Mini-Review

Constructing Mechanochemical Durable and Self-Healing Superhydrophobic Surfaces

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ABSTRACT: Bioinspired superhydrophobic surfaces have attracted great interest due to their special functions and wide applications. However, it is still a big challenge to construct a durable superhydrophobic coating for large-scale applications due to its easy destruction by the mechanochemical attack. In this mini-review, we present the state-of-the-art developments in the rational design of mechanochemical durable and self-healing superhydrophobic surfaces. First, the mechanically durable superhydrophobic surfaces are constructed to endure mechanical damage by adjusting the surface morphology and increasing the binding force between the substrates and the modified materials. Second, chemical damages also have been taken into consideration to develop chemically robust superhydrophobic surfaces, such as chemical etching, ultraviolet (UV)-light irradiation, and bioerosion, etc. Third, endowing superhydrophobic coatings with self-healing function can effectively improve the durability and prolong the lifespan of the coatings by releasing low-surface-energy agents or regenerating topographic structures. Finally, the challenges and future perspectives in developing super durable bioinspired superhydrophobic surfaces by structure design and chemistry control are discussed. The innovative points provided in this mini-review will provide deep fundamental insight for prolonging the lifetime of the superhydrophobic surfaces and enable their practical applications in the near future.

INTRODUCTION

By mimicking the natural organisms, such as lotus leaves and gecko feet, bioinspired surfaces with special wettability have been successfully constructed by rational design, which exhibit a high water contact angle exceeding 150° and a sliding angle lower than 10°. As we know, superhydrophobic surfaces should have two features: (i) appropriate hierarchical micro-/nanostructures and (ii) low surface energy.[1] Due to the unique superhydrophobicity and oleophobicity, the super-wetting surfaces have been widely used in self-cleaning, anti-icing, water collection, and oil−water separation, etc.[2]

Recently, superhydrophobic surfaces have been fabricated not only by "top-down" or "bottom-up" methods on bulk materials but also by multiple steps via creating rough surfaces first and then modified with low-surface-energy materials.[3] The fabrication techniques include the solvothermal/hydrothermal method, chemical vapor deposition, dip/spray-coating technique, electrospinning, etc. However, the development of artificial superhydrophobic surfaces for real-world applications is severely limited by the poor mechanical and chemical stability. The superhydrophobic surfaces usually lose their special wettability due to the destruction of topographic structures and depletion of low-surface-energy agents when subjecting to scratch, washing, and abrasion, or working in an acid/alkali/salt environment, high temperature, and under UV-light irradiation. For example, a facile one-step approach was adopted to fabricate superhydrophobic coatings via the sol−gel processing of long-chain fluoroalkylsilane.[4] However, the coating was easily damaged by a pencil scratch due to the low adhesion between fluoroalkylsilane and glass. Therefore, it is essential to construct mechanochemical robust superhydrophobic coatings by increasing the adhesion with the substrates.

As an alternative to the design of mechanochemical durable surfaces, superhydrophobic surfaces with intelligent self-healing functions have been developed to overcome durability challenges and prolong the lifespan of the superhydrophobic coatings.[5] Inspired by living organisms, releasing low-surface-energy agents and reforming topographic structures are two main techniques to fabricate self-healing superhydrophobic surfaces. The former method can be easily realized by storing the low-surface-energy components inside the rough structures

and then releasing them onto the damaged surface during the self-healing process, while the latter method can be carried out by regenerating the hierarchical micro-/nanostructures again by some catalysts or triggers, which is comparatively difficult and not normally used.[6] However, how to accelerate the migration speed of low-surface-energy components onto the damaged surface and regenerate topographic structures via a facile method is still a critical challenge.

In this mini-review, we present the state-of-the-art developments in the rational design of mechanochemical durable and self-healing superhydrophobic surfaces (Figure 1), including
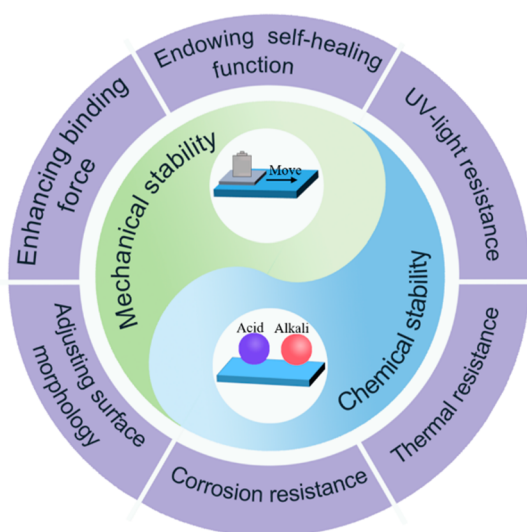

Figure 1. Rational design of mechanochemical durable and self-healing superhydrophobic surfaces by different strategies.

dip/spray coating, electrospinning, wet chemical method, a layer-by-layer assembly technique, 3D printing, and phase separation technique, etc. First, the physical and chemical methods, such as a combination of micro- and nanostructure design, cross-linking technique, forming covalent bonds, and introducing elastic and soft materials, are introduced to synthesize mechanically durable superhydrophobic surfaces. Second, chemical robust superhydrophobic coatings have been developed to endure chemical etching, such as UV-light irradiation, bioerosion, and high-temperature attack, etc. Third, two main strategies to endow superhydrophobic coatings with self-healing function by releasing low-surface-energy agents and regenerating topographic structures are summarized, which can effectively enhance the durability and prolong the lifetime of the superhydrophobic coatings. Finally, the challenges and future perspectives in developing super durable bioinspired superhydrophobic surfaces for practical applications are discussed. This mini-review will provide fundamental insight into understanding the working mechanism of fabricating mechanochemical robust superhydrophobic surfaces, thus inspiring researchers to explore advanced methods for improving the durability and prolonging the lifetime of superhydrophobic surfaces at a low cost.

■ MECHANICALLY DURABLE SUPERHYDROPHOBIC SURFACES

With the rapid development of nanotechnology and materials science, superhydrophobic surfaces have been widely used in our daily life. When compared to low-surface-energy factors,

surface roughness is a more vital element to construct mechanically durable superhydrophobic surfaces, which can be destroyed by mechanical attack. Therefore, keeping the hierarchical structure intact and improving the adhesion with the substrates are two effective ways to endure mechanical damage. It can be realized by adjusting the surface morphology and increasing the binding force between modified materials and substrates via cross-linking, forming covalent bonds, and introducing elastic/soft materials.

■ ADJUSTING THE SURFACE MORPHOLOGY

Encapsulating nanocomposite coatings with a polymer film is an important way to enhance the durability of superhydrophobic surfaces. The incorporated nanoparticles provide rough structures, while the hydrophobic polymers act as nanoparticle binding and provide low tension surface.[7] This process can be easily realized by a simple layer-by-layer assembly technique, dip-/spray-coating, and a wet chemical method. When the nanostructures are fixed stably on the substrate, the combination of superhydrophobicity with the smart surface texture brings a mechanical robust coating (Figure 2a).
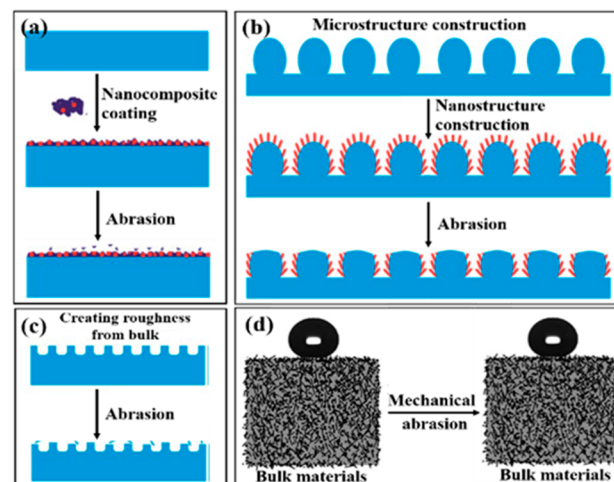

Figure 2. (a) Mechanical durability of nanocomposite coating surface, (b) optimized rough micro-/nanostructure surface, (c) creating micro-/nanostructures on a bulk surface, and (d) filling hydrophobic particles into bulk materials. (a−c) Reproduced with permission from ref 7. (d) Reproduced with permission from ref 10.

Since microstructures can stand more pressure than nanostructures, many researchers have fabricated hierarchical micro-/nanostructures by using the nanostructures to enhance the roughness and/or lower the surface energy and microroughness as a cushion and shelter to improve the mechanical stability and protect fragile nanoscaled topographies. As displayed in Figure 2b, optimizing the hierarchical micro-/nanostructures can effectively enhance their mechanical stability. Our group has fabricated raspberry-like hierarchical structures consisting of micro-/nanoscaled particles by an aerosol-assisted chemical vapor deposition.[8] It displayed superior robustness and retained a similar structure even after tape-peeling and sandpaper abrasion. Benefitting from this design, it can be applied on various substrates to resist chemical damage in any harsh environment.

Besides, creating micro-/nanostructures on inherent bulk materials and filling hydrophobic micro-/nanoparticles into
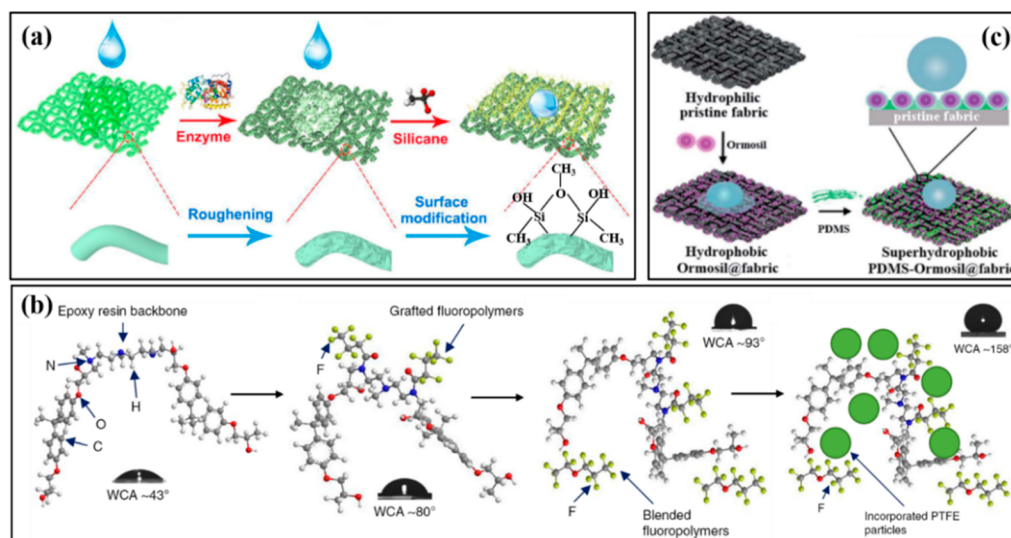
Figure 3. Schematic illustration for fabricating mechanochemical durable superhydrophobic (a) enzyme-etching fabric with a covalent bond, (b) cross-linked fluoropolymers/epoxy/PTEE superhydrophobic coating, and (c) PDMS-modified cotton fabric, respectively. (a) Reproduced with permission from ref 12. (b) Reproduced with permission from ref 1. (c) Reproduced with permission from ref 17.

bulk materials are also effective means to endure the mechanical attack. Roughness could be directly grown on the surface of bulk materials by chemical etching, electrochemical anodization, template method, and 3D printing method. The roughness comes from the substrate matrix as a whole, which avoids the interfacial problems, leading to good adhesion with the substrate and excellent mechanical durability when compared to the nanocoatings deposited on the substrate (Figure 2c). For example, a biomimetic superhydrophobic surface with eggbeater structure was innovatively developed by a 3D printing technique.[9] This method can introduce hierarchical roughness and low-surface-energy agents simultaneously, which is promising for large-scale production with low cost and high speed. Similarly, the modified bulk materials exhibit superior durability of superhydrophobicity and long lifetime because the encapsulated particles inside are difficult to be removed by mechanical abrasion (Figure 2d), such as electrospinning nanofibers, graphene-based aerogels, etc.[10,11]

■ INCREASING THE BINDING FORCE BETWEEN THE MODIFIED MATERIALS AND THE SUBSTRATE

Compared to adjusting the surface morphology, increasing the binding force between the coating layer and substrate is a more facile and promising strategy to improve the mechanical durability of superhydrophobic coatings for commercialization, which can be divided into three routes: forming covalent bonds between the coating layer and substrate, cross-linking with the coating layer, and introducing elastic/soft materials into the coating layer.

■ FORMING COVALENT BONDS BETWEEN THE COATING LAYER AND THE SUBSTRATE

Compared to van der Waals forces, forming covalent bonds between the coating layer and substrate is much stronger to endure the mechanical and chemical attack. However, it requires both the substrate (usually fabric, glass, stone, etc.) and the modified micro-/nanoparticles (usually inorganic such as TiO_2, SiO_2, etc.) to have chemical reactive groups. Cellulose-based fabric, such as cotton, silk fabric, etc., with hydroxyl groups can react with silanol, epoxide, or azide groups

to form covalent bonds first and then are decorated by hydrophobic low-surface-energy agents, resulting in super-hydrophobicity, while for glass, metal, and other synthetic fabrics, they inherently have less reactive groups on their surfaces and must be pretreated by chemical and plasma etching treatment, etc., or be predecorated by mussel-inspired dopamine to form amino and hydroxyl groups on its surface. For example, our group has prepared superhydrophobic cellulose-based fabrics (silk, cotton, and wool fabric) by an environmentally friendly enzyme-etching approach, followed by modification with methyltrichlorosilane via a chemical bond (Figure 3a).[12] It was demonstrated that the superhydrophobic cellulose-based fabrics exhibited excellent mechanical durability against the long cycling abrasion and laundering tests. In short, the covalent bonds can effectively enhance the adhesion with the substrate, making the superhydrophobic surfaces endure a more severe mechanochemical attack than that of the superhydrophobic coating on the substrate only with physical contact.

■ CROSS-LINKING WITH THE COATING LAYER

In addition to forming covalent bonds, cross-linking with the coating layer has been considered as another promising technique to improve the durability of superhydrophobic coatings.[13] This method can be used for both polymer coating and polymeric nanocomposite coating, which can be realized by the addition of cross-linking agents and catalysts or induced by UV-light irradiation and heating treatment, etc. After cross-linking, the polymer can form a stable network, and the micro-/nanoparticles are encapsulated by the network with a strong bond, improving the adhesion between the network and the substrate simultaneously. For example, Liu et al. successfully constructed superhydrophobic coatings on glasses with a three-dimensional continuous network and low-surface-energy hierarchical structure by a facile UV-induced cross-linking vinyltriethoxysilane with silica particles.[14] Making full use of the 3D continuous superhydrophobic network, the robust coating exhibited outstanding stability to both mechanical damage (sandpaper abrasion) and chemical damage (strong acid/alkali/salt solutions). Besides, Tiwari's
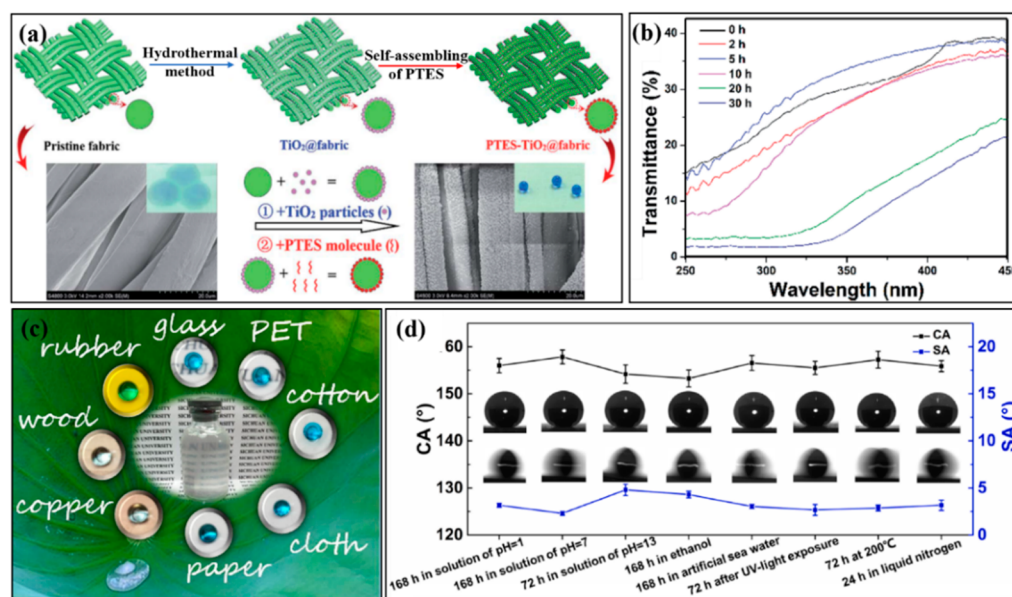
Figure 4. (a) Schematic illustration of the procedure to construct superhydrophobic TiO_2 particle-decorated cotton fabric. (b) UV transmission measurements for various deposition durations of TiO_2 nanoparticles coated onto cotton fabric samples. (c) A fluorine-free superhydrophobic coating with high flexibility on different substrates. (d) Water contact angle (CA) and sliding angle (SA) on the superhydrophobic surface after the treatment at different conditions. (a,b) Reproduced with permission from ref 19. (c,d) Reproduced with permission from ref 22.

group synthesized a robust superhydrophobic all-organic nanocomposite coating by cross-linking fluoropolymers with epoxy backbone, followed by incoporating polytetrafluoroethylene (PTEE) nanoparticles, which not only possessed mechanical and chemical stability but also exhibited excellent resistance to high-speed liquid impact due to the high adhesion with the substrate (Figure 3b).[1] The cross-linking technique can not only increase the binding force between the micro-/nanoparticles and polymer by embedding them inside the network but also enhance the adhesion between the network and the substrate, exhibiting promising prospects in practical applications.

■ INTRODUCING ELASTIC/SOFT MATERIALS INTO THE COATING LAYER

Compared to brittle materials, the nanocomposite coating by incorporating the micro/nanoparticles into elastic and soft materials can endure mechanical damage, and the adhesion with the substrate can even be improved via dispersing energy with deformation.[15] The nanocomposite coating with elastic and soft materials is like a spring. When loaded force is released, the superhydrophobic rough surfaces can recover their morphology as the pristine state. At the same time, the elastic and soft materials are compatible with various substrates, such as fabric, metal, and stone. Benefitting from these advantages, large quantities of elastic and soft materials have been developed to construct mechanical robust superhydrophobic surfaces by a dip-/spray-coating technique, such as PDMS, resin, and polyurethane, etc.[16] Among them, PDMS is the most widely used fluorine-free elastic polymer binder because of its wonderful mechanochemical stability and high adhesion with the substrate, which was widely used to synthesize superhydrophobic coatings by phase separation. For example, our group synthesized superhydrophobic cotton fabrics by first decorating a silica aerogel film, followed by PDMS top-coating via a simple dip-coating technique (Figure 3c).[17] Due to the high binding strength between the

superhydrophobic coating and cotton fabric, it can withstand 100 cycles of abrasion and 5 cycles of accelerated machine wash, without any morphology changes. Besides, it was very stable in strongly acidic and alkaline solutions. Recently, an unstable system for creating hierarchical micro-/nanostructures via phase separation is also raised for constructing superhydrophobicity. Kang et al. successfully fabricated a superhydrophobic coating driven by Marangoni instability and photopolymerization of PDMS.[18] Under UV irradiation, the uneven evaporation and cluster formation of the polymer surfaces in ethanol formed micro- and nanoscale roughness. PDMS acted as a binder and provided low-surface-energy agents simultaneously. This approach provides a rapid and efficient route to the generation of superhydrophobic surfaces, which may be extended to a wide variety of polymers. It is also promising for commercialization of superhydrophobic surfaces with an eco-friendly approach without using any toxic organic solvent solutions.

■ CHEMICALLY ROBUST SUPERHYDROPHOBIC SURFACES

In addition to the physical resistance against mechanical damage, chemical attack should also be taken into consideration in the real-world applications, which destroys the rough structures and exhausts the low-surface-energy agents, resulting in loss of superhydrophobicity. The UV-light irradiation resistance, corrosion resistance, and thermal resistance will be discussed in the following sections.

■ UV-LIGHT IRRADIATION RESISTANCE

The superhydrophobic surfaces outdoors are prone to be destroyed by UV-light irradiation, especially for outdoor-wearing textiles, tents, canopy covers, etc. The UV-light irradiation not only accelerates the aging process of the substrate but also destroys the structure of the superhydrophobic components. Therefore, employing UV-light absorption and inert inorganic micro-/nanoparticles (TiO_2,
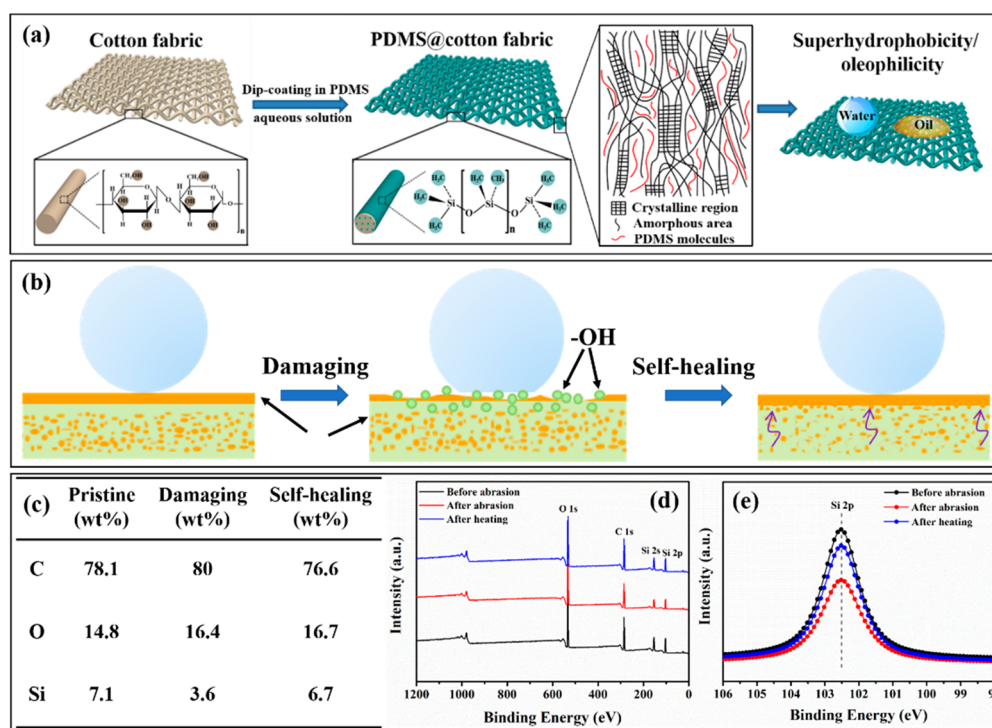
Figure 5. (a) Schematic illustration of the procedure to construct superhydrophobic PDMS@cotton fabric and (b) working mechanism of self-healing process by releasing low-surface-energy agents. (c) Line scan results of content changes of C, O, and Si elements and (d) wide and (e) narrow XPS spectra of PDMS@cotton fabric before abrasion, after abrasion, and after heating treatment. Reproduced with permission from ref 26.

ZnO, SiO_2, etc.) has been adopted to resist UV-light damage. Titanium dioxide has been widely used to fabricate superhydrophobic surfaces with UV-light irradiation resistance due to its excellent UV-light absorption property, chemical stability, and high bond energy between hydrophobic substrates and agents. For example, our group decorated cotton fabric with flower-like TiO_2 nanoparticles by a facile hydrothermal method first, followed by fluoroalkylsilane modification (Figure 4a).[19] The TiO_2@fabric not only exhibited excellent resistance to abrasion but also displayed effectiveness in shielding UV radiation due to high ultraviolet absorbance and the scattering characteristics of flower-like TiO_2 nanostructures (Figure 4b).

Apart from inorganic micro-/nanoparticles, some polymers (like polypyrrole) and carbon materials (like graphene, CNT) also have the ability to resist UV-light irradiation. For example, robust superhydrophobic poly(ethylene terephthalate) fabrics were successfully synthesized by coating with CNT and PDMS by an immersing technique.[20] The as-obtained fabrics possess remarkable mechanical durability to washing and abrasion and chemical stability against different pH solutions and UV light. However, the color of the bulk materials will become black and may not be suitable for fabrics.

CORROSION RESISTANCE

For corrosion resistance, acid, alkaline, and salt environments have a great influence on the durability and lifetime of the superhydrophobic surfaces. When exposed directly to the corrosive solution, the superhydrophobic molecules can easily be destroyed, and the superhydrophobic coating would detach from the substrate due to the hydrolysis of chemical interfacial bonding. Up to now, stabilizing the superhydrophobic coating and keeping a stable air layer between the rough structures and solution are the two main approaches to resist various kinds of

chemical damages. For example, our group coated fabrics with polydopamine first and then modified them with the stearic acid to synthesize superhydrophobic surfaces, exhibiting chemical stability in alkali and salt solutions due to the stability and strong binding force of the superhydrophobic coatings.[21] Besides, a fluorine-free superhydrophobic coating was prepared by functionalized silica nanoparticles and polystyrene-*b*-poly(ethylene-*co*-butylene)-*b*-polystyrene, which has been applied on different substrates by a facile dip/spray-coating technique, such as glass, metal, and polymeric matrix (Figure 4c).[22] The as-created surfaces displayed strong resistance not only to chemical corrosions (immersed in acid, alkaline, and salt solution for a long time) but also to high/low temperature and UV irradiation, without losing superhydrophobic property (Figure 4d).

What's more, the bioerosion (like fungus and bacteria) is also noteworthy. It will destroy the intrinsic structure of the materials, resulting in metal rusting and degradation of cellulose-based fabrics. Therefore, constructing superhydrophobic coatings and incorporating transition metals or noble metals are effective and efficient for bioerosion resistance. Li's group fabricated superhydrophobic cotton-derived carbon fibers modified with nanocopper by a facile and scalable method.[23] It exhibited outstanding antibacterial activity against Escherichia coli and Staphylococcus aureus with antibacterial ratios of >92% due to the superhydrophobicity and strong oxidizing ability of the oxygen reactive radicals by introduction of Cu. Similarly, a superhydrophobic fabric with durable photo-, bio-, and magneto-activities was also developed by in situ synthesis of TiO_2@Fe_3O_4@Ag nanocomposites on its surface.[24] These facile methods are effective and efficient for fabricating superhydrophobic surface with bioerosion property.

THERMAL RESISTANCE

In addition to UV-light irradiation and corrosion damage, thermal resistance has been widely investigated to prolong the lifespan of the superhydrophobic surfaces because the structure and chemical interfacial bonds can be easily destroyed at high temperatures. Recently, modifying the surface coating with inorganic materials, Cl^--containing materials, graphene, CNT, and other novel materials has been adopted to construct fire-resistant superhydrophobic surfaces. For example, our group has synthesized 3D superhydrophobic aluminum hydroxide nanoparticle-coated cellulose composite aerogels.[25] Due to the existence of the inorganic nanoparticles in the bulk materials, the cellulose composite aerogels exhibited remarkable mechanical stability, thermal stability, and fire retardancy.

Except for the aforementioned harsh environmental conditions, some other extreme conditions, such as low temperature, low/high pressure, high humidity, etc., should also be paid much attention to construct chemically robust superhydrophobic surfaces.

INTELLIGENT SELF-HEALING SUPERHYDROPHOBIC SURFACES

Inspired by the naturally superhydrophobic insect wings and plant leaves, always keeping special wettability throughout their lifetime, endowing artificial superhydrophobic surfaces with intelligent self-healing function has been proposed to maintain mechanochemical durability. They are meaningful and promising in practical applications to stand much more severe destructions, such as knife-cut, scratch, etc. The self-healing process can be realized by two means: releasing low-surface-energy agents and regenerating topographic structures.

RELEASING LOW-SURFACE-ENERGY AGENTS

When the hydrophobic leaves are damaged, they can regenerate the hydrophobic epicuticular wax layer and maintain superhydrophobicity throughout its lifetime. By mimicking this phenomenon, releasing low-surface-energy agents inside is the most commonly used technique to construct artificial robust and self-healing superhydrophobic surfaces. Generally speaking, large quantities of low-surface-energy agents (usually fluoroalkylsilane, PDMS, etc.) are injected into the inner space of the rough matrix by the dip-/spray-coating method. Once the superhydrophobic coating is decomposed or degraded by mechanochemical attack (abrasion, washing, plasma damage, etc.), the superhydrophobic molecules will migrate to the damaged surface when triggered by mechanical force, high temperature, and UV-light, which exhibits higher self-healing speed than longtime placement at room temperature. Recently, we fabricated a mechanochemically robust superhydrophobic cotton fabric with intelligent self-healing nature by a "PDMS-in-water" emulsion approach via phase separation (Figure 5a).[26] After dip-coating and drying, PDMS was uniformly dispersed in cotton fiber and grafted on the bulk surface of cotton fabric via a strong interfacial binding force, enhancing inherent hierarchical microstructures and decreasing the surface energy simultaneously. Therefore, the superhydrophobic coatings not only displayed superior chemical stability but also could repair the superhydrophobicity throughout the whole lifetime though damaged by machine washing or abrasion, due to the self-diffusion process of PDMS molecules from the inner part to the outer surface of the cotton fibers to minimize surface free

energy (Figure 5b−e). This environmentally friendly and cost-effective method can be employed on various materials in the nanoscience field for environment and energy applications. Besides, Zhang et al. first fabricated totally waterborne, mechanically robust, and self-healing superhydrophobic coatings by a combination of polyurethane adhesive and hexadecyl polysiloxane-modified SiO_2.[27] Except for the high mechanical stability and chemical stability against corrosive liquids and UV irradiation, the coatings also showed fast and stable self-healing capability owing to migration of the healing agent (hexadecyl polysiloxane) to the damaged surface. However, when the low-surface-energy components are exhausted, the superhydrophobicity cannot be recovered any more. From this view, self-healing superhydrophobic surfaces can be achieved by modifying the rough surfaces with the superhydrophobic agents again via the dip-/spraying-coating method.

REGENERATING TOPOGRAPHIC STRUCTURES

Traditionally, hierarchical micro-/nanoparticles have low adhesion with the substrate, and they can easily peel off from the substrate after mechanical or chemical destruction. Therefore, regenerating topographic structures is another important strategy for constructing robust self-healing superhydrophobic surfaces. Chen's group synthesized a self-healing superhydrophobic surface containing polystyrene, fluorinated SiO_2 nanoparticles, low-surface-energy agents, and TiO_2 nanoparticles.[28] When the micro-/nanostructures were destroyed by mechanical abrasion, polystyrene was decomposed to regenerate the roughness by the photocatalysis of TiO_2 nanoparticles under UV-light irradiation to maintain superhydrophobicity. Recently, shape-memory polymers have appeared to fabricate the mechanochemical robust and self-healing superhydrophobic surfaces.[29] Though it was damaged by mechanical force and the surface became flat, it would recover its original roughness due to the memory effect. However, there are little works on regenerating rough structures. Therefore, it is still a big challenge to be addressed, and more efforts should be put in this area.

CONCLUSIONS AND OUTLOOK

In summary, the state-of-the-art fabrication methods on mechanochemical durable and self-healing superhydrophobic surfaces are presented in this mini-review. The strategies of adjusting surface morphology and increasing the adhesion with the substrate are firstly introduced to synthesize mechanically durable superhydrophobic surfaces. Then, chemically robust superhydrophobic surfaces have been developed to endure chemical etching, such as UV-light irradiation, bioerosion, and high temperature, etc. Endowing superhydrophobic coatings with self-healing function by releasing low-surface-energy agents and reforming topographic structures can also enhance the durability and prolong the lifetime of the super-hydrophobic coatings. These strategies all contribute to constructing mechanochemical robust superhydrophobic surfaces and enable them in practical applications in the following decades.

Though great progress has been achieved in constructing durable and self-healing superhydrophobic surfaces, there still exist some critical challenges. First, the superhydrophobic coatings usually contain fluorine-containing compounds and are often synthesized by the wet chemical method using organic solvent, which is toxic and harmful to human beings. It

is essential to construct artificial robust and self-healing superhydrophobicity via an environmentally friendly technique. Second, though the adhesion with the substrate has been improved by cross-linking or forming covalent bonds, it can only stand mild mechanical damage, such as abrasion, washing, etc. In order to adapt the real-world usage, it is meaningful to construct durable superhydrophobic surfaces against severe mechanical damages (deep scratch, knife-cut, etc.) by forming much stronger interfacial chemical bonds. Third, most of the self-healing processes can be realized by longtime placement or triggered by mechanical force, heating, and UV-light irradiation, etc. Thus, how to accelerate the speed of the migration/mobility of low-surface-energy molecules onto the damaged surface is still a big challenge. Fourth, the traditional methods for fabricating mechanochemical durable and self-healing superhydrophobic materials are limited to large-scale production and cannot meet the industrial requirements. The novel techniques should be developed for mass production by creating roughness and providing low-surface-energy simultaneously, such as 3D/4D printing, an unstable system for constructing superhydrophobicity via phase separation, etc. Finally, various testing methods have been conducted to evaluate the mechanical and chemical stability of the superhydrophobicity, making it difficult to compare the efficiency of different fabrication techniques. Therefore, the testing methods should be unified according to the actual applications, such as abrasion standard, washing standard, etc. We believe that with more efforts put in this field the artificial mechanochemical durable superhydrophobic surfaces can be successfully constructed with multifunctions within a short time. It can also be applied in various environmental and energy applications, such as textile finishing, oil−water separation, anti-icing, catalysis, and lithium-ion batteries, etc.

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Author Contributions

The manuscript was written through the contributions of all authors.

Notes

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■ ACKNOWLEDGMENTS

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