

Corrosion-Resistant Organic Superamphiphobic Coatings

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Abstract: In recent years, organic superhydrophobic coatings have emerged as a promising direction for the protection of metal substrates due to their excellent liquid-repelling properties. Nonetheless, these coatings face challenges such as poor mechanical robustness and short service lives, which have limited their development and garnered attention from numerous researchers. Over time, researchers have gained a deeper understanding of superhydrophobic coatings and have published many related articles. Nevertheless, the lack of logical organization and systematic summarization of research focus in this field hinders its advancement. Therefore, the main purpose of this review is to clarify the design principles and working mechanisms of organic superhydrophobic coatings, as well as to summarize and synthesize the latest research on different aspects of superhydrophobic coatings, including liquid-repellent performance, wear resistance, adhesion, antibacterial properties, and self-healing properties. By employing decoupling mechanisms to study each performance aspect separately, this review aims to provide references for extending the service life of organic superhydrophobic coatings.

Keywords: superhydrophobic; organic coatings; liquid repelling; corrosion-resistant



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1. Introduction

The battle against metal corrosion is a war without smoke. According to statistics [1], the global cost of corrosion was estimated to be approximately four trillion US dollars in 2015. This estimation was derived from the research [2] on international corrosion technologies, applications, and economic measures initiated by NACE International. It accounts for about 3.4% of the global GDP. In order to reduce corrosion costs and extend the service life of substrates, various metal protection strategies, including composite metal substrates, multi-layer casing, corrosion inhibitors, and anticorrosive coatings, have been developed for different environments. Hou et al. [3] demonstrated that anticorrosive coatings constitute the largest proportion (66.15%) of direct corrosion costs. Currently, the anticorrosive coatings in use include (1) graphene coatings [4–6] are primarily used on metal substrates in marine environments or those exposed to high concentrations of chloride ions [7,8]; (2) epoxy coatings [9–11] are mainly applied to metal substrates in industrial warehouses with lower protection needs [12]; (3) polyurethane coatings [13–15], due to their excellent stress–strain properties, are preferred for metal substrates in scratch-prone environments [16,17]; (4) ceramic coatings [18–20], known for their high-temperature resistance, are primarily used on aerospace component surfaces [21,22]; and (5) superamphiphobic coatings [23,24], which act as active protective layers on substrates and can function in a variety of complex environments [25–27].

Superamphiphobic coatings [28] have been emerging as an important class of material in the field of metal material protection in recent years due to their unique properties. The micro/nano-rough structures and low-surface-energy material [29] on the surface of the superamphiphobic coatings render them with high water/oil contact angles (WCAs > 150°)

and low sliding angles (WSAs $< 10^\circ$). These coatings exhibit excellent liquid repellency, effectively isolating corrosive media from contact with the metal substrate and reducing the pathways for corrosive media infiltration.

Researchers have designed corrosion-resistant superamphiphobic coatings with different wettabilities by controlling the surface roughness and energy of the coatings. Inspired by the characteristics of ticks, Wang et al. [30] modified hollow glass microspheres by loading silica nanoparticles on their surface and introduced these microspheres into polyvinylidene fluoride (PVDF) simultaneously. The combination of a rough structure and low-surface-energy functional groups imparts excellent anti-wax performance to the coating, achieving a high wax deposition resistance rate of 82.76%. Xu et al. [31] first utilized small-area cathode moving technology to manufacture a large-area superhydrophobic magnesium alloy surface. Scanning electron microscope (SEM) images show that the magnesium alloy surface has a hierarchically layered, rough structure with low surface energy. During a 180-day immersion in a simulated seawater corrosion environment, the static water contact angle values remained stable in the range of $162.0\text{--}166.5^\circ$. Currently, reviews on superamphiphobic surfaces, both domestically and internationally, mainly focus on the fields of oil–water separation [32–34], anti-icing [35–37] and anti-waxing [30,38,39]. However, there are few reported comprehensive reviews on the crucial aspect of corrosion resistance associated with superamphiphobic surfaces [40].

This review systematically summarizes the latest significant developments in superamphiphobic coatings in the field of corrosion resistance. It introduces the working principles of superamphiphobic coatings and methods for regulating the corrosion resistance and superamphiphobic properties of such coatings. Finally, it addresses the existing issues in current research and proposes feasible improvement solutions, aiming to provide new perspectives for the study of superamphiphobic materials in the field of corrosion prevention.

2. Principles of Superamphiphobicity

2.1. Basic Principles

2.1.1. Young's Equation

In 1805, Thomas Young first proposed the concept of the ideal solid–liquid interface. Different contact angles of liquid droplets on solid surfaces represent varying degrees of hydrophilicity and hydrophobicity. Due to the total energy tending to minimize the state of tension balance [41], the droplet is in a stable state on the solid surface. The tangential component of the surface tension γ_{WA} at the edge of the water droplet is balanced by the interfacial tension, while the normal component of γ_{WA} is balanced by the Laplace force (Figure 1). On an ideally smooth surface, the tension equilibrium of a stationary droplet on a solid surface leads to the derivation of Young's equation [42]:

$$\cos \theta = \frac{\gamma_{sg} - \gamma_{sl}}{\gamma_{gl}}$$

Here, γ_{sg} is the surface tension of the solid/gas interface, γ_{sl} is the surface tension of the solid/liquid interface, γ_{gl} is the surface tension of the liquid/gas interface, and θ is the contact angle on an ideally smooth surface.

The establishment of Young's equation is based on an ideal surface, but solid surfaces are generally endowed with varying degrees of roughness. Therefore, Young's equation cannot fully explain the wetting behavior of droplets on real surfaces.

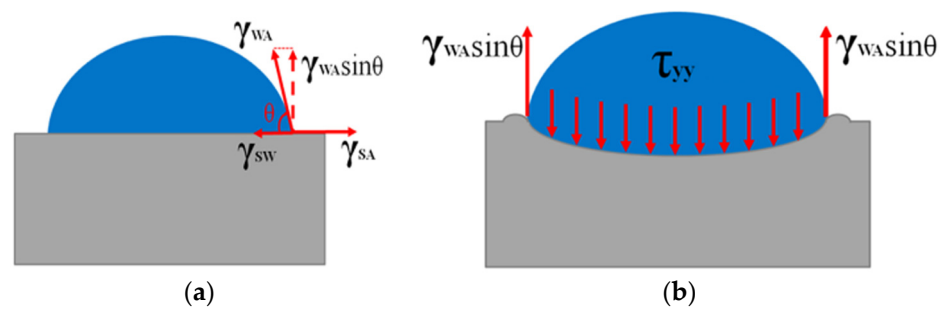


Figure 1. (a) Tangential component of γ_{WA} balanced by interfacial tension; (b) normal component of γ_{WA} balanced by Laplace force [43].

2.1.2. Wenzel Model and Cassie–Baxter Model

Wenzel published a paper in 1936 on the correlation between roughness and wetness [44]. Based on Young's equation, he introduced the concept of surface roughness for the first time. He proposed that the rough structure of the coating surface is capable of enhancing the hydrophilicity of hydrophilic surfaces and, conversely, augmenting the hydrophobicity of hydrophobic surfaces [45]. Because of the different chemical properties and roughness of the coating surface, oil or water may fill or remain in the coating textures on the surface, resulting in a difference between the apparent contact angle and the intrinsic contact angle. He assumed that the droplet completely wets the cavities in the surface structure. The surface in the Wenzel state provides additional solid–liquid contact area (Figure 2a), leading to an increase in surface free energy. Therefore, the equation applied to this state can be described as:

$$\cos \theta_w = \frac{r(\gamma_{SA} - \gamma_{SL})}{\gamma_{LA}} = r \cos \theta_0$$

where θ_0 represents the Young's contact angle, and θ_w represents the Wenzel contact angle. In the Wenzel equation, as r is greater than 1, if θ_0 is less than 90° , then θ_w is less than θ_0 , suggesting that roughness (r) enhances the surface hydrophilicity. If θ_0 is greater than 90° , then θ_w is less than θ_0 . In this case, the introduction of roughness enhances the surface hydrophobicity. Hence, roughness can amplify the hydrophilic/hydrophobic effects of the solid surface. Wenzel elucidated the relationship between the intrinsic contact angle and the apparent contact angle associated with a uniformly rough surface. However, this relationship is not applicable to surfaces composed of multiple chemical components and non-uniform roughness. Cassie and Baxter [46] introduced (Figure 2b) the concept of fractional area coverage in 1994. They proposed that in the presence of strong hydrophobicity, the cavities of surface rough structures cannot be filled, and liquid droplets covering the surface will trap air in the lower cavities. Therefore, the observed solid–liquid contact surface should be composed of solid–liquid–gas three-phase coexistence, representing a composite contact. The equation under this condition is as follows:

$$\cos \theta_{CB} = \frac{\sum_i^n f_i (\gamma_{i,SA} - \gamma_{i,SL})}{\gamma_{LA}} = \sum_i^n f_i \cos \theta_i$$

The roughness of a solid surface can be characterized by the standard roughness factor (RF) and the root mean square roughness (R), where RF is defined as the ratio of the actual solid surface area to the projected solid surface area [47]. Different materials typically display different surface chemical properties and roughness, making it difficult to quantify and compare the impact of roughness on the apparent hydrophobicity of different material surfaces. In terms of most materials, the water contact angle on a flat surface is typically less than 180° [48]. Nevertheless, certain rough surfaces can approach close to 180° [49]. In some cases, a portion of water may fall into the textured structure between rough, solid

surfaces. This is neither the Cassie–Baxter state nor the Wenzel state but rather a special Cassie state under wetting conditions [50] (Figure 2c).

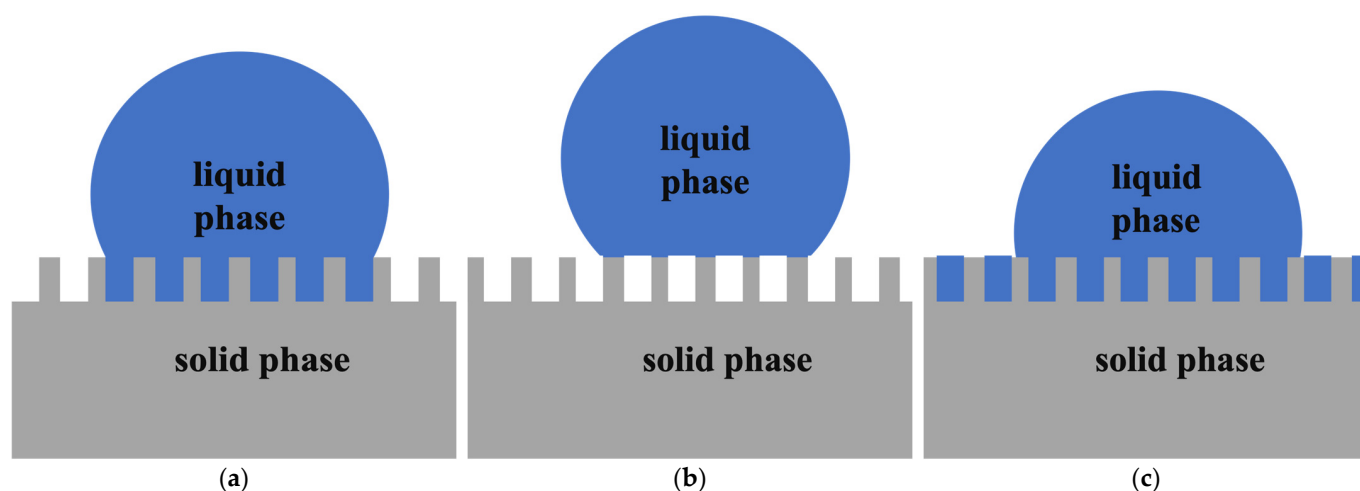


Figure 2. (a) Wenzel Model; (b) Cassie–Baxter Model; (c) Cassie Model under Penetrating State [43].

2.2. Principle of Corrosion Resistance in Superamphiphobic Coatings

One of the significant reasons for metal corrosion is the galvanic cell system formed by the contact of an electrolyte solution with the metal substrate [51]. In the galvanic cell environment, oxidation occurs on the metal surface, leading to the loss of electrons from metal atoms and transforming them into positive ions. On the other hand, the electrolyte on the metal surface undergoes reduction to gain electrons, promoting the continuation of the corrosion process. The micro/nanostructures and ultra-low surface energy of superamphiphobic coatings enable the formation of an air film upon contact with oil or water. This prevents the corrosive medium from further contacting the substrate, achieving functions of corrosion resistance, antiscaling, and antifouling, thereby extending the service life of the metal material. Wu et al. [52] constructed micro/nanostructures on a metal substrate employing spray-coating to capture more air films. Further investigation focuses on the preparation methods of micro/nanostructures in coatings. First, micron-scale thorn-like TiO_2 particles are synthesized using a hydrothermal method. These TiO_2 particles are then immersed in a solution of 1H,1H,2H,2H-perfluorooctyl trichlorosilane (PFOTS) to reduce their surface energy, enhance their liquid repellency, and result in F-thorn-like TiO_2 particles. Second, nano-scale mesoporous silica (F-M- SiO_2) is prepared through a hydrolysis method. Finally, the surface-modified micron-scale TiO_2 and nano-scale SiO_2 particles are mixed with epoxy resin and applied via a one-step spraying method to produce superoleophobic $\text{TiO}_2/\text{SiO}_2$ composite coatings. The micro/nano-rough structures of modified particles and the dendritic geometric shape derived from the spray-coating method result in oil–water contact angles exceeding 155° , endowing the coating with excellent corrosion resistance. Yu et al. [53] prepared (Figure 3a) a simple water-based coating containing only nanoparticles and fluorocarbon surface active agents. The fluorocarbon surface active agent achieved superoleophobicity as a result of reducing the surface energy of the coating. In addition, when heated with silane at 80°C for 15 min, the coating acquires superhydrophobicity. This characteristic allows its application in environments with different temperatures and specific requirements. Zhang et al. [54] constructed a superamphiphobic coating based on double-adhesive layers and aluminum oxide nanoparticles modified with 1H,1H,2H,2H-perfluorodecyltriethoxysilane. This coating exhibits superamphiphobicity to various liquids with different surface tensions, including water, glycerol, ethylene glycol, and peanut oil, showing low wettability, high contact angles, and low sliding angles. Electrochemical tests reveal (Figure 3b) that the corrosion potential shifts positively by more than 250 mV, the corrosion current density decreases by four orders of magnitude, and the

charge-transfer resistance increases by seven orders of magnitude for the coated sample. These results indicate that the coating exhibits enhanced corrosion resistance.

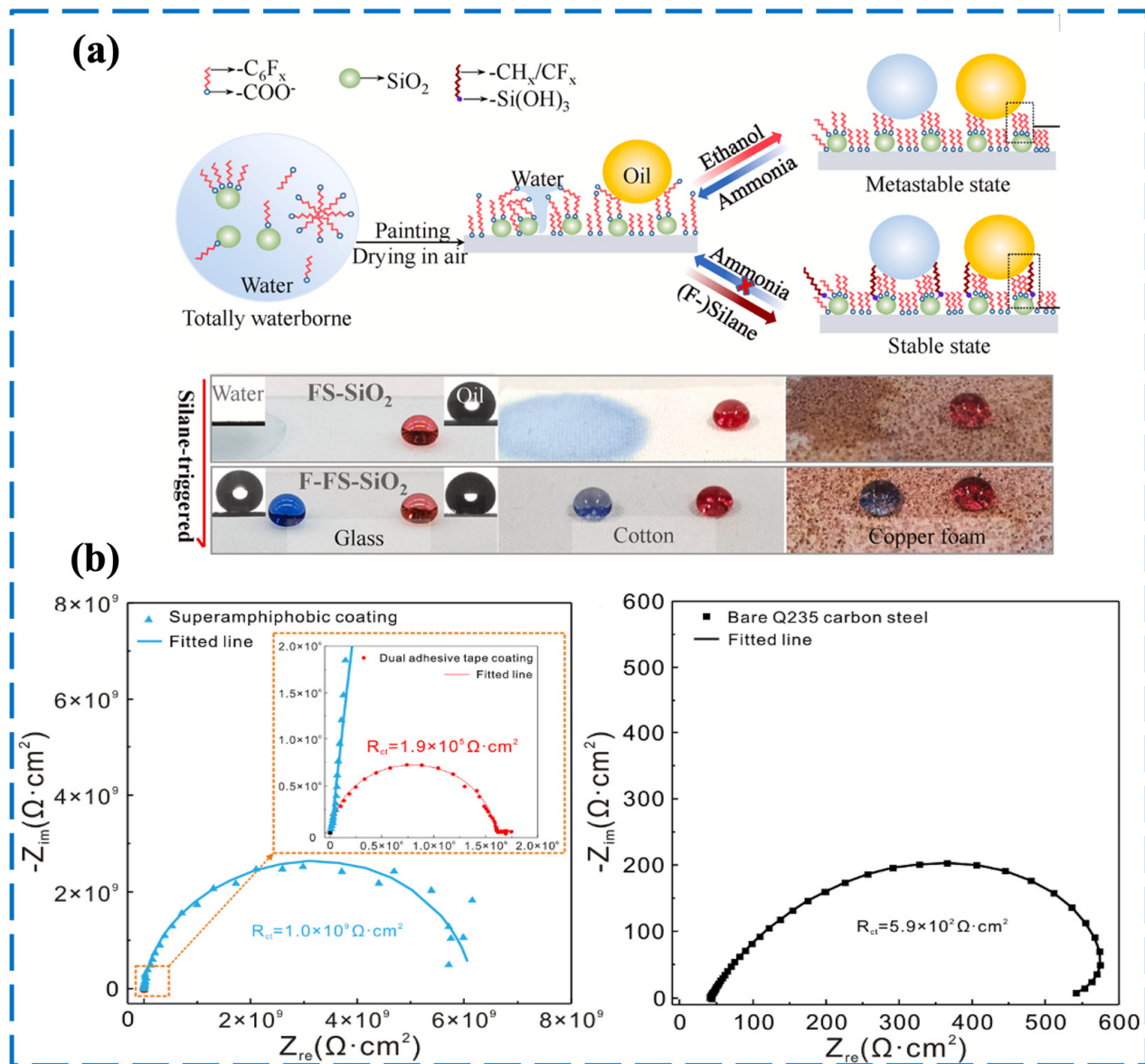


Figure 3. (a) Schematic illustration of the mechanism transition [53]; (b) Nyquist and Bode plots of different surfaces in a 3.5% sodium chloride (NaCl) aqueous solution [54].

3. Regulation Strategies of Corrosion-Resistant Superamphiphobic Coating Performance

Superamphiphobic coatings, due to their active liquid-repelling properties, have greater potential for application in the field of corrosion prevention compared with traditional coatings. However, the corrosion resistance of superamphiphobic coatings is influenced by various coupling factors. This review will use a decoupling mechanism to discuss the individual effects of liquid repellency, wear resistance, adhesion, antimicrobial properties, and self-healing on the corrosion resistance of the coating (Figure 4). The aim is to provide guidance for enhancing the overall performance of superamphiphobic coatings.

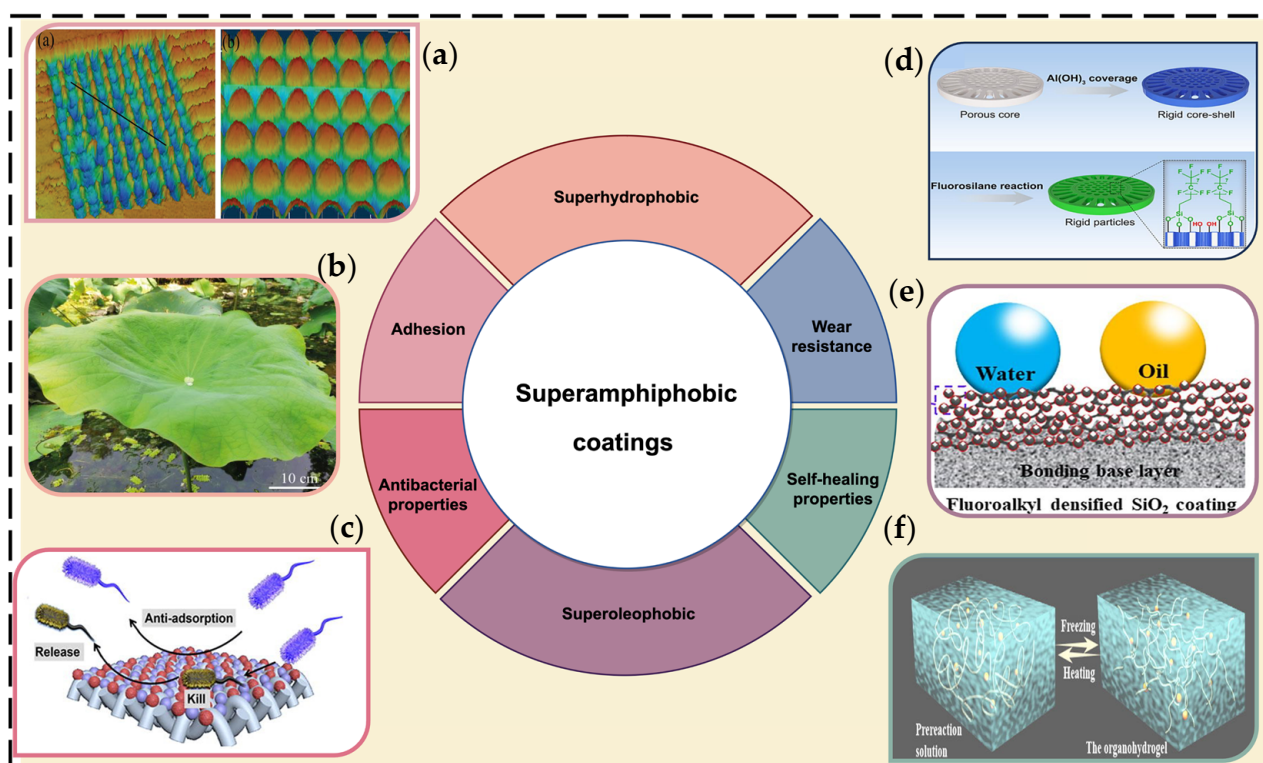


Figure 4. (a) Superamphiphobic surfaces with controllable adhesion fabricated by femtosecond laser Bessel beam on PTFE [55]; (b) biomimetic super “silicate” phobicity and superhydrophobicity of ceramic material [56]; (c) textile coatings configured by double nanoparticles to optimally couple superhydrophobic and antibacterial properties [57]; (d) flexibility and abrasion tolerance of superamphiphobic coatings with rigid core–shell particles [58]; (e) oil-immersion stable superamphiphobic coatings for long-term super liquid-repellency [59]; (f) underwater superoleophobic APTES-SiO₂/PVA organohydrogel for low-temperature tolerance, self-healing, and recoverable oil/water separation mesh [60].

3.1. Liquid-Repellency Performance

The surface of coated coatings is prone to organic contamination and bacterial parasitism, while superamphiphobic surfaces can provide excellent liquid-repelling capability. Upon the intrusion of corrosive media, an air layer spontaneously forms, acting as a barrier against the damage of corrosive media to the metal substrate [61]. Moreover, on account of its liquid-repellent properties, water or oil droplets can be carried away as they roll, reducing the residence time of pollutants on the coating surface [62]. Zhang et al. [63] utilized photolithography to prepare a highly uniform polydimethylsiloxane (PDMS) Dual-T microstructured surface with a downward suspension angle of up to 90° in the horizontal direction. This flexible superhydrophobic Dual-T microstructure surface, without fluorination treatment, is capable of repelling liquids with a surface tension lower than 20 mN m^{−1} in the Cassie–Baxter state. Flexible Dual-T microstructure surfaces were fabricated using a one-step molding process with photoresist templates. To create the Dual-T template, two layers of photoresist and multiple exposures were utilized to form the negative features. The production procedure consisted of five steps: (i) Vacuum deposition of chrome (Cr) film onto a glass wafer, followed by patterning using a wet etching process; (ii) spin-coating two layers of photoresist onto the substrate, with baking after each layer; (iii) performing the first UV exposure, followed by the second UV exposure; (iv) precisely developing the photoresist; and (v) conducting a one-step molding process to produce the Dual-T microstructure surface. Deng et al. [64] prepared a superamphiphobic coating employing photolithography techniques and armor structure design to construct a microscopic inverted-pyramid honeycomb structure on a metal substrate. This coating can withstand more than 1000 Taber wear

cycles, exhibiting ten times the wear resistance compared to traditional superamphiphobic surfaces. Furthermore, it maintains its superamphiphobic properties even after immersion in a mixture of high-concentration HCl/HNO₃ (*v:v* = 3:1) for four hours. Liang et al. [65] utilized phase separation to induce the aggregation of fluorinated silica nanoparticles, resulting in the preparation of superamphiphobic coatings with dense micro/nanostructures. In subsequent tests, the first impact/rebound contact time of water droplets released from a height of one centimeter is 12.75 milliseconds, with the ability to rebound 10–11 times. The excellent liquid-repellent capability is attributed to the dense microstructure on the surface induced by the aggregation of nano-scale particles, which enhances the ability to capture air. They achieved the preparation of low-cost, high-performance superamphiphobic coatings by controlling the density of micro/nanostructures on the coating surface by modulating the initial nanoparticle size. Water droplets released from a height of 1 cm can impact/rebound 19 times. The performance surpasses that of all previously reported superamphiphobic coatings, which is primarily ascribed to the optimal selection of silica raw material particle size (Table 1, 15 nm). The coating exhibits uniform nano-sized pores (30–100 nm) under high magnification.

Table 1. Contact/departure angles of coatings at different particle sizes.

D _{silica} (nm)	10	15	50	100
CA(deg)	162 ± 1.3	163.9 ± 2.3	154.1 ± 1.5	151.4 ± 2.2
SA (deg)	4.0 ± 1.0	2.0 ± 0.6	17.3 ± 2.1	

3.2. Wear Resistance Performance

Currently, most superamphiphobic coatings face challenges related to poor mechanical robustness [66]. In harsh, corrosive environments, addressing the issues of enhancing wear resistance and extending the coating's service life is a current focal point and challenge. Zhang et al. [67] improved the wear resistance of coatings by coordinating the application of primer and topcoat. The coating was prepared by sequentially spray-coating a water-based polyurethane adhesive solution (primer layer) and silica-modified mica (topcoat layer) onto the substrate. The key preparation steps of the topcoat layer are as follows: The PAL@fluoro-POS suspension was prepared through HCl-catalyzed hydrolytic condensation of PFDTES and TEOS in the presence of PAL in water. In the acidic aqueous solution, the ethoxy groups of PFDTES and TEOS hydrolyze immediately, generating hydroxyl groups. Consequently, these silanes can easily couple to the surface of PAL through the condensation of their hydroxyl groups. In wear tests conducted under various loads, the surface retains a well-preserved micro/nanostructure even after 100 cycles of abrasion at 10.5 kPa. This is primarily attributed to the superamphiphobic coating, which serves as the topcoat layer, undergoing random settling, thereby forming a denser micro/nanostructure. Song et al. [68] utilized a combined approach of electrodeposition and spray coating to prepare a non-fluorinated dual-layer micro-arc oxidation superhydrophobic coating. At the lowest frequency, its impedance modulus increased by more than six orders of magnitude, significantly surpassing the values of the original substrate. The corrosion inhibition efficiency reached 99.999%. The enhancement of surface wear resistance and corrosion resistance is attributed to the synergistic effects between the micro-arc oxidation layer and the superhydrophobic layer. The oxide layer formed by the micro-arc oxidation enhances the corrosion resistance of the metal surface, slowing down the oxidation and corrosion of the metal. The hydrophobic nature of the superhydrophobic layer reduces liquid contact on the surface, decreasing the liquid's erosion on the metal surface, thus synergistically improving overall corrosion resistance. This dual-layer coating can maintain its superhydrophobicity even after 180 cycles of wear testing. With the gradual application of superamphiphobic coatings, researchers have been seeking methods to prepare coatings with higher wear resistance while maintaining their superamphiphobic properties. Zhang et al. [58] innovatively applied the concept of “soft overcoming rigid” to create a superamphiphobic composite coating with flexibility and wear resistance. This innovative strategy

combines a flexible polyurethane resin with hard diatomaceous earth/aluminum hydroxide (core-shell) particles, forming a coating structure reminiscent of karst topography. The obtained core-shell coating still exhibits outstanding oleophobicity after 800 Taber wear cycles and 1000 cycles of 360° bending under a 250 g load. Simultaneously, it demonstrates excellent wear resistance and flexibility. It follows that the enhancement of wear resistance in corrosion-resistant superamphiphobic coatings can be approached from the following perspectives: (i) employing a primer plus topcoat strategy for layer-wise spray application of adhesives and superamphiphobic coatings to enhance the coating's wear resistance; (ii) combining a soft adhesive with a rigid fluorinated shell to construct a coating surface with a karst-like structure, protecting the fluorinated coating. When the coating is subjected to external forces, the external protective layer is preferentially consumed, preserving the superamphiphobic capability even after the coating undergoes compressive damage. (iii) Utilizing various methods for composite coating preparation to leverage synergistic effects between different preparation techniques, thereby extending the coating's service life.

3.3. Adhesion Performance

Due to the complexity of corrosive environments, the intrusion of corrosive media into coating gaps can cause coatings with poor adhesion to transition from the "Cassie-Baxter" to the "Wenzel" state, leading to a loss of corrosion resistance. Enhancing adhesion in corrosive environments has become a focal point of research efforts. Pan et al. [69] utilized water-based polyurethane (WPU) and fluorosilica (F-SiO₂) nanoparticles to prepare a WPU/F-SiO₂ superhydrophobic composite coating. To prepare the F-SiO₂ nanoparticle suspension, the following steps were undertaken: (i) TEOS (1.5 mL), along with a specific amount of ammonia-water and high-purity water, was dissolved in 50 mL of ethanol. (ii) The solution was stirred vigorously at 40 °C for 12 h. (iii) After the initial stirring, 1.0 g of FAS was added, and the solution was stirred for another 12 h. (iv) The reaction mixture was then heated to 75 °C for 2 h. (v) Finally, the obtained mixture was ultrasonicated for 30 min to produce a homogeneous and stable F-SiO₂ nanoparticle suspension. The water-based polyurethane emulsion provided stable, strong adhesion for the modified nanoparticles, and in the adhesion test, the adhesion grade of the WPU/F-SiO₂ composite coating to the PC substrate reached 5B. Even after 250 Taber wear cycles, the coating maintained a significantly high water contact angle ($159.2 \pm 0.8^\circ$). Qiao et al. [70] created a dispersion solution for a PTFE/SiO₂@CTMS&Na₂SiO₃-ATP superamphiphobic coating by mechanically dispersing polytetrafluoroethylene emulsion (PTFE), modified silica emulsion (SiO₂@CTMS), sodium silicate (Na₂SiO₃), and modified aluminum metaphosphate (modified ATP). The excellent dispersion capability prevents layering within 180 days. Coatings prepared employing this approach can be immersed for 120 h in simulated strong corrosive environments (hydrochloric acid solution and potassium hydroxide solution) without undergoing any morphological changes. Zhao et al. [71] employed a hydrolysis-condensation reaction to cross-link Si-O-Si bonds with SiO₂/TiO₂ particles and a self-made cross-linking agent (AG). This process connected acrylic ester copolymer (FHA), SiO₂/TiO₂ nanoparticles, and the self-made cross-linking agent (AG) to form a grid structure. An 84 h immersion experiment indicates that the FAFH-SiO₂/TiO₂ coating exhibits excellent chemical stability when exposed to hydrochloric acid (pH = 1), toluene, ethanol, and acetone. This suggests the preparation of a coating with strong corrosion resistance. The underwater superoleophobicity of the coating is typically achieved through underwater superhydrophilicity, but this approach may reduce the adhesion capability of both the substrate and the coating. The amphiphilic polyurethane coating designed by Yu et al. [72] can achieve strong underwater adhesion on various substrates while maintaining superoleophobicity. Its working principle involves a mixture of amphiphilic polyurethane and water-soluble solvents immersed in water. The hydrophobic segments aggregate on the substrate surface through a directional mechanism induced by water permeation, resulting in powerful adhesion. At the same time, the hydrophilic segments undergo physical cross-linking to form a hydrogel

coating, imparting the substrate with underwater superoleophobicity, as shown in Figure 5. By summarizing previous research, we roughly categorize the commonly used research methods into two types: (i) Enhancing the adhesion between the coating and the substrate, introducing high-performance adhesives, and reinforcing the surface microstructure to provide a certain buffering effect under external impact. Commonly used adhesives are polyurethane [73,74], polytetrafluoroethylene [75], and polysiloxane [76]. (ii) Improving the binding interface between organic solvents and modified particles. Due to the poor solubility of modified nanoparticles in organic polymers, phase separation and nanoparticle agglomeration are likely to occur. This can lead to the rapid loss of adhesion and, consequently, the protective effect of the coating on the substrate in corrosive environments. Controlled induction of phase separation can significantly enhance the binding capacity between organic adhesives and modified particles.

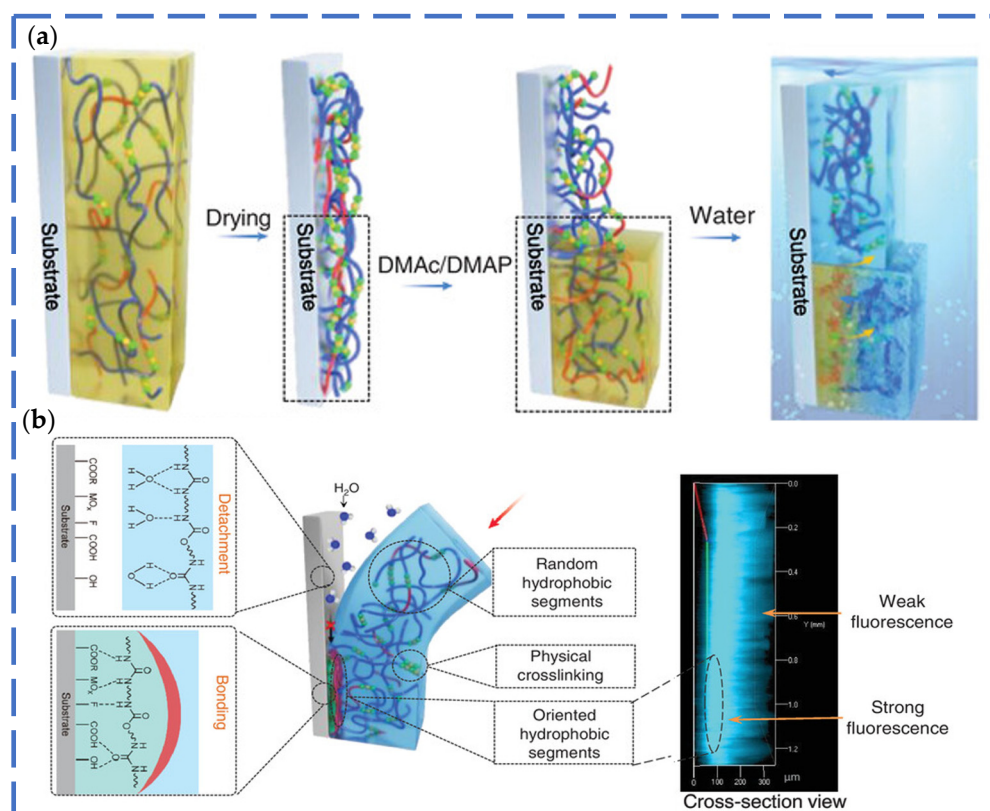


Figure 5. (a) Schematic diagram of the solvent displacement process and the directional mechanism of the hydrophobic/hydrophilic segments; (b) three-dimensional and cross-sectional images of the PCAPU coating obtained by laser scanning confocal microscopy (LSCM) [56].

3.4. Antibacterial Performance

In the face of increasingly complex metal service environments, traditional organic coatings have little resistance to microbial corrosion behavior. Superhydrophobic coatings, as an innovative solution to prevent bacterial adhesion, have attracted increasing attention [77]. Zhang et al. [78] prepared a corrosion-resistant superhydrophobic film on AZ91D magnesium alloy, which exhibited a contact angle of 155° and a low sliding angle of about 2° . First, the pre-treated magnesium alloy is placed at an angle of approximately 60 degrees inside a Teflon-lined stainless steel autoclave and immersed in 9 mL of a 2 mol L^{-1} NaOH aqueous solution. The autoclave is then sealed and maintained at 160°C for 4 h before naturally cooling to room temperature. After the hydrothermal reaction, the sample is submerged in a 0.01 mol L^{-1} ethanolic stearic acid solution and kept at 50°C for 2 h. Subsequently, it is removed from the solution, dried using hot air, and the superhydrophobic film is obtained. Cytotoxicity tests indicated that bacteria tended to adhere

to hydrophilic/superhydrophilic surfaces. On the other hand, the presence of trapped air in the microstructure of the superhydrophobic surface reduced the adherent area. These two primary factors enable the superhydrophobic film to resist bacterial adhesion and demonstrate excellent corrosion resistance in subsequent Hank's solutions. Han et al. [79] designed a superhydrophobic coating using nanodiamonds and siloxane–acrylic resin, demonstrating excellent antibacterial adhesion performance. After culturing *Escherichia coli* for 24 h in the environment, the bacterial colonies on the superhydrophobic coating samples decreased by 73% compared to the blank samples. Electrochemical impedance spectroscopy measurements showed an impedance value of $106 \Omega \text{ cm}^2$ after four hours of immersion, indicating corrosion resistance in the initial hours of immersion. This is mainly attributed to the nano-scale roughness and low-surface-energy substances on the coating surface, hindering the radial flow during bacterial formation, reducing the contact area between cells and the substrate, and minimizing the growth of biofilms [80]. Cui et al. [81] prepared a superhydrophobic antibacterial $\text{SiO}_2@\text{POS}/\text{N}^+$ composite coating, which exhibits outstanding protective performance (99.98%), mechanical properties, and corrosion resistance (21 days). It is noteworthy that the $\text{SiO}_2@\text{POS}/\text{N}^+$ composite material shows excellent antibacterial effects against *Staphylococcus aureus*, achieving a 100% antibacterial effect. This is mainly attributed to hexadecyl trimethyl ammonium bromide, a quaternary ammonium salt, which disrupts the cell wall by contacting the negatively charged cell membrane through N^+ ions, resulting in a bactericidal effect. The prepared coating offers a multifunctional composite coating that is both anti-adhesive and antibacterial. To enhance the antibacterial capability of superhydrophobic coatings, the focus can be directed towards the following two aspects: (i) the construction of nano-scale microstructured surfaces to regulate the surface energy of the coating, hindering radial flow during bacterial formation, and reducing the contact area between bacterial colonies and the coating surface; and (ii) functionalizing superhydrophobic coatings with antibacterial agents such as quaternary ammonium or phosphonium salts to achieve antibacterial and bactericidal effects (Table 2).

Table 2. Comparison of the antibacterial effects of different types of bactericides [82].

Step	Low-Molecular-Weight Biocides	Polymeric Biocides	Dendritic Biocides
Initial adsorption	Low	High	High
Binding to the membrane	Low	Medium	High
Diffusion past the cell wall	High	Low	Medium
Disruption of the membrane	Low	Medium	High

3.5. Self-Repairing Performance

The unique wettability of superomniphobic surfaces enables a wide range of applications, but their relatively short lifespan hinders practical use. Inspired by the self-repairing behaviors in nature, researchers have developed self-healing superomniphobic surfaces to enhance durability and extend their service life [83]. Pan et al. [84] manufactured superhydrophobic aerogels using silver nanoparticles and octadecylamine-decorated alginate-based aerogels. Once the aerogel is scratched or cut, it automatically repairs the layered structure and its structural integrity through wetting or heating processes, restoring superhydrophobicity and mechanical performance. Even after oxygen etching, its superhydrophobic ability can be restored through simple heat treatment. The robust reparability is attributed to the dynamic coordination of catechol–iron and the thermally induced rearrangement of octadecylamine molecules. Sun et al. [85] prepared superamphiphobic coating materials using kaolin, tetraethoxysilane, 1H,1H,2H,2H-perfluorooctyltriethoxysilane, and polydimethylsiloxane. First, the surface of kaolin was modified through a hydrolysis reaction involving ethyl orthosilicate (TEOS) and 1H,1H,2H,2H-perfluorodecyl triethoxysilane (PFDTES). Then, the ZnO antibacterial agent was synthesized in situ using a hydrothermal method. Finally, a durable superamphiphobic coating was achieved by physically blending a polydimethylsiloxane (PDMS) solution with superamphiphobic powder. Experimental results

indicate that perfluorooctyltriethoxysilane migrates from the interior to the surface upon heat exposure, significantly improving the superamphiphobic performance of the coating surface and enhancing its self-healing properties. This was achieved through biomimicry inspired by marine biological skin. Wu et al. [86] first demonstrated the self-assembly of layered microgel spheres modified with hydrophilic polymer chains, achieving an underwater superoleophobic and antibiofouling coating with self-healing properties. Due to its unique three-dimensional ordered structure and hydrophilic components, the surface material based on MHMS exhibits excellent underwater oil resistance and antibiofouling performance. As the surface material is assembled from microgel spheres modified with hydrophilic polymer chains, when the coating undergoes mechanical damage in terrestrial or marine environments (such as abrasion, impact, pressure, etc.), the microgel spheres in the coating expand. Simultaneously, the hydrophilic polymer chains attached to the spheres can replenish the lost hydrophilicity on the liquid/solid interface, as shown in Figure 6. This approach ensures that both the surface structure and hydrophilic polymer chains of the coating can self-heal, significantly extending the coating's lifespan. The above work provides two main directions for extending the service life of coatings: (i) By designing self-responsive repair mechanisms in different environments, utilizing the excellent adsorption capacity of aerogels to carry out dynamic coordination repair with catechol–iron, or using the temperature sensitivity of fluoroalkylsilane to induce migration to specified locations for automatic repair of damaged areas; (ii) by leveraging the tunability and self-assembly capability of microgel spheres, introducing self-healing mechanisms to prolong the service time of coatings.

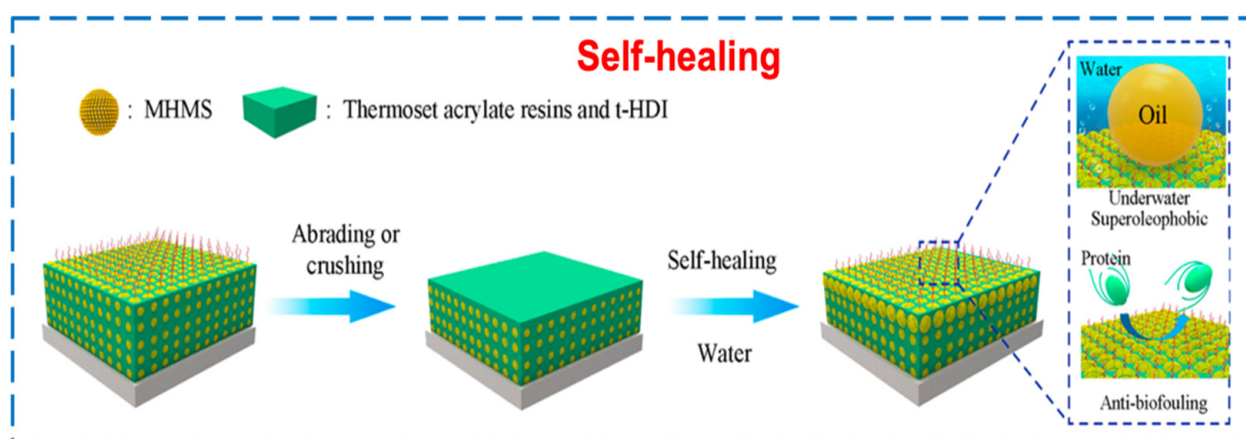


Figure 6. Working principle of a self-healing underwater superhydrophobic/antifouling coating [69].

4. Summary and Recommendations

Superoleophobic coatings, due to their unique wettability, not only serve as protective films between the corrosive medium and the substrate but also actively repel liquids, swiftly carrying away corrosive agents to reduce dwell time. The micro/nanostructures on the coating surface, constructed to leverage the labyrinth effect, effectively delay the onset of corrosion. This makes them highly promising in the field of protecting metal substrates. The construction of superoleophobic surfaces necessitates both low-surface-energy materials and micro/nanostructures on the surface; neither of these elements can be omitted. However, the design principles of superoleophobic coatings are in conflict with their mechanical performance. Increasing the roughness of a superoleophobic surface unavoidably compromises its wear resistance, and enhancing wear resistance requires more curing agents. The cross-linking action of curing agents forces a reduction in the micro/nanostructures on the coating surface. The loss of the structure that supports the air shield leads to a decrease in superoleophobic ability, thereby affecting the coating's corrosion resistance. Therefore, there is an urgent need for a preparation method that can balance superoleophobic performance with excellent mechanical properties. The self-repair

ability of coatings after corrosion is a key means to extend their service life. However, most superoleophobic coatings exhibit slow repair rates in the absence of external stimuli. The use of slow-release repair agents faces challenges such as uneven dispersion and limited repair cycles, restricting their self-repair capability. The use of toxic fluorinating agents also hinders the development of superoleophobic coatings in large-scale production. This review outlines various strategies for enhancing the liquid repellency, durability, adhesion, antibacterial properties, and self-healing capabilities of superhydrophobic coatings. However, the absence of experimental data prevents the establishment of a universally applicable preparation method for these coatings, making it challenging to assess and compare the cost-effectiveness of different preparation techniques. Therefore, future research directions include (i) developing a “decoupling mechanism” to enhance various properties of superoleophobic coatings; (ii) exploring self-responsive repair mechanisms to prolong the corrosion resistance of superoleophobic coatings; and (iii) seeking non-toxic and environmentally friendly low-surface-energy materials to replace fluorides in the preparation of corrosion-resistant superoleophobic coatings.

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