REVIEW

Recent Advances in Superhydrophobic and Antibacterial Cellulose-Based Fibers and Fabrics: Bio-inspiration, Strategies, and Applications

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Abstract

Cellulose-based fabrics are ubiquitous in our daily lives. They are the preferred choice for bedding materials, active sportswear, and next-to-skin apparels. However, the hydrophilic and polysaccharide characteristics of cellulose materials make them vulnerable to bacterial attack and pathogen infection. The design of antibacterial cellulose fabrics has been a long-term and on-going effort. Fabrication strategies based on the construction of surface micro-/nanostructure, chemical modification, and the application of antibacterial agents have been extensively investigated by many research groups worldwide. This review systematically discusses recent research on super-hydrophobic and antibacterial cellulose fabrics, focusing on morphology construction and surface modification. First, natural surfaces showing liquid-repellent and antibacterial properties are introduced and the mechanisms behind are explained. Then, the strategies for fabricating super-hydrophobic cellulose fabrics are summarized, and the contribution of the liquid-repellent function to reducing the adhesion of live bacteria and removing dead bacteria is elucidated. Representative studies on cellulose fabrics functionalized with super-hydrophobic and antibacterial properties are discussed in detail, and their potential applications are also introduced. Finally, the challenges in achieving super-hydrophobic antibacterial cellulose fabrics are discussed, and the future research direction in this area is proposed.

Keywords Cellulose fiber · Super-hydrophobic · Antibacterial · Fabrication strategies · Applications

Introduction

Bacterial or viral infection can cause potentially life-threatening conditions, massive social unrest, and even global health crisis [1, 2]. Smallpox and 'Spanish' flu caused by viral infections killed around 300–500 million and 5–100 million people worldwide, respectively [3, 4]. It is estimated that around 2.8 million cases of cholera and more than 90,000 deaths arise from cholera annually [5]. In modern society, personal protective materials, such as respiratory masks, protective clothing, and medical textiles, have made important contributions to preventing the spread of viruses, reducing cross-infection, improving the success rate of surgery, and medical progress [6–14]. In recent years, the COVID-19 pandemic has caused global crisis and produced substantial impacts on the world economy, which also intensified the emphasis on health protection textiles.

Cellulose is a fibrous material of plant origin and the basis of all natural and man-made cellulosic fibers. Cellulose is a linear homo-polysaccharide made of β-1,4-anhydro-D-glucopyranose rich in three hydroxyl groups in each monomer, showing both excellent physical processability and high chemical reactivity. Cellulose-based fabric products have been ubiquitous in human life, of which cotton fibers occupy a large share of the textile market attributable to their flexibility, comfort, water/hygroscopicity, and breathability [11, 15, 16]. With the growing demand for eco-friendly fibers

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with good safety and comfort attributes, the consumption of cellulosic fabrics is expanding beyond the limit of natural cellulosic fibers like cotton, flax, hemp, jute, and ramie. Man-made cellulose-based fibers, e.g., Rayon, Modal, Lyocell, and (Tri-)acetate produced by regeneration of dissolved cellulose are also of great interest. The cellulose-based nonwovens consist of cellulose fibers originating from the most abundant natural resources in the world, which are used in paper materials, personal protective textiles, high-efficiency air filters for aircrafts and vehicles, etc. [17–25]

Textiles have been realized as host media for the growth of bacteria. Cellulose fibers are inherently hydrophilic and nutritious, which make them a suitable breeding ground for bacteria [26, 27]. In general, the life cycle of bacteria on fibers can be divided into four stages: adhesion, proliferation, drying, and washing-off. Bacteria arising from sweat are initially drawn into the hydrophilic fibers. The bacteria quickly attach to the fiber surface and form a biofilm. When the fibers become dry, the bacteria on the surface irreversibly adhere to the fiber surface by capillary action and are hard to be removed completely by laundry [27, 28]. The accumulation of bacterial biofilms can wreak havoc. For instance, bacteria, fungi, and virus can cause odors in intimate clothing and active sportswear, allergic reactions, stains on upholstery and other household items, and cross-contamination in medical settings, which adversely impact on personal and public health. Antibacterial finishes of textiles have been used to maintain their hygiene and avoid the infection from pathogens at home and especially in hospitals, nursing homes, schools, hotels, and crowded public areas [6, 22, 29–33].

Basically, the antibacterial finishing of fibers can be divided into anti-biofouling and bactericidal. Among them, anti-biofouling is considered to be related to the non-adhesive super-hydrophobic fiber surfaces, and the bactericidal performances depend on the post-treatment of cellulose fibers with antibacterial agents. Figure 1 summarizes some natural surfaces with super-hydrophobic and antibacterial properties, the main fabrication strategies and antibacterial properties of developed super-hydrophobic antibacterial cellulose fabrics and their potential applications. There are mainly five approaches to prepare super-hydrophobic cellulose fiber surfaces, e.g., polymerization, wet-chemical coating, sol–gel, layer-by-layer assembly (LbL), and chemical vapor deposition (CVD). Their antibacterial functions can be realized via anti-biofouling, contact killing, biocide leaching, and self-cleaning.

Previous reviews have discussed the preparation strategies and potential applications of antibacterial surfaces [39–44]. Zhan et al. [45] reviewed the recent progress of antibacterial super-hydrophobic coatings focusing on bacterial adhesion and infections. Bhandari et al. [46] summarized the use of metal nanoparticles for antimicrobial
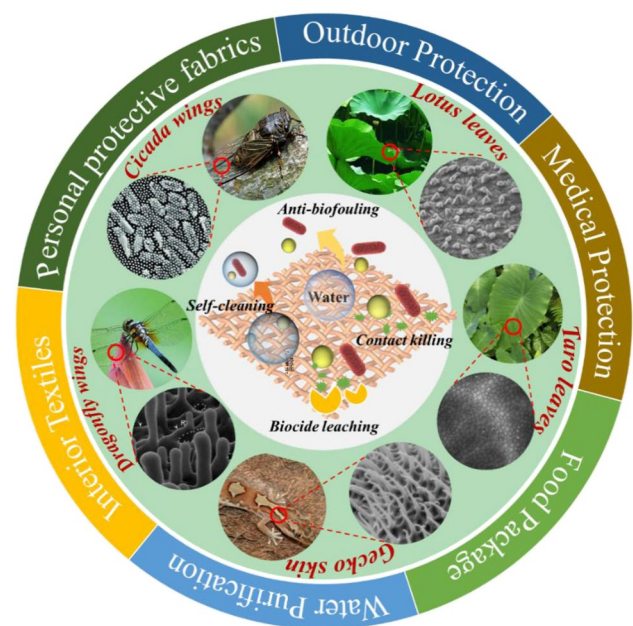
Fig. 1 Schematic illustration showing natural super-hydrophobic and antibacterial surfaces (i.e., taro leaves, cicada wings, gecko skin, and dragonfly wings), the fabrication strategies and antibacterial properties of super-hydrophobic antibacterial cellulose fabrics and their potential applications as summarized in this review. Reproduced with permission from ref [34] copyright 2012 Wiley. Reproduced with permission from ref [35]. Copyright 2015 Elsevier. Reproduced with permission from ref [36]. Copyright 2016 Royal Society of Chemistry. Reproduced with permission from ref [37]. Copyright 2017 American Chemistry Society. Reproduced with permission from ref [38]. Copyright 2020 Natural Publishing Group

finishes on textiles. The developments of anti-biofouling surfaces prepared by combining surface topography and chemistry have also been summarized [47, 48]. The works on cellulose fabrics with super-hydrophobic surfaces imparting unique anti-biofouling and contact killing or biocide leaching properties are seldom reviewed. This review presents the recent progress on super-hydrophobic antibacterial cellulose fabrics. Some classic examples of natural plants and insects showing super-hydrophobic and antibacterial surfaces are introduced, and their respective mechanisms are explained. Then, the recent development of super-hydrophobic cellulose surfaces and the main strategies for fabricating superhydrophobic antibacterial cellulose fabrics are categorized and discussed in detail. The contribution of superhydrophobicity to reducing live bacteria adhesion is elucidated, and the related antibacterial mechanisms of superhydrophobic surfaces are also discussed. The potential applications of superhydrophobic antibacterial cellulose fabrics will be briefly summarized. Finally, the challenges and future research directions of superhydrophobic antibacterial cellulose fabrics are proposed.

Antibacterial Super-Hydrophobic Surfaces in Nature

Naturally occurring plant and insect surfaces have evolved diversified strategies to minimize contamination from foreign particles. The encounter of surfaces with nanoscale pollutants is unavoidable in nature. Therefore, their antifouling properties against particulates have practical implications in many applications, e.g., self-cleaning, antibiofouling or antibacterial [34, 49–64]. Superhydrophobicity is closely related to both hierarchical roughness structures and low surface energy species of the surfaces. Studies have shown that contaminant particles hardly affect the superhydrophobicity of the surface as long as their sizes exceed the surface pore size or the contaminant thickness is lower than the height of the protrusions on the rough surface [63]. Natural surfaces with antibacterial properties can be classified into antibiofouling and bactericidal surfaces [34, 65]. Antibiofouling means some natural surfaces can prevent or limit the settlement of early microbial contaminants. This is because the air layer retained on the superhydrophobic surface can inhibit the cell attachment. However, this air layer gradually disappears with the prolongation of immersion time, losing the anti-biofouling ability. Therefore, the resistance to biofouling in the fully wet state has attracted attention. Bactericidal property refers to the ability of some natural micro-/nano-structured topologies to inactivate bacteria by physically puncturing or stretching bacterial cells. [34, 35, 37, 34, 35] Many creatures in nature have multifarious surfaces or skins that show effective anti-bio-fouling and bactericidal properties via ingenious and robust designs through evolution,

which can inspire researchers to develop super-hydrophobic antibacterial surfaces.

Taro Leaves

Ma et al. [66] revealed the biofouling-resistant surface of taro leaves under completely wet conditions. Hexagonal epidermal cells on taro leaves formed microscopic "papillary bumps"; the taro leave surface was also unevenly covered by nanoscale wax crystals, with denser coverage of the "papillary bumps" than the edges. As a result, taro leaves are super-hydrophobic with a water contact angle (WCA) of ~ 155° as shown in Fig. 2a and b [66, 76]. A few abiotic (carboxylate-modified poly(styrene-co-acrylic acid)) PSA particles and biotic cells (*P. aeruginosa*) were applied to observe the antifouling performances. Under dry conditions, an air layer was trapped on the surface and the surface showed no obvious adhesion in the papillary bumps area, as shown in Fig. 2c and d. Similar phenomenon was observed after wetting the surface with ethanol to remove the trapped air layer, as shown in Fig. 2e and f. The adhesion mainly happened at the boundary because nanoscale wax crystals, which have higher distribution density in the central part than at the boundary. Therefore, it can be affirmed that the nanoscale wax crystals of taro leaves are responsible for their antibiofouling properties in both "non-wet" and "submerged" states. As for the "submerged" state antifouling, Parkin et al. [77] proposed that the rough structure of the superhydrophobic surface may provide a suitable environment for bacterial attachment, serving as reservoirs for bacteria and enhancing the adhesion of bacteria.
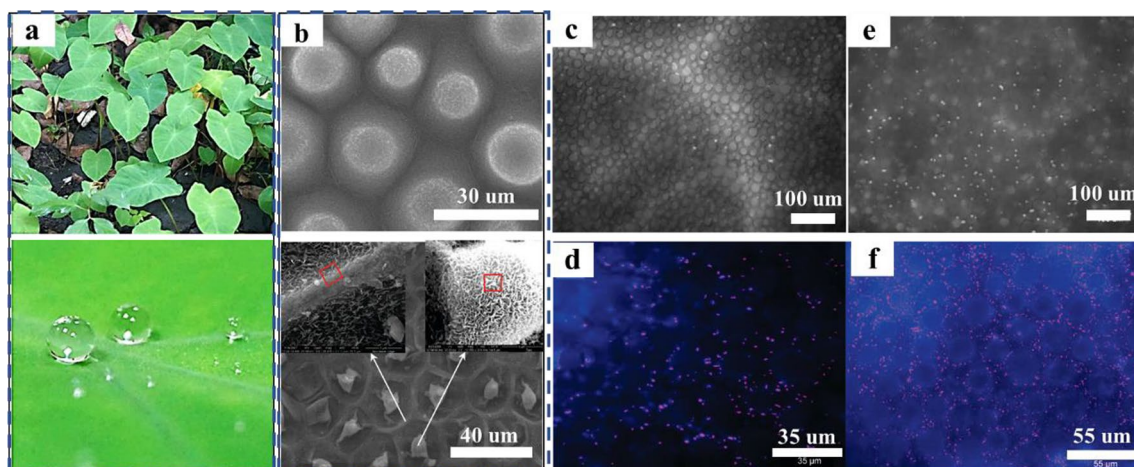
Fig. 2 a Photo of taro leaves and water droplets on the surface. Reproduced with permission from ref [38]. Copyright 2020 Natural Publishing Group. **b** SEM images of the taro leaf treated by liquid substitution (top) and air-dry sputter coating (bottom). **c–d** PSAA microsphere and *P. aeruginosa* adhesions on the taro leaf under non-wet conditions, respectively. **e** and **f** PSAA microsphere and *P. aeruginosa* adhesions on the taro leaf under wet conditions, respectively. Reproduced with permission from ref [66]. Copyright 2011 American Chemical Society
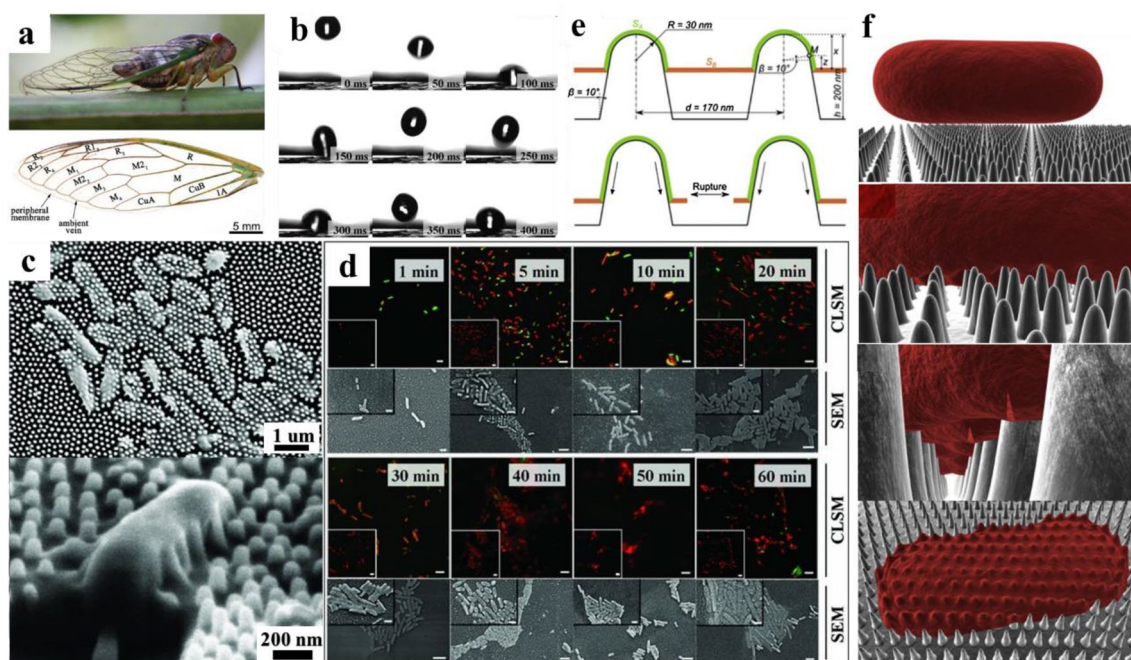
Cicada Wings

The cicada (*Psaltoda claripennis*) wing is the first reported natural surface with physico-mechanical bactericidal properties. Unlike antifouling taro leaves that only repel bacterial cells, cicada wings are able to stay clean through sustained bactericidal action [34, 65, 67, 70]. The surface of cicada wings is covered by an array of nano-scale superhydrophobic pillars with approximately hexagonal spacing, and water droplets could easily bounce off the surface to achieve self-cleaning as seen in Fig. 3a and b. It has been found that these nanopillars on the surface of cicada wings are so lethal to bacteria that the individual bacterial cell can be killed in about 3 min (Fig. 3c). Confocal laser scanning microscopy (CLSM) and scanning electron microscopy (SEM) images in Fig. 3d show that the nanopillars started penetrating the *P. aeruginosa* cells immediately after they were attached, with most of the cells killed within 5 min and an "attachment/kill cycle" occurring within 20 min.

It has also been revealed that the bactericidal property of cicada wings is mainly due to their surface topology. After the super-hydrophobic surface of cicada wings was gold-coated, the surface became hydrophilic, but the topographical structure was reserved, which continued to provide the cicada wings with bactericidal property [34]. The rigidity of bacterial cells was found to be a key factor in determining their resistance/sensitivity to the bactericidal function of cicada wings [67]. Gram-negative cells (*B. subtilis, P. maritimus, and S. aureus*) are more sensitive to the cicada wing surface morphology than Gram-positive cells because Gram-positive cells have a thicker peptidoglycan layer and are generally more rigid than Gram-negative cells. The biophysical sterilization model of cicada wings has been proposed as follows: when bacteria attach to the nanopillar structures on the wing surface, their cell membranes stretch in the area suspended between the pillars causing the bacterial cells to rupture and the eventual direct penetration of bacterial membranes as seen in Fig. 3e and f [65].

Since the first discovery of mechanical killing of *Pseudomonas aeruginosa cells* by the cicada *Psaltoda claripennis* wing nanoarchitecture, surfaces with bioinspired geometries have been developed. A self-organized nanotextured surface with patterns of black silicon (bSi) provided a direction for large-scale fabrication of nanotextured surfaces with mechano-bactericidal effect against bacterial cells [68, 78, 79]. In addition, the superhydrophobic gecko skin has a similar morphological structure to the convex array on the cicada wing [35]. The surface features of the gecko skin were shown in Fig. 4a and b. These micro-/nanostructures composed of thorns with submicron spacing and curvature radius of less than 20 nm exhibited very low adhesion to pollen and silica particles. As a result, the gecko skin also

Fig. 3 a Photograph of cicada and cicada forewing structure. **b** SEM image of *P. aeruginosa* cells on the surface of a cicada wing. **c** Water droplets contact the wing surface. **d** CLSM and SEM images of *P. aeruginosa cells* attached to the cicada wing surface. Live cells: in green and dead cells: in red. Reproduced with permission from ref [34]. Copyright 2012 Wiley. **e** and **f** The biophysical model of the interaction between cicada wing nanopillars and the penetration of bacterial membranes. Reproduced with permission from ref [65]. Copyright 2013 Elsevier
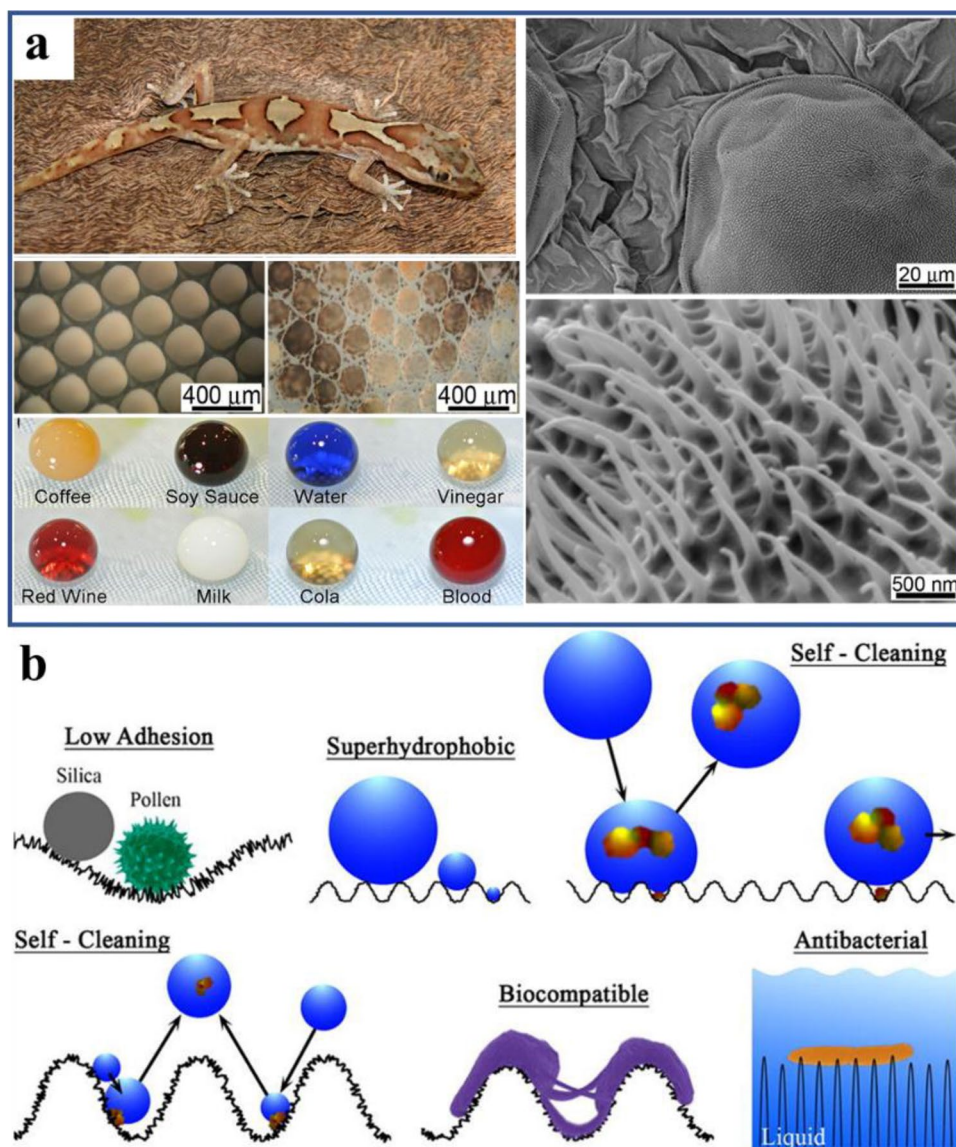
Fig. 4 a Photos, SEM images and the super-hydrophobicity of gecko skin surface. **b** Surface feature illustration of the gecko skin. The low adhesion and super-hydrophobicity of the surface facilitate self-cleaning by impingement and droplet rolling. Reproduced with permission from ref [35]. Copyright 2015 Elsevier

exhibited antibacterial properties derived from the biophysical sterilization, by contacting (adsorbing and stretching) the bacterial cell wall of Gram-negative bacteria (*Porphyromonas gingivalis*).

Dragonfly Wings

Surfaces featured with high-aspect ratio nanostructures are common in nature. Dragonfly wings have been found to have disordered nanostructures with graded topographical characteristics [36, 68, 71, 72, 75]. These surface nanostructures exert deformation stress on the peptidoglycan cell wall and inner membrane of bacterial cells resulting in

efficient bactericidal activity, analogous to the sterilization mechanism of cicada wing nano-pillars [68]. The surface of dragonfly wing showed super-hydrophobic property with a WCA of around 155–164° as seen in Fig. 5a, which can be attributed to the combination of the rough topography and chemical components of amides from protein, chitin, and fatty acids [36]. Figure 5b shows the nano-protrusions and optical profiles of the surface, which consists of numerous randomly arranged nano-pillars with average heights of ∼ 200 nm (Fig. 5c). It was found that these disordered nanostructures on the dragonfly wing were more effective in reducing protein adhesion than the ordered cicada replicated surface, thus the dragonfly wing surface was highly effective
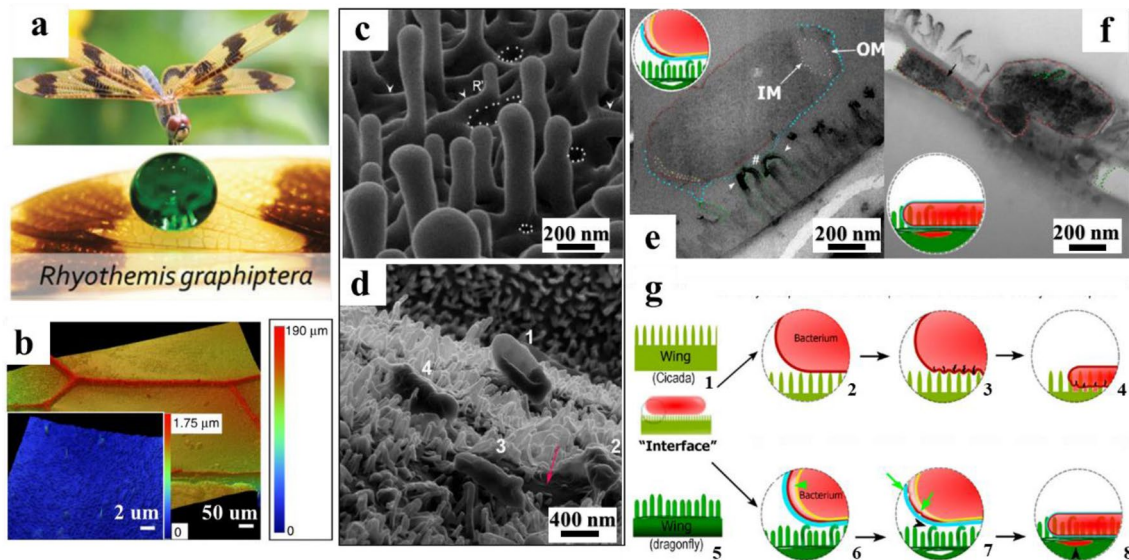
Fig. 5 **a** The dragonfly wings and the super-hydrophobic surface property. Reproduced with permission from ref [36]. Copyright 2016 Royal Society of Chemistry. **b** Optical profile of nano-protrusions in the dragonfly forewing. Reproduced with permission from ref [68]. Copyright 2013 Nature Publishing Group. **c** Helium ion micrograph of the nanopillar arrangement on the dragonfly wing. **d** Four stages of gradual death of *E. coli* attached to the nanopillar surface of dragon-fly wing. **e**, **f** TEM images showing bacteria–nanopillar interaction: **e** Separation of the inner membrane and outer membrane of the *E. coli* bacteria, and the bending of the nanopillars beneath the bacteria. **f** Bacteria deformed and shrank to the nanopillar matrix. **g** Mechanisms of bactericidal activity models based on cicada wing structure (1–4) and dragonfly wing (5–8), respectively. Reproduced with permission from ref [75]. Copyright 2017 American Chemical Society

in inhibiting the adhesion of bacteria (i.e., Gram-negative and -positive) due to the disruption of cell surface in contact with the nanostructures [80–83]. Figure 5d shows the four stages of *E. coli* death process, respectively. Bacteria numbered 1, 2, and 3 were in the primary and intermediate stages of death with gradual wrinkling of their cellular morphology, but the bacterial membrane remained intact on the nanopillars. The height of dead bacteria 4 became comparable to that of nanopillars indicating the cytoplasm exuded completely and bacteria sank into the nanopillars [35]. In addition, Bandara et al. [75] believed that the bacteria settled into the nanopillars only after their membranes were damaged. The bacteria attached to nanopillars on the surface of the dragonfly wings through secreted extracellular polymers, which can avoid direct contact between the cell membrane and nanopillars. When bacteria tried to move on the nanopillars, the cell membrane was stretched due to the strong van der Waals and shear forces between the bacterial membrane and the nanopillars. As a result, the inner and outer membranes were separated and the nanopillars were bent as shown in Fig. 5e and f, and the bacterial cells were damaged and lost their integrity with cytoplasm leaking and flowing to the surrounding nanopillars. Figure 5g illustrates the antibacterial mechanisms based on the nanostructured surfaces of cicada wing and dragonfly wing, which confirms the membrane perforation theory of the bactericidal mechanism of cicada wings. However, the dragonfly wing derives

its antibacterial property from the synergistic effect of strong bacteria–nanopillar adhesion and shear force when immobilized bacteria move on the nanopillar surface.

Inspired by the dragonfly wing surface, Ivanova et al. [68] developed biomimetic bSi surface with disordered nanopillar systems and compared its antibacterial ability with dragonfly wings. As seen in Fig. 6, both bSi and dragonfly wing surfaces were effective not only against the Gram-negative *P. aeruginosa* cells but also against Gram-positive *S. aureus and B.* subtilis cells. Gram-positive cells are more rigid and resistant than Gram-negative cells. The results indicated that similar antibacterial activities can be found on the biomimetic nanostructured bSi surfaces, which were resulted from mechanical and structural responses to the deformational stresses imposed by the surface nano-texture on the cell wall of peptidoglycan and inner membrane of bacterial cells. Later, the same group fabricated biomimetic bSi surfaces with controlled height of bSi nanopillars between 200 and 600 nm by plasma etching [72]. The resulted nano-textured bSi surfaces were superhydrophilic with a WCA of only 8°. After modification with trichloro(1H, 1H, 2H, 2H-perfluorooctyl) silane (PFTS), the bSi surfaces became superhydrophobic with a WCA of over 160°, while the PFTS coating did not change the original surface morphology. The antibacterial effects of nanotextured bSi surfaces with different wettabilities ranging from superhydrophilicity to superhydrophobicity were all evaluated. It has been found that
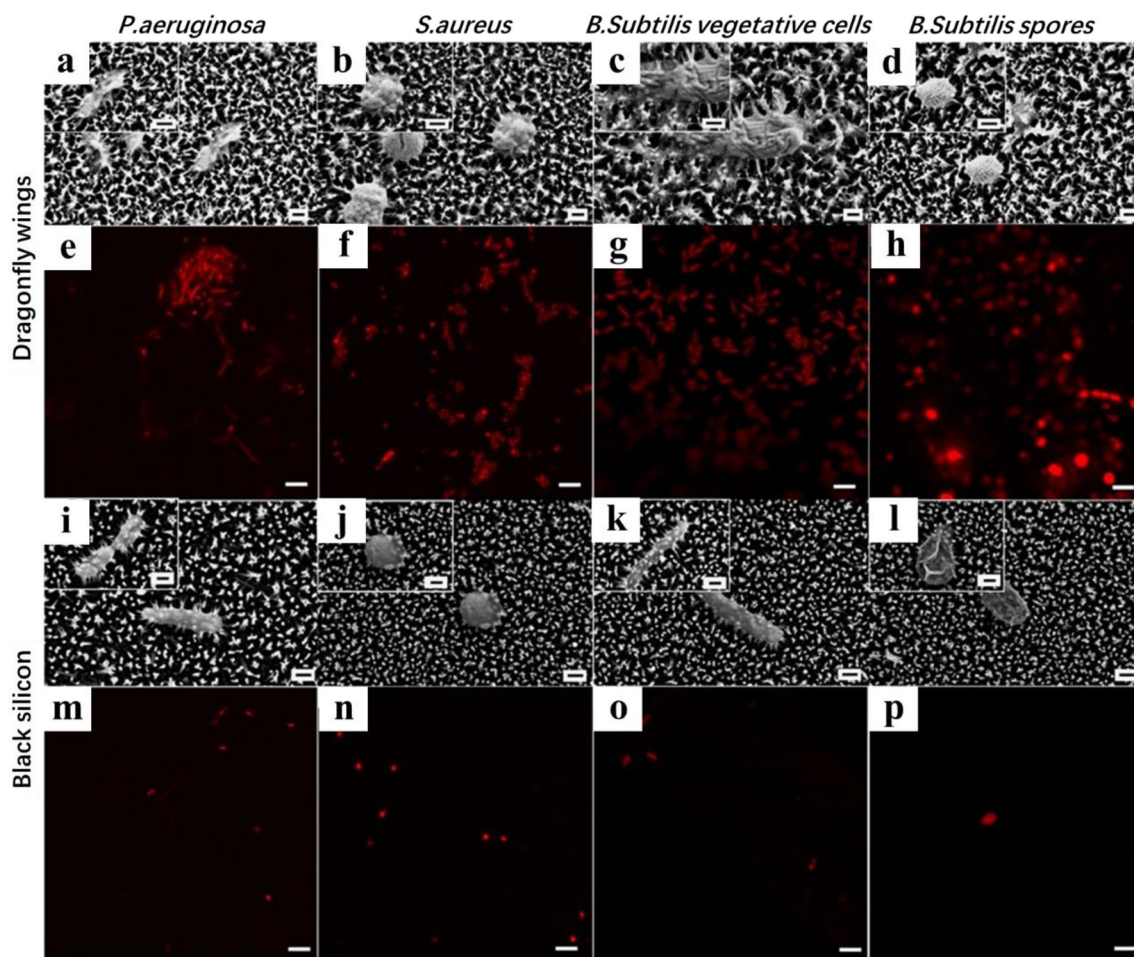
Fig. 6 SEM images of different bacterial cells disrupted through interaction with the dragonfly wing (**a–d**) and bSi **i–l**. (Scale bars = 200 nm) Confocal laser scanning images showed the disruption by dragonfly wing (**e–h**) and bSi (**m–p**) was lethal to the cells. Nonviable bacterial cells and spores were red colored, indicating the inactivating the bacteria. (Scale bars = 5 μm) Reproduced with permission from ref [68]. Copyright 2013 Nature Publishing Group

superhydrophobic and superhydrophilic bSi surfaces showed similar antibacterial capability against Gram-negative (*P. aeruginosa* and Gram-positive (*S. aureus*). Furthermore, bSi surfaces with longer and more pronounced nanopillars exhibited enhanced bacterial cell deformability compared to dragonfly wings. The results indicated that such mechanobactericidal behavior was not affected by the change of surface chemical properties.

Lotus Leaves

Lotus leaf surfaces, one of the most well-known superhydrophobicity examples in nature, have been extensively studied for their self-cleaning and anti-biofouling properties [50]. The lotus leaf surface has a micro–nano-hierarchical structure composed of micro-scale protrusions and nanoscale wax tubes, which render the surface with super-hydrophobicity with a WCA of over 150° and a small rolling angle of around 5º as shown in Fig. 7a and b. The air cushion trapped at liquid/solid interfaces resists the bacterial adhesion at the initial stage, but it may gradually lose the antibacterial-adhesion efficiency due to the impairment of air cushion with the prolonged immersion time [74, 77]. The lotus leaf surface has also demonstrated bactericidal properties in addition to anti-biofouling property. The CLSM images of *E.coli* cells attached on lotus leaves and the corresponding SEM images are shown in Fig. 7c and d. The results indicated that the lotus leaf surface showed excellent resistance against biofouling with nearly no bacterial attachment at the initial 3 h incubation due to the anti-bacterial adhesion of the super-hydrophobic surface, and the air cushion trapped on the surface gradually disappeared with the prolonged soaking time. Although more bacteria adhered to the surface for over 24 h (Fig. 7c), almost all the bacteria attached were killed (stained in red) during the full observation period from 3 to 24 h, verifying the bactericidal
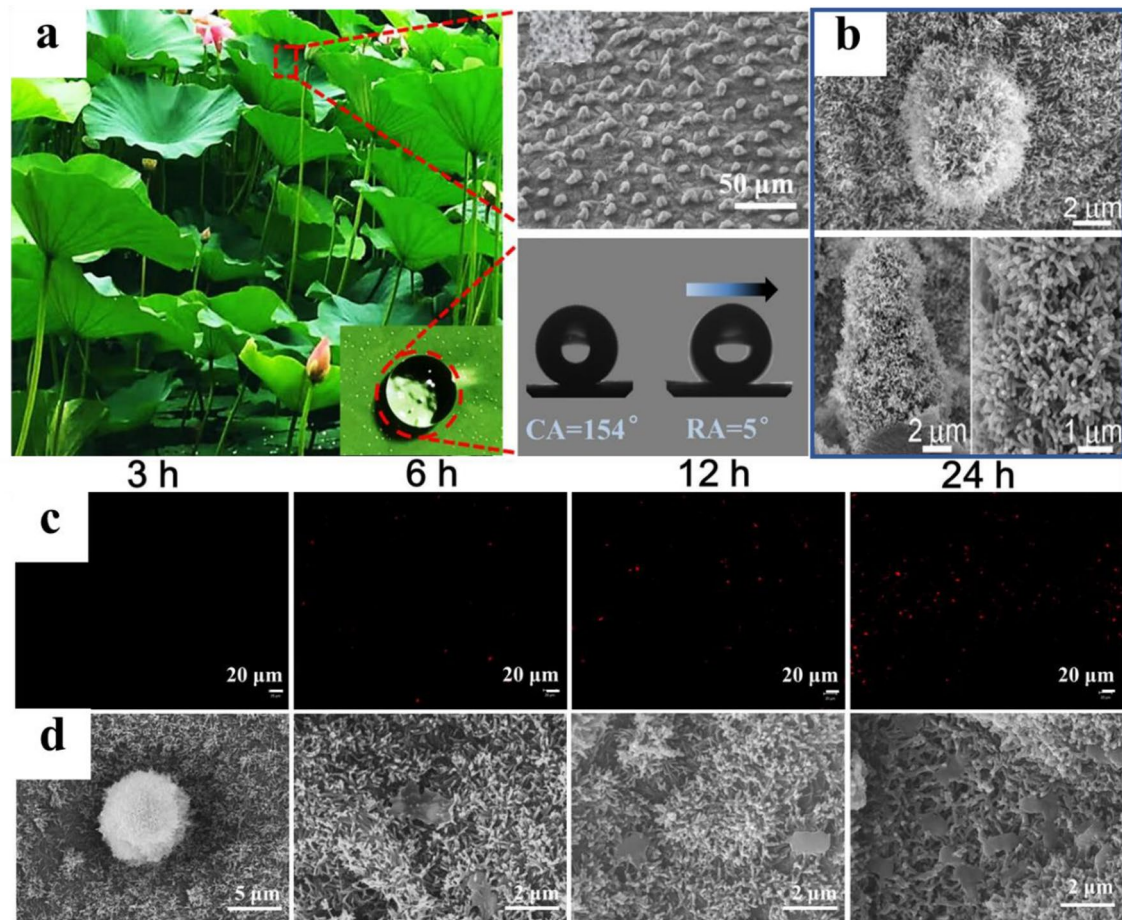
Fig. 7 **a** Photo of lotus leaf and a water droplet on its surface, typical SEM image, and the WCA and SA of the lotus leaf surface. Reproduced with permission from ref [74]. Copyright 2020 Elsevier. **b** SEM images of low and high magnification of lotus leaf surface. **c** and **d** Representative CLSM and SEM images of *E. coli* on the surface of fresh lotus leaves after incubation for 3 h, 6 h, 12 h, and 24 h. Live bacteria (green) and dead bacteria (red). Reproduced with permission from ref [37]. Copyright 2017 American Chemistry Society (color figure online)

properties of lotus leaf surfaces. To further explore the bactericidal mechanism of the lotus leaf surface, the morphologies of attached bacterial cells were evaluated by the SEM images shown in Fig. 7d; the bacterial film attached to the surfaces was highly deformed and gradually sank into the nanotube gaps, eventually the bacterial cell membrane was engulfed by the spaces between randomly oriented nanotubes. Such mechanical killing strategy is quite similar with those of cicada and dragonfly wings [74].

Inspired by the lotus leaf surface morphology and function, many super-hydrophobic surfaces with micro-/nano-topographic structures have been developed and their anti-adhesion to microorganisms has been demonstrated [84–86]. Zouaghi et al. [85] developed different biomimetic surfaces (i.e., slippery lubricant liquid-infused surface and atmospheric plasma-sprayed silane-based nano-rough thin films on stainless steel substrates), and compared their anti-fouling and anti-bacterial properties. A lotus like surface was first prepared by femtosecond laser ablation treatment and silanized with 1H,1H,2H,2H-perfluorodecyl, a perfluorinated oil was then used as lubricant oil to infuse the textured surface to obtain slippery superhydrophobic surface. The nano-rough plasma spraying coated surface was prepared by spraying hexamethyldisiloxane (HMDSO) on the stainless steel substrate. Anti-biofouling tests were conducted in a pilot pasteurizer with 1.0 wt% whey protein, a complete suppression of fouling can be observed on the slippery surface. Moreover, the plasma coating reduced the number of adhered *L.monocytogenes* and *S.enterica* by 58% and 80%, respectively.

In addition, some insect wings, such as planthopper wings, exhibiting similar surface chemistry and nano-textured morphology with lotus leaf surfaces, also showed bactericidal properties [37, 51, 87]. Figure 8a shows photos of a plant-hopper resting on a twig, and its hindwing and forewings. Figure 8b shows the dynamic super-wettability
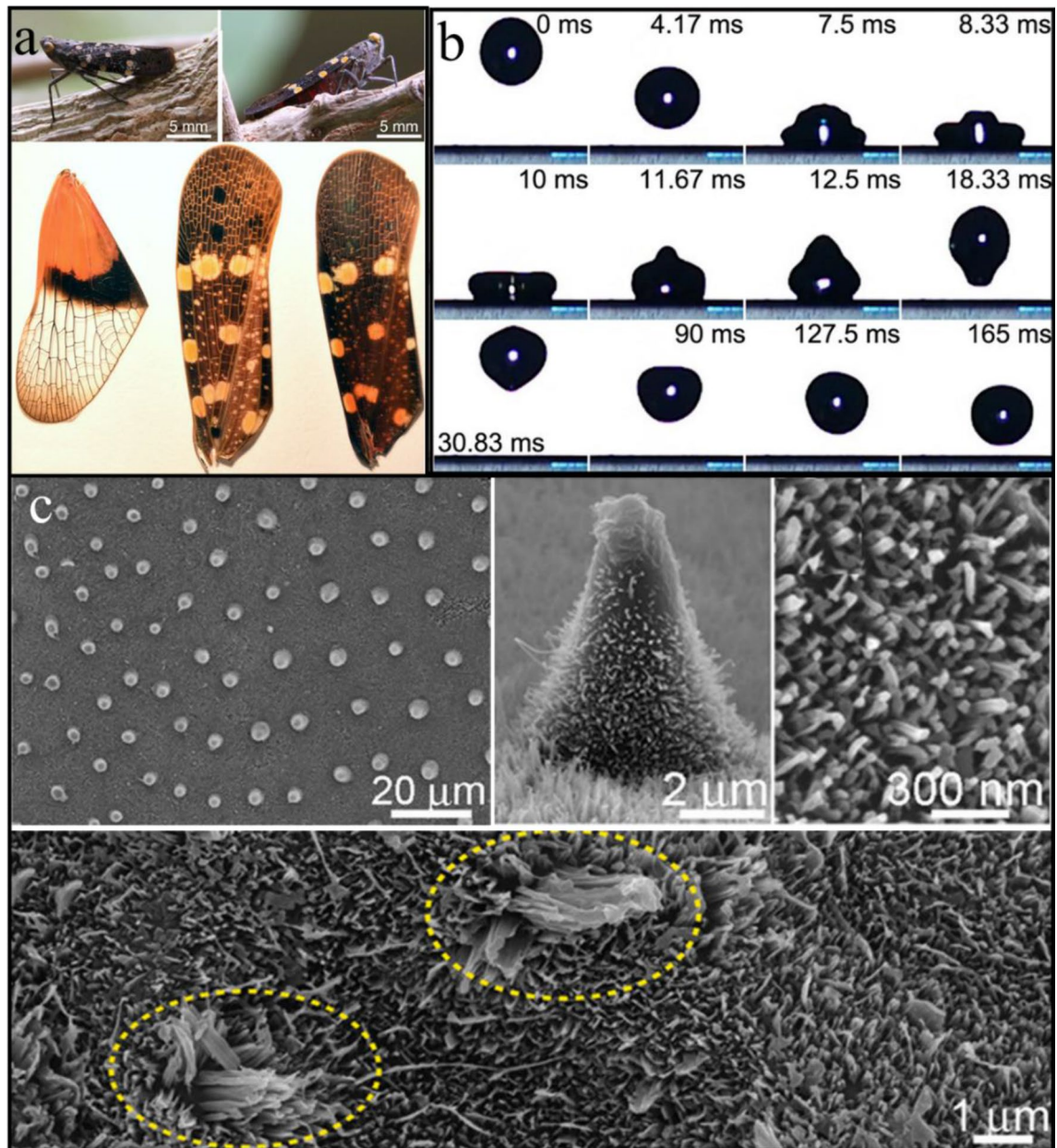
Fig. 8 **a** Photos of native planthoppers (*D. danae*) living in the Australian and Northern Queensland and their hindwing and forewing. **b** Dropping water on the forewing surface of planthopper. **c** SEM images of plant-hopper hindwing at various magnifications, the high-lighted regions in the bottom image show the upper-tier topography displaying isolated clumps protruding from the surface. Reproduced with permission from ref [37]. Copyright 2017 American Chemistry Society

of the plant-hopper's wing surface. When small water droplets (~2.2 mm in diameter) fall onto the wing surface, they can bounce on the surface without sticking to it, indicating the excellent super-hydrophobicity. Such anti-wetting surface may be able to inhibit microbial growth. A similar topographical configuration to the lotus surface architecture can be seen in Fig. 8c, the planthopper wing surface is also covered with randomly arranged small papillae, which were proved to impair bacterial function [37].

In conclusion, many natural creatures have multifarious surfaces with super-hydrophobic, antifouling or antibacterial properties. The surface anti-wetting property plays key roles in dictating mechanisms used by these creatures to interact with their environment. For a bio-inspired super-hydrophobic surface, the air layer trapped within the distinctive rough structure of the anti-wetting surface may help to keep the surface free from bacteria attachment at the early stage and thus inhibit the growth of many micro-organisms.

Moreover, the morphology of a super-hydrophobic surface limits its contact with solid materials, and also facilitates the removal of interactions with a range of bacterial fouling. The micro–nano-engineered surface structure has contributed to bactericidal properties, although mechanical puncture is normally believed to cause bacterial cell stretching and rupture, cell motion shear force and capillary action are also responsible for the surface's bactericidal properties. Inspired by the extraordinary superhydrophobic and antibacterial surfaces from natural species, superhydrophobic cellulose fabrics that could prevent the attachment and growth of bacteria have been developed. Mimicking the natural surfaces, specifically designed surface topography, such as nanopillars, nanotubes, micro-/nano-spikes, may cause mechanical stress on the adhered bacteria and organisms, resulting in cell membrane failure and cell death [39, 48, 88, 89].

Super-Hydrophobic Antibacterial Cellulose Fabrics

Super-hydrophobic surfaces with a water contact angle (WCA) over 150° and a sliding angle (SA) less than 10˚ have received extensive interest since the elucidation of the "lotus effect" on lotus leaves by Barthlott and Neinhuis in 1997 [50, 90]. Inspired by the extraordinary special wettability surfaces of natural examples, considerable efforts have been made to develop artificial super-hydrophobic surfaces and identify the mechanism between surface structure and special wettability. The fabricated super-hydrophobic surfaces showed high potential in diverse applications including self-cleaning [91–93], anti-bacterial [94], anti-corrosive [95, 96], anti-fogging [97], anti-icing [98, 99], water harvesting [100], drag reduction [101–103], water/oil separation [104–106], and wound dressing. [92, 107] In the last decade, bacteria and biofilm contamination has attracted great attention because of cross-infections by pathogens, foul smell, discoloration of contaminated cellulose-based textiles [27, 108], and hard dislodgement of the formed biofilm on the textiles. Since cellulose fiber materials are closely related to our daily lives, especially for fabric and paper applications, once they are contaminated by bacteria, they may directly or indirectly spread pathogenic microorganisms to humans, causing health problems. The antibiosis mechanisms of fabrics with antibacterial properties have been intensively studied [6, 22, 30]. However, the improper and excessive application of bactericide may cause bacteria to develop resistance to antibiotics. Therefore, development of anti-microbial cellulose fabrics based on special non-wetting properties has attracted considerable research interests, which aim to impart cellulose fabrics with the self-cleaning property, enhance their antibacterial efficiency, and effectively reduce the possibility of developing resistance to antibiotics, thus

resulting in the greatly extended product service life and a healthy environment.

Super-Hydrophobic Fabrics

The fabrication of super-hydrophobic textiles has primarily relied on the employment of coating comprising micro-nanoparticles and low surface energy agents. The ubiquitously dispersed micro–nano-particles on the fiber surface can not only improve the surface roughness but also easily impart fibers with special functionalities. Many chemical materials have been employed to reduce the surface energy of cellulose fabrics, such as 1H,1H,2H, 2H-perfluorododecyl-1-thiol (PFDT), polytetrafluoroethylene (PTFE), fluorinated polyhedral oligomeric silsesquioxane (F-POSS), poly(vinylidene fluoride-co-hexafluoropropylene) (PVDF-HFP), FAS, fluorinated polyurethane (FPU), fluoroalkylsilane (1H,1H,2H,2H-perfluorooctyltriethoxysilane) (POTS), PDMS, octadecylamine (ODA), methyltrimethoxysilane (MTMS), and hexadecyltrimethoxysilane (HDTMS) [102, 109–122]. Polymerization, wet chemical coating, sol–gel, layer-by-layer method, chemical vapor deposition, etc. have generally been employed to reduce the surface energy of cellulose fibers.

Polymerization

By virtue of the hydroxyl groups present, cellulose polymers can partially or fully react with many functional groups. Polymerization is commonly used to form covalent bonds between cellulose fibers and low surface energy molecule chains to endow the surfaces with durable super-hydrophobic properties. This method involves grafting polymerization [123], atom transfer radical polymerization (ATRP) [115], plasma-induced polymerization [124], solution polymerization [102], etc. Deng et al. [123] grafted 1H, 1H, 2H, 2H-nonafluorohexyl-1-acrylate onto cellulose fabrics by the radiation-induced polymerization method. This imparted the cotton fabrics with superhydrophobicity (WCA > 160°), excellent chemical stability over the entire pH range, and high durability of over 250 times of home laundry. Xu et al. [125] synthesized poly[(methyl methacrylate)-b-(trifluoroethyl methacrylate)] (PMMA-b-PTFMA) copolymer by the polymerization and atomization method as shown in Fig. 9a. The obtained copolymer was then dissolved and atomized onto cotton fabrics; the PMMA blocks in the copolymers were covalently linked to cotton molecules through a transesterification reaction, while the PTFMA segments endowed fabrics with low surface energy. In addition, the copolymer coating has been found to efficiently increase the surface roughness, which is attributed to its self-assembly feature. As a result, the coated cotton fabrics showed self-healing ability and high stability against
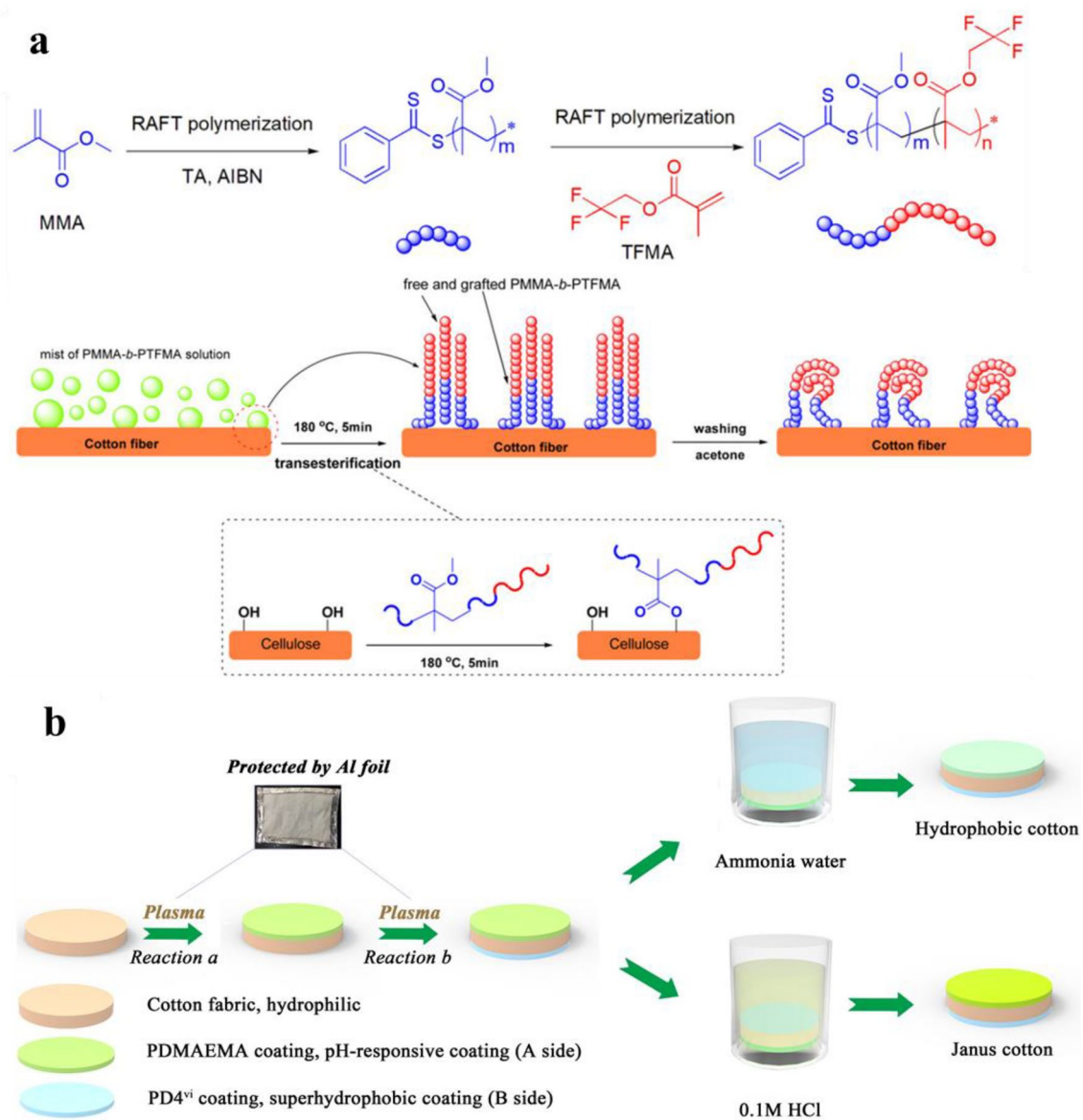
Fig. 9 a Synthesis of PMMA-b-PTFMA and schematic illustration of surface modification of cellulose fibers. Reproduced with permission from ref [125]. Copyright 2020 Elsevier. **b** The preparation of pH-responsive Janus cellulose fabrics. Reproduced with permission from ref [124]. Copyright 2017 Elsevier

60 washes and 2,000 mechanical abrasions. The developed atomization coating method in this work can avoid the influence of long-term solution immersion on the softness, air permeability, and abrasion resistance of the coated fabrics, and also reduce the amount of unreacted monomers. Li et al. [115] prepared superhydrophobic cellulose fibers by grafting methacryl POSS (MA-POSS) and PFDT onto cotton through ATRP and thiol-Michael addition reaction followed with fluorination reaction. The coated cellulose fabrics exhibited a WCA of 160°, a SA of 10° and self-healing ability against plasma etching through heat treatment. Long-chain fluorine-containing precursors in the coating process have

excessive environmental stability, bioaccumulation, and may cause environmental and human health issues. Therefore, their application has been prohibited in a growing number of countries, and non-fluorinated chemical agents have become more and more popular in preparing liquid-repellent surfaces. Yan et al. [124] prepared pH-responsive Janus coating on cellulose fabrics via plasma-induced polymerization of 1,3,5,7-tetravinyl-1,3,5,7-tetramethylcyclotetrasiloxane, and the treated cotton fabric exhibited Janus super-wettability in acid condition and hydrophobicity in alkali condition (Fig. 9b). Fu et al. [102] grafted polyaniline (PANI) nanofibers onto cotton fibers by solution polymerization to obtain

Fabric@PANI, which formed super-hydrophilic fabric@PANI@Ag after in situ oxidation. After modification with octadecylthiol (thiol), the fabric turned super-hydrophobic with a WCA of 160° and a SA less than 10°.

The polymerization method allows grafting of low surface energy polymer molecules onto the surface of substrates, thereby improving the durability of the treated surface against physical and chemical damages such as repeated washing and physical abrasion. However, the polymerization process involves tedious reactions and prolonged solution polymerization process, and might also affect the softness, breathability, and wear ability of the treated cellulose fabrics.

Wet chemical Coating

Wet chemical coating applies a coating suspension onto fabric substrates through different coating methods, such as dip-coating, [122, 126] spray-coating, [127] electrospray-coating [128, 129], and spin-coating [130, 131], followed by a drying and/or curing process. The super-hydrophobic coating suspensions normally consist of micro-/nano-particles that can increase the surface roughness and fluorocarbons or siloxanes to reduce the surface free energy. Generally, polymeric binders are applied to enhance the coating durability. Lu et al. [132] prepared an ethanolic suspension containing perfluorooctyltriethoxysilane-coated dual-scale TiO_2 nanoparticles (NPs), which can be applied onto various substrates, e.g., cotton fabric, cellulose paper, metal or glass, by either spraying, dip-coating, or painting method. The coated surfaces remained super-hydrophobic even after being contaminated with oil.

Nowadays, for the concern of environment protection and health, aqueous coating systems have become prevalent for fabricating super-hydrophobic surfaces. However, the uniform and stable dispersion of low free energy agents in the aqueous solution together with the low durability of coating are the two major challenges for waterborne coating systems. Zhou et al. [126] developed a ternary dispersion system with hydrophobic nanoparticles, 1H,1H,2H,2H-perfluorodecyltrimethoxysilane (FAS), and fluorinated copolymers uniformly dispersed in an aqueous solution, which were successfully applied to fabricate water-based superamphiphobic coating on a variety of substrates, including fabrics, sponge, wood, glass, and metal through the dip-coating method as shown in Fig. 10a and b. The coating was durable and could not only withstand various severe actions and environments, such as physical abrasion, washing, boiling water, and strong acid/alkali, but also exhibited self-healing property to chemical and physical damages. Ren et al. [117] designed a colorful superhydrophobic coating adapted to both hard and soft substrates. The coating solution was comprised of a certain proportion of cellulose, chitosan, zeolite, polytetrafluoroethylene (PTFE), three primary color dyes (methyl blue, methyl orange, Rhodamine), FAS, and [3-(trimethoxy silyl) propyl] ethylenediamine (AS). The coating solution can be applied onto substrates by spraying, dip-coating or painting method to prepare colorful coatings, which exhibited excellent durability and can withstand sandpaper abrasion, tape-peeling, salt spray test, and UV irradiation due to the formation of siloxane covalent bonds and hydrogen bonds between the coating and substrates (Fig. 10c).

Sol–Gel

Sol–gel method is a representative and simple technology to synthesize a nanoscale material from sol to gel through gelation including hydrolysis, condensation, molding, and drying. It has the ability to control the particle size and morphology through systematic monitoring of reaction parameters, and is often used in conjunction with the coating method to fabricate superhydrophobic surfaces. Lin et al. [133] reported a superhydrophobic and flame-resistant cotton fabric prepared by the one-pot sol–gel method (Fig. 11a). The fabric was first treated by O_2 plasma and then dip-coated in the solution containing tetraethoxysilane (TEOS), hydroxyl-terminated polydimethylsiloxane (HPDMS), and ammonium polyphosphate (APP). With the addition of ammonia, polydimethylsiloxane–silica hybrid (PDMS–silica) was generated via in situ sol–gel reaction of TEOS and HPDMS. The coated cotton fabric showed durable superhydrophobicity with a WCA of 160° and excellent flame resistance by virtue of the dense intumescent PDMS–silica layer. Qi et al. [120] prepared superhydrophobic flame-retardant cellulose fabrics by a sol–gel process and the brush-coating technique using P, P-diphenyl-N-(3-(triethoxysilyl)propyl) phospinic amide (DPTES) and polydimethylsiloxane@silicon dioxide ($PDMS@SiO_2$) as the coating materials (Fig. 11b). The coated cotton fabric had a WCA of 154° and a SA of 8°, and exhibited not only anti-washing and self-cleaning ability and superior friction resistance, but also a self-extinguishing feature.

Layer-by-Layer Assembly

The layer-by-layer (LbL) assembly technique is capable of fabricating thin films with tailored composition and properties on various substrates, which relies on alternating sequential adsorption of polycations and polyanions onto a charged surface. However, the substrates need to be dipped cyclically in different solutions to get a coating with desirable thickness and the LbL coating usually has low stability and is vulnerable to chemical and physical damages. Zhao et al. [134, 135] alternately deposited HMBS
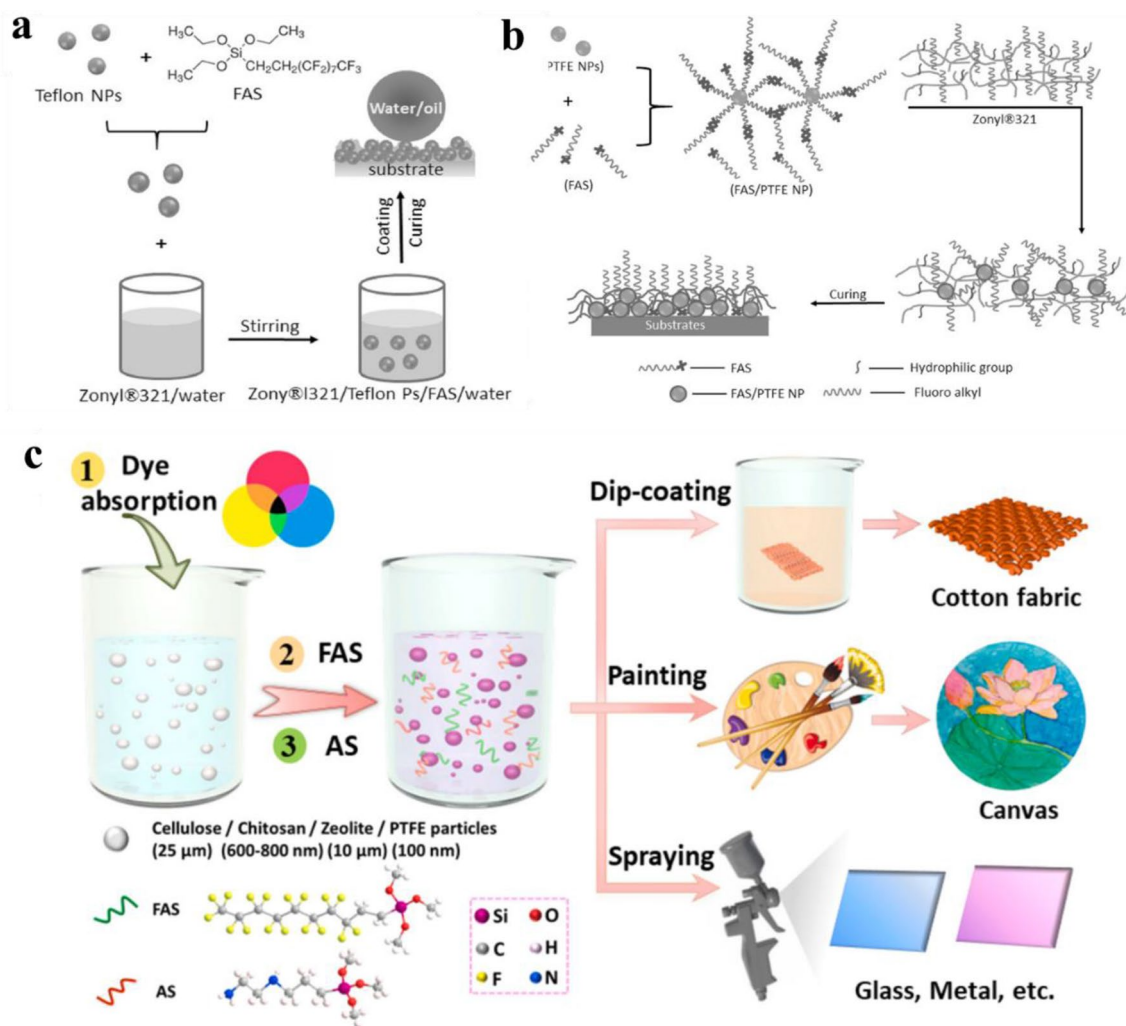
Fig. 10 a Schematic illustration of the coating treatment and **b** A possible interaction among PTFE NPs, FAS, and Zonyl321. Reproduced with permission from ref [126]. Copyright 2017 Elsevier. **c** The fabrication process of the multicolored superhydrophobic coating. Reproduced with permission from ref [117]. Copyright 2021 Elsevier

(2-hydroxy-4-methoxybenzophenone-5-sulfonic acid) and LDH (layered double hydroxide) onto the cotton surface via layer-by-layer (LbL) method to fabricate multi-functional cellulose fabrics showing both superhydrophobic and UV-blocking properties. Herein, the LDH nanoplatelets provided hierarchical surface roughness while the application of fluoroalkylsilane contributed to low surface free energy of coating (Fig. 12a). Li et al. [136] alternately immersed cellulose fabrics in poly(allylamine hydrochloride) (PAH) aqueous solution and $(NH_4)_2PdCl_4$ aqueous solution to obtain LbL self-assembled $(PAH/(NH_4)_2PdCl_4)*n$ film-coated cellulose fabrics. Then Cu and fluoride were deposited on the self-assembled surface to prepare superhydrophobic cellulose fibers with electrical conductivity and self-healing properties (Fig. 12b) [136].

Chemical Vapor Deposition

Chemical vapor deposition (CVD) is to deposit coating materials onto substrate surfaces in the vapor phase under a controlled chemical reaction. This process allows the precise control of coating thickness on substrates at the nanoscale without the use of solvents and can avoid complex multi-step processes [96, 137]. It is suitable for the uniform coating of substrates of complex shapes or surface with nanoscale features. Cheng et al. [138] fabricated superhydrophobic cotton by the enzymatic etching and CVD methods (Fig. 12c). The enzymatic etching process increased the fiber surface roughness, while the grafted methyltrichlorosilane (MTCS) containing hydrophobic groups efficiently reduced the surface energy. The obtained fabrics exhibited a WCA of 156.7° and a SA of 8.5°. Song et al. [137] synthesized antibacterial

Fig. 11 **a** Schematic illustration of the one-pot sol–gel method to obtain superhydrophobic cotton. Reproduced with permission from ref [133]. Copyright 2018 Academic Press Inc. **b** Schematic illustration of the superhydrophobic and flame-retardant coating on cellulose fabrics. Reproduced with permission from ref [120]. Copyright 2022 Academic Press Inc
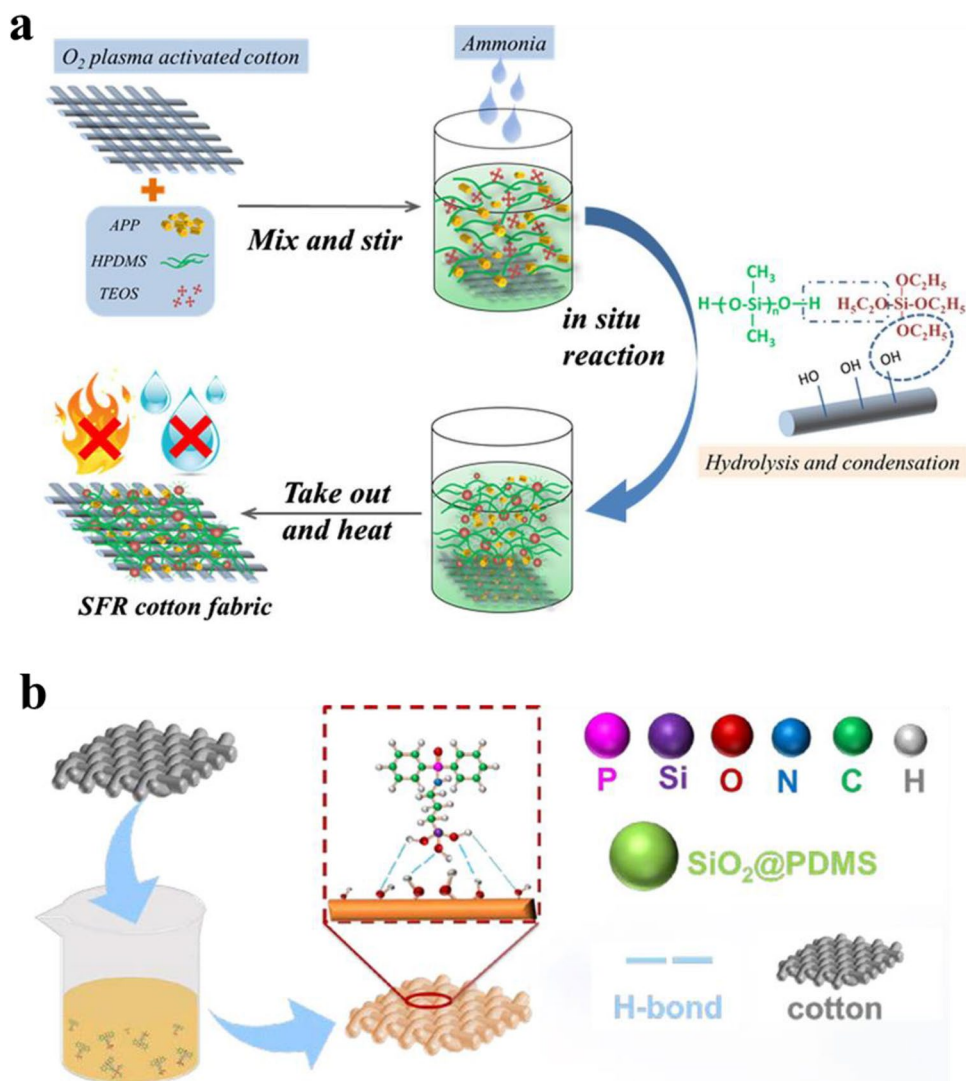
fabrics via CVD with fluorinated poly-cationic coating. The hydrophobic poly(dimethylaminomethylstyrene-co-1H,1H,2H,2H-perfluorodecyl acrylate) (P(DMAMS-co-PFDA), PDP) coating composed of fluorine-containing PFDA and DMAMS was prepared by CVD method. The coating can render the fabrics with super-liquid-repellency with antifouling capability against blood, paraffin oil, honey, and milk. The CVD method does not require expensive equipment, high reaction temperatures, and organic solvents, and is suitable for substrates requiring fine modification of nanoscale pores and uniform coatings.

Table 1 summarizes the strategies for developing superhydrophobic cellulose fabrics, their performances and applications reported in the last decade. So far, the reported works on superhydrophobic modification of cellulose fabrics have mainly focused on cotton fibers. With the fast growing market for regenerated cellulose fibers, it is now imperative to address the super-hydrophobic modification of regenerated

cellulose fabrics (e.g., Viscose, Modal, and Lyocell). Some pioneering works have already been conducted [139–141].

Super-Hydrophobic Anti-biofouling Surfaces

Biofouling is closely related to the wettability of solid–water–biomass systems. Superhydrophobic lotus leaves with special surface roughness structures are self-cleaning and anti-biofouling. Inspired by the lotus leaf effect, Marmur et al. [162] proposed that superhydrophobic surfaces can guide the development of anti-biofouling surfaces in two different directions. One strategy is to develop a "mirror image" of the lotus leaf surface structure. The mirrored super-hydrophilic surface has higher affinity for water than for biological entities, and water can fully penetrate into the rough grooves. While the size of the rough structure is smaller than the biological entity, which prevents the biological entity from settling in the rough groove. The other

Fig. 12 **a** Schematic illustration of fabrication process of HMBS/LDH bilayer coated cotton. Reproduced with permission from ref [135]. Copyright 2013 Elsevier. **b** Fabrication procedure of the conductive and self-healing superhydrophobic cotton fabric, and the chemical structures of POTS and F-POSS. Reproduced with permission from ref [136]. Copyright 2018 American Chemistry Society. **c** Schematic illustration of the process of constructing fluorine-free superhydrophobic coating on fabric surface by enzymatic etching and CVD. Reproduced with permission from ref [138]. Copyright 2019 Elsevier
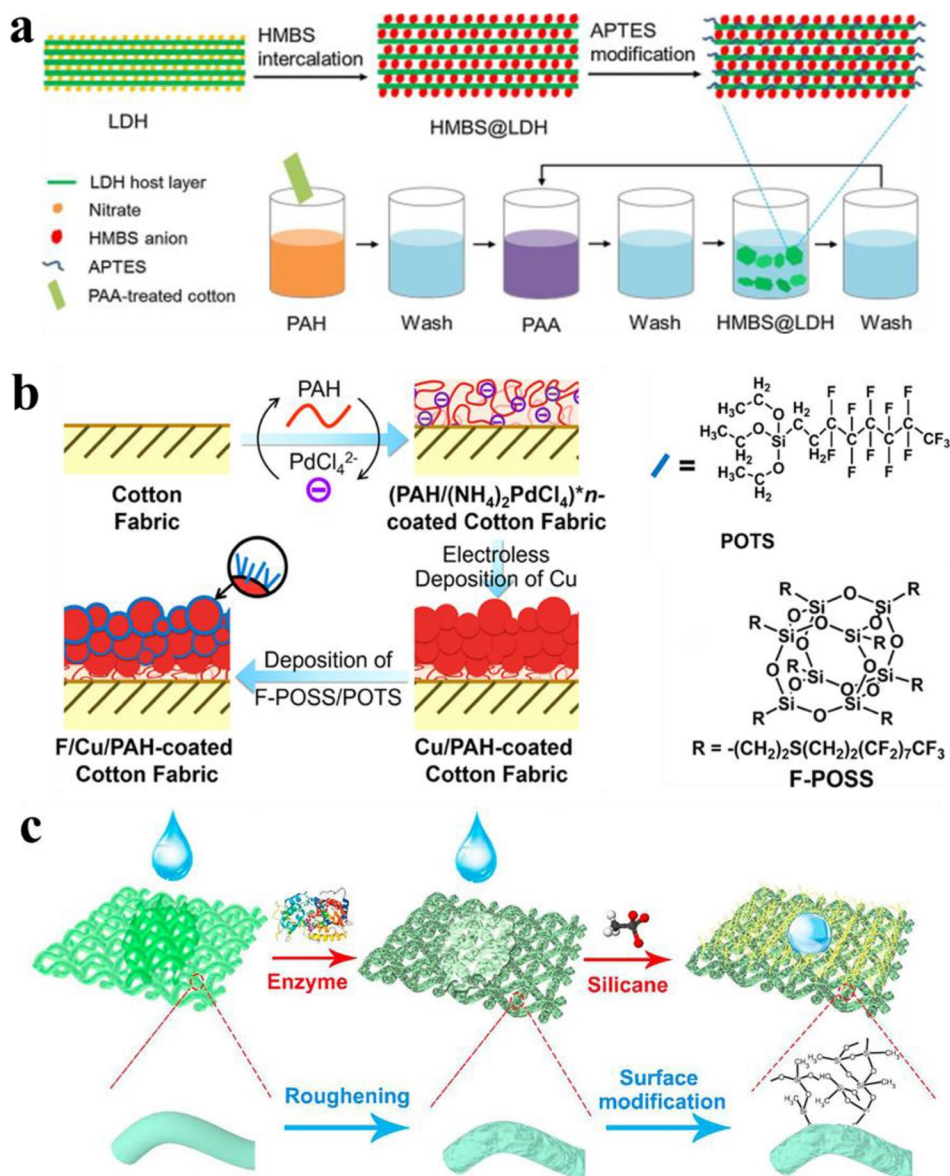
strategy is to create underwater super-hydrophobicity, which can form a plastron layer when immersed in water, and this air layer then reduces the probability of biological entities touching the solid surface. Meng et al. [163] systematically explored the effect of surfaces with wetting gradients ranging from super-hydrophilicity to super-hydrophobicity on cell adhesion (Fig. 13a); NIH/3T3 mouse embryonic fibroblast cell was applied as the model cell line, and the results showed that higher cells adhesion could be observed in the superhydrophilic and the fully wetted hydrophobic substrates with hierarchical surface roughness. However, the number of adhered cells decreased sharply on smooth surfaces or superhydrophobic substrates having enhanced surface roughness. The former was owing to the limited contact area derived from the lack of the effectively nano-topographic interaction between the smooth surface and the nanoscaled surface of

cells, the latter was probably because the trapped air layer in the superhydrophobic surface can efficiently prevent cell adhesion as seen in Fig. 13b and c. Those results are consistent with the hypothesis by Marmur et al. [162] However, the antibiofouling properties of superhydrophobic surfaces are transient due to the instability of the plastron. Therefore, it is essential to impart bactericidal properties to superhydrophobic surfaces with prolonged antibacterial properties.

Antibacterial agents can control the growth of microbes based on various mechanisms, including inhibition of cell reproduction, blocking of enzymes, reaction with cell membranes for their disruption, and poisoning the cells from within [29]. There are generally three ways to achieve antibacterial effect on fabrics with regard to superhydrophobic modification: (1) Designing superhydrophobic surfaces to reduce bacteria adhesion and breeding

Table 1 Fabrication strategies of super-hydrophobic cellulose fabrics

| Coating materials	Preparation methods	WCA	Durability				Application	References
			Laundering (cycles)	Abrasion (cycles)	Organic solvents(h)	Acidic/alkaline(h)		
PGMA, PTFEMA	ATRP	163°	40	40 (3.9 kPa, 5 cm s^{-1})	60 (>150°)	60 (>150°)	Self-cleaning, waterproof and air-breathable textiles, water–oil separation	[142]
1H,1H,2H,2H-nonafluorohexyl-1-acrylate	Polymerization	160°	250 (AATCC 61–2006)	–	–	–	–	[123]
TEOS, PDA, SiO$_2$	Copolymerization reaction	>150°	–	50 (200 g loading, 800 mesh sandpaper, >150°)	7 days (>150°)	–	Water–oil separation	[143]
MPTES, 8-MAPOSS, FMA	Polymerization	~161°	24 (ISO 105-C10:2006C)	160 (100 g loading, 1,000 mesh sandpaper, >150°)	–	12 (>150°)	Antifouling, self-cleaning, dehydration resistance, buoyancy boost, drag reduction performance	[144]
Coral-reef like silica particles, OTES	Dip-coating	167°	–	50 (100 g loading, slight change of WCA), finger wiping, 50 cycles of bending/unbending, tap peel off test, neither of above affected the super-hydrophobicity	2 h in DMF, THF, hexane, chloroform and ethyl acetate (>163°)	24 h in 3.5% NaCl (>160°)	Self-cleaning, anti-mycotic, anti-fouling	[145]
PMMA, PTFMA	Polymerization	160°	60 (143°)	2000 (>145°)	–	–	–	[125]
PFDT, FSPU	Thiol-ene click chemistry	158.2°,	30 (~150°)	600 (150.2°)	–	–	Oil–water separation	[146]
FCMFO NPs, VPDMS	UV light irradiation	157.1°	1 (153.2°)	80 (150.3°)	24 (>154°)	24 (>152°)	Oil–water separation, self-cleaning	[147]
Diamond-like carbon	CVD	169.3 ± 2.2°	20	–	–	72 (166–172°)	Water–oil separation	[148]
PANI, PTES	CVD	156°	–	600 (260 g, 4 cm s^{-1})	–	72 (143–156°)	Water–oil separation	[149]
F-POSS, POTS, (NH$_4$)$_2$PdCl$_4$	CVD	>150°	–	–	–	100 (>150°)	Self-cleaning, electromagnetic interference shielding	[136]
TiO$_2$, PTES, POTS	One-pot hydrothermal reaction	160°	5	30 (GB/T 3920-2008)	–	–	UV shielding, self-cleaning, water–oil separation	[150]
PDMS, ZnO	Enhanced hydrothermal technique	>170°,	–	–	–	–	Anti-icing performance	[151]

Table 1 (continued)

Coating materials	Preparation methods	WCA	Durability				Application	References
			Laundering (cycles)	Abrasion (cycles)	Organic solvents(h)	Acidic/alkaline(h)		
PDMS, organically modified silica aerogel (ormosil)	Sol–gel method, dip-coating	>160°	5 (AATCC 61–2006)	100 (3.5 kPa, 3 cm s^{-1})	–	24 (144°)	Self-cleaning, water–oil separation	[152]
bPEI, APP, F-POSS	Dip-coating, deposition	160°	–	1000 (44.8 kPa, 30 cm s^{-1})	–	–	Flame-resistant, waterproof, and self-cleaning, self-healing	[153]
POTS, SiO$_2$ NPs, PVDF	Dip-coating	158°	30	10 (200 g, 2 cm s^{-1}, 100 mm, >150°)	–	12 (>150)	Self-cleaning, self-healing	[154]
APP/PER, SiO$_2$ NPs, PDMS, HMDS	Dip-coating	160°	–	500 (>150°)	–	–	Organic solvent/oil and water separation	[155]
PDMS, silica NPs	Dip-coating	161°	40 (150.4°)	130 (sandpaper, 148.2°)	–	24 (>150°)	Oil–water separation, self-Cleaning	[156]
SiO$_2$ NPs, HDTMS	Dip-coating	160°	40 (>130°)	40 (>120°)	–	48 (>150°)	Wearable electronic devices	[155]
Ag NPs, PFDT, poly-dopamine	Dip-coating	160°	–	–	–	–	E-textile	[113]
A-POSS, PA, PDMS, TiO$_2$	Spray-coating	>160°	5 (>160°)	50 (>139°)	–	–	Oil–water separation	[157]
PDA, rGO, STA, Cu NPs	Dipping and in situ growth method	153°	2 (ultrasonic, 145–155°)	–	–	24 (140–150°)	Sensor for underwater intelligent devices	[158]
OTMS, TCPP, TiO$_2$	Step-wise deposition	156°	10 (AATCC 190–2003)	–	–	–	Photocatalytic self-cleaning	[159]
Polystyrene NPs, AP, PDMS	LbL self-assembly	>150°	35 (150.7°)	750 (151°); 2000 (>130°)	72 (160°)	72 (>155°)	Self-cleaning	[160]
PA, FeCl$_3$, PDMS	LbL self-assembly, dip-coating	152.9° ± 1.8°	–	30 (100 g, 600 mesh sandpaper, 142.5° ± 2.5°)	7 days (>150°)	–	Water–oil separation	[161]

PGMA poly(glycidyl methacrylate), *PTFEMA* poly(2,2,2-trifluoroethyl methacrylate), *PTES* 1H,1H,2H,2H-per-Fuorodecyltriethoxysilane, *bPEI* branched poly(ethylenimine), *APP* ammonium polyphosphate, *FAS* fluorinated alkyl silane, *DTMS* dodecyltrimethoxysilane, *OTMS* trimethoxy(octadecyl)silane, *TCPP* meso-tetra(4-carboxyphenyl)porphyrin, *PDA* polydopamine, *MPTES* 3-Mercaptopropyltriethoxysilane, *MAPOSS* Methacryl polyhedral oligomeric silsesquioxane, *FMA* 2-(perfluorooctyl) ethyl methacrylate, *PVDF* vinylidene fluoride-co-hexafluoropropylene, *AEAPS* 2-aminoethylaminopropyl trimethoxysilane, *PTFMA* trifluoroethyl methacrylate, *HMDS* hexadecyltrimethoxysilane, *STA* stearic acid, *rGo* reduced graphene oxide, *AP* aluminum phosphate, *FCMFO* Co$_{0.2}$Mg$_{0.8}$Fe$_2$O$_4$, *VPDMS* vinyl-containing poly(dimethylsiloxane), *FSPU* fluorosilicone polyurethane
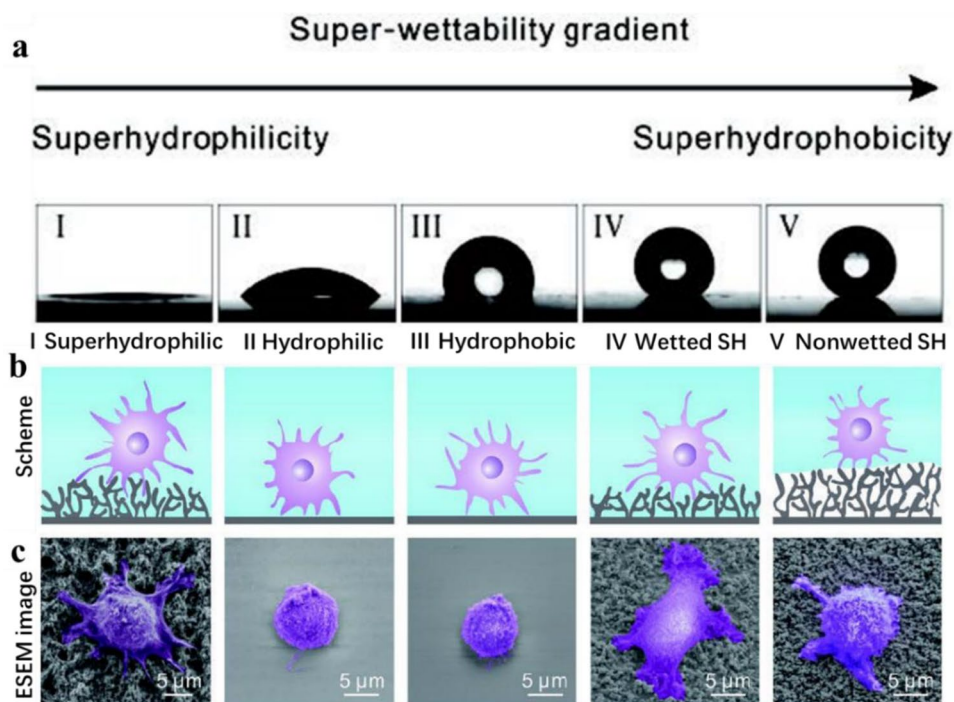
[164–166]. (2) Applying bactericidal materials including metal-based antibacterial agents (e.g., Au, Ag, TiO_2, ZnO, CuO, metal–organic framework (MOF), and imidazolate framework) [28, 88, 167–176], organic chemicals (e.g., guanidine-based polymer, quaternary ammonium salt cationic compounds, peptides, zwitterionic betaine compounds, bacteriophages, and abietanes) [177–180], and natural bactericidal substances (e.g., chitosan, cyclodextrin, sericin, alginate) [181–183]. (3) The combination of antimicrobial agents and superhydrophobic modification. In general, antibacterial agents can be classified into two types, leaching and non-leaching. Leaching agents work under the regulated release mechanism via leaching on the fabric surface or at the interior of fibers, which can efficiently repel microbes bound at the fiber surface or in the surrounding environment until the antimicrobial agents are fully released. Non-leaching antimicrobial agents bind to the fiber surfaces and can only have an effect on the microbes contacting the fiber surface.

Superhydrophobic Fabrics with Antibacterial Properties

It has been proved that a superhydrophobic surface based on the combination of low surface free energy and nano-/micro- surface structure can reduce the adhesion force between bacteria and surface [164]. However, due the interaction between bacterial and superhydrophobic surfaces and gradual surface contamination, it is impossible for fabrics to indefinitely prohibit bacterial attachment. It is imperative to impart cellulose fabrics with superhydrophobicity and antibacterial function simultaneously for a prolonged antibacterial effect. Superhydrophobic antibacterial cellulose fabrics can be prepared by one-step fabrication strategies, e.g., dip-coating [184–188], copolymerization [26], spray-coating,[189] and multi-step fabrication strategies, e.g., in situ growth/self-assembly/dip-coating [190], free radical polymerization/spraying coating [191], heterogeneous transesterification/dip-coating [192], dip-coating/sequential deposition [193], dual-stage silanization [194], polymerization/dip-coating [195], dip-coating/thermal evaporation [196], and dip-coating/in situ growing [28]. The materials applied and fabrication strategies for developing super-hydrophobic antibacterial cellulose fabrics have been summarized in Table 2.

Figure 14 shows the mechanism of antibacterial super-hydrophobic surfaces, which includes anti-biofouling, bactericidal, and easy removal of the dead or alive biofilm attached [45]. In recent years, many efforts have been made to prepare antibacterial super-hydrophobic surfaces. Metal elements (e.g., silver, copper, zinc, titanium, gold and their metallic oxides), quaternary ammonium compounds, and *N*-halamines are the three main types of antibacterial additives in the super-hydrophobic field. The silver-related antibacterial agents have been applied in textile industry for a long time. The intensity order of killing and inhibiting bacteria is listed as: Ag > Hg > Cu > Cd > Cr > Ni > Pb > Co > Au > Zn > Fe > Mn > Mo > Sn [197–200]. The quaternary ammonium compounds have potent antibacterial capability

Table 2 Fabrication strategies, superhydrophobic and antibacterial properties of cellulose fabrics

Material	Fabrication strategy	WCA (°)	Bacteria	Test method	Durability	Other properties	References
AgNO$_3$, gum arabic, ascorbic acid (C$_6$H$_8$O$_6$), OTES	In situ growth, self-assembly, dip-coating,	151°	S. aureus, E. coli	Disk diffusion	–	Self-cleaning	[190]
HDTMS	Dip-coating	>150°	E. coli	Inhabitation zone	1 h washing in detergent solution, hot water, and benzene	Self-cleaning, stain-resistant	[184]
QAS functionalized fluorinated copolymers, PUF NPs	Free radical polymerization, spraying coating	162.3±3.2°	S. aureus, E. coli	Plate count	16 abrasion and 20 cross-cut tape cycles	Self-cleaning and liquid-repellent	[191]
Gen, ODA	Heterogeneous transesterification, dip-coating	~145°	S. aureus, E. coli	Disk diffusion, colony count	10 cycles of standard washing	–	[192]
PEI, Ag NPs, F-POSS	Dip-coating, sequential deposition	~169°	E. coli, B. subtilis	OD method	5000 abrasion cycles, 80 cycles of dry cleaning, 8 cycles of O$_2$-plasmaetching / healing process	Tunable color, self-healing, washing and abrasion durability	[193]
Ag nanowire, Danasylan® F 8815	Dip-coating	156.2°±3.2°	S. aureus, E. coli	AATCC 100–2004	–	UV protection	[185]
Styrene, PCL–HEMA, M-PHGC	Copolymerization	140.26°	S. aureus, E. coli	Colony count and OD	50 cycles of standard washing	Thermal stability	[26]
Copper acetate monohydrate, ascorbic acid, stearic acid	Dip-coating	159°	S. aureus, E. coli	Disk diffusion	10 washing cycles, all pH ranges	Self-cleaning	[186]
ZnO NPs, APTES, HDTMS	Dual-stage silanization approach	154°	S. aureus, E. coli	AATCC 100–2004	800 cycles of abrasion, 2 h ultrasonic, 24 h in all pH ranges	UV-blocking, durability against abrasion, ultra-sonic washing	[194]
AgNO$_3$, PDA, PDMS	Polymerization, dip-coating	154°	S. aureus, E. coli	Halo	8 h in acidic (pH=2) and alkaline (pH=12)	Microwave shielding, self-cleaning, acid–alkali corrosion resistance, electrical conductivity	[195]
SiO$_2$, nanoporous cellulose acetate, silver, PDMS	Dip-coating, thermal evaporation	161°	S. aureus, E. coli	Inhibition zone	–	Infrared reflector, heater	[196]
SiO$_2$ NPs, PDMS, ZIF-8	Dip-coating, in situ growing	151.36°	S. aureus, E. coli	AATCC100-2004	300 cycles of abrasion and 5 cycles of washing	Abrasion resistant, washing durability, oil–water separation	[174]
TiO$_2$ NPs, chitosan	Dip-coating	161°	S. aureus, E. coli	Plate count	96 h in NaCl and HCl solutions, 4 h of sonication	UV-block, antibacterial garment and masks	[187]
ZnO NPs, PVSQ	Dip-coating	158.93°	S. aureus, E. coli	AATCC100-2004	40 washing time	Durable against repeated washing, water-proof, UV shielding	[188]

Table 2 (continued)

Material	Fabrication strategy	WCA (°)	Bacteria	Test method	Durability	Other properties	References
TFOA, QDEMA, HDI, DMAEMA, HEMA, PUF NPs	Spray-coating	>150°	E. coli	Plate count	–	Self-cleaning, wettability conversion, brilliant durability	[189]
Ag NPs, HDTMS	Dip-coating	157.3°±1.6°	E. coli	Disk diffusion	10 times water washing	Conductive	[242]

OTES octyltriethoxysilane, QAS alkyl-dimethyl tertiary amine, Gen gentamicin, PCL-HEMA polycaprolactone-2-hydroxylethyl methacrylate, APTES aminopropyltriethoxysilane, TFOA Tridecafluorooctyl acrylate, HDI hexamethylene diisocyanate, HEMA 2-hydroxyethyl methacrylate, DMAEMA 2-(dimethylamino)ethylmethacrylate, PUF poly(urea–formaldehyde)

against Gram-positive, Gram-negative bacteria, fungi, and multidrug-resistant strains due to the positive charge on the N atom [201–204]. N-halamines are antimicrobial, being active against various bacteria, fungi and viruses [45, 201]. The design of antibacterial surfaces mainly relies on bio-adhesion resistance, contact killing of bacteria, and leaching of antibacterial agents as shown in Fig. 14. [45, 205, 206]

Adhesion Resistance

The adhesion and proliferation of bacteria on the fiber surface may lead to unpleasant odors, cross-infection, disease or fiber deterioration, and a series of health concerns [27]. Bacterial surface adhesion and colonization are one of the pressing problems in public health, and surfaces where colonies or biofilms form can lead to serious health and hygiene consequences. Unlike their natural cellulose counterparts like hemp [207, 208], bamboo [209] that inherently possess antibacterial properties (due to presence of lignin in hemp and bamboo), cotton fabrics do not have ability of preventing or killing bacteria and other microorganism, therefore it is imperative to develop antibacterial cotton fabrics for long-time wearing comfort, removal of odor, and health and hygiene applications.

It is vital to understand the interaction between bacteria and a substrate, and how bacteria actually adhere to the substrate materials. The first stage of bacterial adhesion is a rapidly reversible initial interaction process between the bacterial surface and the substrate. The second stage is an irreversible interaction between adhesion proteins on the bacterial surface and binding molecules on the substrate material (Fig. 15a). The factors affecting bacterial adhesion are complex, mainly including the surface characteristics of the Gram-positive or Gram-negative bacterium cell envelope, substrate roughness, charges, hydrophobicity, Lewis acid–base properties, and hydrogen bonding [205]. Reducing protein adsorption is indisputably the first step to prevent fiber surface from microorganism pollution. The super-hydrophobic modifications of fabric surfaces have been effective strategies to prevent attachment/adhesion of bacterial pathogens on the surfaces by significantly diminishing available anchor sites for bacterial adhesion, making it difficult for bacteria to adhere and colonize [6, 205, 210–212]. The air layer trapped in the super-hydrophobic surface can prevent protein from attaching in the initial stage because the protein cannot adhere to the gas–liquid interface [210, 213–215].

Huang et al. [213] applied green fluorescence intensity as the qualitative measure of amount of bound protein and observed that the green fluorescence contrast between super-hydrophobic and super-hydrophilic surfaces was obvious in the presence of entrapped air. They found almost no protein bound on the super-hydrophobic surface (Fig. 15 b, A and C
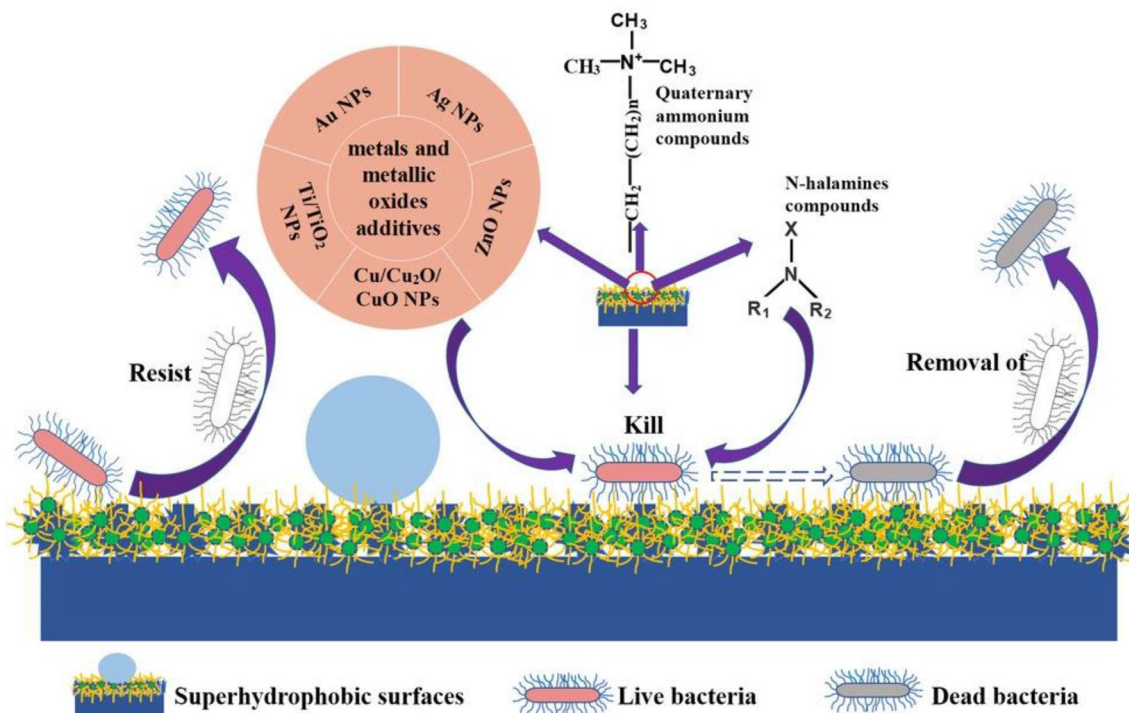
Fig. 14 The mechanism of antibacterial super-hydrophobic surface. Reproduced with permission from ref [45]. Copyright 2021 Wiley

net grid) and a large number of labeled proteins bounded on the super-hydrophilic surface (Fig. 15 b, A and C , squares). Interestingly, the fluorescence contrast between the super-hydrophobic and super-hydrophilic surfaces was slightly blurred after removal of the trapped air layer by sonication (Fig. 15 b, B and D), because the surface can absorb some protein after the removal of air layer [210, 213, 214]. Furthermore, Koke et al. [216, 217] found that protein binding was affected by the surface curvature by comparing protein adsorption on the super-hydrophobic surfaces with different roughness features, but it is still difficult to predict precisely which surface features are most effective in eliciting a particular cellular response.

The mechanism of adhesion and growth of bioactive components on super-hydrophobic surfaces is complex and influenced by the physicochemical properties of fabrics and bacterial surfaces. Super-hydrophobic surfaces can only decrease the bacterial adhesion to a limited extent because proteins are surface active molecules, and denatured protein can gradually adsorb onto the surface, thus increasing the surface energy of the material. This will eventually result in the air layer shift and surface moisture, thereby reducing the lifespan of the barrier [210]. Figure 15c shows that cells attached to the surface from the fetal bovine serum (FBS) solution transitioned from a hydrophilic surface to a hydrophobic surface and eventually covered the entire surface, and the trapped air cannot impede the transitional growth of cells after 4 days of culture [213]. In pursuit of long-term

and effective antibacterial effect, it is favorable to combine super-hydrophobic surfaces with antimicrobial agents [6, 176, 212, 218].

Contact Killing

The contact killing surfaces act on bacteria through direct surface contact by constructing cationic polymers, reactive oxygen species (ROS), and antimicrobial peptides [26, 122, 137, 187, 206, 218]. Most bacteria cells are negatively charged, so positively charged materials can easily interact with them and disrupt their cell structure leading to disintegration. The positively charged chitosan, guanidine-based polymer, poly(hexamethylene biguanide) hydrochloride, and quaternary ammonium salt cationic compounds have been widely employed in the antibacterial research [26, 218–220].

Han et al. [26] grafted environmentally friendly nanogels onto cotton fabrics, which improved both the hydrophobicity and anti-adhesion properties, and imparted cotton with durable antibacterial properties. The nanogels contain polyhexamethylene guanidine hydrochloride (M-PHGC) chains with long-lasting bacteriostatic activity, polystyrene with nanosphere features, and biodegradable hydrophobic polycaprolactone (PCL) chains (Fig. 16a). The antibacterial rate of nanogel-grafted cotton fabrics against *E. coli* and *S. aureus* was 91.30% and 97.23%, respectively. This was because the high hydrophobicity of fabric surface repelled bacteria at the initial stage. The negatively charged bacteria interacted with
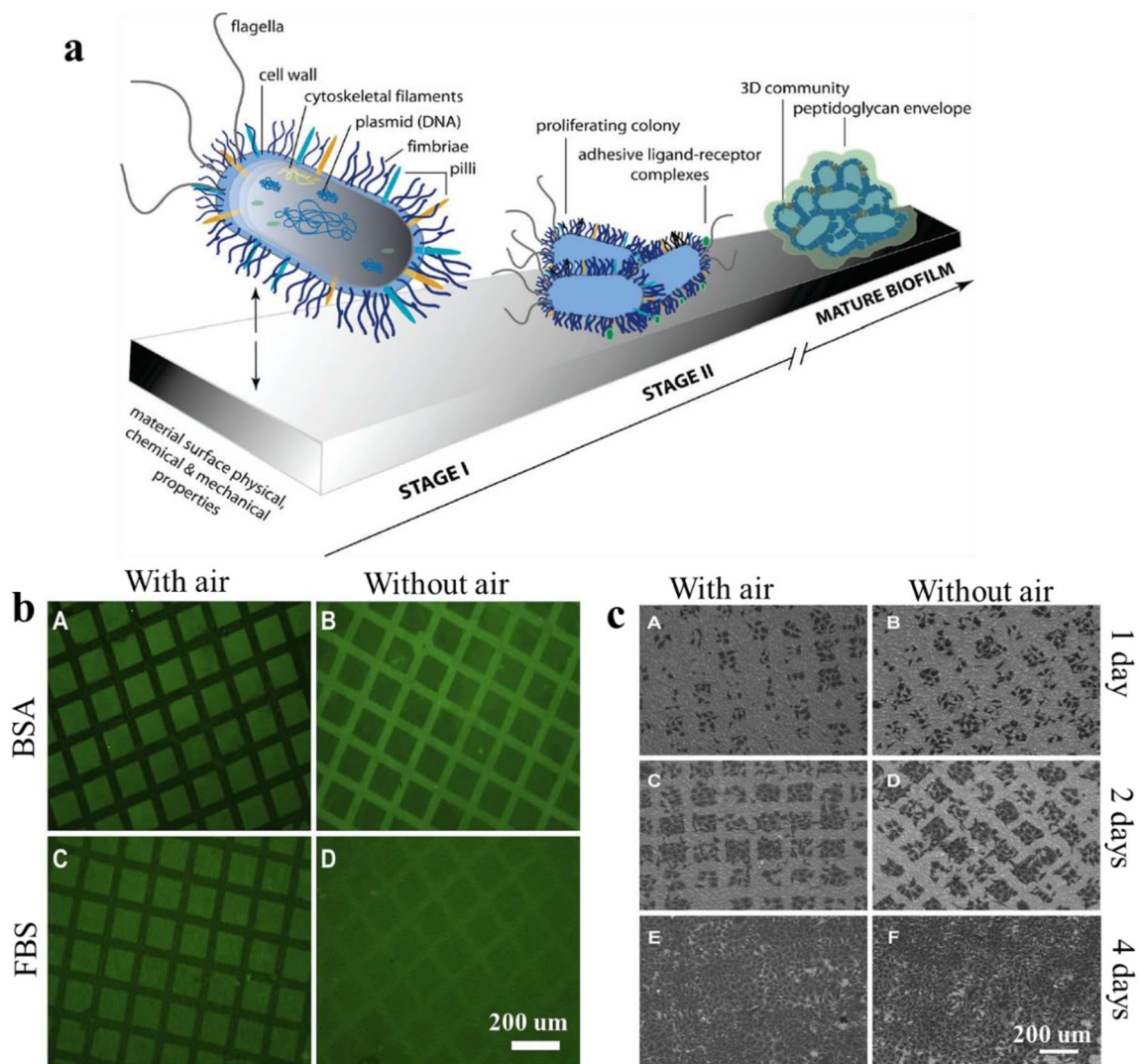
Fig. 15 **a** Two-stage bacterial adhesion model. Reproduced with permission from ref [205]. Copyright 2009 American Chemical Society. **b** Fluorescence microscopy images of fluorescein isothiocyanate (FITC-labeled proteins bound on the super-hydrophilic (squares)/super-hydrophobic (net grid) micro-templates. (**A** and **C** corresponded to FITC-labeled bovine serum albumin (BSA and FBS with air, respectively; **B** and D correspond to FITC-labeled BSA and FBS without air, respectively)). **c** SEM images of the number of HeLa cells attached to super-hydrophobic/super-hydrophilic micro-templates from FBS solution over time with and without air entrapment. Reproduced with permission from ref [213]. Copyright 2012 Elsevier

the positively charged PHGC chains and the bacterial structure was disrupted leading to the bacteria death. In addition, the hydration shell formed by the PHGC chains on the fabric surface can act as an effective barrier to prevent the surface from direct contact with dead bacteria (Fig. 16b) [26]. Ye et al. [218] fabricated double nano-roughness coated fabrics by immobilizing anti-adhesion fluorinated mesoporous silica nanoparticles(F-MSNs), contact killing quaternary ammonium-functionalized MSNs (Q-MSNs), and low surface energy PDMS onto the fibers as shown in Fig. 16c and d. The coated cotton fabric showed superhydrophobicity and strong ability to prevent protein adsorption, the antibacterial rate of F/Q-MSNs (4:2) cotton fabric was 99.9%. In addition

to the bacterial barrier function, the coated cotton fabric exhibited breathability, mechanical properties, and high coating durability as well.

N-halamine is a nitrogen–halogen bond containing compound with broad-spectrum activity against bacteria, fungi, and viruses. The oxidative N–Cl bond was reduced to an N–H bond when it came into contact with bacteria; the strong oxidizing property caused bacterial damage and death. The reduced N–H bond could be sterilized by a suitable chlorine bleaching process to convert the N–H bond back to N–Cl bond, showing recyclable antibacterial properties [221, 222]. Figure 17 shows biocidal mechanisms of *N*-halamines in three ways: the first is contact killing by
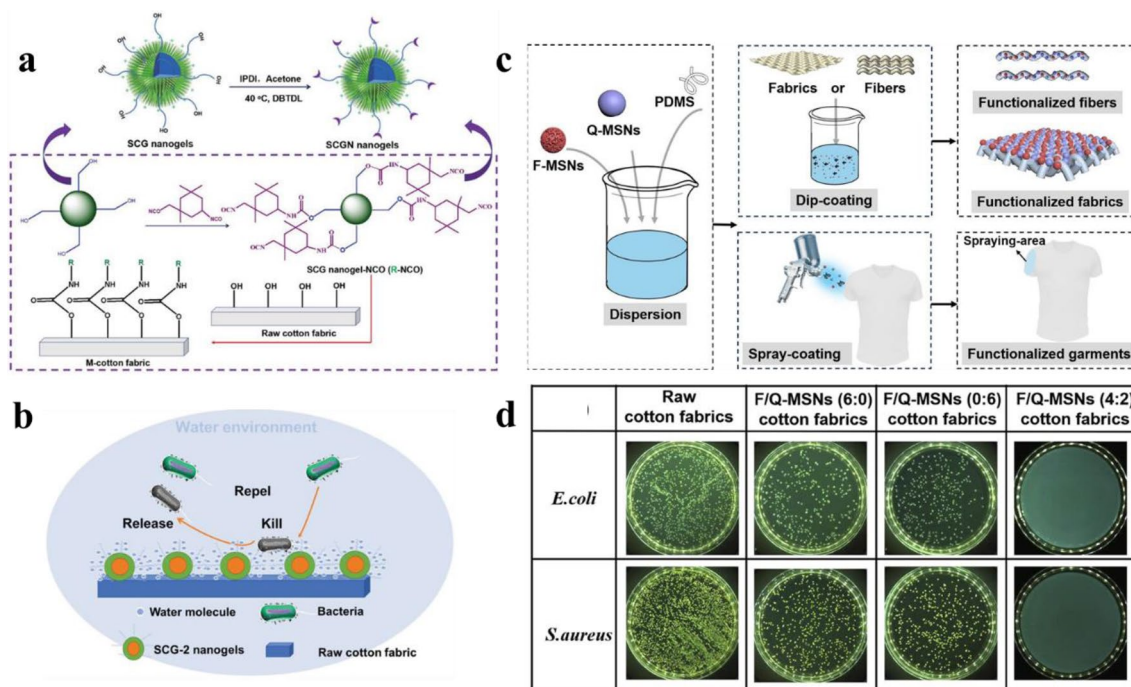
Fig. 16 **a** Grafting nanogels to the surface of cellulose fabrics. **b** Bacterially antiadhesive action model of M-cellulose fabrics. Reproduced with permission from ref [26]. Copyright 2019 Elsevier. **c** Schematic illustration of the configuring process of functionalized textiles. **d** Bacterial colonies on agar plates for different cellulose fabrics. Reproduced with permission from ref [218]. Copyright 2021 Elsevier
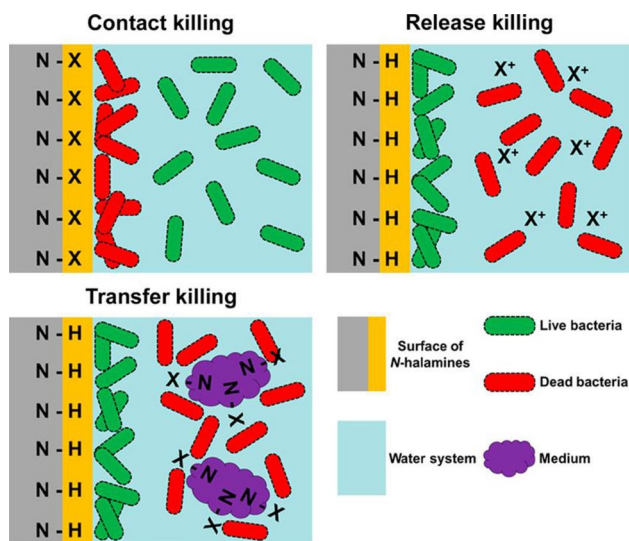
Fig. 17 Schematic illustration of three biocidal mechanisms of *N*-halamines [221]

direct transfer of positive halogen to bacteria; the second is release killing by releasing halogen to the solutions with subsequent inactivation; the third is transfer killing by transferring halogen to medium constituents, hence inhibiting bacterial growth and viability [221]. Among those biocidal mechanisms, contact killing and release killing play dominant roles.

Wang et al. [223] prepared silica nanoparticles by first treating silica with (3-(trimethoxysilyl) propyl methacrylate), and then grafting with an *N*-halamine precursor, 3-(3′-hydroxypropyl)-5,5-dimethylhydantoin, the prepared silica nanoparticles were applied onto the cotton fiber surfaces, the coated cotton showed high hydrophobicity and good antibacterial properties against Gram-positive bacteria, Gram-negative bacteria, and fungi. Sun et al. [224] synthesized hydantoin-based *N*-halamines using chlorinated poly(3–4′-vinylbenzyl)-5,5-dimethylhydantoin-*co*-vinyl acetate) and applied them onto the fabric via a coating process, as there was sufficient contact between the fabrics and bacteria, the *N*-halamines killed the bacteria by direct contact killing. Li et al. [225] further studied the contact killing mechanism of the *N*-halamines by designing a series of new biocides with both hydantoin *N*-halamines and quaternary ammonium moieties and demonstrated the transfer of *N*-halamines when they were in full contact with bacteria. Ahmed et al. [226] confirmed the contact killing ability of *N*-halamines by directly contacting the bacteria suspension with *N*-halamines in the absence of any media to mediate free halogen release, and found that the *E.coli* and *S.aureus* were effectively killed.

Photodynamic therapy (PDT) contact killing works based on light-induced reactive oxygen species, which

has also been widely used for the treatment of antibacterial fabrics [88, 122, 173, 227]. Graphene nanomaterials, ZnO, TiO$_2$, Ag NPs, MOF, and anthraquinone compounds are the commonly used antibacterial materials in PDT [88, 167, 170–173, 176]. Song et al. [88] reported a synergistic antibacterial strategy based on super-hydrophobic bacteriostatic and photodynamic sterilization, and found that surface super-hydrophobicity can significantly improve the bactericidal activity of PDT by reducing the number of surface-adhering bacteria. The super-hydrophobic cotton surface was fabricated by depositing silica nanoparticles, grafting hydrophobic photosensitizer Chlorin-e6 (Ce6), and dip-coating with perfluoroalkylsilane, which could effectively reduce bacterial adhesion. Meanwhile, the introduced photosensitive Ce6 damaged bacteria by virtue of high cytotoxic ROS generated under visible light irradiation, with bactericidal efficiencies of 93% and 100% against *E. coli* and *S. aureus*, respectively. Furthermore, the PDT bactericidal strategy mediated by cytotoxic ROS under visible light irradiation can avoid the risk of antimicrobial resistance.

Although cationic polymers, ROS and antimicrobial peptides are selected to explain the contact-killing phenomena here, we note that the contact-killing effect is hard to characterize experimentally and other mechanisms may take effect. For example, detection of bacteria as branched-polymer matrices, intercalation into bacterial external structures, and binding to surface proteins or modulating their activity through ion chelation show the ability of bacteria to extract lipopolysaccharides in various ways, ultimately causing cell death. Enzymatic degradation of cellular membrane components or disruption of eDNA as well as limitation of the nutrient reservoir may also contribute to cell mortality.

Biocide Leaching

The biocide leaching materials can slowly release biocides, including antibiotics, nanoparticles, nitrogen oxides, etc. and kill bacteria adhered to the material surface showing antibacterial effects. It should be noted that bacteria may gradually develop resistance. Therefore, the antibacterial effect will gradually decline with time during the application of biocide leaching agents [218]. In addition, the killed bacteria may adhere to the surface preventing further release of antimicrobial agents [211]. Therefore, biocide leaching surfaces need to be created with super-hydrophobic and self-cleaning properties to achieve long-term antibacterial behaviors.

Nanoparticles have been popular materials for the fabrication of super-hydrophobic antimicrobial cotton by virtue of their unique morphological and physicochemical properties, such as nanoscale zero-dimension structure, high surface area-to-volume ratio, especially excellent biocidal activities toward a range of microorganisms without developing resistant strains. Biocidal nanoparticles made of silver, copper

oxide, zinc oxide, titanium dioxide, zero-valent iron, carbon nanotubes, and bio-materials like chitosan are currently under extensive investigation [46, 228]. The general modes of action and possibility of multiple simultaneous antimicrobial activities of nanoparticles are summarized as follows:

- Reactive oxygen species (ROS) generation: ROS (e.g., superoxide anions, hydroxyl radical, and hydrogen peroxide) are produced after exposure to nanomaterials such as metal oxides, which can induce the peroxidation of the polyunsaturated phospholipids in the bacterial cells to damage DNA, and cause cell death eventually.
- Physical damage: The large curvature of nanostructured materials can damage bacterial cell wall membranes.
- Binding: When being bound with nanomaterials, bacterial cell walls may lose cell membrane integrity and cause efflux of cytoplasmic materials.
- Release of metal ions: Metal ions released from the nanomaterials into culture media can inhibit the ATP production and DNA replication to destroy the cells. The possible bactericidal mechanisms of antibacterial nanoparticles, especially metal ions, are shown in Fig. 18a [229].

Silver nanoparticles (Ag NPs) are one of the most popular high-efficiency broad-spectrum antibacterial agents. The proposed mechanism of Ag NPs-related toxicity using Gram-negative bacteria as the model bacteria is illustrated in detail in Fig. 18b [230]. (1) Silver ions can directly damage bacterial membrane by blocking the respiratory chain, collapsing the membrane potential and stopping ATP production. (2) Silver can promote ROS formation to damage both the membrane lipids and DNA. (3–4) Ag$^+$ ions may enter the cytosol-binding bacterial chromosome and intracellular proteins, thus affecting metabolic activity and replication. In addition, negatively charged bacteria can attract positively charged Ag NPs leading to higher local concentration of NPs. Bu et al. [231] introduced tannins rich in catechol groups into viscose fabrics which can reduce Ag ions to Ag NPs in situ. The chelation of tannic acid with Ag ions firmly anchored the formed Ag NPs onto the fiber surface. Low surface energy PFDT was assembled on viscose textile surfaces via metal thiol coordination and Michael addition or Schiff base reaction to get super-hydrophobic antibacterial textiles. The treated viscose fibers showed long-lasting antibacterial properties after 50 washing. Despite many advantages of Ag NPs, the leaching of Ag NPs is a problem in the textile industry. AgNPs may accumulate in tissues after absorption, and induce normal human cell damage and apoptosis [206, 230, 232–234]. Wu et al. [193] developed a tunable colored cotton fabric by sequentially depositing branched poly(ethylenimine)(PEI), Ag NPs and F-POSS with improved slow low release of Ag$^+$. Wherein, Ag NPs

Fig. 18 **a** Schematic representation of antimicrobial mechanisms of metal ions. Reproduced with permission from ref [235]. Copyright 2016 Elsevier. **b** Photographs of Ag NPs of different colors, cellulose fabrics dyed with different Ag NPs solutions, and antibacterial property of modified cellulose fabrics against *E. coli* and *S. aureus*. Reproduced with permission from ref [193]. Copyright 2016 Elsevier. **c** Proposed mechanisms of Ag NPs-related toxicity. Reproduced with permission from ref [230]. Copyright 2014 Royal Society of Chemistry
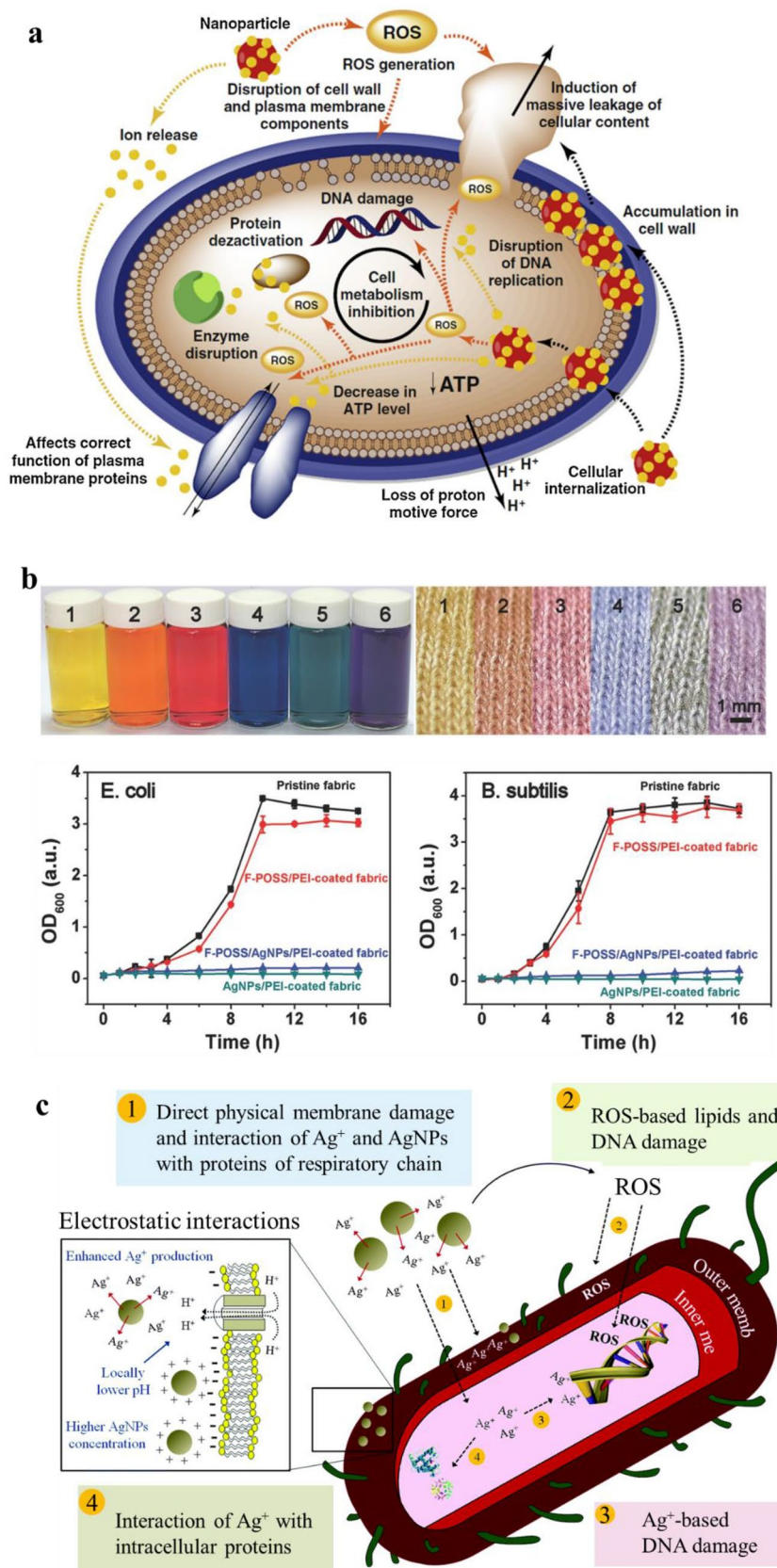
were used as dyes to obtain colored cellulose fabrics via surface plasmon resonance effect, while maintaining their antibacterial properties (Fig. 18c). The treated cotton fabric showed a WCA of 169° and antibacterial activity against *E. coli* and *B. subtilis*. The super-hydrophobic coating prevented the cotton surface from attachment of bacteria, and the slow release of Ag$^+$ endowed cotton with better bactericidal properties.

MOF and imidazole framework (ZIF) nanoparticles can slowly release antibacterial metal ions and have been widely applied as antibacterial materials to finish textiles in recent years [168, 169, 168, 169, 236]. The unique large porosity and specific surface area of MOF/ZIF can effectively increase the interaction of MOF/ZIF with biological cells, which adversely affects bacterial cell membranes. Furthermore, the controlled release of metal ions or guest molecules preloaded into the pores produced a synergistic antibacterial effect. In addition, MOF/ZIF can create multi-scale rough surfaces on various substrates, which is important for improving the hydrophobicity of the substrates [169, 174, 175]. Yang et al. [174] designed a super-hydrophobic antibacterial fabric by in situ growth of zeolitic imidazolate framework-8 (ZIF-8) followed by dip-coating in PDMS. ZIF-8 nanoparticles increased the surface roughness and PDMS reduced the low surface energy, resulting in superhydrophobic cellulose fabrics. The coated fabric exhibited 100% antibacterial activity against *E. coli* and *B. subtilis*, which can be attributed to the synergistic effect of the superhydrophobicity and slow release of ZIF-8 coatings. According to release of metal ions in the NPs suspensions, their antibacterial effect can be summarized into three classes: ZnO NPs' antibacterial effect originated from the released Zn^{2+}, CuO NPs' antibacterial effect originated from both the released Cu^{2+} and the CuO particles, and antibacterial effects of Fe$_2$O$_3$, Co$_3$O$_4$, Cr$_2$O$_3$, and NiO originated from metal oxide NPs themselves [237].

Ahmed et al. [238] investigated *N*-halamines modified silica gels with different N–X bonds (X was Cl, Br, or I) in barbituric acid, and the resultant inhibition zones illustrated that all three kinds of *N*-halamines showed obvious antibacterial properties, which confirmed the release killing mechanism of *N*-halamines. Hu et al. [239] developed functional materials and oxidative chlorine using oxazolidinone containing *N*-halamines and quaternary ammonium salt, and verified the release killing mechanism of *N*-halamines. Dong and coworkers [240] successfully prepared antibacterial amine *N*-halamine-labeled silica nanoparticles (ANHLS NPs) with a core–shell structure based on 2,2,6,6-tetramethyl-4-piperidinol for fighting against human pathogen bacteria *E. coli* and *S. aureus*. Due to release of positive chlorine, ANHLS NPs resulted in obvious inhibition region and sparsely distributed bacterial colonies outside the inhibition region compared with the control plate as seen in Fig. 19a and b.

In general, the antibacterial mechanism of superhydrophobic fabrics is complex and the result of the synergistic effect of antibacterial agents and surface superhydrophobicity. The strategies for developing antibacterial cellulose fabrics based on anti-biological adhesive superhydrophobic coating are summarized in Table 2. Moreover, it is commonly accepted that coating durability is another significant factor on determining their real-life applications. Therefore, micro–nano-roughness features, super-hydrophobic coatings, and durability of antimicrobial agents on cellulose fabrics must also be fully considered. In addition, the coating cannot reduce the original flexibility, breathability, and moisture permeability and wearing comfort of cellulose fabrics.

By employing octyltriethoxysilane (OTES) modified silica nanoparticles with a coral-reef like morphology, Anjum et al. [145] developed a robust superhydrophobic coating that had a large WCA of 167° and excellent durability against different types of damages, e.g., abrasion, UV irradiation, organic solvents, corrosive fluid, as

Fig. 19 Photos of inhibition zones of (**a**) the control sample and **b** ANHLS NPs against *E. coli*. Reproduced with permission from ref [240]. Copyright 2015 Elsevier
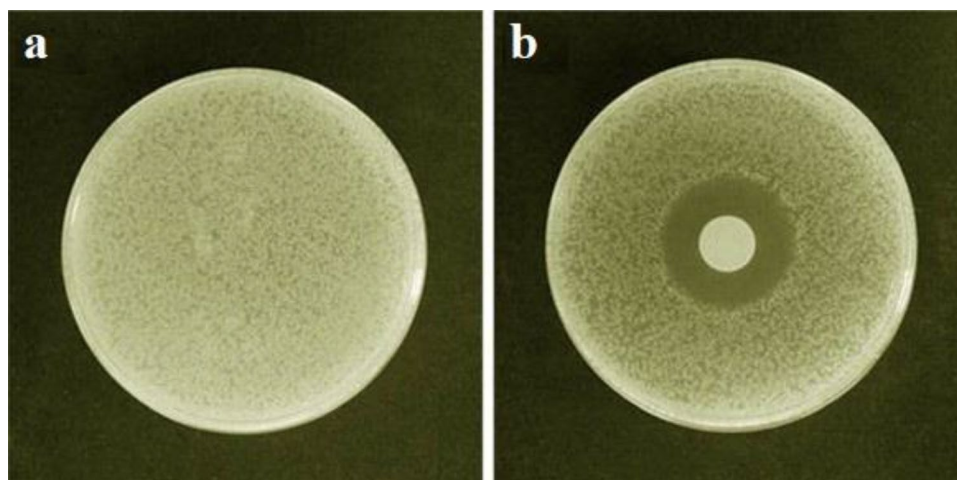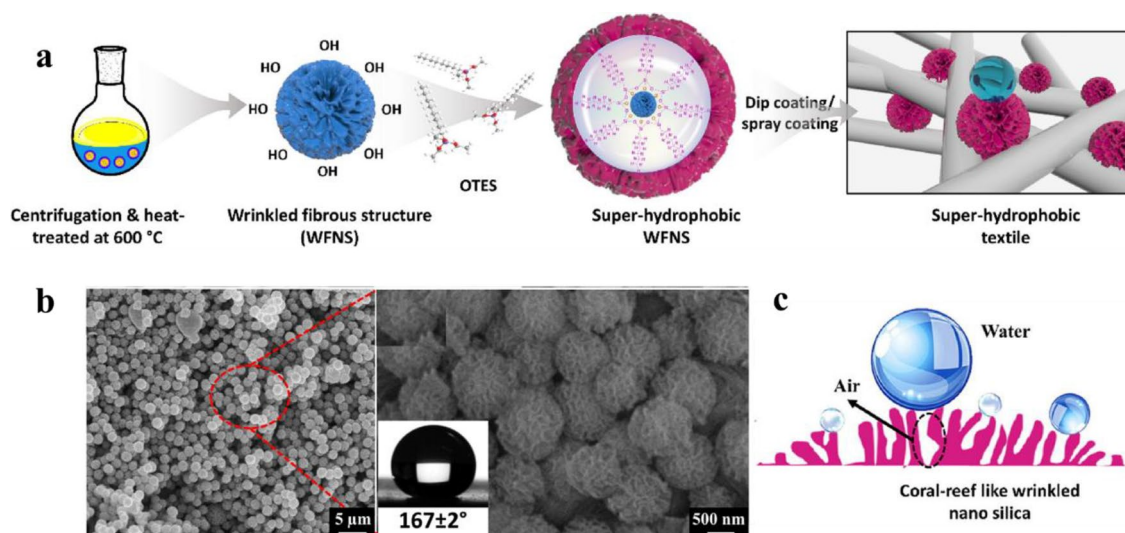
Fig. 20 a Procedure for fabrication of superhydrophobic fabric via dip-coating with coral-reef like silica NPs. **b** SEM images of the fabricated wrinkled fibrous coral-reed inspired silica NPs, and the WCA of superhydrophobic surface coated with the fabricated NPs. **c** Illus-tration of the hierarchical surface of wrinkled silica trapped with a layer of air. Reproduced with permission from ref [145]. Copyright 2020 Elsevier

shown in Fig. 20a and b. The coating also showed anti-mycotic property, probably because its hierarchical and hydrophobic surface can prohibit microorganism adhesion and contact with moisture and nutrients essential for microbe growth (Fig. 20c). Seth et al. [241] in situ fabricated broccoli-like hierarchical nickel stearate on cotton fabrics followed by stearic acid treatment to achieve superhydrophobic fabrics, which had a WCA of ~ 160° and exhibited antibacterial and antifungal activities against Gram-negative bacteria *E.coli*, Gram-positive bacteria *S. aureus*, and fungi *C. albicans*. The Ni^{2+} release behavior and super-hydrophobic surface together were believed to contribute to their antibacterial properties.

The Potential Applications

Viruses and bacteria are ubiquitous in our daily lives. The growth of bacteria and microorganisms and their large reproduction under humid environment may not only cause huge economic losses, but also harm people's health. Cellulose fabrics are widely used and indispensable in our daily life due to their moisture absorption ability, which also breeds bacteria and viruses. Therefore, superhydrophobic antibacterial cellulose fabrics may have practical applications in various fields, such as oil–water separation, anti-fouling coating, medical diagnostic devices, wound dressing, food-related fouling management, and paper-based electronic devices. Figure 21 summarizes the potential applications of functional cellulose-based fabrics showing super-hydrophobic and antibacterial properties.
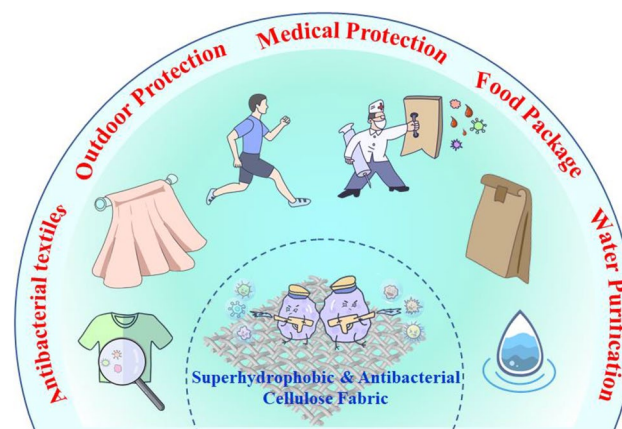
Fig. 21 Potential applications of super-hydrophobic antibacterial cellulose fabrics

Personal Protective Textiles

Interest in functional textiles has grown rapidly due to the need for additional properties, such as antibacterial, water-proof, and easy cleaning to improve life qualities [243]. With the increase of consumer awareness, safe and environmentally friendly products ensuring a comfortable and healthy life, are now in great demand [194]. Triggered by the growing market demand, researchers have focused their attention on the development of multi-functional textiles using unconventional technologies [244]. The protective products of super-hydrophobic antibacterial cellulose fabrics continue to emerge endlessly. Rilda et al. [245] reported a

superhydrophobic cotton fabric with high antibacterial properties against *Staphylococcus epidermidis* and *Escherichia coli* through first coating with ZnO/SiO$_2$ NPs via hydrothermal method, followed by the coating of additional layers of HDTMS. Similarly, Riaz's team modified nano-silicas with different concentrations of 3-(trimethoxylsilyl) propyl-N,N,n-dimethoxylammonium chloride and 3-glycylyoxyl-propyl) trimethoxylsilane, which were then applied onto cotton fabrics through pad–dry–curing coating technique. The obtained fabric showed durable superhydrophobic and antibacterial performance, which is a good choice in the application of personal protection [246]. In addition, Li and co-workers deposited zinc oxide (ZnO) onto polydopamine (PDA)-pretreated cotton fabric by atomic layer deposition method [16]. The coated fabric displayed antibacterial capability against *Staphylococcus aureus* and *Escherichia coli* of over 95% within 10 min. This work provided reference for the rapid production of antibacterial textiles.

Fabrics showing superhydrophobic antibacterial properties are also popular in outdoor sports and smart wearable textiles. Ou et al. [247] prepared superhydrophobic fabrics by sequential deposition of PDA, Ag$_2$O and 1H,1H,2H,2H-perfluorinated dodecanethiol, respectively. Liu et al. [196] prepared superhydrophobic fabrics by introducing SiO$_2$ NPs/PDMS layer, followed by depositing a thin layer of silver. The obtained super-hydrophobic and antibacterial fabrics can be used in home textiles, outdoor outerwear, and other facilities. Chauhan et al. [184] developed super-hydrophobic and antibacterial cotton fabrics with stable mechanical properties using fluoro-free cetyltrimethoxylsilane solution through dip-coating method. The resulting cotton fabric exhibited self-cleaning, stain-resistant, and antibacterial properties, and can be good choice for domestic and industrial fabric applications.

Medical Protective Textiles

Antibacterial cellulose fabrics have been in great demand in medical field due to the natural fibers advantages of lightness, softness, and non-stickiness.[8, 10]. Raeisi et al. [187] prepared super-hydrophobic cotton fabric using chitosan/TiO$_2$ nanocomposites. The super-hydrophobic fabric showed an antibacterial rate of 99.8% and 97.3% against *Escherichia coli* and *Staphylococcus aureus*, respectively. Such fabrics have potential use as antibacterial clothing and mask in clinical environment. Moreover, Wu et al. [193] used a simple leaching method to deposit branched PEI, silver NPs and F-POSS on cotton fabrics to prepare colored cotton fabrics with high color fastness, durable antibacterial, self-healing, and excellent super-hydrophobic properties. The fabric can maintain good super-hydrophobic and antibacterial properties upon repeated wear and washing. Similarly, Ye et al. [248] developed multifunctional cotton

fabric using silver–silica hybrid nanoparticles (Ag-MSNPs) and fluorinated MSNs (F-MSNs), and the treated fabric had good moisture retention and antibacterial activity against *Escherichia coli* and *Staphylococcus aureus*. Medical protection requires not only passive antibacterial property, but also active anti-infection and anti-blood adhesion ability. Suryaprabha and colleagues reported a super-hydrophobic cotton fabric with good self-cleaning and antibacterial activity prepared by copper and stearic acid deposition. The obtained fabric also showed anti-blood and self-cleaning ability [186]. Liu et al. [249] deposited chitosan (CS), gallic acid-modified silver NPS (GA@Ag NPs) and low surface energy PFDT onto cotton fabrics successively to make cotton yarns with super-hydrophobic and antibacterial properties. The developed cotton yarns can be used in wound recovery to reduce the risk of bacterial infection.

Food-Related Packaging Textiles

Food waste happens every day with negative social, economic, and environmental impacts [250]. In order to better protect the quality of food or extend the shelf life, it is necessary to cut off the possible contact between bacteria and food [166, 251]. Cellulose fiber-based papers have gained considerable interest as barrier and enhancement fillers in food packaging materials due to their interesting properties, including biodegradability, low density, non-toxicity, and low cost. The super-hydrophobic and antibacterial treatment of cellulose fiber papers can keep them away from potential pathogenic bacteria and affirmatively extend their service life [25, 179, 252–255]. Zhang et al. [179] used zwitterionic sulfopropylbetaine and quaternary ammonium salt, active isocyanate group as antibacterial finishing agents, and applied impregnation–filling–drying process to treat cotton fabric in a simple and environmentally friendly manner. As expected, the modified cotton fabric showed good antifouling and effective bactericidal activity, which can be used for food product packaging. Qiu et al. [253] also applied the layer-by-layer self-assembly technique, using γ-metha cryloxypropyltrimethoxysilane (KH570) and anionic polymer ε-polylysine (ε-PL)-modified TiO$_2$, to modify cellulose papers, which inhibited the colonization of *E. coli* (98.8%) and *S. aureus* (99.2%). Yang et al. [254] coated cellulose papers by layer-by-layer assembly of carvacrol-loaded ZnMOF-74 and polydimethylsiloxane (PDMS), to impart both antibacterial adhesion effect and bactericidal ability to the papers.

Water Purification

Traditional water treatment methods are mostly chemically and operationally intensive, non-renewable, expensive, time-consuming, and ineffective. Thus, it is important to

develop efficient, low-cost, and environmentally friendly water purification materials [256]. Fu et al. [189] prepared a super-hydrophobic antibacterial fabric with controllable wettability. The resulting cotton fabric was used in oil–water separation and showed high separation efficiency of over 95% even after 20 times of repeated use. Moreover, the coated fabric showed good self-cleaning performance after oil contamination, and its bactericidal rate was up to more than 80% no matter how the pH value was varied, which can reduce the surface biological pollution caused by bacterial proliferation. Heydarifard et al. [257] developed water-repellent cellulose foam paper. In the presence of an appropriate number of fine fiber particles, adding starch-modified guanidine-based polymer or antibacterial thermoplastic starch into the fiber network can significantly improve the quality of purified water. This work expanded the boundaries for cellulose applications and also pointed the way for future research.

Conclusions and Outlooks

The antibacterial or bactericidal superhydrophobic surfaces of cicada wings, dragonfly wings, lotus leaf, etc., have ensured their survival in fiercely competitive environments or formidable natural conditions. Superhydrophobic surfaces combining low surface energy surfaces and high roughness structures exhibit many remarkable properties, such as self-cleaning surfaces, oil/water separation, antifogging surfaces, anticorrosion surfaces, and antifouling surfaces. As one of the widely used fiber materials, cellulose fiber-based fabrics have been ubiquitous in our daily lives, but their polysaccharide characteristic makes them vulnerable to attack from microorganisms. Top-down and bottom-up strategies have been employed to fabricate super-hydrophobic cellulose fabrics possessing anti-biofouling, antibacterial, or bactericidal properties.

The super-hydrophobic cellulose-based fabrics show antifouling and self-cleaning properties, because the air layer on the super-hydrophobic surface can reduce effective contact between proteins and the surface, limit the initial adhesion of proteins, thereby inhibiting bacterial proliferation. However, the interaction of natural microorganisms with the super-hydrophobic surface and the tiny defects on the super-hydrophobic surface makes it impossible for air layer to stabilize for a long time and bacterial attachment happens eventually. Super-hydrophobic fabric surfaces with contact killing or biocide leaching functions that demonstrate multiple antibacterial strategies are highly desirable. The super-hydrophobic surface inhibits the initial adhesion of bacteria, and the biocide kills the bacteria on the surface or nearby environment, and then the super-hydrophobic self-cleaning effect removes most of the dead bacteria, reducing the adhesion of dead bacteria on the surface of the antibacterial agent, thereby increasing the life of the biocide.

In recent years, huge amounts of personal protective textiles, such as protective clothing and face masks, have been used due to the COVID-19 [258]. On account of environmental sustainability and economic benefits, developing bio-based antibacterial textiles is of extreme importance. Moreover, it can be anticipated that the rising environment pollution caused by COVID-19-related wastes can be reduced using degradable cellulose-based fabric materials, and the nano-/micro-plastics can be significantly decreased by replacing the polypropylene-based materials with cellulose-based ones [259].

Although different strategies have been developed to manufacture super-hydrophobic antibacterial cellulose fabrics, there are still many challenges: (1) Complex chemical composition and a lack of systemic studies on all the chemical materials. Fluorine-containing low surface energy components, especially those containing long alkyl chains, metal-containing antimicrobial agents, are still used extensively in developing super-hydrophobic and antibacterial fabrics. It is vital to design and synthesize environmentally friendly low surface energy fluorine-free chemicals. (2) Inspired by natural examples, super-hydrophobic and antibacterial surfaces can be also achieved via constructing nano-/microstructures on the fabric surfaces, so that the air trapped in the surface structures can prevent the surface from bio-fouling adhesion. However, in practical application, such fragile construction could be easily damaged physically. Therefore, there is still a need for further explorations on super-durable materials with substantial benefits for practical applications, especially for complex and harsh conditions. (3) The antibacterial or bactericidal performances of cellulose fabrics in moist environments or underwater scenarios should also be considered. (4) The self-healing function can impart cellulose fabrics with prolonged super-hydrophobicity, while the antibacterial effect of most leaching antibacterial agents will gradually diminish. Hence, fabrics demonstrating self-healing and antibacterial agent recycling functions are also critical and beneficial. (5) So far, *S. aureus*, *E. coli*, Gram-negative, and Gram-positive cells have been used to evaluate the performance of antibacterial surfaces, future research might focus on antiviral properties of protective fabrics. Moreover, the cost-effective production, non-toxic finishing process, product longevity strategies need to be considered for practical applications. Overall, antibacterial super-hydrophobic cellulose fabrics may be promising candidates for expanded applications in the fields of personal protection, sportswear, food-/paper-related packaging materials, medical protective fabric materials, liquid separation, and purification.

Data availability The data are available from the corresponding author upon request.

Declarations

Conflict of Interest The authors declare no conflict of interest.

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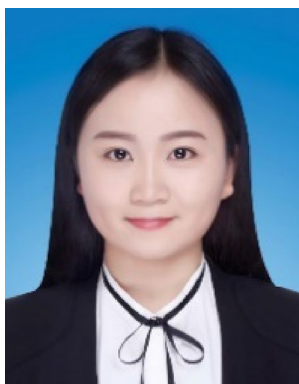
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