [52] successfully prepared superhydrophobic/superoleophobic modified composite fabric using cotton fabric as substrate through a simple two-step dipping strategy. Firstly, the modified SiO_2 is dipped on the fabric surface to obtain micro/nano level rough structure, and then the fluoropolymer is combined by dipping to improve the durability of the composite material. The modified fabric has a double dimensional hierarchy structure, which not only has excellent superhydrophobic/superlipophilic properties, but also maintains good washable durability and self-cleaning ability. Although the modified materials with excellent properties can be easily obtained by the two-step dipping method, and the fluorine-containing substances have exceptionally good durability and hydrophobic effect, the use of fluoropolymers still has no small harm to the environment.

Liu et al. [53] used a low-cost and environmentally friendly dipping method to coat polyester fabric with a combination of polydimethylsiloxane, stearic acid, and silicon dioxide. The resulting PDMS/STA/SiO_2 coated fabric exhibited improved hydrophobicity, reduced surface energy, and increased surface roughness. The modified polyester exhibits excellent hydrophobic properties, with a water contact angle (WCA) of 163°. In addition, after 700 friction experiments, the contact angle is remains above 150°, indicating its potential for practical applications as an environmentally friendly and durable means. Fig. 4

shows the SEM diagram, self-cleaning performance, and preparation process of superhydrophobic fabrics.

Nahar et al. [54] utilized eco-friendly materials, including boric acid, silica, and polyvinyl alcohol to modify cotton fabrics. By using a two-step dip coating and drying method and leveraging the unique cross-linking properties of three different materials, a dense, microporous, and rough structure is formed on the surface of cotton fabric. Additionally, the use of PDMS helps to reduce surface energy and results in the creation of a durable, environmentally friendly, and self-healing superhydrophobic cotton fabric. The preparation process is simple and rapid, providing a novel approach for the development of stable and durable superhydrophobic materials that can be applied to various substrates and easily produced on a large scale.

Fig. 4 is to be inserted here

Through the previous introduction, we learned that the process of combining nanoparticle materials with fabric surfaces often requires the use of adhesives. Unfortunately, many of the widely used adhesives are not environmentally friendly, such as vinyl chloride copolymer, polymethyl methacrylate (PMMA), phenolic resin, and so on. [55, 56] For the sustainable development of ecology, the level of environmental friendliness of materials has been given increasing attention. Cheng et al. [57] prepared the superhydrophobic and environmentally friendly composite fabric by using

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cheap and environmentally friendly materials, such as renewable fabric as the base, biodegradable diacid curable epoxidation soybean oil thermosetting material as the adhesive of ZnO nanoparticles attached to the fabric surface, and combined with a two-step dip coating method. The modified material is not only environmentally friendly, but also exhibits excellent superhydrophobic properties through immersion in the oil/water mixture for up to a week. Inspired by the superhydrophobic properties of lotus leaves, He et al. [58] successfully reduced the surface energy of stearicacid, a hydrophobic substance on the surface of lotus leaves, by grafting it onto the fabric surface. In addition, the pre-treated cotton fabric was immersed in a prepared non-toxic ZnO nanoparticle seed solution to obtain a micro-nano structure fabric surface. The prepared superhydrophobic fabric still maintains excellent separation efficiency even under harsh acid-alkaline conditions, and because of the combination of ZnO, the fabric has a self-cleaning effect and can resist ultraviolet radiation.

As is well known, TiO_2-based materials possess self-cleaning ability, high acid and alkali resistance, and the ability to degrade pollutants in wastewater under light conditions. Therefore, they are widely used in composite materials for oil/water separation and wastewater treatment. [59] In addition to the oil/water mixture, the wastewater also contains many organic dyes. Feng et al. [60] employed a fast and environmentally friendly

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dip coating approach to fabricate micro-nano level roughness of carboxylic acid-modified TiO_2 on the surface of nonwoven fabric. The resulting coated fabric exhibited excellent superhydrophobic properties and displayed efficient degradation of organic pollutants under light conditions. Pal et al. [61] utilized fluoride-free, environmentally friendly, and economical TiO_2 and 3-(trimethoxysilyl)propyl methacrylate materials to impregnate the surface of cotton fabrics and successfully produced a highly efficient, strong acid and alkali-resistant, self-cleaning, and superhydrophobic cotton fabric. Tudu et al. [62] developed a rapid method for creating superhydrophobic fabrics by applying a combination of TiO_2 nanoparticles and silane coupling agents onto the surface of cotton fabrics. The silane coupling agent used in this study was perfluorodecyl triethoxysilane (PFDTS), which not only reduced the fabric's surface energy, but also imparted a certain degree of roughness. The presence of TiO_2 on the surface of the coated fabric also brings high antibacterial properties. The preparation schematic diagram and antibacterial performance of modified superhydrophobic fabric are shown in Fig. 5.

Fig. 5 is to be inserted here

CuO nanoparticles are frequently utilized for air and water purification owing to their exceptional photocatalytic properties. With the rapid development of industry, many researchers have also combined CuO's photocatalytic properties with oil/water separation materials for

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application in sewage treatment. [63] Moreover, nanoparticles of CuO and Ag exhibit excellent antibacterial properties, are more cost-effective, and have a good market potential. [64] Cao et al. [65] prepared a superhydrophobic nanocoated fabric using a low-cost impregnation strategy, which involved immersing the fabric in a Cu ion solution, binding CuO onto the fabric surface, and reducing the surface energy using a silane coupling agent. This fabric not only exhibits excellent separation efficiency for a variety of oil/water and organic compound mixtures, but also has a good photocatalytic degradation effect on organic dyes present in sewage.

The bonding stability of crosslinkers is crucial for the adhesion of nanoparticles to fabric surfaces, but the environmental impact of most crosslinkers needs further consideration. [66] Agrawal et al. [67] adopted an environmentally friendly and simple dipping method to attach CuO nanoparticles to the fabric surface using a fluorosilan-free coupling agent as a crosslinking agent, effectively improving the durability of the hydrophobic coating. Additionally, the successful combination of metal oxides imparts superhydrophobic, antibacterial, and anti-fouling properties to the fabric.

Despite the relatively high cost of silver ions compared to other nanoparticles, the outstanding antibacterial effect of silver-containing composites is beyond doubt. Additionally, their good stability ensures that the separation performance of silver-treated separation materials is not

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compromised even in harsh solution environments. Silver ions have been widely employed in recent years to enhance the efficiency of oil/water separation by inhibiting the growth of bacteria and microorganisms in wastewater.[68] Zhu et al.[69] utilized a simple dipping process to densely and roughly coat the fabric surface with nano-silver, followed by fluorination to create a repairable and stable superhydrophobic fabric. Even when the hydrophobic properties are lost after repeated use, they can be easily restored through simple repairs, which not only extends the material's lifespan, but also significantly reduces costs. Although the modified fabric requires a certain amount of time and technology for subsequent maintenance and repair, it can still be widely used in many fields in the future. In light of the aforementioned limitations, Liu et al.[70] successfully developed a superhydrophobic cotton fabric by combining Ag/AgCl particles on the fabric surface through dip coating and electrostatic adsorption, followed by modification with polydimethylsiloxane for hydrophobicity. The resulting fabric exhibited excellent mechanical stability even after undergoing 50 cycles of friction. Additionally, the fabric displayed excellent self-cleaning properties under ultraviolet irradiation. It is worth noting that the incorporation of Ag/AgCl particles onto the fabric surface is known to enhance the anti-bacterial and anti-fungal properties of the fabric, making it useful for various applications in the healthcare and textile industries.

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In general, the dip coating method is a cost-effective and efficient surface modification technology with a low barrier to entry. However, achieving a uniform coating is crucial yet challenging, as the coating process requires careful adjustment of parameters such as material concentration, temperature, and pH to achieve the desired coating effect.

3.2. Deposition

The deposition method has been widely used for the preparation of superhydrophobic and superoleophilic fabrics, and both chemical vapor deposition and solution deposition methods are commonly employed. The difference from the dip coating method lies in the various means of deposition, and the compounds are deposited onto the fabric surface through external conditions. [71] The use of plasma technology enables the state of a substance to be altered, allowing for uniform, controllable, and effective deposition onto the surface of a material. [72] Electrophoretic deposition is a process that enables the rapid and uniform deposition of charged particles onto an electrode surface under the influence of an electric field. [73] The chemical vapor deposition (CVD) method is primarily used for the preparation of thin films. This process occurs under high-temperature conditions, where molecules of the raw material in the gas phase react chemically on the surface of the material to form a coating. [74]

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In recent years, researchers have found that functionalizing material surfaces with SiO_2 nanosol particles have effectively improved the material's friction resistance and stability during use. And, this treatment can be combined with the excellent characteristics of fabric, such as environmental protection and renewability. The SiO_2 nanosol particles firmly combine with the fabric, reducing the possibility of loose particles falling off and improving the fabric's resistance to washing. [75]

The wear resistance is an important consideration in the design of hydrophobic surfaces, as the surface roughness of modified fabrics without fluorine material is easily damaged by friction, leading to a reduction in hydrophobic properties. Lahiri et al. [76] used non-toxic and environmentally friendly materials, such as silicone polymer, to create micro-nano structures on the fabric surface using a deposition strategy involving boric acid, alkyl silane polymer, and silica composite material. They then achieved superhydrophobic cotton fabric with excellent durability through hydrophobic modification using PDMS. After being subjected to 40 rounds of sandpaper grinding and 80 rounds of tape bonding, the cotton fabric still retains its superhydrophobic properties. This cost-effective and eco-friendly approach holds great potential for industrial applications. The SEM diagram of the coated fabric, the process for preparing fluorine-free superhydrophobic cotton fabric, and the volumes of oil and water after multiple cycles are shown in Fig. 6.

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Shaheen et al. [77] applied a chemical in-situ deposition method to combine a SiO_2/TiO_2 nanoparticle sol mixture onto cotton fabric. They then treated the surface with octamethyltrisiloxane to create an ultra-hydrophobic cotton fabric with UV resistance and effective antibacterial properties. The experimental results showed that the modified material had a high bactericidal effect even against the most pathogenic Gram-positive bacteria at high nanosol concentrations.

Fig. 6 is to be inserted here

Medical textiles are a common infrastructure for health care, because of the low cost of fabric renewable, as well as superior comfort. The medical field requires medical textiles with excellent antibacterial properties to prevent the spread of infection. [78] By employing plasma deposition, Irfan et al. [79] successfully fabricated a green ultra-hydrophobic medical cotton cloth by incorporating a silver nanoparticle coating onto the surface of the cloth. The deposition efficiency was high, and the issue of agglomeration and uneven dispersion of silver nanoparticles was effectively addressed. The modified fabric exhibited long-lasting antibacterial properties.

As a bionic adhesive, polydopamine (PDA) exhibits strong adhesion ability and excellent durability when combined with various materials, making it an ideal candidate for fabric and nanoparticle binding. However, the efficiency of commonly used polydopamine deposition methods is

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often low. [80, 81] Zhang et al. [82] discovered that the deposition process of polydopamine on cotton fabric surfaces could be accelerated by using external ultraviolet irradiation and photosensitizer treatment. The fabric is treated with silver nanoparticles that have a rough structure and modified with alkyl to achieve hydrophobicity. This results in the preparation of a photothermal responsive superhydrophobic fabric. This strategy significantly reduces the reaction time and has strong practical application value.

Electrophoretic deposition is a commonly used deposition method with good results, but the limitation of the need for conductive substrate is also obvious. Kim et al. [83] solved this headache by utilizing a combination of nanoparticle self-assembly and electrophoretic deposition. Through this approach, a multi-layer mixed structure of ZnO and SiO_2 nanoparticles is formed on the surface of the fabric, which minimizes the adhesion and survival of bacteria on the fabric surface. Then, by hydrophobic modification of water repellent agent, superhydrophobic cotton fabric with effective antibacterial properties is successfully prepared.

TiO_2 and ZnO typically exhibit a rod-like morphology, whereas CuO tends to form a flower-like structure. Ming et al. [84] utilized a simple and cost-effective acoustic chemical deposition method to deposit copper oxide nanoparticles onto polyester fabric surfaces, resulting in the creation of environmentally friendly and durable superhydrophobic textiles. The entire

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process utilizes non-fluorinated, harmless coatings, maximizes the use of environmentally friendly solvents, and boasts high efficiency in separating oil and water, as well as long-lasting durability.

The deposition method is highly efficient and produces satisfactory results in terms of quality. However, it often requires complex auxiliary acceleration means that can be difficult to control, limiting its scope of application. As people's environmental protection requirements continue to evolve, there is a growing need for more environmentally friendly deposition methods to be developed and designed.

3.3. Spraying

Spraying is considered a promising method for surface modification due to its simplicity, efficiency, and ability to be used on a large scale. The process involves preparing a mixture of materials with both hydrophobic and lipophilic properties, which is then sprayed onto the surface of the fabric using a specialized device, resulting in a uniform and firmly bonded coating.

The coating of superhydrophobic surface is easy to fall off during use, and has weak mechanical stability, which has become a short board for practical application to a large extent. Kong et al. [85] utilized a simple and efficient two-step spraying method to apply an elastic rubber adhesive as the first layer coating onto the fabric surface. Subsequently, hydrophobic modified vapor SiO_2 nanoparticles were sprayed onto the adhesive coating

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to prepare a superhydrophobic modified fabric with strong resistance to wear and pollution. After various friction tests, the coating produced by the spraying method on the fabric surface has proven to be stable and durable. In the self-cleaning test, the coating has shown excellent anti-fouling performance, and it is believed that this fluoride-free, environmentally friendly coating has a good application market in the field of outdoor wear. The SEM images after wear and the process of preparing the coating on the polyester fabric surface are shown in Fig. 7.

It is difficult to bind TiO_2 nanoparticles to the fabric surface by impregnation. He et al. [86] used a more efficient method to modify the nanoparticles and fabric respectively by using silane coupling agent in a two-step process. The modified nanoparticles were firmly bonded to the fabric by spraying, and superhydrophobic TiO_2 composite cotton fabric was prepared. The material can be used to decompose pollutants by photocatalysis while separating sewage, which is of great significance for environmental restoration.

Fig. 7 is to be inserted here

The application of circuits has penetrated into all aspects of human life, but the electrical conductivity of aging circuits will decline in harsh environments. As we all know, silver has excellent electrical conductivity and good chemical stability, and is widely used in the field of conductive film. [87] Wang et al. [88] adopted a spraying strategy, spraying rough silver

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nanoparticles on the surface of the fabric, and then using polydimethylsiloxane bonding effect to make the nanoparticles bond more firmly. The water contact angle of the modified conductive composite fabric is as high as 163°, and it maintains efficient ice-breaking performance and electrical conductivity even in a humid environment. The excellent performance shown can be well developed and applied in electromagnetic shielding materials.

Although ZnO has excellent ultraviolet irradiation resistance and antibacterial properties, the durability of ZnO nanoparticle coatings is still a headache. Song et al. [89] solved the problem of poor durability by studying different proportions of ZnO and APESP siloxane sprayed on fabric surface. When the ratio of ZnO to APESP is 1: 2, the modified fabric has the lowest washing loss rate.

CuO nanoparticles have high specific surface area and satisfactory photocatalytic degradation of pollutants. In recent years, CuO nanoparticles have been widely used in the field of pollutant removal and photocatalysis. Long chain fatty acids are commonly used as modifiers for low surface energy modification of fabrics. [90] Ghashghaee et al. [91] first modified CuO nanoparticles with stearic acid, although this can already make the fabric have certain hydrophobic properties, and then formed a nanocomposite layer of polymethyl methacrylate and modified nanoparticles on the surface of the fabric through a simple spraying

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strategy, and prepared by mixing CuO/PMMA in different proportions. The resulting superhydrophobic modified fabric with optimal properties exhibits a water contact angle of up to 161°. At the same time, the photocatalytic degradation effect of the fabric is good, but after multi-layer coating, the degradation time of pollutants becomes longer. The state of water droplets on the surface of modified fabrics, the mechanism of photocatalytic degradation, and effect of CuO/PMMA ratio on the contact angle of the textile surface are shown in Fig. 8.

Fig. 8 is to be inserted here

Although the spraying method is the most widely used because of its high efficiency, there are still some practical shortcomings. For example, the dispersion uniformity of nanomaterials in mixed solutions should pay attention to the stability of particles. Durability is also worth paying attention to, after all, the modified fabric in the process of use can’t avoid the test of wind blowing and various harsh water environment.

3.4. Sol-gel method

Sol-gel method is often combined with other methods, the main purpose is to make raw materials such as nanoparticles form sol-gel with excellent uniformity after a series of reactions in a liquid environment, and then can be combined with impregnation, spraying, coating and other ways on the fabric surface. [92]

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Hao et al. [93] formed a thin film coating on the fabric surface by sol-gel method. The surface has nano-scale roughness, and re-treated the material with a new fluoroalkyl siloxane polymer as a hydrophobic agent, and successfully prepared fluorine-containing wear-resistant superhydrophobic fabric. Yang et al. [94] adopted a low-cost and environmentally friendly one-step sol-gel strategy. First, TiO_2 sol was catalyzed by acetic acid, and then micro-nano rough coating containing TiO_2 sol was combined on the fabric surface. The coated fabric showed excellent self-cleaning performance through testing in harsh environments, expanding the application range of superhydrophobic materials.

In recent years, the performance synthesis of nanomaterials has attracted the attention of many scholars. By combining the excellent properties of at least two materials, some defects of modified materials can be made up. For example, compared with a single nanoparticle, the composite nanoparticle combined fabric has higher self-cleaning performance. [95] It is difficult to adjust the content ratio of nanoparticles through ion and sputtering technology. Li et al. [96] adopted a process combining sol-gel and impregnation with high efficiency and controllable composition to coat $AgNO_3$ and SiO_2 nanoparticles on the surface of cotton/linen fabric through a coating machine, which not only has a high contact angle after repeated friction, but also has a high contact angle due

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to the presence of silver ions. The antibacterial properties of the material are also surprising.

In the process of hydrophobic fabric oil water treatment, the fabric will be polluted and produce bad smell due to the presence of bacteria and other microorganisms in the sewage. [97] Shaban et al. [98] used a sol-gel strategy to prepare ZnO nanoparticles, which are then loaded on the fabric surface by means of coating. The micro and nano structures formed on the surface of the composite not only improve the hydrophobic properties of the fabric, but also give the fabric excellent antibacterial effect and self-cleaning ability. The antibacterial process of photocatalytic antibacterial process is particularly effective in inhibiting gram-positive and Gram-negative bacteria. Formula for the reaction structure of ZnO on the surface of fabrics, its inhibitory effect on different types of bacteria, antibacterial mechanism, and hydrophobic effect are shown in Fig. 9.

Fig. 9 is to be inserted here

The annual consumption of medical textiles is very surprising, and antibacterial performance is the basis of medical textiles and the most critical link, copper metal has a low adverse reaction to human skin, and is conducive to wound healing. [99] Khani et al. [100] first combined CuO and TiO_2 nanoparticles on the fabric surface to build a rough structure through an easy-to-operate sol-gel strategy, and then used acids and alcohols to improve the bonding strength of the nanoparticles and reduce the adhesion

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of bacteria during the use of the material. The prepared medical antibacterial dressing has good hydrophobic properties and excellent durability.

The sol-gel strategy has prepared materials that can be used under harsh conditions and are suitable for larger scale production. However, because special materials and more process equipment are often used in the production process, the preparation cost is higher, and solvents that are not conducive to environmental protection will be used. In the future, the process of sol-gel means and the materials used need to be further designed and improved.

3.5. Other methods

In recent years, many scholars have made good progress in the field of antibacterial fabrics, although the antibacterial performance of medical textiles is excellent, but the durability is still a short board, when more bacteria are killed on the surface of the fabric will form accumulation, so the anti-fouling self-cleaning effect of the fabric needs to be improved. When the fabric has low surface energy and rough surface structure, it can effectively solve the accumulation and adhesion of bacteria. [101]

Cheng et al. [102] adopted a green economy preparation method, using the strong adhesion properties of the biopolymer polydopamine to bind silver nanoparticles in-situ on the surface of the fabric, and then grafted hydrophobic octachylamine on the surface through addition reaction and

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Schiff base reaction. The modified fabric obtained hydrophobic and antibacterial properties, and the inhibition effect against Escherichia coli and Staphylococcus aureus reached 99%, and the PDA/AgNPs/ODA coating showed excellent adhesion fastness after multiple wear tests and acid-base tests.

Fu et al. [103] adopts the strategy of free radical polymerization combined with sol-gel method. Firstly, a large number of silica nanoparticles are combined with tetraethyl orthosilicate and 3-mercaptopropyl triethoxysilane on the surface of the fabric through hydrolytic condensation reaction, and the micro-nano structures formed have preliminary hydrophobic effect. Subsequently, the surface energy was reduced by grafting 2, 2, 3, 4, 4, 4-hexafluoromethacrylate on the rough surface. And the formed chemical bond energy is large, ensuring the stability of the polymer, which makes the fabric even in harsh conditions and after multiple oil/water mixture cycles, the separation efficiency is still guaranteed to be more than 98%.

Abd El-Hady et al. [104] combines ZnO/SiO_2 nanocomposites on the fabric surface through the electrostatically layer-by-layer self-assembly technology, and the prepared composite materials have a multi-layer structure. Firstly, the cotton fabric is cationized to facilitate the construction of the film layer by layer. Then, ZnO/SiO_2 nanocomposites were deposited on the fabric surface by electrostatic adsorption, and the

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surface energy of the material was reduced by stearic acid. By UPF method, the modified fabric showed excellent UV resistance, and the tensile properties and air permeability of the treated material were also improved.

Due to the flexible and adjustable structural aperture, highly ordered, and excellent surface contact ability, metal-organic framework determines that this material has excellent adsorption capacity, and has been widely used in storage, filtration, separation and other fields in recent decades. Previous MOFs applications have mainly been studied in powder form, which has brought many disadvantages to the performance and recycling of materials. The surface of the fabric can provide sites to facilitate MOFs binding. [105] The fabric modified by MOFs can not only remove pollutants and harmful heavy metals while separating in water treatment, but also improve the reuse capacity of MOFs, facilitate the reuse of materials, and reduce the burden on the environment. [106] Long et al. [107] adopted an in situ growth strategy to generate compact ZIF-8 nanocrystals on the fabric surface. Firstly, PDA@Cotton and ZIF-8 nanomaterials were prepared. On the one hand, the durability and impact resistance of the materials were further improved by PDA. On the other hand, the nanoparticles could be well bonded by PDA as an intermediate layer. The materials were immersed in a mixed solution and ZIF-8 nanocrystals were in-situ bonded on the surface of PDA@Cotton at room temperature. The overall material separation efficiency is remarkable, up to 97%, and after repeated use and

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testing under harsh chemical conditions, it shows excellent durability and stability. SEM diagram, corresponding contact angles, and preparation process of superhydrophobic fabrics are shown in Fig. 10.

Fig. 10 is to be inserted here

4. Hydrophilic and oleophobic fabrics

Superhydrophobic and superoleophilic fabrics have certain anti-pollution and self-cleaning capabilities, but after repeated recycling, the oil stains and bacteria in the oil-water mixture can still block the surface of the fabric to some extent, greatly reducing the oil/water separation efficiency the material. Even after the subsequent surface treatment, the original performance is restored, but it also wastes a certain amount of human resources, and hydrophilic underwater superoleophobic fabric can solve this problem. [108]

Cotton fabrics have hydrophilic and oleophilic properties. In order to give cotton fabrics oil/water separation ability, they are modified to havehydrophilic and underwater superoleophobic properties. Inspired by the hydrophilic and oleophobic characteristics of the surface of fish and shrimp in water, it has been confirmed by many studies that the hydrophilic/underwater oleophobic surface should have hydrophilic substances and a certain multi-dimensional structure. [109] When hydrophilic/underwater superoleophobic fabric is pre-wetted, it forms a water film, which effectively blocks the adhesion of oil substances and

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reduces the risk of being clogged by oil. So far, researchers have also used nanoparticles and other substances to form micro and nano structures on the fabric surface by various means, and modified by hydrophilic chemical components to effectively improve the hydrophilic and oleophobic properties of the fabric. It can be used for the separation of mixtures of light oil and water, where the denser water stays at the bottom and the light oilulates at the top. The water can pass through the fabric due to its hydrophilic and oleophobic properties, thus achieving oil/water separation. Table 2 lists the materials, methods, and efficiency of a typical hydrophilic and oleophobic fabric.

Table 2 is to be inserted here

ZnO nanoparticles have excellent surface area and photocatalytic properties, and are often used in pollution treatment. Yang et al. [110] used zinc chloride aqueous solution as microsolubilizing agent and zinc source, ammonia gas as base source, and adopted in-situ growth strategy to uniformly bind zinc ions on the surface of fabric fibers. Under ultraviolet conditions, the ZnO semiconductor material produces holes that can improve the ability to adsorb water molecules, thus optimizing the water absorption of the fabric. The modified cotton fabric has superhydrophilic/underwater superoleophobic, and the separation efficiency is still as high as 99.2% after multiple cycles of separation. It has excellent degradation ability for dyes in oil/water mixture, and

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excellent oil resistance allows fabrics to maintain durability, and the material is expected to be effectively applied in the field of separation.

Nowadays, most separation materials have excellent processing capacity for dispersed oil/water mixtures, but because the emulsion is mixed by liquids of different particle sizes, these separation methods are difficult to have a good effect on the emulsion, but to use demulsifier to achieve the separation effect. [111, 112] Zhang et al. [113] prepared a solution with aqueous glutaraldehyde as the crosslinking agent and H_2SO_4 as the pH, and immersed the fabric in the solution. Then the PVA solution was poured onto the fabric, and the crosslinking reaction of PVA was controlled by to ensure the firm combination of PVA and the fabric while retaining the hydrophilicity of PVA. In addition, because of the capillary effect of the fabric on water and the hydrophilic effect of the coating, the oil droplets in the emulsion contact and fuse with each other to form large oil droplets, resulting in the demulsification effect. The separation efficiency of oil-in-water emulsion by surface fabric can reach more than 96%. In addition, the composite material does not lose its effect in the strong acid and alkali environment, and the hydrophilic and oleophobic ability ensures the anti-fouling and self-cleaning effect.

In view of the problem that some oleophobic materials need to be pre-treated before use, Li et al. [114] inspired by the special infiltration of natural insects, adopted a simple spraying strategy and constructed high

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and low surface energy coatings on the surface of ball milled microcrystalline cellulose by using perfluorooctanoic acid, successfully prepared fabrics with stable superhydrophilic and superoleophobic properties.

In previous literature, a highly oleophobic and superhydrophilic coating was prepared on the fabric using a short fluoroalkyl acrylate. [115, 116] Chi et al. [117] uses a UV-induced polymerization strategy to prepare coatings on the surface of polyester fabrics using environmentally friendly short-chain fluorinated acrylates, hydrophilic monomers and crosslinkers as raw materials. The results show that the coated fabric is superoleophobic and hydrophilic, the contact angle of most oils is as high as 150°, the fabric quickly absorbs water droplets within 70 milliseconds, effectively prevents the oil droplets from permeating, and improves the anti-fouling ability of the fabric during the separation process. And the fabric shows good antistatic property, so the coated fabric is expected to be used in the occasion of dealing with oil. Schematic diagram of the formation of hydrophilic and hydrophobic coatings, fiber SEM diagram, photos of water and oil drops on the coated polyester fabric, the repellency of fluorinated chains to oil and the hydration of water molecules into the hydrophilic subsurface are shown in Fig. 11.

The hydrogels with three-dimensional network structure have strong water absorption and water retention ability, which can be attributed to the

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fact that there are more hydrophilic groups on the surface. Unlike traditional materials, which are difficult to recycle and have poor anti-pollution ability, the application of hydrogels in water-absorbing materials has been widely concerned. [118, 119] Kordjazi et al. [120] successfully prepared oil/water separation and filtration materials covered by hydrogel by in-situ synthesis of chitosan/acrylamide hydrogels on fabric surface with the strategy of thermal polymerization. The whole experimental process is simple and green, and the results show that this material has excellent acid-alkali resistance and stable separation efficiency, and its excellent cost and environmental benefits are considered to be the product of industrialization.

Fig. 11 is to be inserted here

Fabrics have the advantages of natural environmental protection and low price. Although the oil/water separation materials based on fabrics have been widely studied and put into practical application in recent years, they still have obvious disadvantages in terms of external force resistance and wear resistance compared with other rigid materials such as stainless-steel mesh. The mechanical properties of nonwovens can be greatly improved by mixing polyester fibers with different melting points. [121] Sun et al. [122] blends polypropylene PP and polyester fiber LPET in different proportions to effectively strengthen the tensile strength of nonwovens, and then modifies the fabric surface hydrophilically with

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N-isopropylacrylamide (PNIPAM) by dipping coating strategy. The modified nonwovens showed hydrophilic/underwater superhydrophobic properties and improved mechanical properties.

5. Intelligent response oil/water separation fabric

There are quite a few types of oil/water mixture, such as suspended oil and emulsion, which are more complex and difficult to deal with. Besides, due to the uncontrollable external environment, separated materials with one-way processing capacity are faced with the limitation of low efficiency. Therefore, the research and development of switching wettability materials is also bound to be the major trend and result of separated materials. At present, the research of intelligent controllable oil/water separation fabric has made some progress. [123] Table 3 lists the materials, methods, and efficiency of a typical intelligent response fabrics.

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Yan et al. [124] adopted an easy-to-operate impregnation strategy, mixing Fe_3O_4 nanoparticles with TiO_2 composites modified by lauric acid to make a solution. In the soaking process, composite nanoparticles were combined on the fabric surface, and successfully prepared fabrics with changeable wettability under the influence of acid and alkali. The material exhibits excellent hydrophobic properties in acidic environment. When pH value exceeds 11 and becomes alkaline, the material's wettability will change from hydrophobic to hydrophilic. After repeated use of the material

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and the external conditions of ultraviolet light, the material still maintains a separation efficiency of up to 98%. In addition, the modified fabric is magnetic and easy to recycle after use.

The preparation process of many switchable wettability materials is too complicated, [125] and the excessive use of organic solvents in the preparation process also brings a certain burden to the environment, which is not conducive to the wide market application. [126] Liu et al. [127] modified TiO_2 particles and cellulose mixture in water with a variety of silane coupling agents, and successfully prepared coated cotton fabrics with switching wettability under heat treatment and ultraviolet irradiation. Under normal air conditions, the modified fabric has a super hydrophobic wetting effect, and after a period of ultraviolet irradiation, the adsorption property of the material to water gradually increases. And this wettability switch is reversible, by 120 ℃ treatment, the material's wettability can be restored to the original. Due to the photocatalytic effect of TiO_2 particles, the fabric can also effectively degrade pollutants in water during the separation process, which greatly improves the anti-fouling ability and durability of the fabric. This ability to treat oil/water mixtures on demand greatly improves the separation efficiency of separated materials. Flow chart for preparation of switchable wettability coatings, underwater OCA of different organic solutions, and mechanism diagram of wettability switching are shown in Fig. 12.

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Fig. 12 is to be inserted here

The intelligent separation membrane has a good separation efficiency for the oil/water mixture with stable surface activity and still doesn't want to miscible oil/water emulsion, but the intelligent response material still has the pain points of difficult migration and inadequate response. [128] Compared to the trigger conditions of other responsive materials, changing the wettability of materials with gases is safer, cheaper, and doesn't produce additional products that harm the environment. [129]

Wang et al. [130] adopted a self-assembly strategy, first using poly (diethylaminoethyl methacrylate)-methyl comethacrylate to prepare a CO_2 and N_2 responsive polymer, and then in situ self-assembly inside and on the surface of the fabric, successfully obtaining a switching wettable fabric with a gas response. The material is driven by capillary diffusion force and has excellent treatment efficiency especially for unevenly dispersed emulsion mixtures. After CO_2 treatment, the material's wettability changes from superhydrophobic to superhydrophilic, and then CO_2 is removed by N_2, and the material's wettability returns to the original superhydrophobic state.

6. Conclusions

In this work, we review three types of fabric-based oil/water separation applications that have emerged in recent years, including superhydrophobic and oleophilic fabrics, hydrophilic oleophobic fabric,

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and intelligent response oil/water separation fabric. The majority of scholars employ a technique that involves the integration of multiple nanoparticles onto the fabric surface to create micro and nanostructures with a rough texture. Subsequently, the hydrophobic properties are further enhanced through the application of hydrophobic materials.

While these separation materials exhibit excellent oil/water treatment effects under laboratory conditions, it is challenging to obtain more comprehensive preparation results. The sustainable and green development of oil/water separation fabrics, including simple fabrication, rapid fabrication, low-cost fabrication; good adhesion, and the separation of emulsified oil/water samples by novel methods such as rapid in situ complexation between fatty acid ligands/ metal ions/ surface - to form hierarchical rough and superhydrophobic fabric surfaces in a facile way is the future development direction. [131] Since there is a risk of damage to the substances forming the rough structures on the fabric surface during actual use, which may lead to reduced oil-water efficiency, how to obtain fabrics with good corrosion resistance and mechanical durability is also a future research direction.

Declaration of competing interest

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

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References

1. Zhai, Y.; Yuan, X.; Weber, C. C.; Varley, R. J.; Henderson, L. C. Review of plant cellulose-based aerogel materials for oil/water mixture separation. *Journal of Environmental Chemical Engineering* **2024,** *12* (5), 113716.

2. MohammadAlizadeh, A.; Elmi, F. Flame retardant and superoleophilic polydopamine/chitosan-graft (g)-octanal coated polyurethane foam for separation oil/water mixtures. *International Journal of Biological Macromolecules* **2024,** *259*, 129237.

3. Kanu, I.; Achi, O. Industrial effluents and their impact on water quality of receiving rivers in Nigeria. *Journal of Applied Technology in Environmental Sanitation* **2011,** *1*, 75-86.

4. Singh, R. L.; Singh, P. K. Global environmental problems. In *Principles and Applications of Environmental Biotechnology for a Sustainable Future*, Singh, R. L., Ed. Springer Singapore: Singapore, 2017; pp 13-41.

5. Zhao, C.; Huang, H.; Li, Z.; Li, J.; Li, Y.; Xiang, D.; Wu, Y.; Chen, J.; Chen, X.; Qin, M. 3D superhydrophobic/superoleophilic sponge with hierarchical porous structure and robust stability for high-efficiency and continuous separation of oily wastewater. *Separation and Purification Technology* **2022,** *299*, 121820.

6. Zhao, C.; Li, J.; Chen, Z.; Huang, H.; Cheng, J.; Xiang, D.; Li, Z.; Li, Y.; Wu, Y. Simple preparation superhydrophobic melamine sponge via one-step emulsion polymerization for continuous oil/water separation in harsh environment. *Colloids and Surfaces A: Physicochemical and Engineering Aspects* **2023,** *676*, 132267.

7. Tomkowiak, K.; Mazela, B.; Szubert, Z.; Perdoch, W. Hydrophobic cellulose-based sorbents for oil/water separation. **2024,** *29* (19), 4661.

8. Matabola, K. P.; Mokhena, T. C.; Bambo, M. F.; Mokhothu, T. H.; Modise, J. S.; Mochane, M. J. PVDF-based electrospun nanofibers for oil/water separation: A review. **2024,** *309* (8), 2300390.

9. Beagan, A.; Chen, C.; Mohamed, M. E. Bio-copper nanoparticle-based superhydrophobic membranes for sustainable oil/water separation. *Water Science and Technology* **2024,** *89* (3), 799-810.

10. Li, Q.; Yan, K.; Li, S.; Wang, M.; Liu, K.; Xia, M.; Cheng, Q.; Xu, J.; He, S.; Zhao, Y.; Li, M.; Wu, Y. Robust and multifunctional 3D superhydrophilic/superoleophobic sponge for rapid oil/water separation and water purification. *Progress in Organic Coatings* **2024,** *192*, 108427.

11. Zhang, X.; Li, Z.; Liu, K.; Jiang, L. Bioinspired multifunctional foam with self-cleaning and oil/water separation. *Advanced Functional Materials* **2013,** *23* (22), 2881-2886.

View Article Online
DOI: 10.1039/D4SU00674G

12. Liu, Y. Q.; Han, D. D.; Jiao, Z. Z.; Liu, Y.; Jiang, H. B.; Wu, X. H.; Ding, H.; Zhang, Y. L.; Sun, H. B. Laser-structured Janus wire mesh for efficient oil–water separation. *Nanoscale* **2017,** *9* (45), 17933-17938.

13. Baig, U.; Faizan, M.; Dastageer, M. A. Polyimide based super-wettable membranes/materials for high performance oil/water mixture and emulsion separation: A review. *Advances in Colloid and Interface Science* **2021,** *297*, 102525.

14. Bai, X.; Yuan, Z.; Lu, C.; Zhan, H.; Ge, W.; Li, W.; Liu, Y. Recent advances in superwetting materials for separation of oil/water mixtures. *Nanoscale* **2023,** *15* (11), 5139-5157.

15. Zhang, N.; Qi, Y.; Zhang, Y.; Luo, J.; Cui, P.; Jiang, W. A review on oil/water mixture separation material. *Industrial & Engineering Chemistry Research* **2020,** *59* (33), 14546-14568.

16. Zheng, L.; Su, X.; Lai, X.; Chen, W.; Li, H.; Zeng, X. Conductive superhydrophobic cotton fabrics via layer-by-layer assembly of carbon nanotubes for oil-water separation and human motion detection. *Materials Letters* **2019,** *253*, 230-233.

17. Yamamoto, M.; Nishikawa, N.; Mayama, H.; Nonomura, Y.; Yokojima, S.; Nakamura, S.; Uchida, K. Theoretical explanation of the lotus effect: Superhydrophobic property changes by removal of nanostructures from the surface of a lotus leaf. *Langmuir* **2015,** *31* (26), 7355-7363.

18. Gondal, M. A.; Sadullah, M. S.; Dastageer, M. A.; McKinley, G. H.; Panchanathan, D.; Varanasi, K. K. Study of factors governing oil–water separation process using TiO_2 films prepared by spray deposition of nanoparticle dispersions. *ACS Applied Materials & Interfaces* **2014,** *6* (16), 13422-13429.

19. Zhang, L.; Li, H.; Lai, X.; Su, X.; Liang, T.; Zeng, X. Thiolated graphene-based superhydrophobic sponges for oil-water separation. *Chemical Engineering Journal* **2017,** *316*, 736-743.

20. Chen, B.; Qiu, J.; Sakai, E.; Kanazawa, N.; Liang, R.; Feng, H. Robust and superhydrophobic surface modification by a "paint + adhesive" method: applications in self-cleaning after oil contamination and oil–water separation. *ACS Applied Materials & Interfaces* **2016,** *8* (27), 17659-17667.

21. Wang, J.; Wang, H. Multilayered chitosan/kaolin@calcium carbonate composite films with excellent chemical and thermal stabilities for oil/water filtration realized by a facile layer-by-layer assembly. *Separation and Purification Technology* **2022,** *289*, 120738.

22. Sun, F.; Li, T. T.; Zhang, X.; Shiu, B. C.; Zhang, Y.; Ren, H. T.; Peng, H. K.; Lin, J. H.; Lou, C. W. In situ growth polydopamine decorated polypropylen melt-blown membrane for highly efficient oil/water separation. *Chemosphere* **2020,** *254*, 126873.

23. Upadhyay, R. K.; Waghmare, P. R. Underwater oil drop storage, guided transport, and oil/water separation using surfaces with wettability contrast prepared through a vapor-based etching method. *ACS Applied Materials & Interfaces* **2020,** *12* (9), 11144-11154.

24. Wang, B.; Guo, Z. Superhydrophobic copper mesh films with rapid oil/water separation properties by electrochemical deposition inspired from butterfly wing. *Applied Physics Letters* **2013,** *103* (6), 063704.

25. Qiao, B.; Liang, Y.; Wang, T.; Jiang, Y. Surface modification to produce hydrophobic nano-silica particles using sodium dodecyl sulfate as a modifier. *Applied Surface Science* **2016,** *364*, 103-109.

26. Xu, M.; Wang, G.; Zeng, Z.; Chen, J.; Zhang, X.; Wang, L.; Song, W.; Xue, Q. Diverse wettability of superoleophilicity and superoleophobicity for oil spill cleanup and recycling. *Applied Surface Science* **2017,** *426*, 1158-1166.

27. Hu, J.; Zhan, Y.; Zhang, G.; Feng, Q.; Yang, W.; Chiao, Y. H.; Zhang, S.; Sun, A. Durable and super-hydrophilic/underwater super-oleophobic two-dimensional MXene composite lamellar membrane with photocatalytic self-cleaning property for efficient oil/water separation in harsh environments. *Journal of Membrane Science* **2021,** *637*, 119627.

28. Binks, B. P.; Clint, J. H. Solid wettability from surface energy components: relevance to pickering emulsions. *Langmuir* **2002,** *18* (4), 1270-1273.

29. Wan Ikhsan, S. N.; Yusof, N.; Aziz, F.; Ismail, A. F.; Jaafar, J.; Wan Salleh, W. N.; Misdan, N. Superwetting materials for hydrophilic-oleophobic membrane in oily wastewater treatment. *Journal of Environmental Management* **2021,** *290*, 112565.

30. Kovalchuk, N. M.; Trybala, A.; Starov, V.; Matar, O.; Ivanova, N. Fluoro- vs hydrocarbon surfactants: Why do they differ in wetting performance? *Advances in Colloid and Interface Science* **2014,** *210*, 65-71.

31. Xiang, B.; Sun, Q.; Zhong, Q.; Mu, P.; Li, J. Current research situation and future prospect of superwetting smart oil/water separation materials. *Journal of Materials Chemistry A* **2022,** *10* (38), 20190-20217.

32. Xue, Z.; Cao, Y.; Liu, N.; Feng, L.; Jiang, L. Special wettable materials for oil/water separation. *Journal of Materials Chemistry A* **2014,** *2* (8), 2445-2460.

33. Liu, H.; Zhang, L.; Huang, J.; Mao, J.; Chen, Z.; Mao, Q.; Ge, M.; Lai, Y. Smart surfaces with reversibly switchable wettability: Concepts, synthesis and applications. *Advances in Colloid and Interface Science* **2022,** *300*, 102584.

34. Yong, J.; Huo, J.; Chen, F.; Yang, Q.; Hou, X. Oil/water separation based on natural materials with super-wettability: recent advances. *Physical Chemistry Chemical Physics* **2018,** *20* (39), 25140-25163.

35. Lin, T. C.; Chang, J. S.; Lee, D. Cotton fabrics modified with tannic acid/1-eicosanamine grafting layer for oil/water separation. *Chemosphere* **2024,** *355*, 141703.

36. Ramaiah, G.; Simeno, Z.; Negawo, T. A.; Baraki, S. Y.; Legese, R.; Asfaw, D. Extraction of ensete fibers and its woven fabric green composite development for ceiling board applications. *Industrial Crops and Products* **2025,** *223*, 120189.

37. Abu Elella, M. H.; Abu-Thabit, N. Y.; Uwaezuoke, O. J.; Azad, A. K. Superwetting cotton textiles for separation of oil/water mixtures. *Cellulose* **2023,** *30* (12), 7427-7462.

View Article Online
DOI: 10.1039/D4SU00674G

38. Xiang, B.; Liu, Q.; Sun, Q.; Gong, J.; Mu, P.; Li, J. Recent advances in eco-friendly fabrics with special wettability for oil/water separation. *Chemical Communications* **2022,** *58* (97), 13413-13438.

39. Xia, S.; Yu, Z.; Pang, Y.; Chen, Z.; Chen, Y.; Zhang, X.; Guo, S. Advances in the application of superhydrophobic fabric surfaces for oil-water separation and extension of functionalization. *Journal of Environmental Chemical Engineering* **2024,** *12* (6), 114156.

40. Meiron, T. S.; Marmur, A.; Saguy, I. S. Contact angle measurement on rough surfaces. *Journal of Colloid and Interface Science* **2004,** *274* (2), 637-644.

41. Zhou, X.; Koh, J. J.; He, C. Robust oil-fouling resistance of amorphous cellulose surface underwater: A wetting study and application. *Langmuir* **2019,** *35* (4), 839-847.

42. Volpe, C. D.; Maniglio, D.; Morra, M.; Siboni, S. The determination of a 'stable-equilibrium' contact angle on heterogeneous and rough surfaces. *Colloids and Surfaces A: Physicochemical and Engineering Aspects* **2002,** *206* (1), 47-67.

43. Whyman, G.; Bormashenko, E.; Stein, T. The rigorous derivation of Young, Cassie–Baxter and Wenzel equations and the analysis of the contact angle hysteresis phenomenon. *Chemical Physics Letters* **2008,** *450* (4), 355-359.

44. Wenzel, R. N. Resistance of solid surfaces to wetting by water. *Industrial & Engineering Chemistry* **1936,** *28* (8), 988-994.

45. Cassie, A. B. D.; Baxter, S. Wettability of porous surfaces. *Transactions of the Faraday Society* **1944,** *40* (0), 546-551.

46. Decker, E. L.; Frank, B.; Suo, Y.; Garoff, S. Physics of contact angle measurement. *Colloids and Surfaces A: Physicochemical and Engineering Aspects* **1999,** *156* (1), 177-189.

47. McHale, G.; Newton, M. I.; Shirtcliffe, N. J. Water-repellent soil and its relationship to granularity, surface roughness and hydrophobicity: a materials science view. *European Journal of Soil Science* **2005,** *56* (4), 445-452.

48. Zhu, N. X.; Wei, Z. W.; Chen, C. X.; Wang, D.; Cao, C. C.; Qiu, Q. F.; Jiang, J. J.; Wang, H. P.; Su, C. Y., Self-generation of surface roughness by low-surface-energy alkyl chains for highly stable superhydrophobic/superoleophilic MOFs with multiple functionalities. *Angewandte Chemie International Edition* **2019,** *58* (47), 17033-17040.

49. Bano, S.; Zulfiqar, U.; Zaheer, U.; Awais, M.; Ahmad, I.; Subhani, T. Durable and recyclable superhydrophobic fabric and mesh for oil–water separation. *Advanced Engineering Materials* **2018,** *20* (1), 1700460.

50. Tang, X.; Yan, X. Dip-coating for fibrous materials: mechanism, methods and applications. *Journal of Sol-Gel Science and Technology* **2017,** *81* (2), 378-404.

51. Li, X.; Du, X.; He, J. Self-cleaning antireflective coatings assembled from peculiar mesoporous silica nanoparticles. *Langmuir* **2010,** *26* (16), 13528-13534.

52. Lin, J.; Zheng, C.; Ye, W.; Wang, H.; Feng, D.; Li, Q.; Huan, B., A facile dip-coating approach to prepare SiO_2/fluoropolymer coating for superhydrophobic and superoleophobic fabrics with self-cleaning property. *Journal of Applied Polymer Science* **2015,** *132* (1).

View Article Online
DOI: 10.1039/D4SU00674G

53. Liu, X.; Gu, Y.; Mi, T.; Wang, X.; Zhang, X Dip-coating approach to fabricate durable PDMS/STA/SiO_2 superhydrophobic polyester fabrics *Coatings*, 2021.

54. Jannatun, N.; Taraqqi-A-Kamal, A.; Rehman, R.; Kuker, J.; Lahiri, S. K. A facile cross-linking approach to fabricate durable and self-healing superhydrophobic coatings of SiO_2-PVA@PDMS on cotton textile. *European Polymer Journal* **2020,** *134*, 109836.

55. Magalhães, S.; Alves, L.; Medronho, B.; Fonseca, A. C.; Romano, A.; Coelho, J. F. J.; Norgren, M. Brief overview on bio-based adhesives and sealants. **2019,** *11* (10), 1685.

56. Imam, S. H.; Gordon, S. H.; Mao, L.; Chen, L. Environmentally friendly wood adhesive from a renewable plant polymer: characteristics and optimization. *Polymer Degradation and Stability* **2001,** *73* (3), 529-533.

57. Cheng, Q. Y.; An, X. P.; Li, Y. D.; Huang, C. L.; Zeng, J. B. Sustainable and biodegradable superhydrophobic coating from epoxidized soybean oil and ZnO nanoparticles on cellulosic substrates for efficient oil/water separation. *ACS Sustainable Chemistry & Engineering* **2017,** *5* (12), 11440-11450.

58. He, Y.; Wan, M.; Wang, Z.; Zhang, X.; Zhao, Y.; Sun, L., Fabrication and characterization of degradable and durable fluoride-free super-hydrophobic cotton fabrics for oil/water separation. *Surface and Coatings Technology* **2019,** *378*, 125079.

59. Nakata, K.; Fujishima, A. TiO_2 photocatalysis: Design and applications. *Journal of Photochemistry and Photobiology C: Photochemistry Reviews* **2012,** *13* (3), 169-189.

60. Feng, L.; Hou, Y.; Hao, Q.; Chen, M.; Wang, S.; Hu, X.; Yang, W. A multi-function textile with pH-induced switch wettability transition for controllable oil−water separation. *Textile Research Journal* **2021,** *92* (9-10), 1357-1368.

61. Pal, S.; Mondal, S.; Pal, P.; Das, A.; Pramanik, S.; Maity, J. Fabrication of durable, fluorine-free superhydrophobic cotton fabric for efficient self-cleaning and heavy/light oil-water separation. *Colloid and Interface Science Communications* **2021,** *44*, 100469.

62. Tudu, B. K.; Sinhamahapatra, A.; Kumar, A. Surface modification of cotton fabric using TiO_2 nanoparticles for self-cleaning, oil–water separation, antistain, anti-water absorption, and antibacterial properties. *ACS Omega* **2020,** *5* (14), 7850-7860.

63. Ighalo, J. O.; Sagboye, P. A.; Umenweke, G.; Ajala, O. J.; Omoarukhe, F. O.; Adeyanju, C. A.; Ogunniyi, S.; Adeniyi, A. G. CuO nanoparticles (CuO NPs) for water treatment: A review of recent advances. *Environmental Nanotechnology, Monitoring & Management* **2021,** *15*, 100443.

64. Ahamed, M.; Alhadlaq, H. A.; Khan, M. A. M.; Karuppiah, P.; Al-Dhabi, N. A. Synthesis, characterization, and antimicrobial activity of copper oxide nanoparticles. **2014,** *2014*, Article 17.

65. Cao, C.; Wang, F.; Lu, M. Superhydrophobic CuO coating fabricated on cotton fabric for oil/water separation and photocatalytic degradation. *Colloids and Surfaces A: Physicochemical and Engineering Aspects* **2020,** *601*, 125033.

View Article Online
DOI: 10.1039/D4SU00674G

66. Tambe, S. P.; Jagtap, S. D.; Choudhari, R. N.; Mallik, B. P. Influence of cross-linking agents and curing condition on the performance of epoxy coating. *Pigment & Resin Technology* **2016,** *45* (5), 354-362.

67. Agrawal, N.; Low, P. S.; Tan, J. S. J.; Fong, E. W. M.; Lai, Y.; Chen, Z. Durable easy-cleaning and antibacterial cotton fabrics using fluorine-free silane coupling agents and CuO nanoparticles. *Nano Materials Science* **2020,** *2* (3), 281-291.

68. De la Rosa-Gómez, I.; Olguín, M. T.; Alcántara, D. Antibacterial behavior of silver-modified clinoptilolite–heulandite rich tuff on coliform microorganisms from wastewater in a column system. *Journal of Environmental Management* **2008,** *88* (4), 853-863.

69. Zhu, X.; Zhang, Z.; Yang, J.; Xu, X.; Men, X.; Zhou, X. Facile fabrication of a superhydrophobic fabric with mechanical stability and easy-repairability. *Journal of Colloid and Interface Science* **2012,** *380* (1), 182-186.

70. Liu, H.; Yang, L.; Zhan, Y.; Lan, J.; Shang, J.; Zhou, M.; Lin, S. A robust and antibacterial superhydrophobic cotton fabric with sunlight-driven self-cleaning performance for oil/water separation. *Cellulose* **2021,** *28* (3), 1715-1729.

71. Du, C.; Wang, J.; Chen, Z.; Chen, D. Durable superhydrophobic and superoleophilic filter paper for oil–water separation prepared by a colloidal deposition method. *Applied Surface Science* **2014,** *313*, 304-310.

72. Martinu, L.; Poitras, D. Plasma deposition of optical films and coatings: A review. *Journal of Vacuum Science & Technology A* **2000,** *18* (6), 2619-2645.

73. Besra, L.; Liu, M. A review on fundamentals and applications of electrophoretic deposition (EPD). *Progress in Materials Science* **2007,** *52* (1), 1-61.

74. Carlsson, J. O.; Martin, P. M. Chapter 7 - Chemical Vapor Deposition. In *Handbook of Deposition Technologies for Films and Coatings (Third Edition)*, Martin, P. M., Ed. William Andrew Publishing: Boston, 2010; pp 314-363.

75. Zhang, N.; Xu, M.; Cai, L. Improvement of mechanical, humidity resistance and thermal properties of heat-treated rubber wood by impregnation of SiO_2 precursor. *Scientific Reports* **2019,** *9* (1), 982.

76. Lahiri, S. K.; Zhang, P.; Zhang, C.; Liu, L. Robust fluorine-free and self-healing superhydrophobic coatings by H_3BO_3 incorporation with SiO_2–Alkyl-Silane@PDMS on cotton fabric. *ACS Applied Materials & Interfaces* **2019,** *11* (10), 10262-10275.

77. Shaheen, T. I.; Salem, S. S.; Zaghloul, S. A new facile strategy for multifunctional textiles development through in situ deposition of SiO_2/TiO_2 nanosols hybrid. *Industrial & Engineering Chemistry Research* **2019,** *58* (44), 20203-20212.

78. Fei, Z.; Liu, B.; Zhu, M.; Wang, W.; Yu, D. Antibacterial finishing of cotton fabrics based on thiol-maleimide click chemistry. *Cellulose* **2018,** *25* (5), 3179-3188.

79. Irfan, M.; Polonskyi, O.; Hinz, A.; Mollea, C.; Bosco, F.; Strunskus, T.; Balagna, C.; Perero, S.; Faupel, F.; Ferraris, M. Antibacterial, highly hydrophobic and semi transparent Ag/plasma polymer nanocomposite coating on cotton fabric obtained by plasma based co-deposition. *Cellulose* **2019,** *26* (16), 8877-8894.

80. Lee, H.; Dellatore, S. M.; Miller, W. M.; Messersmith, P. B. Mussel-inspired surface chemistry for multifunctional coatings. **2007,** *318* (5849), 426-430.

81. Huang, Q.; Chen, J.; Liu, M.; Huang, H.; Zhang, X.; Wei, Y. Polydopamine-based functional materials and their applications in energy, environmental, and catalytic fields: State-of-the-art review. *Chemical Engineering Journal* **2020,** *387*, 124019.

82. Zhang, H.; Ou, J.; Fang, X.; Lei, S.; Wang, F.; Li, C.; Li, W.; Hu, Y.; Amirfazli, A.; Wang, P. Robust superhydrophobic fabric via UV-accelerated atmospheric deposition of polydopamine and silver nanoparticles for solar evaporation and water/oil separation. *Chemical Engineering Journal* **2022,** *429*, 132539.

83. Kim, J.; Lee, H.; Joung, Y. S. Antibacterial fabric with contradictory functions of water repellency and absorbency realized by electrophoretic deposition of hydrophobic SiO_2 and hydrophilic ZnO nanoparticles. *Progress in Organic Coatings* **2021,** *161*, 106455.

84. Ming, H. P.; Chan, C. Y.; Mutalik, S.; Younas, M. W.; Pragya, A.; Noor, N. Sonochemical routes to superhydrophobic soft matter coatings: Comparing silica and copper oxide coatings on polyester fabric. *Industrial & Engineering Chemistry Research* **2022,** *61* (51), 18729-18743.

85. Kong, X.; Zhu, C.; Lv, J.; Zhang, J.; Feng, J. Robust fluorine-free superhydrophobic coating on polyester fabrics by spraying commercial adhesive and hydrophobic fumed SiO_2 nanoparticles. *Progress in Organic Coatings* **2020,** *138*, 105342.

86. He, T.; Zhao, H.; Liu, Y.; Zhao, C.; Wang, L.; Wang, H.; Zhao, Y.; Wang, H. Facile fabrication of superhydrophobic Titanium dioxide-composited cotton fabrics to realize oil-water separation with efficiently photocatalytic degradation for water-soluble pollutants. *Colloids and Surfaces A: Physicochemical and Engineering Aspects* **2020,** *585*, 124080.

87. Sun, D.; Feng, Y.; Sun, S.; Yu, J.; Jia, S.; Dang, C.; Hao, X.; Yang, J.; Ren, W.; Sun, R.; Shao, C.; Peng, F. Transparent, self-adhesive, conductive organohydrogels with fast gelation from lignin-based self-catalytic system for extreme environment-resistant triboelectric nanogenerators. *Advanced Functional Materials* **2022,** *32* (28), 2201335.

88. Wang, Q.; Zhu, S.; He, H.; Du, J.; Li, W.; Kang, Z.; Chen, D. Conductive and superhydrophobic Ag/PDMS films with high stability for passive de-icing and electromagnetic shielding. *Progress in Organic Coatings* **2022,** *169*, 106919.

89. Song, T.; Liu, L.; Xu, F.; Pan, Y.; Qian, M.; Li, D.; Yang, R. Multi-dimensional characterizations of washing durable ZnO/phosphazene-siloxane coated fabrics via ToF-SIMS and XPS. *Polymer Testing* **2022,** *114*, 107684.

90. Fallahah, M.; Rabiee, A.; Ghashghaee, M.; Ershad-Langroudi, A. Enhanced procedure for fabrication of an ultrahydrophobic aluminum alloy surface using fatty acid modifiers. *Physical Chemistry Research* **2017,** *5* (2), 339-357.

91. Ghashghaee, M.; Fallah, M.; Rabiee, A. Superhydrophobic nanocomposite coatings of poly(methyl methacrylate) and stearic acid grafted CuO nanoparticles with photocatalytic activity. *Progress in Organic Coatings* **2019,** *136*, 105270.

92. Guo, X.; Zhang, Q.; Ding, X.; Shen, Q.; Wu, C.; Zhang, L.; Yang, H. Synthesis and application of several sol–gel-derived materials via sol–gel process combining

View Article Online
DOI: 10.1039/D4SU00674G

with other technologies: a review. *Journal of Sol-Gel Science and Technology* **2016,** *79* (2), 328-358.

93. Hao, L. F.; An, Q. F.; Xu, W.; Wang, Q. J. Synthesis of fluoro-containing superhydrophobic cotton fabric with washing resistant property using nano-SiO_2 sol-gel method. *Advanced Materials Research* **2010,** *121-122*, 23-26.

94. Yang, M.; Liu, W.; Jiang, C.; He, S.; Xie, Y.; Wang, Z. Fabrication of superhydrophobic cotton fabric with fluorinated TiO_2 sol by a green and one-step sol-gel process. *Carbohydrate Polymers* **2018,** *197*, 75-82.

95. Gordon, T.; Perlstein, B.; Houbara, O.; Felner, I.; Banin, E.; Margel, S. Synthesis and characterization of zinc/iron oxide composite nanoparticles and their antibacterial properties. *Colloids and Surfaces A: Physicochemical and Engineering Aspects* **2011,** *374* (1), 1-8.

96. Li, H.; Zhuang, Y.; Li, H.; Bracamontes, K. C.; Wang, D.; Sun, Y.; Sun, D.; Kong, L.; Gao, S.; Song, M. Preparation, characterization, antibacterial properties and hydrophobic evaluation of SiO_2/Ag nanosol coated cotton/linen fabric. *The Journal of The Textile Institute* **2020,** *111* (1), 75-83.

97. Liu, Y.; Dong, Q.; Shi, H. Distribution and population structure characteristics of microorganisms in urban sewage system. *Applied Microbiology and Biotechnology* **2015,** *99* (18), 7723-7734.

98. Shaban, M.; Mohamed, F.; Abdallah, S. Production and characterization of superhydrophobic and antibacterial coated fabrics utilizing ZnO nanocatalyst. *Scientific Reports* **2018,** *8* (1), 3925.

99. Davies, A. 16 - Healthcare textiles. In *Waterproof and Water Repellent Textiles and Clothing*, Williams, J., Ed. Woodhead Publishing: 2018; pp 447-471.

100. Khani, A.; Talebian, N. In vitro bactericidal effect of ultrasonically sol–gel-coated novel CuO/TiO_2/PEG/cotton nanocomposite for wound care. *Journal of Coatings Technology and Research* **2017,** *14* (3), 651-663.

101. Xiong, D.; Liu, G.; Duncan, E. J. S. Diblock-copolymer-coated water- and oil-repellent cotton fabrics. *Langmuir* **2012,** *28* (17), 6911-6918.

102. Cheng, W.; Liu, W.; Wang, Q.; Wang, P.; Zhou, M.; Yu, Y. Durable hydrophobic and antibacterial textile coating via PDA/AgNPs/ODA in situ assembly. *Cellulose* **2022,** *29* (2), 1175-1187.

103. Fu, X.; Wang, S.; Liu, Z.; Luo, Y.; Du, X.; Wang, H.; Zhou, M.; Cheng, X.; Du, Z. Robust superhydrophobic fabric for durability, self-cleaning, and oil/water separation via thiol–acrylate polymerization. *Macromolecular Materials and Engineering* **2021,** *306* (1), 2000504.

104. Abd El-Hady, M. M.; Sharaf, S.; Farouk, A. Highly hydrophobic and UV protective properties of cotton fabric using layer by layer self-assembly technique. *Cellulose* **2020,** *27* (2), 1099-1110.

105. Liu, C.; Wang, J.; Wan, J.; Yu, C. MOF-on-MOF hybrids: Synthesis and applications. *Coordination Chemistry Reviews* **2021,** *432*, 213743.

106. Yu, S.; Pang, H.; Huang, S.; Tang, H.; Wang, S.; Qiu, M.; Chen, Z.; Yang, H.; Song, G.; Fu, D.; Hu, B.; Wang, X. Recent advances in metal-organic framework

View Article Online
DOI: 10.1039/D4SU00674G

membranes for water treatment: A review. *Science of The Total Environment* **2021**, *800*, 149662.

107. Long, Z.; Yuan, L.; Chen, J.; Luo, L.; Shi, C.; Wu, C.; Qiao, H.; Wang, K. A durable fluorine-free MOF-based self-cleaning superhydrophobic cotton fabric for oil-water separation. *Advanced Materials Interfaces* **2022,** *9* (13), 2102427.

108. Ragesh, P.; Anand Ganesh, V.; Nair, S. V.; Nair, A. S. A review on ‘self-cleaning and multifunctional materials’. *Journal of Materials Chemistry A* **2014,** *2* (36), 14773-14797.

109. Yue, R.; An, C.; Ye, Z.; Owens, E.; Taylor, E.; Zhao, S. Green biomass-derived materials for oil spill response: recent advancements and future perspectives. *Current Opinion in Chemical Engineering* **2022,** *36*, 100767.

110. Yang, P.; Yang, J.; Wu, Z.; Zhang, X.; Liu, Y.; Lu, M. Facile fabrication of superhydrophilic and underwater superoleophobic surfaces on cotton fabrics for effective oil/water separation with excellent anti-contamination ability. *Colloids and Surfaces A: Physicochemical and Engineering Aspects* **2021,** *628*, 127290.

111. Zhang, W.; Liu, N.; Cao, Y.; Lin, X.; Liu, Y.; Feng, L. Superwetting porous materials for wastewater treatment: from immiscible oil/water mixture to emulsion separation. *Advanced Materials Interfaces* **2017,** *4* (10), 1600029.

112. Kang, W.; Jing, G.; Zhang, H.; Li, M.; Wu, Z. Influence of demulsifier on interfacial film between oil and water. *Colloids and Surfaces A: Physicochemical and Engineering Aspects* **2006,** *272* (1), 27-31.

113. Zhang, Y. R.; Meng, B. W.; Hao, B.; Ma, P. C. Aggregation-induced demulsification triggered by the hydrophilic fabric for the separation of highly emulsified oil droplets from water. *Aggregate* **2022,** *3* (1), e131.

114. Li, X.; Peng, Y.; Zhang, F.; Yang, Z.; Dong, Z. Fast-response, no-pretreatment, and robustness air-water/oil amphibious superhydrophilic-superoleophobic surface for oil/water separation and oil-repellent fabrics. *Chemical Engineering Journal* **2022,** *427*, 132043.

115. Takayanagi, T.; Yamabe, M. Progress of fluoropolymers on coating applications: Development of mineral spirit soluble polymer and aqueous dispersion. *Progress in Organic Coatings* **2000,** *40* (1), 185-190.

116. Korzeniowski, S. H.; Buck, R. C.; Newkold, R. M.; kassmi, A. E.; Laganis, E.; Matsuoka, Y.; Dinelli, B.; Beauchet, S.; Adamsky, F.; Weilandt, K.; Soni, V. K.; Kapoor, D.; Gunasekar, P.; Malvasi, M.; Brinati, G.; Musio, S. A critical review of the application of polymer of low concern regulatory criteria to fluoropolymers II: Fluoroplastics and fluoroelastomers. *Integrated Environmental Assessment and Management* **2023,** *19* (2), 326-354.

117. Chi, H.; Xu, Z.; Ma, Y.; Tang, T.; Zhang, T.; Zhao, Y. Multifunctional highly oleophobic and superhydrophilic fabric coatings prepared by facile photopolymerization. *Adv. Sustainable Syst.* **2020,** *4* (7), 2000049.

118. Zhang, W.; Shi, Z.; Zhang, F.; Liu, X.; Jin, J.; Jiang, L. Superhydrophobic and superoleophilic PVDF membranes for effective separation of water-in-oil emulsions with high flux. *Advanced Materials* **2013,** *25* (14), 2071-2076.

119. Loo, S. L.; Vásquez, L.; Athanassiou, A.; Fragouli, D. Polymeric hydrogels—a promising platform in enhancing water security for a sustainable future. *Advanced Materials Interfaces* **2021,** *8* (24), 2100580.

120. Kordjazi, S. Kamyab, K.; Hemmatinejad, N. Super-hydrophilic/oleophobic chitosan/acrylamide hydrogel: an efficient water/oil separation filter. *Advanced Composites and Hybrid Materials* **2020,** *3* (2), 167-176.

121. Kubo, S.; Kadla, J. F. Lignin-based carbon fibers: Effect of synthetic polymer blending on fiber properties. *Journal of Polymers & the Environment* **2005,** *13* (2), 97-105.

122. Sun, F.; Li, T. T.; Zhang, X.; Shiu, B. C.; Zhang, Y.; Ren, H. T.; Peng, H. K.; Lin, J. H.; Lou, C. W. Facile fabrication of hydrophilic-underwater superoleophobic poly(N-isopropylacrylamide) coated PP/LPET nonwoven fabrics for highly efficient oil/water separation. *Prog. Org. Coat.* **2020,** *148*, 105780.

123. Li, J. J.; Zhou, Y. N.; Luo, Z. H. Polymeric materials with switchable superwettability for controllable oil/water separation: A comprehensive review. *Progress in Polymer Science* **2018,** *87*, 1-33.

124. Yan, T.; Chen, X.; Zhang, T.; Yu, J.; Jiang, X.; Hu, W.; Jiao, F. A magnetic pH-induced textile fabric with switchable wettability for intelligent oil/water separation. *Chemical Engineering Journal* **2018,** *347*, 52-63.

125. Ye, H.; Zhu, L.; Li, W.; Liu, H.; Chen, H. Simple spray deposition of a water-based superhydrophobic coating with high stability for flexible applications. *Journal of Materials Chemistry A* **2017,** *5* (20), 9882-9890.

126. Zhou, H.; Wang, H.; Niu, H.; Zhao, Y.; Xu, Z.; Lin, T. A waterborne coating system for preparing robust, self-healing, superamphiphobic surfaces. *Advanced Functional Materials* **2017,** *27* (14), 1604261.

127. Liu, X.; Wei, Y.; Tao, F.; Zhang, X.; Gai, L.; Liu, L. All-water-based superhydrophobic coating with reversible wettability for oil-water separation and wastewater purification. *Progress in Organic Coatings* **2022,** *165*, 106726.

128. Rana, D.; Matsuura, T. Surface modifications for antifouling membranes. *Chemical Reviews* **2010,** *110* (4), 2448-2471.

129. Dong, L.; Zhao, Y. CO_2-switchable membranes: structures, functions, and separation applications in aqueous medium. *Journal of Materials Chemistry A* **2020,** *8* (33), 16738-16746.

130. Wang, Y.; Yang, S.; Zhang, J.; Chen, Z.; Zhu, B.; Li, J.; Liang, S.; Bai, Y.; Xu, J.; Rao, D.; Dong, L.; Zhang, C.; Yang, X. Scalable and switchable CO_2-responsive membranes with high wettability for separation of various oil/water systems. *Nature Communications* **2023,** *14* (1), 1108.

131. Abu-Thabit, N. Y.; Azad, A. K.; Mezghani, K.; Hakeem, A. S.; Drmosh, Q. A.; Akhtar, S.; Adesina, A. Y. Facile and green fabrication of superhydrophobic polyacrylonitrile nonwoven fabric with iron hydroxide nanoparticles for efficient oil/water separation. *ACS Applied Polymer Materials* **2022,** *4* (11), 8450-8460.

132. Lin, D.; Zeng, X.; Li, H.; Lai, X.; Wu, T. One-pot fabrication of superhydrophobic and flame-retardant coatings on cotton fabrics via sol-gel reaction. *Journal of Colloid and Interface Science* **2019,** *533*, 198-206.

View Article Online
DOI: 10.1039/D4SU00674G

133. Jiang, B.; Chen, Z.; Sun, Y.; Yang, H.; Zhang, H.; Dou, H.; Zhang, L. Fabrication of superhydrophobic cotton fabrics using crosslinking polymerization method. *Applied Surface Science* **2018,** *441*, 554-563.

134. Cheng, D.; Zhang, Y.; Bai, X.; Liu, Y.; Deng, Z.; Wu, J.; Bi, S.; Ran, J.; Cai, G.; Wang, X. Mussel-inspired fabrication of superhydrophobic cotton fabric for oil/water separation and visible light photocatalytic. *Cellulose* **2020,** *27* (9), 5421-5433.

135. Liang, L.; Dong, Y.; Xu, W.; Meng, X. Fabrication of hydrophobic/oleophilic cotton fabric based on thiol–ene click reaction for oil/water separation. *The Journal of The Textile Institute* **2022,** *113* (9), 1838-1844.

136. Liang, L.; Su, M.; Zheng, C.; Li, J.; Zhan, H.; Li, X.; Meng, X. Fabrication of hydrophobic/oleophilic cotton fabric by mussel-inspired chemistry for oil/water separation. *Fibers and Polymers* **2017,** *18* (12), 2307-2314.

137. Meng, X.; Dong, Y.; Arias, M. J. L.; Mu, S.; Liang, L. RAFT reaction modified cotton fabric and its application for oil/water separation. *Fibers and Polymers* **2022,** *23* (2), 396-403.

138. Meng, X.; Song, C.; Xing, Z.; Lis Arias, M. J.; Yan, J.; Ren, Q.; Xiang, Y.; Liang, L. ZIF-8/GO/PDMS modified cotton fabric to form a hierarchical-structure coating for fast oil/water separation. *Journal of Water Process Engineering* **2024,** *60*, 105158.

139. Ghorbani, L.; Caschera, D.; Shokri, B. Effect of oxygen plasma pre-treatment on the surface properties of Si-modified cotton membranes for oil/water separations. **2022,** *15* (23), 8551.

140. Mohamed, M. E.; Abd-El-Nabey, B. A. Fabrication of durable superhydrophobic/oleophilic cotton fabric for highly efficient oil/water separation. *Water Science and Technology* **2020,** *83* (1), 90-99.

141. Nabipour, H.; Wang, X.; Song, L.; Hu, Y. Hydrophobic and flame-retardant finishing of cotton fabrics for water–oil separation. *Cellulose* **2020,** *27* (7), 4145-4159.

142. Lin, T. C.; Chang, J. S.; Lee, D. J. Hydrophobic cotton fabric with 3-mercaptopropyltriethoxysilane/polyhedral oligomeric silsesquioxane/1-octadecanethiol modification for oil/water separation. *International Journal of Biological Macromolecules* **2023,** *253*, 126748.

143. Li, N.; Chen, J.; Li, J.; Wu, H.; Li, Z.; He, X.; Cai, L. Facile construction of versatile cotton fabrics with robust hydrophobicity, self-cleaning and oil–water separation. *Fibers and Polymers* **2024,** *25* (2), 565-575.

144. Liu, X.; Chen, X.; Bian, H.; Ni, S.; Li, Z.; Liu, N.; Qin, M.; Zhang, F. Highly hydrophobic cotton fabric by in-situ co-deposition of lignin/metal particles for oil/water separation. *Industrial Crops and Products* **2023,** *204*, 117393.

145. Zhang, Y. R.; Meng, B. W.; Hao, B.; Ma, P. C. Aggregation ‐ induced demulsification triggered by the hydrophilic fabric for the separation of highly emulsified oil droplets from water. *Aggregate* **2021,** *3*.

146. Yu, B.; Hou, K.; Fan, Z.; Jin, K.; Cai, Z. Design fiber-based membrane with interfacial wettability rapidly regulated behavior by pH for oily wastewater high-efficient treatment. *Progress in Organic Coatings* **2024,** *189*, 108326.

147. Li, J.; Yang, L.; Liu, H.; Li, G.; Li, R.; Cao, Y.; Zeng, H. Simple preparation method for hydrophilic/oleophobic coatings. *ACS Applied Materials & Interfaces* **2020,** *12* (40), 45266-45273.

148. Wang, Y.; You, C.; Kowall, C.; Li, L. A nanometer-thick, mechanically robust, and easy-to-fabricate simultaneously oleophobic/hydrophilic polymer coating for oil–water separation. *Industrial & Engineering Chemistry Research* **2018,** *57* (45), 15395-15399.

149. Xiao, M.; Huang, Y.; Xu, A.; Zhang, T.; Zhan, C.; Hong, L. On-demand oil–water separation by environmentally responsive cotton fabrics. *ACS Omega* **2019,** *4* (7), 12333-12341.

150. Fu, J.; Yang, F.; Guo, Z. Fabrication of switchable surface wettability with UV-triggered on cotton fabric. *Materials Letters* **2021,** *283*, 128767.

151. Chen, J.; Shen, C.; Yang, S.; Rana, M.; Ma, P. C. Acid and temperature dual-responsive cotton fabrics with polymer coating. *Composites Communications* **2017,** *4*, 10-15.

152. Wang, Y.; Xiao, Y.; Fu, X.; Jiang, L.; Yuan, A.; Xu, H.; Wei, Z.; Lei, Y.; Lei, J. Facile preparation of cotton fabric with superhydrophilicity–oleophobicity in air and superoleophobicity under water by using branched polyethyleneimine/perfluorooctanoic acid composites. *New Journal of Chemistry* **2021,** *45* (34), 15321-15327.

153. Zhang, G.; Liu, Y.; Chen, C.; Long, L.; He, J.; Tian, D.; Luo, L.; Yang, G.; Zhang, X.; Zhang, Y. MOF-based cotton fabrics with switchable superwettability for oil–water separation. *Chemical Engineering Science* **2022,** *256*, 117695.

154. Qiuyu, Y.; Xinyue, L.; Qing, R.; Jiang, T.; Peng, W.; Ming, L.; Hang, X. Anionic ammonium polyphosphate coated underwater superoleophobic cotton fabric for effective oil/water separation. *Industrial Crops and Products* **2023,** *202*, 117080.

155. Zhang, Y. R.; Chen, J. T.; Hao, B.; Wang, R.; Ma, P. C. Preparation of cellulose-coated cotton fabric and its application for the separation of emulsified oil in water. *Carbohydrate Polymers* **2020,** *240*, 116318.

156. Wang, M.; Peng, M.; Zhu, J.; Li, Y. D.; Zeng, J. B. Mussel-inspired chitosan modified superhydrophilic and underwater superoleophobic cotton fabric for efficient oil/water separation. *Carbohydrate Polymers* **2020,** *244*, 116449.

157. Qu, M.; Pang, Y.; Li, J.; Wang, R.; He, D.; Luo, Z.; Shi, F.; Peng, L.; He, J. Eco-friendly superwettable functionalized-fabric with pH-bidirectional responsiveness for controllable oil-water and multi-organic components separation. *Colloids and Surfaces A: Physicochemical and Engineering Aspects* **2021,** *624*, 126817.

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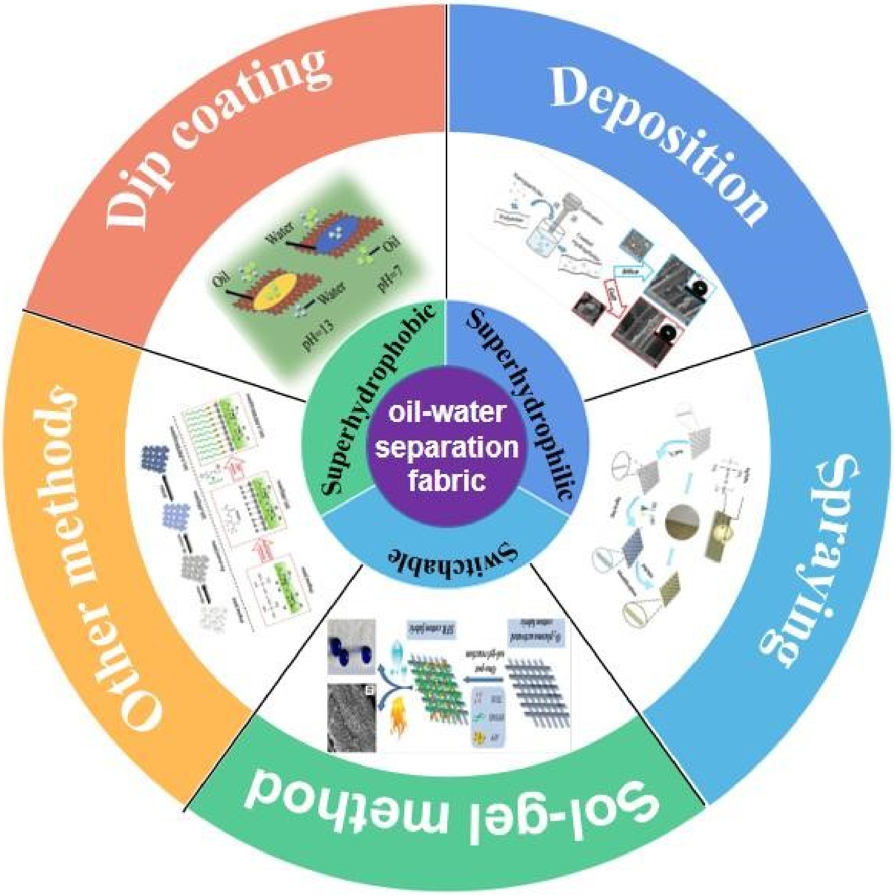

Fig. 1 Schematic illustration of the fabrication method of oil/water separation cotton fabrics, such as dip coating [60], deposition [84], spraying [88], sol-gel method [132], other methods [103].

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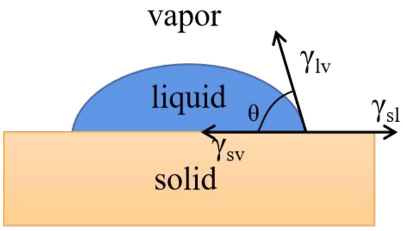

Fig. 2 Contact angle measured on a solid surface.

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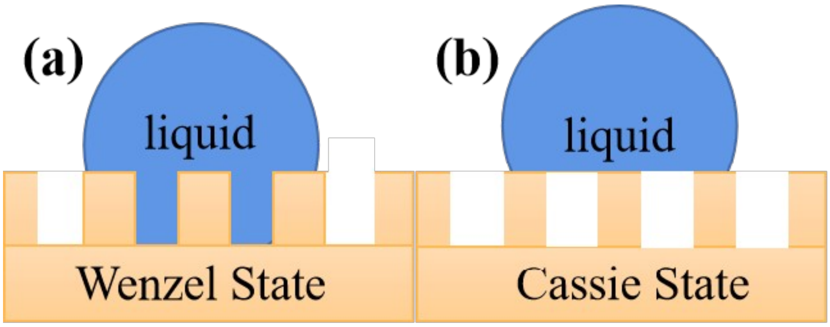

Fig. 3 (a) Wenzel model, (b) Cassie-Boxter model.

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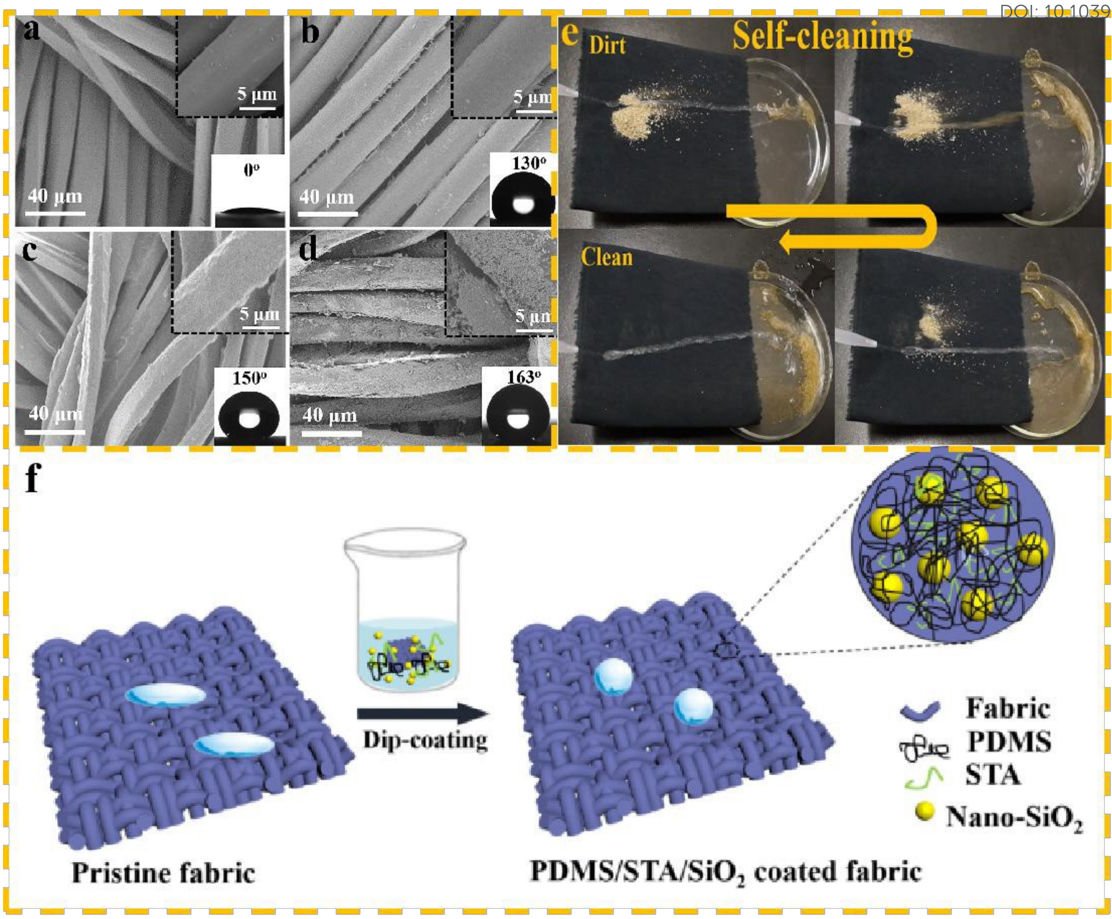

Fig. 4 Scanning electron microscopy (SEM) images of the fabrics: (a) pristine fabric, (b) PDMS coated fabric, (c) PDMS/STA-coated fabric, (d) PDMS/STA/SiO_2-coated fabric. Insets are differently magnified SEM images and WCAs of corresponding fabrics, (e) the self-cleaning property of the PDMS/STA/SiO_2-coated fabric, and (f) fabrication scheme of the superhydrophobic coating on the fabric.

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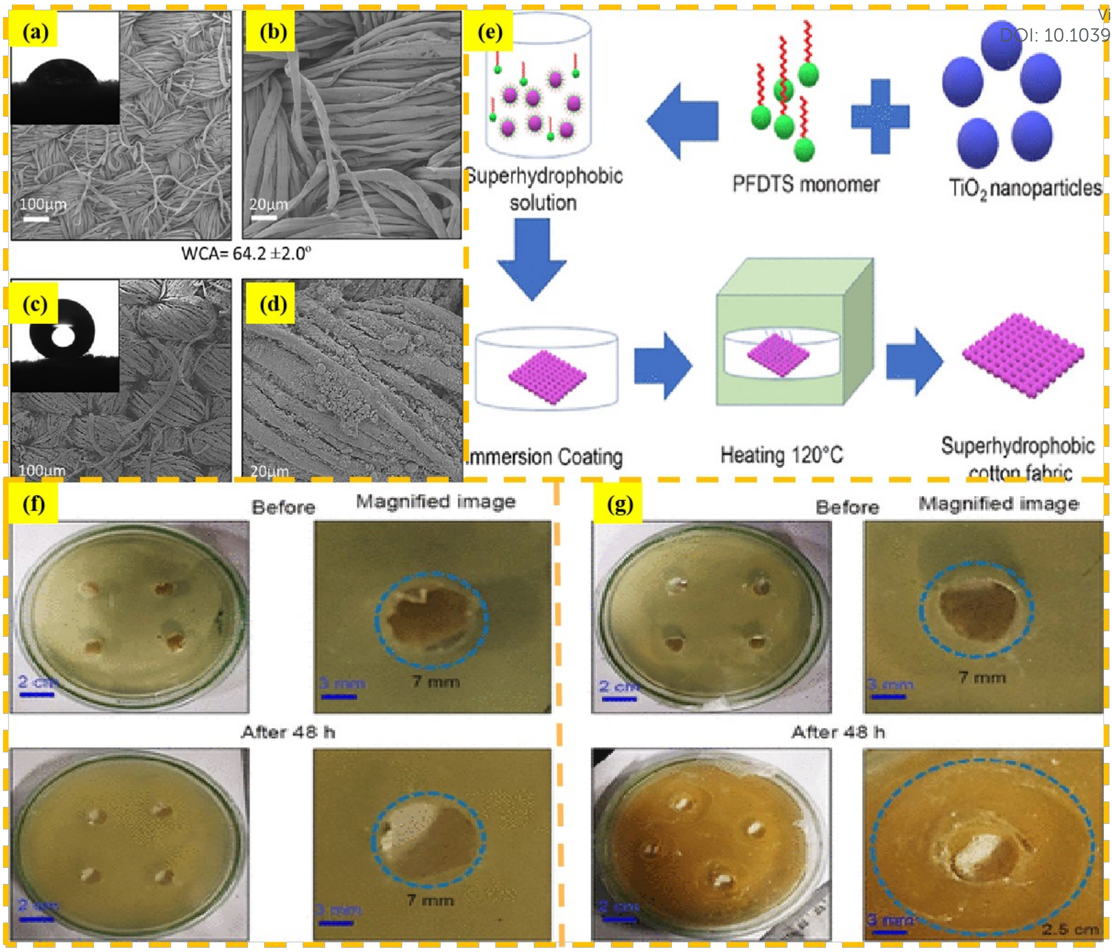

Fig. 5 SEM images of (a, b) coated and (c, d) uncoated cotton fabric with static contact angle of water, (e) schematic diagram of the preparation of superhydrophobic cotton fabric using PFDTS and TiO_2 nanoparticles in toluene, (f), (g) on behalf of optical images of uncoated and coated cotton fabric before and after 48 h incubation with E. coli bacteria at 37 °C. Inhibition zone (no growth of bacteria) near the coated fabric shows its antibacterial property.

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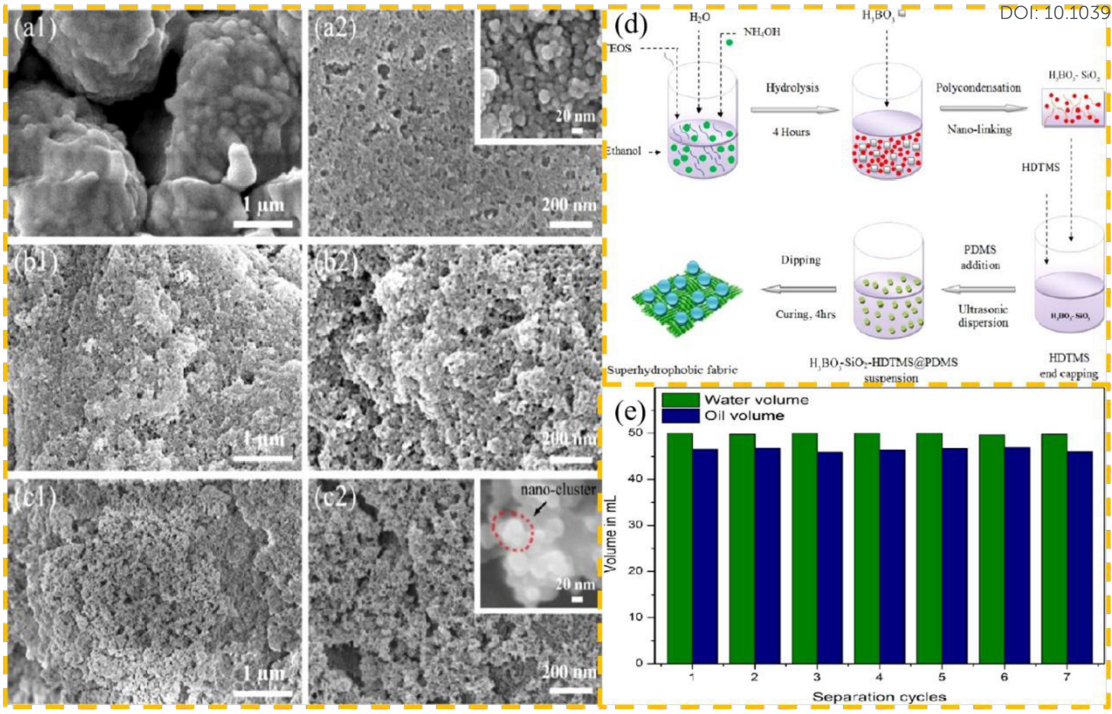

Fig. 6 SEM images of the coated fabric surface under different magnifications. Surface containing 21 wt% of SiO_2 (a1, a2), 9 wt% of H_3BO_3-SiO_2 (b1, b2), and 18 wt% of H_3BO_3-SiO_2 (c1, c2), (d) modification process of fluorine-free superhydrophobic cotton fabric, (e) oil/water volume (mL) after seven separation cycles by the coated superhydrophobic fabric.

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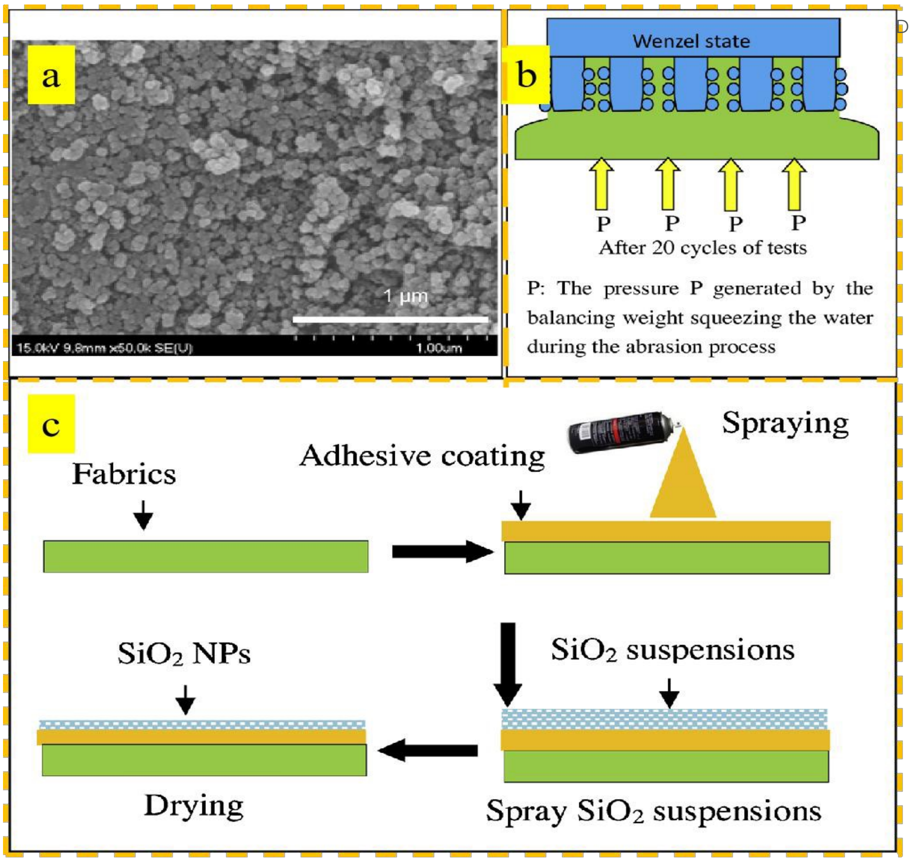

Fig. 7 (a) SEM images of the SH textile after 20 cycles of mutual abrasion tests in water under 0.68 kPa, (b) water wettability changed from a Cassie state into a Wenzel state after 20 test cycles, (c) process for the preparation of SH coatings on polyester fabrics.

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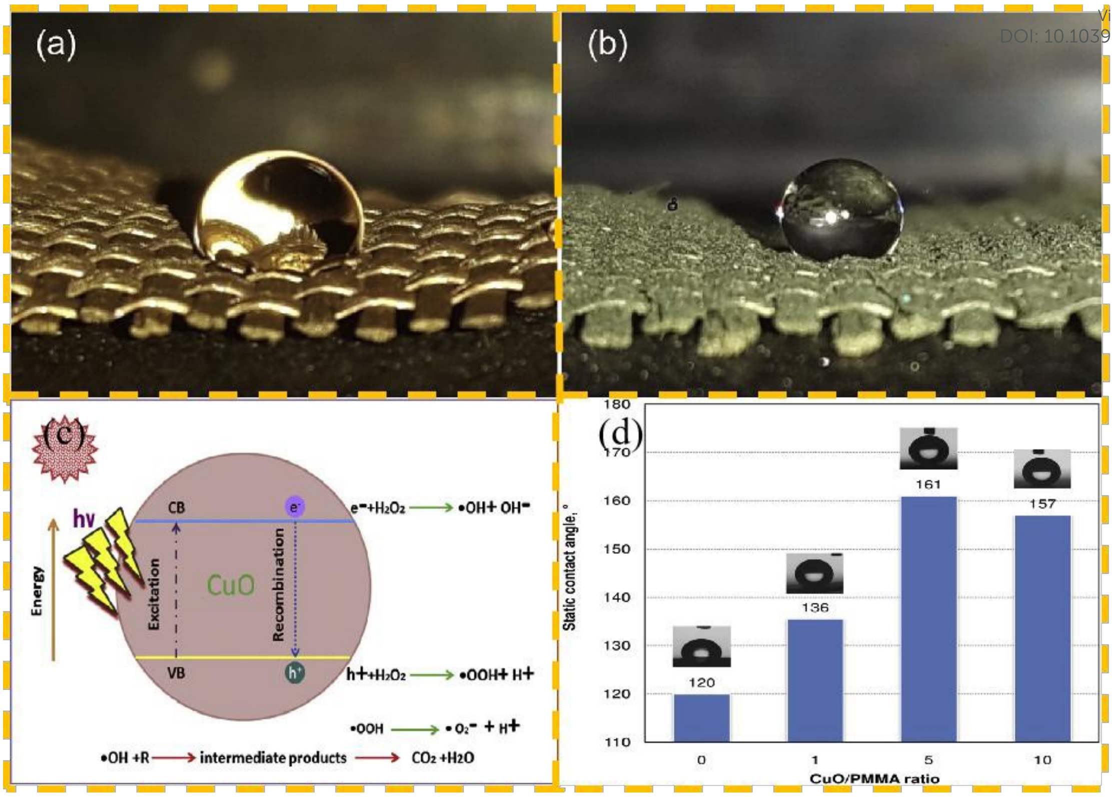

Fig. 8 Photographs of the water droplet on (a) the textile surface coated with the nanocomposite layer of unmodified CuO nanoparticles and PMMA, (b) the textile surface coated with the nanocomposite layer of modified CuO nanoparticles and PMMA, (c) mechanism of color change of aqueous solutions of rhodamine B upon the exposure to UV irradiation in time, (d) effect of CuO/PMMA ratio on the contact angle of the textile surface, and the contact angles were accurate within ± 0.2°.

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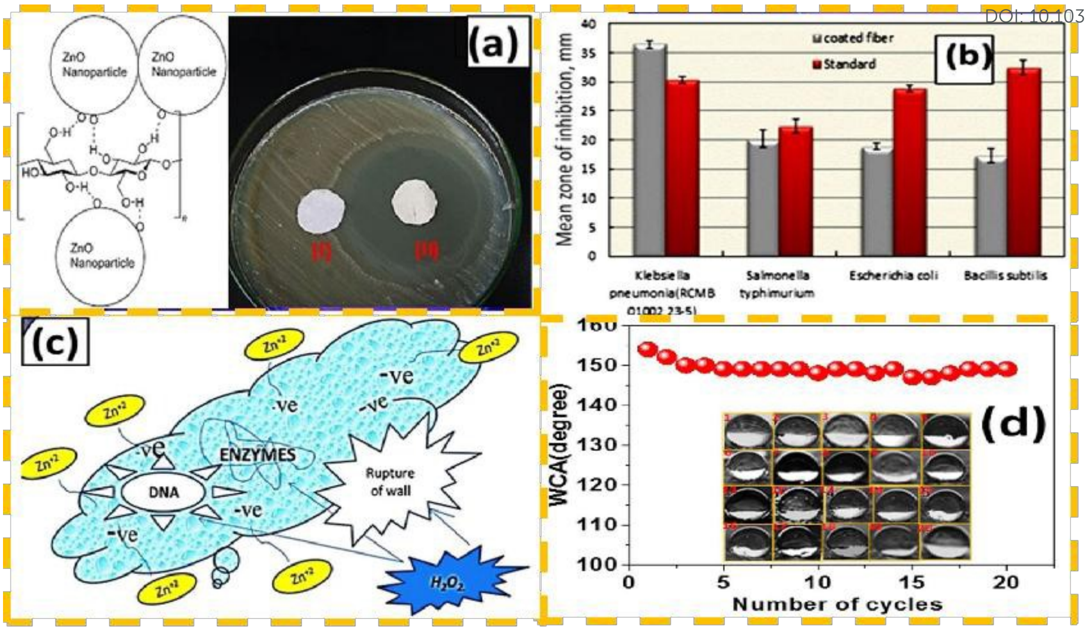

Fig. 9 (a) Antibacterial activity of uncoated cotton fabric and ZnO-coated fabric against Klebsiella pneumonia. The inset is the structural formula of ZnO-coated cellulosic cotton fabric, (b) effect of ZnO-coated fabric on zones of growth inhibition (mm) of different species of bacteria, (c) schematic diagram of the different antibacterial mechanisms of ZnO-coated fiber, (d) number of polishing cycles for the optimized ZnO-coated fabric. Insets: the photographs of the water droplets on the optimized ZnO-coated fabric.

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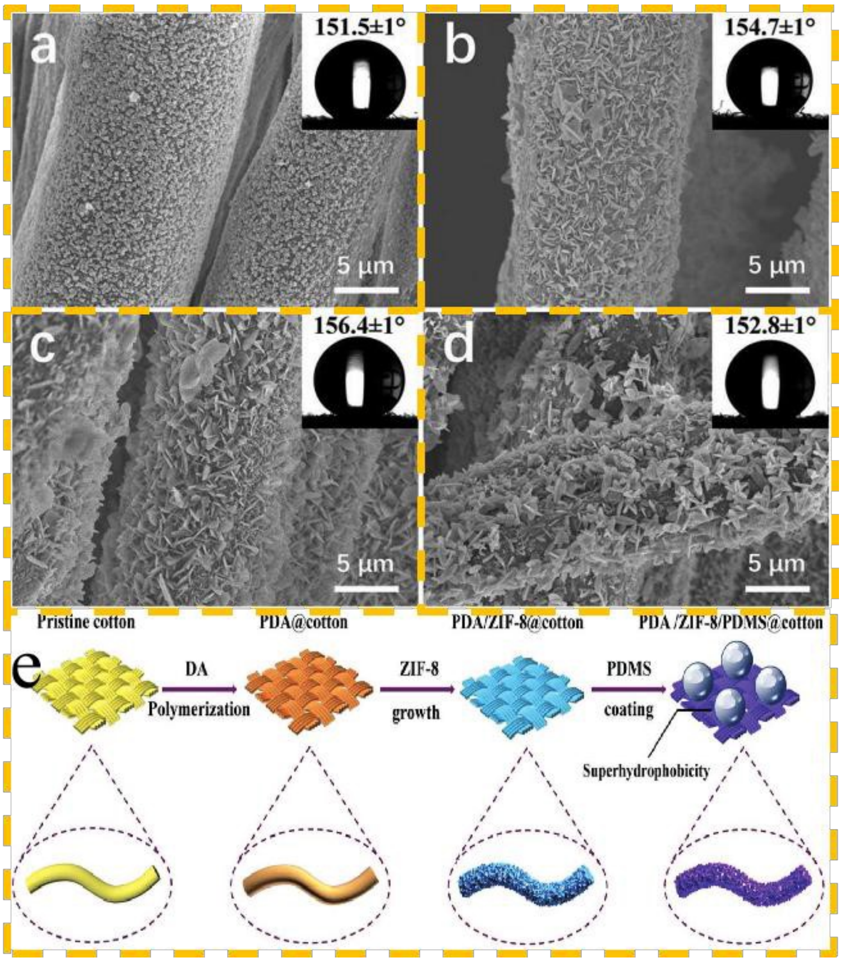

Fig. 10 SEM images (inset on the top right were the static behavior of the water droplet on the fabric) of PDA/ZIF-8/PDMS@cotton fabric with (a) n (Zn^{2+}): n (Hmim) = 1: 16, (b) n (Zn^{2+}): n (Hmim) = 1: 10, (c) n (Zn^{2+}): n (Hmim) = 1: 8, (d) n (Zn^{2+}): n (Hmim) = 1: 6, and (e) schematic illustration of the preparation of superhydrophobic PDA/ZIF-8/PDMS@cotton fabric.

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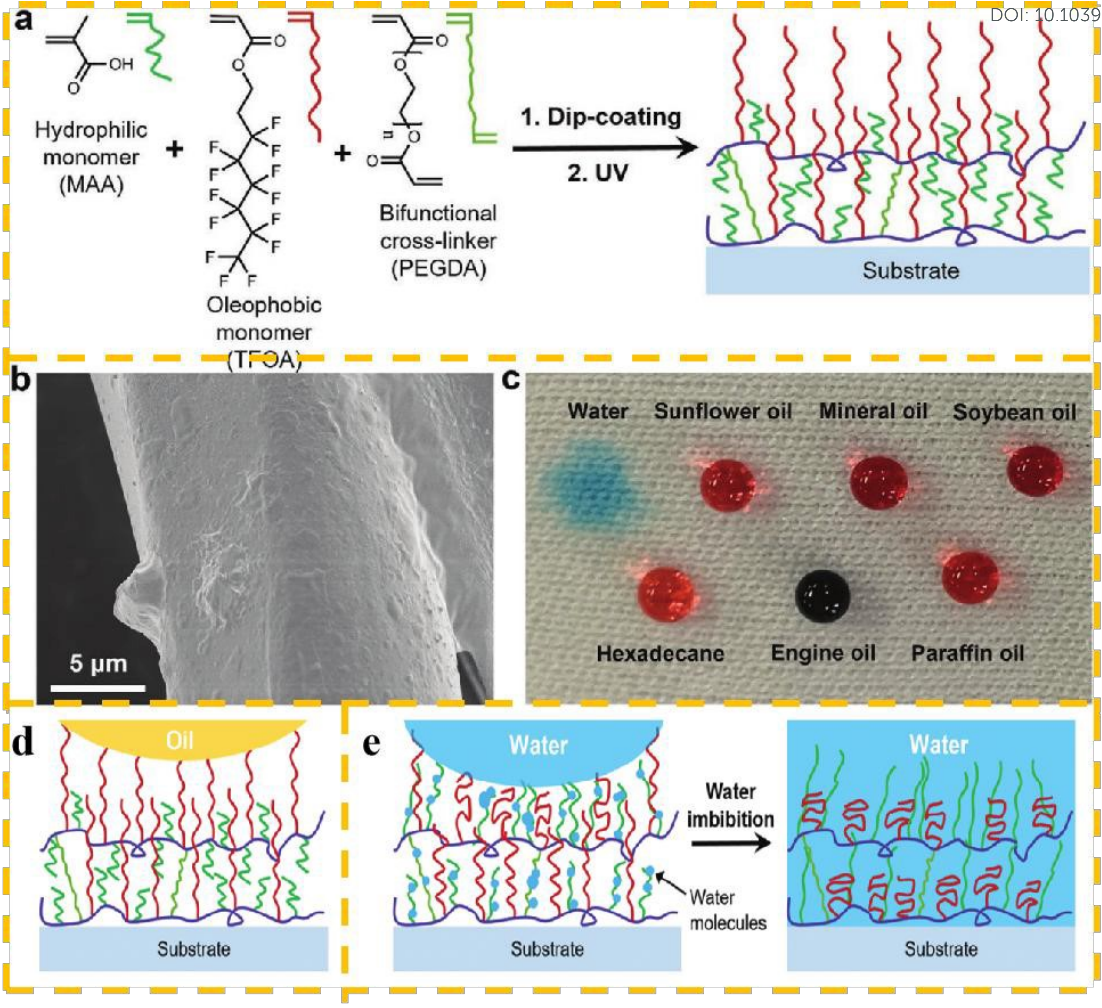

Fig. 11 (a) Schematic illustration for the formation of the oleophobic and hydrophilic coating, (b) SEM image of the coated polyester fiber, (c) photos of water and oil drops on the coated polyester fabric. Water and clear oils were colored with methylene blue and oil red, respectively, for easy observation, (d) the repellency of fluorinated chains to oil, and (e) the hydration of water molecules into the hydrophilic subsurface as well as the following water imbibition.

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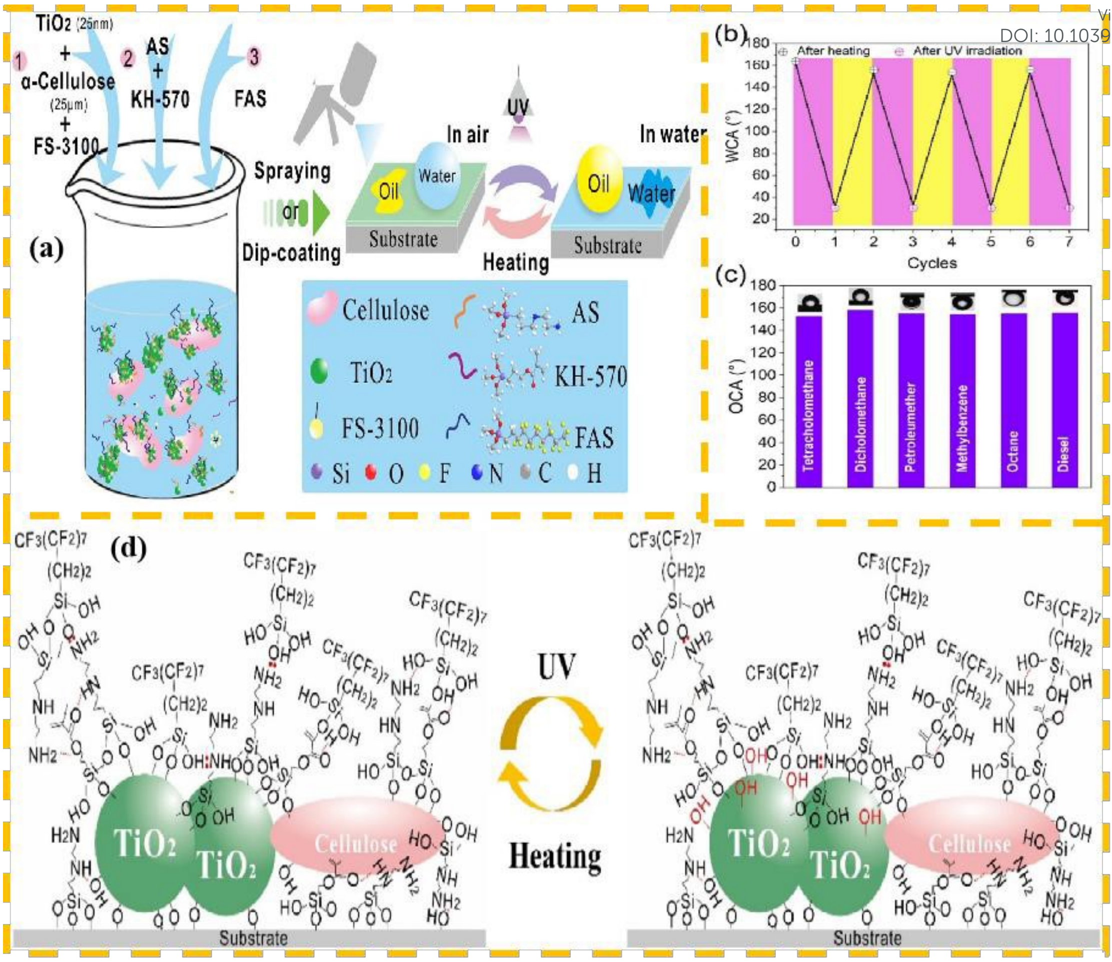

Fig. 12 (a) Schematic illustration of the fabrication of all-water-based superhydrophobic coating with reversible wettability, (b) the reversible wettability cycles of the coated cotton fabrics, (c) underwater OCA of UV-treated cotton fabric for different organic liquids, and (d) mechanistic diagram of wettability conversion of the coating under UV irradiation and heating treatment.

Table 1. Materials, methods, and efficiency of hydrophobic oleophilic fabrics.

Substrate	Material	Method	Efficiency	Refs.
cotton fabrics	$(Cu(NO_3)_2$, NaOH, 1-dodecanethiol	dipping	96.0%	65
cotton fabrics	Polydopamine, $AgNO_3$, BPO	deposition	96%	82
cotton fabrics	TiO_2, VTMS, 3-MPTMS	spraying	96.7%	86
cotton fabrics	TETA, TMC, Al_2O_3	crosslinking polymerization method	99%	133
cotton fabrics	PDA, DDT, $FeCl_3 \cdot 6H_2O$	in-situ	98%	134
cotton fabrics	MPTES, octadecyl methacrylate	grafting reaction	94%	135
cotton fabrics	DMA, Octadecyl acrylate	grafting reaction	94%	136
cotton fabrics	octadecyl methacrylate	grafting reaction	97%	137
cotton fabrics	PDMS	dipping	95%	138

cotton fabrics	PDMS	dipping	90%	139
cotton fabrics	palmitic acid	grafting reaction	95%	140
cotton fabrics	NH_4-HMP, LAP, hexadecyltrimethoxysilane	finishing	-	141
cotton fabrics	POSS, MPTES	grafting reaction	-	142
cotton fabrics	polyacrylates	dip-coating	-	143
cotton fabrics	lignin/metal ion	dip-coating	99.9%	144

Table 2. Materials, methods, and efficiency of hydrophilic oleophobic fabrics.

Substrate	Material	Method	Efficiency	Refs.
cotton fabrics	$ZnCl_2$, ammonia	in-situ	99.3%	110
basalt fibre fabric	CCl_4, H_2SO_4, HCl, NaOH	coating	99.4%	145
MCC	PFOA, TEMPO	spraying	98%	114
cotton fabrics	Chitosan, APS, MBA	coating	98%	120
cotton fabrics	HDTMS, 12-aminodododecanedioic acid	grafting reaction	97.3%	146
cotton fabrics	1H,1H,2H,2H-perfluorooctyltriethoxysilane	grafting reaction	97%	147
cotton fabrics	PFPE	dip-coating		148
cotton fabrics	ABC miktoarm star terpolymers	dip-coating	99.4%	149

cotton fabrics	STA, TiO_2, Al_2O_3	dip-coating	-	150
cotton fabrics	Copolymer of isopropylacrylamide and acrylic acid	dip-coating	-	151
cotton fabrics	polyethyleneimine, perfluorooctanoic acid	grafting reaction	96.5%	152
cotton fabrics	MOF	dip-coating	98.6%	153
cotton fabrics	Anionic ammonium polyphosphate	micro-dissolution	-	154
cotton fabrics	cellulose	dip-coating	93.2%	155
cotton fabrics	chitosan	in-situ surface deposition	99 %	156

Table 3. Materials, methods, and efficiency of intelligent response fabrics.

Response condition	Material	Method	Efficiency	Refs.
pH	TBT, LA, $NH_3 \cdot H_2O$	dipping	99%	124
UV irradiation, heating treatment	TiO_2, FAS, AS, KH-570	spraying	95%	127
CO_2/N_2	PMMA, PDEAEMA	self-assembly	99.9%	130
pH	VTMS, AA, HDTMS, DEAMP	free radical polymerization	97.5%	157

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Data Availability Statement

All data included in this work are available upon request by contact with the corresponding author.