

Review

Preparation and Applications of Superhydrophobic Coatings on Aluminum Alloy Surface for Anti-Corrosion and Anti-Fouling: A Mini Review

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Abstract: Aluminum alloy is widely used in many fields for its excellent performance. However, in practical application, aluminum alloy is easy to become corroded and be invalidated, owing to the influence of environmental factors. Some pollutants or corrosive media on its surface adhesion will further deepen the failure rate of aluminum alloy, resulting in a large amount of workforce waste, waste of material and financial resources, and energy consumption. So the anti-corrosion and anti-fouling treatment of aluminum alloy surfaces to expand their practical application range and reduce energy loss are of great significance. In this paper, we first summarize the types of anti-corrosion and anti-fouling coatings on aluminum alloy surfaces. We found that superhydrophobic coating has attracted widespread attention recently because of its surface's non-wetting and low surface energy characteristics. The superhydrophobic coating refers to a coating with a water contact angle on its surface greater than 150° and a sliding angle of less than 10°. Preparing superhydrophobic coatings on the surface of aluminum alloy can improve the corrosion resistance and anti-pollutant adhesion ability at the same time. Therefore, in the following part of the review, the preparation methods of superhydrophobic coatings on aluminum alloy surfaces for anti-corrosion and anti-fouling and the significant problems encountered in the practical application of the superhydrophobic coating on aluminum alloy surfaces, such as poor anti-corrosion durability and poor mechanical stability, and the existing methods to solve these critical problems, are summarized and discussed in detail. Finally, we put forward some new solutions to solve these vital issues.

Keywords: aluminum alloy; superhydrophobic coating; anti-corrosion; anti-fouling; durability



Citation: Zhu, Q.; Du, X.; Liu, Y.; Fang, X.; Chen, D.; Zhang, Z. Preparation and Applications of Superhydrophobic Coatings on Aluminum Alloy Surface for Anti-Corrosion and Anti-Fouling: A Mini Review. *Coatings* **2023**, *13*, 1881. <https://doi.org/10.3390/coatings13111881>

Academic Editor: Mariola Robakowska

Received: 13 October 2023
Revised: 30 October 2023
Accepted: 31 October 2023
Published: 1 November 2023



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1. Introduction

Aluminum and its alloys are widely used in aviation, military, transportation, decoration, and other fields owing to their light weight, good thermal conductivity, electrical conductivity, flexibility, and recyclability [1,2]. At the same time, aluminum is rich in crustal reserves, ranking third in annual production and second only to steel [3,4]. However, a large number of existing studies and applications have found that, due to the influence of external environmental factors (such as high temperature, high humidity, or a marine environment containing high content of corrosive particles), aluminum and its alloys will undergo local corrosion phenomena such as pitting corrosion, stress corrosion cracking, intergranular corrosion, and layered corrosion during their service time [5], resulting in a shortened service life and limited application range. Furthermore, the adhesion of some corrosive media or pollutants to the surface of the aluminum and its alloys will also lead to their accelerated corrosion and affect their service performance, such as increasing the running resistance and weight of vehicles, increasing energy consumption, etc. [6]. Therefore,

carrying out surface anti-corrosion and anti-fouling treatment of aluminum and its alloys is enormously significant.

In recent years, superhydrophobic coatings have attracted widespread attention because of their unique low surface energy characteristics and non-wettability to aqueous media. Surface wettability is one of the characteristics of solid surfaces, and the static water contact angle (WCA) is defined as the included angle between the liquid–air and the solid surface [7]. When the $WCA > 90^\circ$, the solid surface exhibits hydrophobicity. At the same time, the water droplets are repelled by the solid surface and shrink into a spherical shape. Apart from WCA, the sliding angle (SA) is another criterion for describing the behavior of solid surfaces. The SA is defined as the inclination angle of a solid surface when water droplets first roll. Generally, the characteristics of superhydrophobic surfaces are $WCA > 150^\circ$ and $SA < 10^\circ$. In addition, the wetting behavior of solid surfaces has been deeply studied. Three wetting models exist: Young’s, Wenzel, and Cassie–Baxter [8]. Their diagrammatic sketches are shown in Figure 1, and θ is the contact angle and γ_{SA} , γ_{SL} , and γ_{LA} represent the interfacial tension at the solid–air, solid–liquid, and liquid–air interfaces, respectively. Young’s model describes droplets on an ideal smooth, solid surface. Wenzel’s model, rather, describes droplets on a rough solid surface and assumes that the droplets fully penetrate the groove. However, both of the above assumptions are uniform infiltration states. The droplet on the solid surface, explained by the Cassie–Baxter model, is a heterogeneous wetting state, because the air is trapped in a rough groove below the droplet, preventing it from penetrating the groove and forming an air cushion. The preparation of superhydrophobic coatings on the surface of aluminum alloys can not only effectively prevent direct contact between the corrosive medium and the matrix but also significantly shorten the residence time of pollutants or corrosive media on its surface, and inhibit the formation of continuous electrolyte film on the surface of the coating, thereby playing the dual role of anti-corrosion and anti-fouling (self-cleaning) on the aluminum alloy substrate [9–11].

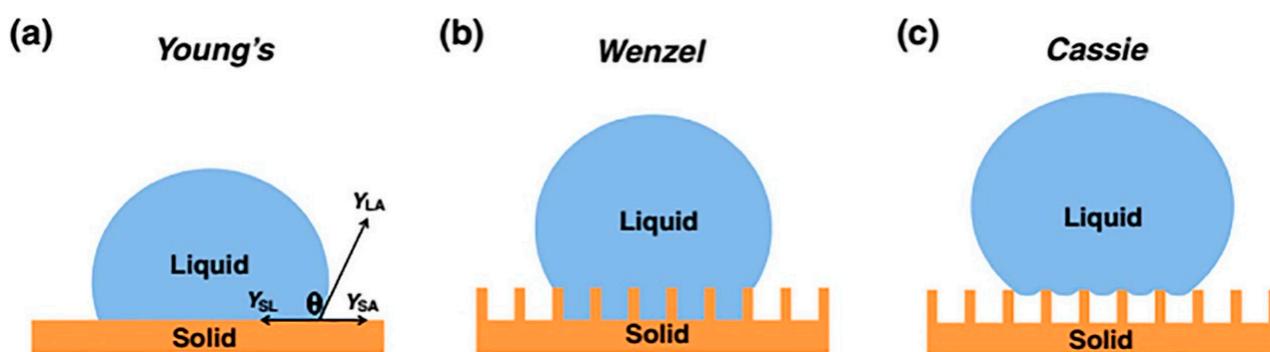


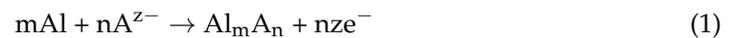
Figure 1. A liquid droplet on different surfaces: (a) Young’s model, (b) Wenzel’s model and (c) the Cassie–Baxter model [8].

In this review, we first summarized the types of anti-corrosion and anti-fouling coatings on aluminum alloy surface and their advantages and disadvantages. It is found that the superhydrophobic coatings not only have an excellent anti-corrosion effect on the aluminum substrate but also can effectively remove the stain on the surface of the substrate and play a self-cleaning role. Therefore, the following part of the review will mainly focus on the preparation methods and application status, such as the significant problems encountered in the practical application of the superhydrophobic coating on aluminum alloy. The existing strategies to improve the performance of anti-corrosion and anti-fouling superhydrophobic coatings will also be discussed in detail. Finally, we will propose new ideas or methods to enhance the comprehensive properties of superhydrophobic anti-corrosion and anti-fouling coatings on aluminum alloys.

2. Coating Types on the Surface of Aluminum Alloy for Anti-Corrosion and Anti-Fouling

2.1. Chemical Conversion Film Coatings

Preparing a protective film or coating on the surface of aluminum alloy can effectively improve its durability. The inorganic conversion coating is one of the protective coatings which has been studied for decades. Its preparation process is mature and has been widely used. The chemical conversion coating of aluminum alloy is a stable and sound adhesion of a thin layer formed by the chemical or electrochemical reaction between anions in solution and the aluminum or alumina substrate surface [12]. The following reaction equation can represent the typical reaction:



In the above reaction equation, A^{z-} represents the anions with a valence state of $-z$ in the medium. During recent decades, Chromate conversion coatings (CCC) have been widely used in aerospace due to their advantages, such as good corrosion resistance, simple preparation process, and low cost. When the conversion film is damaged, Cr (VI) in the film layer will transfer to the broken place to passivate the substrate, thereby effectively protecting the substrate material [13]. However, Cr (VI) is carcinogenic and environmentally unfriendly and has been restricted by many countries, so finding an alternative coating for CCC is urgent [14–16]. The trivalent chromium process (TCP) is one of the replacements. Kim et al. [17] put 3003 aluminum alloy in hexafluoro-zirconate solution containing trivalent chromium for 18 min to obtain a conversion coating. Then, immerse the above conversion coating in 3.5 wt.% NaCl solution for 24 h, or perform a 24 h simulated acid rain test, expressing good corrosion resistance. We are cautious about whether TCP will replace CCC as an environmentally friendly conversion coating. As Peltier et al. [14] mentioned, the conversion baths generally contain Cr^{3+} salts, fluoride (ZrF_6^{2-}), and H_2SO_4 or NaOH . Therefore, Cr (III) may be oxidized to Cr (VI) during production.

Except for CCC, there is also a traditional conversion coating called phosphate conversion coating (PCC). The formation of PCC is mainly divided into two parts: the dissolution of Al substrate and surface alumina and the deposition of insoluble phosphate [18]. Huang et al. [19] prepared PCC on the surface of 2A12 Al alloy by chemical conversion. The corrosion current density of PCC is three orders of magnitude lower than that of the 2A12Al matrix after immersing the PCC in 3.5 wt.% NaCl solution for 30 min, and the corrosion inhibition efficiency of PCC can be as high as 99.91%. The highest oxidation state of transition metals can form cations in aqueous solutions, which can be reduced by electrochemical reactions to form insoluble oxides, similar to the formation of chromate conversion coatings [18]. Therefore, permanganate (MnO_4^-), molybdate (MoO_4^{2-}), and vanadate (VO_4^{3-}) conversion coatings are also trends in chemical conversion coatings. Janqour et al. [20] prepared a vanadium-based conversion coating by immersing 2024 aluminum alloy in sodium vanadate salt. They found that the conversion coating markedly enhanced the corrosion resistance of 2024 aluminum alloy. Even after soaking the vanadium-based conversion coating in 3.5 wt.% NaCl solution for 15 min, the corrosion current density (I_{corr}) of vanadium-treated 2024 aluminum alloy was still lower by one order of magnitude than that of untreated, while the corrosion potential (V_{corr}) and the charge transfer resistance (R_{ct}) of the vanadium-treated were still more positive and larger than the untreated, respectively. Dmitry et al. [18] indicated that alumina's catalytic and poor conductivity inhibits the reduction of V^{5+} adsorbed on the surface of aluminum alloys, playing a corrosion-inhibiting role.

In addition to the alternative mentioned above to hexavalent chromate conversion coatings, cerium ions show an inhibitory effect on the corrosion of aluminum alloys, so rare earth conversion coatings have also become an effective surface anti-corrosion coating for aluminum alloys. It is reported that rare earth metal ions, such as Ce, La, Pr, Nd, and Y, have a specific resistance to the local corrosion of aluminum alloy by forming an insoluble hydroxide/oxide layer [21]. Valdez et al. [22] immersed 6061-T6 aluminum alloy in cerium

chloride solution to obtain a cerium-based conversion coating. Then, soak the coating in 3 wt.% NaCl solution for one hour; the coatings' I_{corr} , being $1 \times 10^{-7} \text{ A}\cdot\text{cm}^{-2}$, was almost two orders of magnitude lower than that of the bare aluminum alloy ($5.1 \times 10^{-6} \text{ A}\cdot\text{cm}^{-2}$). At the same time, the R_{ct} of the cerium-based was much higher than that of the untreated. Although conversion coatings have the advantages of simple preparation, low cost, and large-scale application in industrial production, there is currently no aluminum alloy chromium-free conversion coating comparable to chromate conversion coatings since Cr (VI) was banned.

In addition, the film layer, which is composed of layered double hydroxides (LDH) on the metal substrate surface, exhibits an excellent physical barrier effect and interlayer anion exchange on the metal substrate (such as aluminum and its alloys) due to its unique layer structure and ionic composition [23], thereby blocking or trapping aggressive anions (such as Cl^-), so that the aggressive ions cannot be in direct contact with the substrate metal to achieve an anti-corrosion effect. Moreover, due to the weak interaction between the anions and the interlaminae of LDH films, interlayer anions can also be exchanged by corrosion inhibitor ions to prepare LDH films containing corrosion inhibitor ions. Then, the modified LDH can release corrosion inhibitor ions when the surface film layer erodes, slowing the corrosion process of substrate metals [24]. Figure 2 shows the exchange process of corrosion inhibitor ions in the LDH film layer. He et al. [25] prepared a Zn-Al LDH- VO_x film with vanadium ion intercalation on the surface of AA2024 aluminum alloy by combining electrodeposition and ion intercalation. The experiment found that the exposed AA2024 aluminum alloy suffered severe corrosion after being immersed in 3.5 wt.% NaCl solution for two hours. However, the aluminum alloy covered with Zn-Al LDH- VO_x film did not have noticeable corrosion pits even after one month of immersion, which indicated that, compared with the traditional hydrothermal method, the LDH film prepared by electrodeposition is thicker, but it has better corrosion resistance. Besides, there are also studies [11] using a hydrothermal method and ion exchange method to obtain methionine-modified Al-Li-Met LDH film on the surface of 6063 aluminum alloy, which found that this film layer reduces the corrosion current density of aluminum alloy in 3.5 wt.% NaCl by one order of magnitude compared with blank aluminum alloy. However, it can also automatically restore its original morphology and structure after external mechanical scratches, showing particular self-healing performance and improving its corrosion protection durability.

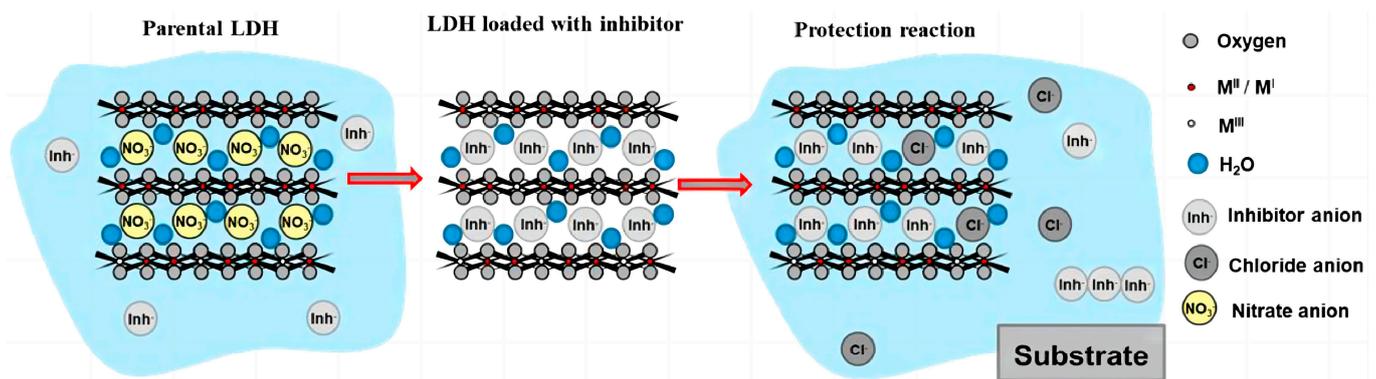


Figure 2. Structure of LDH, exchange of anion and corrosion inhibitor ion, and schematic diagram of corrosion prevention triggered by the release of corrosion inhibitor ion [24].

2.2. Anodizing Film Coatings

In the natural environment, aluminum is easy to passivate with oxygen to form a raw alumina film due to its chemical activity, which can prevent further contact oxidation between the aluminum substrate and the atmosphere, thereby reducing the corrosion rate of the substrate. The stability of the naturally formed alumina oxide film is very high in the service environment, with a pH of 5–8. Still, it is not ideal in extreme environments

(such as strong acids, alkali, and ultraviolet irradiation) [26]. Therefore, to improve the anti-corrosion stability of the alumina film in most environments, an anodic oxidation method has been adopted to fabricate the oxide film on the surface of aluminum and its alloys. Anodizing is an electrochemical process in which aluminum oxide grows on the aluminum anode plate through current action, continuously consuming the aluminum anode. Figure 3 shows a schematic diagram of the anodic oxide film, which has a self-assembled and highly ordered hexagonal cell structure in which a hole in the center of each cell structure extends from the substrate to the top. Still, the part of the hole bottom connecting the substrate is closed [27]. Since most of the surface of the anodized alumina film is porous, the corrosive medium can readily enter the film through these pores, thus affecting the protective performance of the entire film layer. Given the problem of the film's poor corrosion resistance due to the porous structure of the alumina film layer, there are two leading solutions. One is to change the surface structure of the film layer by changing the anodic oxidation parameters during the preparation process [28] to improve the film layer's compactness. The other is to seal the resulting film layer after the end of anodizing [29] to reduce the penetration of the corrosive medium into the film layer and ultimately provide the corrosion resistance of the film layer. However, the current mainstream research direction is more inclined to the latter because adding sealing materials gives aluminum or aluminum alloys more diverse functions.

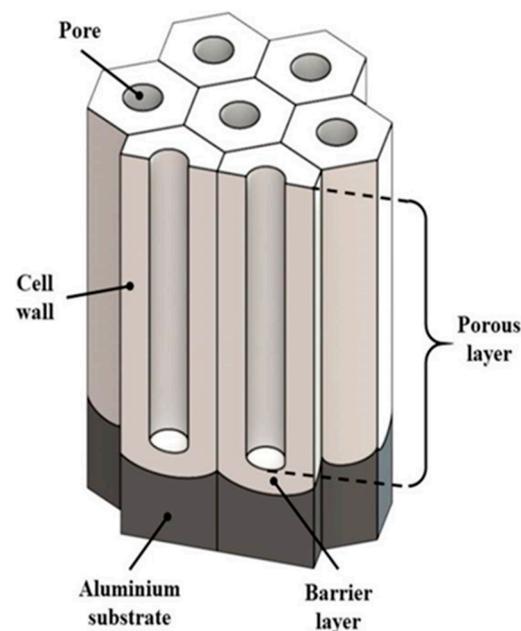


Figure 3. Schematic diagram of hexagonal cell structure in anodized liner substrate [27].

Khan et al. [30] anodized 2024 aluminum alloy, followed by pore sealing treatment using boiling distilled water. The results showed that the corrosion current density of the anodic oxide film on the surface of 2024 aluminum alloy is reduced by four orders of magnitude compared with the blank 2024 aluminum alloy in 3.5 wt.% NaCl solution, indicating an excellent anti-corrosion property of the anodic oxide film. Yu et al. [31] anodized 6061 aluminum alloy in a citric acid–sulfuric acid system and then sealed the hole with a potassium dichromate solution. It is found that, after soaking in 3.5 wt.% NaCl solution for nine days, the corrosion current density of the aluminum alloy samples treated by anodization and hole sealing decreased by two orders of magnitude compared to blank aluminum alloys. Also, it fell by one order of magnitude compared with the samples only undergoing anodization, indicating that post-sealing treatment can further improve the corrosion resistance of the film layer.

2.3. Organic Painting Coatings

At present, the anti-corrosion organic coatings for aluminum alloy mainly include alkyd resin coatings, phenolic resin coatings, epoxy resin coatings, polyurethane coatings, polyvinyl alcohol coatings, perchloroethylene resin coatings, furan resin coatings, organo-silane coatings, etc. [32–35]. There are five main anti-corrosive paths for organic coatings on aluminum alloys: [36,37]: (1) the organic coatings' simple shielding effect on the substrate; (2) the coatings' slow-release impact on the passivation of the substrate; (3) the enhanced wet adhesion between the organic film and the substrate; (4) the increased circuit resistance between the coating and the substrate; (5) adding metal powder (such as zinc [37]) to the paint plays a cathodic protection role at the expense of the anode. Ge et al. [38] sprayed epoxy resin primer containing aluminum tripolyphosphate (ATP) and polyurethane topcoat sequentially on the surface of 5083 aluminum alloy to form a double-layer organic coating. Its charge transfer resistance increased ten times compared with blank aluminum alloy after soaking in 3.5 wt.% NaCl solution for 15 days. It was found that there were no bubbles and evident rust around the scratches after exposing the scratched coating to a neutral salt spray environment for ten days, indicating that the organic coating could act as an effective barrier against the corrosion of aluminum alloys. Gad et al. [39] pointed out that, when the surface of 2198-T851 aluminum alloy coated with epoxy resin was sprayed with a layer of vinyl chloride (PVC) coating containing cerium ion further, after five weeks of exposure to a salt spray environment, the modified coatings' charge transfer resistance increased by two orders of magnitude. The increased charge transfer resistance means that adding cerium ions provides an idea for improving the durability of the coating.

However, aluminum and its alloy products will not only be corroded by environmental media during actual use but also become fouled due to the adhesion of environmental pollutants, such as microorganisms, dust, or other contaminants. For example, ships serving in the ocean often slow their operation and increase energy consumption due to the attachment of marine organisms. As microorganisms grow, they will destroy the hull's anti-corrosion coating, accelerating its corrosion. Thus, improving the anti-pollutant adhesion ability of aluminum and its alloy surface is also essential to enhance its protection ability and expand its application range. Currently, widely used self-polishing coatings can release toxic compounds (such as TBT [40] or copper oxide [41]) through the hydrolysis reaction of the resin to achieve the effect of killing microorganisms. But TBT has been restricted worldwide as early as 2008. At the same time, the long-term use of low-toxicity copper oxide also has potential environmental hazards, and the galvanic reaction of copper ions with aluminum or aluminum alloy substrates will affect the anti-corrosion performance [42]. If non-toxic compounds can replace the toxic compounds in self-polishing coatings, the problem of environmental hazards would disappear. Sha et al. [43] prepared a self-polishing anti-fouling coating polymer based on eugenol methacrylate, which solved the problem of microbial attachment by interpreting eugenol with antibacterial and anti-algae properties through phenolic ester-based water. In addition, except for the self-polishing coatings that achieve anti-fouling through chemical reactions, the stain-release anti-fouling films with low surface energy can also make the pollutants challenging to adhere to solid surfaces [44,45]. Zhao et al. [46] prepared a silicone anti-fouling coating containing a fluorinated side chain of natural ice chips, which was able to resist 95.6% of marine bacteria while reducing the coverage area of the biofilm by 86.9%, effectively preventing the adhesion of diatoms.

Finding environmentally friendly organic painting coatings has always been one of the research directions for organic painting coatings on aluminum alloy surfaces, and it is also necessary to address the mechanical durability and thermal stability of organic painting coatings in corrosive environments. Due to the complexity of organic painting coating failure, it is only discovered after the complete failure of the coating. Therefore, a new and effective failure criterion is necessary, which can detect and guide maintenance work in the early stages of coating failure. Based on the low surface energy characteristics of the fouling release coating, the bionic superhydrophobic coating is also an anti-fouling coating that has been studied more often and has a good application prospect. Moreover,

the non-wettability and the unique micro-nano structure of the superhydrophobic layer also provide it with two simultaneous functions: anti-corrosion and anti-fouling.

2.4. Superhydrophobic Coatings

In general, a biomimetic superhydrophobic surface is defined when the static water contact angle (WCA) on the coating surface is greater than 150° , the sliding angle (SA) is less than 10° [7,8], and the Cassie–Baxter model is mainly used to explain the surface-wetting state of existing superhydrophobic coatings. Superhydrophobic coating surfaces with the above wetting properties are often prepared by constructing a graded rough structure and then modifying it with low surface energy substances. When a superhydrophobic surface with a graded rough structure is just immersed in the electrolyte, the air trapped in the hierarchical structure can temporarily leave it in an unstable Cassie–Baxter wetted state, thereby preventing direct contact between the metal matrix and the corrosive medium, reducing the actual contact area between the corrosive electrolyte and the substrate surface, thereby improving the corrosion resistance of the substrate. At the same time, the low surface energy of the superhydrophobic coating can reduce its adhesion to water droplets. When the coating is in contact with an aqueous solution or pollutants, the interaction force between the pollutants is much higher than the adhesion force between the contaminants and the coating. Therefore, the superhydrophobic coating can make it difficult for the polluting liquid–solid mixture to adhere to the surface. The low adhesion between the liquid–solid mixture and the coating surface can effectively shorten the maintenance time of the liquid–solid mix on the surface of the coating, inhibit the formation of a continuous electrolyte film on the surface of the coating, and finally enhance the anti-corrosion and anti-fouling effect of the coating [47]. Additionally, owing to the superhydrophobic coating having a low SA (less than 10°), the liquid contaminants are easy to roll off from the surface and carry away the other solid pollutants during the rolling process to achieve the effect of anti-fouling enhancement. Therefore, bionic superhydrophobic coatings' inherent rough structure and low surface energy can give aluminum or aluminum alloy surfaces good anti-corrosion and anti-fouling (self-cleaning) performance.

Cao et al. [48] prepared layered double hydroxide (LDH) on 5052 aluminum alloy by in situ growth and then impregnated it into stearic acid (STA) to obtain STA–LDH coating. The WCA of the coating obtained was 151° , and the SA was less than 5° . After immersing the sample covered with STA–LDH in 3.5 wt.% NaCl solution for 20 min, the charge transfer resistance increased from $5.105 \times 10^4 \Omega\cdot\text{cm}^2$ (blank aluminum alloy sample) to $8.46 \times 10^6 \Omega\cdot\text{cm}^2$. The corrosion current density also decreased by about two orders of magnitude. The above results indicate that the STA–LDH superhydrophobic coating has an excellent anti-corrosion effect on 5052 aluminum alloy. Li et al. [49] obtained a coating with a WCA of 162° by chemical etching and impregnation modification of 1H, 1H, 2H, 2H-perfluorodialkyltriethoxysilane. The obtained coating's corrosion current and potential were approximately the same as the coating without immersion after immersing the coating in 3.5 wt.% NaCl solution for 72 h. It is not difficult to see that the above studies all adopted a “two-step” method to prepare superhydrophobic coatings. Therefore, to simplify the preparation process of superhydrophobic coatings, the study began to explore the use of a “one-step” method to prepare superhydrophobic coatings. Zhang et al. [50] used perfluorooctanoic acid (PFOA) as raw material to obtain a superhydrophobic coating on the surface of 5083 aluminum alloy by a one-step hydrothermal method, and its WCA could reach $167.2 \pm 2^\circ$. The superhydrophobic modified 5083 aluminum alloy was soaked in a simulated seawater solution, and it was found that the corrosion inhibition efficiency reached 94.45%. At the same time, simple anti-fouling and self-cleaning tests also proved that the coating has water repellency and low surface adhesion.

Rasitha et al. [51] prepared a superhydrophobic coating composed of hexamethyl-disilazane-modified silica and (3-glycyloxy propyl) trimethoxy-silane (GPTMS) on the surface of pure aluminum by a dipping and pulling method. The study shows that the WCA of the superhydrophobic coating can be as high as 170° , and the SA is less than 1° .

The modified aluminum sample's corrosion current density is smaller than the blank pure aluminum sample in 3.5 wt.% NaCl aqueous solution and the corrosion inhibition efficiency is as high as 99%, effectively realizing the aluminum's corrosion protection. Eventually, Table 1 summarizes the different types of coatings on aluminum alloy surfaces and their characteristics, advantages, and drawbacks.

Table 1. Different types of coatings and their characteristics, advantages, and drawbacks.

The types of Coatings	Characteristics	Advantages	Drawbacks	References
Chemical Conversion Film Coatings	A few micrometers in thickness As a connecting layer between the topcoat and the aluminum alloy substrate, it increases the adhesion of the coating.	Low cost Simple production process Wear resistant Good corrosion resistance Suitable for large-scale industrialization	Cr (VI) is poisonous and carcinogenic. Phosphorus-containing wastewater could cause eutrophication of water bodies.	[11–25]
Anodizing Film Coatings	Thickness in micrometers Composed of alumina in the shape of nano-channels Post-treatment for sealing holes or use with coatings.	Good adhesion on the substrate Suitable for large-scale industrialization	Once the coating is damaged, the substrate is corroded	[26–31]
Organic Painting Coatings	Consists of three layers, with the layer in contact with the aluminum alloy being a priming coat, the top layer being a topcoat, and the intermediate coat connecting the priming coat and topcoat. Suitable for substrates of any size and shape.	Good weather resistance Anti-fouling and preventing adhesion of marine organisms Suitable for large-scale industrialization	It may degrade in aqueous solution. Environmentally unfriendly Poor adhesion on the substrate	[32–46]
Superhydrophobic coatings	Non-infiltration. Synergistic effect of micro-nano graded rough structure and low surface energy material modification.	Good anti-corrosion and anti-fouling Self-healing	Poor chemical stability Poor durability	[7,8,47–51]

3. Preparation Methods for Superhydrophobic Coating on the Surface of Aluminum Alloy for Anti-Corrosion and Anti-Fouling

Numerous studies on a natural superhydrophobic surface show that the two essential factors for the design and preparation of a superhydrophobic surface are: (i) the substrate surface has a micro-nano level rough structure, (ii) the substrate surface has low surface energy substance [52]. Generally, there are two main methods for preparing superhydrophobic coatings. One is the “top-down” method, that is, to construct a suitable micro-nano rough structure on the surface of hydrophobic materials (low surface energy substances). The other is a “bottom-up” approach, in which surfaces with micro-nano rough structures are modified with low surface energy substances. Many studies [53–65] have reported several methods for preparing superhydrophobic coatings on aluminum alloy surfaces, including impregnation, spray method, anodization, electrodeposition, etching, and hydrothermal processes.

3.1. Impregnation Method

Impregnation is the most common method for preparing superhydrophobic coatings on aluminum alloys. The impregnation method generally involves immersing the pre-treated aluminum alloy in a soluble compound solution containing active components and then removing it after impregnation for some time. The active ingredients are attached to the aluminum alloy in the form of compounds and result in the superhydrophobic coating on the surface of the aluminum alloy [48,49,51,53–55]. Ma et al. [53] immersed 6061 aluminum alloy in a mixed solution of bis-[3-(triethoxy-silyl)propyl]-tetra-sulfide (BTESPT) and reduced graphene oxide (rGO) and then cured them to obtain a BTESPT/rGO composite film on the aluminum alloy surface. The testing results showed that, after immersing

in 3.5 wt.% NaCl solution for 30 min, the corrosion current density of the aluminum alloy covered with BTESPT/rGO composite film was three orders of magnitude smaller than that of the blank aluminum alloy, and the anti-corrosion efficiency could reach 99.59%. The enhanced anti-corrosion efficiency ascribed to the doping of graphene could effectively cover the micro-pores and micro-cracks in the BTESPT film. However, corrosion appeared on the edge of the specimen on the 7th day of exposure to the neutral salt spray test, and the cracks on the surface began to increase after continued exposure on the 10th day, indicating that the chemical durability of the BTESPT film was still unsatisfactory. Kumar et al. [54] used hydrochloric acid and nitric acid to etch the surface of aluminum alloy. Then they impregnated the surface into cetyl trimethoxy-silane to obtain a superhydrophobic coating, with WCA of $162.0 \pm 4.2^\circ$ and SA of $4 \pm 0.5^\circ$. The coating loses its superhydrophobicity after soaking in strong acid (pH = 2) or strong base (pH = 13) for 10 min and two hours, respectively, which means its chemical stability is not ideal.

The superhydrophobic coating obtained by the impregnation method has unsatisfactory chemical durability, and its mechanical wear resistance is also far from meeting the requirements of practical applications. Lomga et al. [55] prepared a superhydrophobic coating by sodium hydroxide etching and lauric acid impregnation. The coating lost its superhydrophobicity after 15 viscous peel tests. Rasitha et al. [51] prepared a superhydrophobic coating of hexamethyl-disilazane modified silica compounded with (3-glycidioxypropyl) trimethoxy-silane (GPTMS) on the surface of pure aluminum by an impregnation method. In the wear test (ASTM D4060), the coating loses its superhydrophobicity after being loaded with a weight of 50 g and dragged for a distance of 150 cm. The poor durability of the above superhydrophobic coating obtained by the dipping method may be because the bonding force between the superhydrophobic coating and the substrate is mainly through the physical adsorption force and the chemical van der Waals force. The resulting bonding force between the coating and the substrate is often not ideal. As a result, the low surface energy material of the surface layer is readily polished off and loses its superhydrophobic property when external forces wear it. At the same time, most of the low surface energy compounds adsorbed on the surface of the substrate by physical/chemical adsorption are long-chain aliphatic alkane, fluoro-silane, and other mixtures. These compounds always have poor stability in the corrosive medium (usually aqueous solution), and long-term immersion makes it easy for the coating to absorb water and swelling, resulting in the coating falling off from the substrate and losing its original special anti-fouling effect in long-term immersion. Therefore, most of the superhydrophobic films obtained by the impregnation method show poor mechanical wear resistance and chemical stability.

3.2. Spraying Method

The spraying method generally refers to using a spray gun or dish atomizer, with the help of pressure or centrifugal force, to disperse the paint into uniform and fine droplets and then coat it on the substrate surface. Peng et al. [56] prepared a superhydrophobic coating composed of polystyrene microspheres (PS) and polydimethylsiloxane (PDMS) on the surface of aluminum alloy by a simple and effective one-step spraying method. The obtained film has a WCA of $158.1^\circ \pm 0.6^\circ$ and an SA of $5.9^\circ \pm 1.4^\circ$ and also exhibits a certain degree of self-cleaning while resisting the adhesion of a variety of droplets. However, the PS/PDMS coating's SA increases to more than 10° after immersing in neutral and alkaline (pH > 10) solution for one hour and also lost its superhydrophobicity after the coating was heated to 260°C , which indicates that the chemical stability and high-temperature stability of this polymer coating are not ideal. Nevertheless, Gong et al. [57] reported that superhydrophobic coatings prepared by spraying showed better corrosion durability. They sprayed a slurry made of epoxy resin, fluoro-silicone paint, fluorinated polyurethane, and F-SiO₂ nanoparticles on the surface of the 2A12-T4 aluminum alloy to form a coating with a WCA of 161.35° and an SA as low as 2.0° . In addition, after 20 days of neutral salt spray accelerated corrosion test, the corrosion current density of the coating was $5.26 \times 10^{-8} \text{ A}\cdot\text{cm}^{-2}$, which was still four orders of magnitude smaller

than the blank aluminum alloy. The spraying process is relatively simple and can prepare superhydrophobic coatings with good durability on a large scale. However, the spraying method always requires a specific spray gun device, and the fluidity and dispersion of the paint are high. Moreover, due to the limitation of spraying technology, the method is always used to prepare coatings with low surface precision requirements. It is unsuitable for the surface treatment of small precision instruments.

3.3. Anodization Method

In the metal anodization process, metal ions and water react electrochemically in the presence of protons to produce an oxide layer with a micro-nano-level rough structure on the metal surface. However, preparing superhydrophobic coating on an aluminum alloy surface by the anodization method generally requires two steps. Because anodizing treatment can only construct micro-nano structures on the surface of aluminum alloy, further post-sealing treatment or modification with low surface energy substances is needed to make aluminum alloy obtain super hydrophobic anti-corrosion and anti-fouling properties [29]. Zang et al. [58] used a phosphoric acid system to anodize 6061 aluminum alloy. Then, they performed lauric acid modification treatment; the WCA of the coating was about 160° , and the SA was less than 2° . It exhibits an excellent self-cleaning ability, as shown in Figure 4. At the same angle of roll, the water droplets carried away the dust on the superhydrophobic surface, while the dust still on the blank aluminum alloy surface. Simultaneously, the corrosion current density of the aluminum alloy covered with this coating immersed in 3.5 wt.% NaCl solution is three orders of magnitude smaller than the untreated aluminum alloy. The corrosion potential also shifts in a positive direction, indicating that the superhydrophobic coating improves the corrosion resistance of the substrate aluminum alloy. However, the coating lost its superhydrophobicity after rubbing 300 cm on the surface of 2000 mesh sandpaper under 1 N load, which indicates that the mechanical wear resistance of the coating needs to be further improved. Zhang et al. [59] conducted anodic oxidation on the surface of 6061 Al alloy. They investigated the effect of TiO_2 nanoparticles introduction and impregnation with octadecyl-trimethoxy-silane (OTS) modification on the corrosion resistance of the coating. We know that the corrosion resistance of superhydrophobic coatings comes from the presence of an air cushion on the surface of the coating when it comes into contact with the solution, reducing the contact area between the corrosive solution and the substrate. They found that the air cushion gradually disappeared due to the degradation of OTS during the process of immersion corrosion solution, along with the corrosion ion Cl^- attacking the Al substrate.

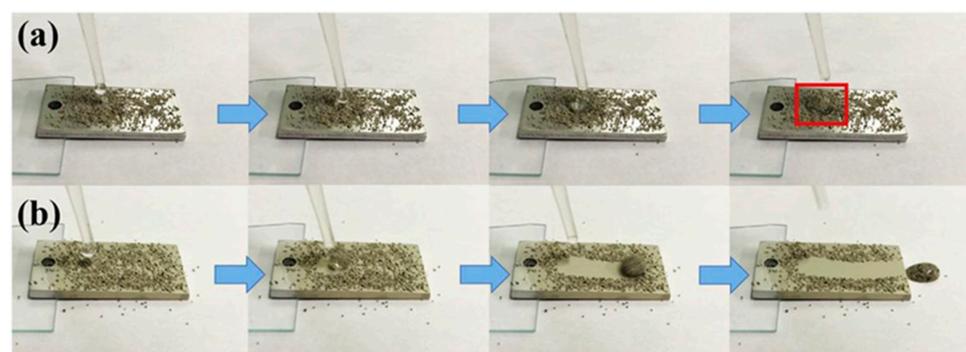


Figure 4. (a) Blank aluminum alloy substrate and (b) self-cleaning process of aluminum alloy surface after superhydrophobic modification [58].

Therefore, the existing research showed that the anodic oxidation method can only obtain a rough micro-nano structure with good mechanical properties on the surface of aluminum alloy. To finally obtain an anti-fouling superhydrophobic coating, combining the anodization method with other surface modification methods, such as the impregnation method and the physical/chemical vapor deposition method, is necessary. The superhy-

drophobic coating obtained by “two-times”-surface modification also has the characteristics of a low surface energy substance and rough structure. However, the low surface energy substance always has low hardness, wear resistance, long-term water immersion resistance, etc., which eventually leads to the anti-corrosion and anti-fouling durability of the coating, which cannot meet the actual application needs.

3.4. Plasma Electrolytic Oxidation Method

With the deepening of the research on anodic oxidation methods in recent years, a technique that can produce high-hardness ceramic coatings on the surface of different metals (such as Al, Mg, Ti, and their alloys), i.e., plasma electrolytic oxidation (PEO), has been developed. The electrolyte used in the PEO method is environmentally friendly, and the equipment is easy to operate. The coating made by PEO also has higher corrosion, wear and oxidation resistance than ordinary anodized coatings [60,61]. Fu et al. [62] obtained a ceramic-based superhydrophobic coating (T-PEO-S) composed of porous ceramic coating and fluorinated silica nanoparticles on the surface of pure aluminum by the PEO and chemical vapor deposition method. The preparation process is shown in Figure 5. The result showed that the T-PEO-S coating has a WCA of up to 160.5° , and the PEO ceramic coating improves the mechanical durability of the superhydrophobic coating. However, the coating's SA is greater than 10° after 14 times of tape stripping tests. When the number of times reaches 20, the hydrophobicity of the coating also disappears, which indicates that the mechanical wear resistance of the superhydrophobic coating is not stable enough due to the poor stability of the outer organic layer. Yang et al. [63] prepared a superhydrophobic alumina coating by PEO technology, and the WCA of the coating was $150 \pm 3^\circ$. Unfortunately, on the second day of the salt spray test, the coating lost superhydrophobicity and only maintained hydrophobicity. Although the PEO method can achieve one-step preparation of superhydrophobic coatings, the durability of superhydrophobic coatings does not seem ideal. In addition, PEO is similar to anodizing, except that the voltage of PEO is higher, resulting in the preparation of inorganic ceramic coatings. Therefore, PEO coatings have higher hardness than anodized coatings, resulting in better wear resistance. Nonetheless, most PEO coatings still require low surface energy substances to seal their pores in order to achieve the superhydrophobic coating we need. Therefore, improving the durability of PEO superhydrophobic coatings is urgent.

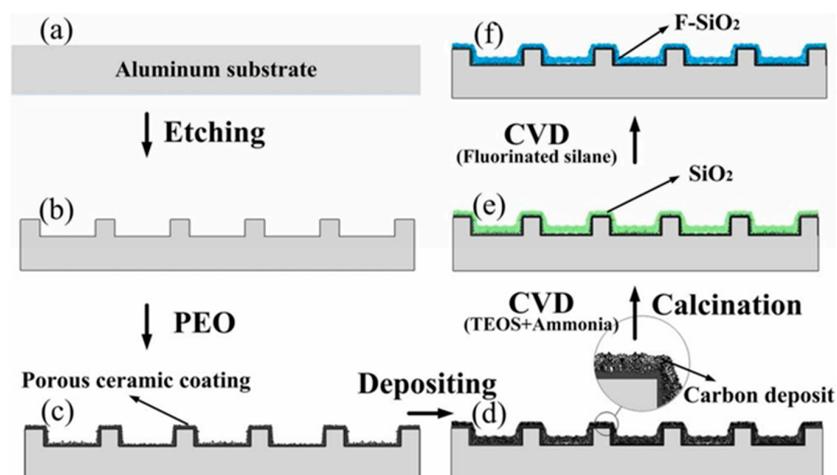


Figure 5. Schematic diagram of the preparation of T-PEO-S superhydrophobic surface: (a) aluminum substrate, (b) etched aluminum substrate, (c) porous ceramic coating, (d) deposited carbon coating, (e) formed hydrophilic silica network and (f) superhydrophobic surface [62].

3.5. Electrodeposition Method

The electrodeposition method refers to the process/method of directional movement of metal ions or composite ions in the electrolyte under the action of the external elec-

tric field. Then, the ions are reduced on the cathode surface to form the corresponding metal or alloy coating. By adjusting the electrodeposition parameters (such as electrolyte composition, pH, temperature, voltage and current, etc.), the formation of micro-nano structures and the thickness of the coating can be easily controlled. Yang et al. [64] fabricated a coating composed of mastoid structures of different sizes by simple electroplating and octa-decanthiol impregnation modification. The WCA of the obtained coating could reach 153.6° . Still, the research did not study the coating's mechanical wear resistance and chemical stability. Zhao et al. [65] prepared a superhydrophobic film (WCA approx. 167°) by electrodepositing Ni-Co alloy coating on the surface of 5052 aluminum alloy in 6-(N-allyl-1,1,2,2-tetrahydroperfluorodialkyl) amino-1,3,5-triazine-2,4-dithiol monosodium (AF17N). The corrosion current density of the aluminum alloy covering this film layer is three orders of magnitude smaller than that of the bare aluminum alloy when immersed in 3.5 wt.% NaCl solution. Also, the aluminum alloy covering this film layer shows a specific self-cleaning effect. However, this study only reported the anti-corrosion and anti-fouling stability of the film layer in the air. It did not conduct in-depth research on the film layer's chemical stability and mechanical wear resistance. The examples above show that the early electrodeposition method synthesized superhydrophobic coatings in two steps. Today, many studies [66–69] have optimized the existing electrodeposition method and proposed that only a one-step process can achieve superhydrophobicity on the surface of aluminum alloys, which we will discuss in the next section.

3.6. Other Methods

The aluminum alloy surface with superhydrophobic properties is widely used in aerospace, marine, construction, automotive, and other fields due to its outstanding anti-corrosion and anti-fouling properties. However, diversifying application fields leads to different functional requirements for the aluminum alloy surface. Some applications even require the aluminum alloy surface to have two or more properties, such as anti-corrosion, anti-fouling, self-cleaning, and suitable mechanical wear resistance. So, in preparing a superhydrophobic coating on the surface of aluminum alloy, it is necessary to optimize a single preparation method or combine various ways to obtain a multi-functional superhydrophobic coating on the aluminum alloy surface. Table 2 briefly summarizes the preparation methods of superhydrophobic coating on the surface of aluminum or aluminum alloy and the relative properties of the obtained coating, in which the corrosion medium is 3.5 wt.% NaCl solution. Each preparation method has advantages and disadvantages (Table 3). For example, the coating prepared by the spraying method is suitable for exterior applications, i.e., textile, architectural, and glass applications. Superhydrophobic coatings obtained by rotary spraying have lower roughness and thinner thickness, making them suitable for smaller surfaces. Lithography is highly demanding and challenging to implement on an industrial scale [7]. The hydrothermal method [70–74] is to prepare coatings in sealed pressure vessels using water as a solvent and powder dissolution and recrystallization. It is usually used to construct micro-nano scale rough structures on aluminum alloy surfaces and cannot achieve superhydrophobic surface properties in one step. Therefore, a second modification step with low surface energy substances is required. Moreover, hydrothermal methods' high temperature and high-pressure conditions have always troubled practical applications. The superhydrophobic coating prepared by the sol-gel method is generally a polymer coating, one part of which is a polymer modified by organic silicon, the other part is a polymer modified by organic fluorine, and the last part is a polymer modified by organic fluorine/silicon [7,8,75,76]. We know that fluorine is not environmentally friendly. Some siloxanes will degrade after being immersed in a solution for a long time, and some polymers will inevitably crack due to heat. Table 3 compares the advantages and disadvantages of several commonly used methods for preparing superhydrophobic coatings. Of course, in practical applications, the choice of preparation method for superhydrophobic coatings ultimately depends on the service environment and its specific use.

Table 2. Preparation methods of superhydrophobic coating on aluminum and aluminum alloy surface and performance characteristics of the obtained coating.

Al alloy Type	Preparation Method	Coating Composition	The Performance of the Coating				References
			Water Contact Angle (WCA)	Sliding Angle (SA)	E_{corr} (V)	I_{corr} ($A \cdot cm^{-2}$)	
6061	Hydrothermal method + impregnation modification	Ni-Al LDHs, Stearic acid (SA)	$162.1 \pm 0.4^\circ$	$1.9 \pm 0.3^\circ$	−0.28	2.88×10^{-8}	[70]
4037	Hydrothermal method + impregnation modification	ZnO, Poly-dimethyl-siloxane (PDMS) Mg-Al LDHs,	161.1°	3.3°	−0.59	8.12×10^{-8}	[71]
6061	Hydrothermal method + impregnation modification	Triethoxy-1H,1H,2H,2H-trideca-fluoro-n-octylsilane (FAS-13)	160°		−0.33	7.94×10^{-6}	[72]
3003	Hydrothermal method+ impregnation modification	Zn-Al LDHs, Cetyl trimethoxy-silane, hexadecyl-trimethoxy-silane	156.3°		−0.66	3.87×10^{-8}	[73]
6061	Hydrothermal method	Zn-Al LDHs, Sodium dodecyl sulfate (SDS)	161°		−0.26	2.01×10^{-8}	[74]
6020	Shot peening + chemical etching + impregnation modification	Methyl trichlorosilane (MTCS)	$153 \pm 2^\circ$	$8 \pm 2^\circ$			[10]
5083	Thermo-mechanical and microwave-assisted hydrothermal processing+ Chemical vapor deposition	1H,1H,2H,2H-Perfluorooctyltrieth-oxysilane (FOTES)	$162 \pm 1^\circ$	$1 \pm 2^\circ$			[77]
6061	Coating method+ Ultrasonic spray hydrolysis deposition	Polyvinylidene fluoride (PVDF), hydrolyzed methyl-trimethoxy-silane (HMTMS)	167°	$7 \pm 1^\circ$	−0.61	2.87×10^{-8}	[78]
6061	Anodization + sealing treatment+ impregnation modification	TiO ₂ nanoparticles, Octadecyl trimethoxy-silane (OTS)	$154.2 \pm 1.7^\circ$	8°	−0.348	4.78×10^{-11}	[59]
2024	Anodization + spraying	Triethoxy-silane containing ZnO nanoparticles	151.2°	7°	−0.503	3.79×10^{-6}	[79]
Pure Al	Plasma electrolytic oxidation+ electrodeposition	Al ₂ O ₃ , Cerium hexa-decylate	165.5°	5.2°	−0.73	1.8×10^{-7}	[80]
6061	Electrodeposition	Cobalt stearate	$161 \pm 1^\circ$	2°	−0.706	0.8×10^{-8}	[66]
Pure Al	Two-step electrodeposition	SiC particles, Cetyl-trimethyl-cerium, Palmitic acid	162.3°	1.5°	−0.755	5.224×10^{-9}	[81]
6063	water treatment + impregnation modification	Stearic acid	154.1°		−0.96	5.01×10^{-5}	[82]
5052	chemical etching + Deposition + impregnation modification	Zn-Al LDH, Stearic acid	$164 \pm 3^\circ$	$1 \pm 0.5^\circ$	0.49	3.5×10^{-7}	[83]
Pure Al	laser etching+ impregnation modification	1H,1H,2H,2H-Perfluoro-octane triethoxy-silane (PFOTES)	152.8°	0.6°			[84]
Al-Si alloy	Sol-gel process	Heptadecafluoro-1,1,2,2-tetrahydrodecyl trichlorosilane (HDFS)	166.04°	8.86°			[75]
2024-T3	Spraying	SiO ₂ nanoparticles, Dodecyl-trimethoxy-silane, Epoxy resin	165°	$\approx 0^\circ$	−0.63	2.66×10^{-9}	[85]

Table 3. Advantages and disadvantages of different methods for preparing superhydrophobic coating on aluminum or aluminum alloy surfaces.

Preparation Method	Advantages	Disadvantages	References
Spraying	Simple one-step preparation; Low cost; Large-scale preparation.	The adhesion of the coating is low, and it is easy to fail after being damaged by external forces, resulting in unsatisfactory durability. Organic coatings are potentially harmful to humans and the environment.	[86–89]
Impregnation	Simple and economical; Suitable for large-scale preparation; Suitable for surfaces of all shapes or sizes; Able to realize one-step preparation.	The uniformity of the coating is uncontrollable; The adhesion of the coating to the substrate is poor, and it is easy to fall off. The use of fluorine-containing organic reagents contradicts environmental protection.	[7,76,90,91]
Anodization	Low cost and simple and fast manufacturing; Precise control of surface roughness and nanostructure; Better adhesion to substrates; Not prone to aging and wear.	The prepared film layer has pores; subsequent pore-sealing treatment or modification of low surface energy substances is required. Obtaining a relatively uniform coating is challenging.	[92–94]
Electrodeposition	Easy control of coating topography and thickness; The process is relatively simple; Suitable for industrial-scale production; Environmentally friendly; Able to realize one-step preparation.	Limited cathode metal area leads to low preparation efficiency; low surface energy substances in the electrolyte are not used efficiently; The coating adhesion is poor, and the surface is easy to wear.	[95–97]
Chemical etching	The preparation process is simple; Low cost; Able to precisely select the processing area.	The micro-nanostructure of the coating is not easy to control; The solutions or by-products used are not environmentally friendly; Subsequent modification of low surface energy substances is required.	[52,98,99]
Laser etching	It is possible to obtain regular and controllable micro-nano structures; The preparation process is environmentally friendly.	Special equipment is required; it is costly. Subsequent modification with low surface energy substances is needed.	[84,92,100]
Hydrothermal method	The microscopic size of the coating is relatively uniform; Able to realize one-step preparation.	High temperature and high-pressure conditions are required; High requirements for preparation equipment.	[8,101,102]
Sol-gel process	High temperature and pressure conditions are not required; Suitable for surfaces of different shapes or sizes.	There is thermal cracking behavior, Inaccurate coating thickness, and Expensive and environmentally unfriendly.	[7,75,76,103]

Based on the previous research, we can generally conclude that the methods for preparing superhydrophobic coatings consist of a “one-step method” and a “two-step method.” Most existing superhydrophobic coatings must go through a two-step process: first, build a rough structure on the substrate surface, then use low surface energy substances to modify the rough structure. Of course, there are also studies to obtain superhydrophobic coatings by one-step-spray low surface energy substances to make them have anti-corrosion and anti-fouling effects. Still, this method requires the synthesis of specific paints at first, the synthesis process is also relatively complex, and the spraying process has high requirements for the spray gun [86]. Also, there are studies on obtaining superhydrophobic coatings by one-step impregnating low surface energy substances. However, the coating obtained by this method has poor stability in aqueous media and quickly loses its superhydrophobic properties when subjected to external mechanical friction [76]. Therefore, the

“one-step method” to prepare superhydrophobic coatings is also limited to a certain extent in practical applications.

In the study of the “two-step method” preparation of superhydrophobic coatings, the electrodeposition process is one of the common steps. The existing electrodeposition process is generally to construct the rough structure of the substrate surface by electrodeposition of nanoparticles and then obtain the superhydrophobic coating by modifying low surface energy substances, such as fatty acids [104,105], halothane [65] and siloxane [106]. Other methods also exist to obtain micro-nano rough structures on the substrate surface. Then, deposit low surface energy substances on this structure by electrodeposition to obtain superhydrophobicity. The above research shows that the electrodeposition process can construct the micro-nano rough structure and prepare low surface energy substances to modify the hierarchical structure in order to obtain a superhydrophobic surface. Thus, if low surface energy substances can be added directly to the electrolyte solution, metal ions will react with the functional groups of these substances during the electrodeposition process. Metal compounds with low surface energy will form on the cathode surface directly. The metal compound deposition layer can obtain different micro-nano structures by adjusting the electrodeposition parameters [94]. In this way, the electrodeposition method can obtain a superhydrophobic coating of the metal substrate surface in one step. In a recent study, Xu et al. [67] used one-step electrodeposition to prepare a superhydrophobic film layer on the surface of 6061 aluminum alloy in an ethanol solution of Ni^{2+} and stearic acid (SA) at a direct current voltage of 20 V. They found that the WCA of the obtained film layer was $160 \pm 1^\circ$, and the contact hysteresis angle was $2.13 \pm 1.42^\circ$. Jiang et al. [68] used ethanol solution composed of $\text{Ce}(\text{NO}_3)_3 \cdot 6\text{H}_2\text{O}$ with myristic acid, palmitic acid, and stearic acid as electrolytes, respectively, to obtain different superhydrophobic coatings on the surface of 2024 aluminum alloy by electrodeposition, realizing the synergistic effect of low surface energy substances and rough surface structure. The cerium stearate coating had the highest WCA ($\approx 173.02^\circ$) and the lowest SA ($\approx 1.5^\circ$) among the obtained films for the cerium stearate coating with lower polarity, reducing the electrostatic interaction between water and the surface. Zhang et al. [69] explored the performance of superhydrophobic coating on the surface of aluminum alloy by electrodeposition method at different voltages, using an ethanol solution of ferric (III) chloride hexahydrate and myristic acid as electrolyte. Xiong et al. [66] prepared a cobalt stearate superhydrophobic coating by one-step electrodeposition on the surface of 6061 aluminum alloy. They studied the effect of different Co/stearic acid contents on the corrosion resistance of the film. However, the anti-corrosion, anti-fouling stability, and mechanical wear resistance of superhydrophobic coatings obtained by the “one-step electrodeposition method” have rarely been systematically studied.

4. Application of Superhydrophobic Coating on the Surface of Aluminum Alloy for Anti-Corrosion and Anti-Fouling

4.1. Application Status and Problems of Superhydrophobic Coating on Aluminum Alloy Surface for Anti-Corrosion and Anti-Fouling

The application of aluminum alloy mainly focuses on transportation, aerospace, household appliances, article packaging, building materials, and other fields. The chemical stability (such as corrosion resistance) of aluminum alloys mainly comes from its spontaneous surface oxide layer (Al_xO_y) in the atmospheric environment, which is about a few nanometers thick and can significantly prevent the continued oxidation of the aluminum alloy. However, in some practical application scenarios, aluminum alloy products need to be exposed to harsh environments, such as a strong acid, strong alkali, high humidity, high/low-temperature environments, or an aqueous medium containing many corrosive particles (such as chloride ions), etc. At this time, the oxide layer (Al_xO_y) on the surface of the aluminum alloy will be partially dissolved and lose its defensive performance [8]. Therefore, reasonable surface modification of aluminum alloy is one of the most common ways to improve its defensive performance and expand its application field. Superhydrophobic coatings consist of micro/nano rough structures and chemicals with low surface energies, whose micro/nano-scale hierarchical structures trap air and hinder the transfer of electrons

and ions between the metal substrate and electrolyte. The low surface energy substances on the coating surface can reduce the adhesion of pollutants and the fouling caused by the external contaminants [107]. Thus, the superhydrophobic surface treatment of aluminum alloy can improve the substrate's wetting resistance to corrosive substances and enhance the substrate's corrosion resistance. Also, it can reduce the accumulation of dust, dirt, corrosive media, or the adhesion of marine organisms on the surface of aluminum alloy in practical applications and improve the anti-fouling ability of the substrate. At the same time, the high WCA (greater than 150°) and low SA (less than 10°) of the superhydrophobic surface can also make the substrate have a specific self-cleaning effect [108].

Lv et al. [109] prepared a leaf-like copper oxide superhydrophobic coating on the surface of 6061 aluminum alloy by direct thermal oxidation of copper and stearic acid in the air. The results show that the obtained copper oxide superhydrophobic coating has good self-cleaning, chemical stability, and corrosion resistance. The corrosion current density of aluminum alloy covered with copper oxide coating (7.7×10^{-8} A/cm²) in 3.5 wt.% NaCl aqueous solution is an order of magnitude lower than that of blank aluminum alloy substrate (7.18×10^{-7} A/cm²). Zhang et al. [110] prepared a superhydrophobic coating on the surface of 5083 aluminum alloy by ammonia etching and 1H,1H,2H,2H-perfluorodecyltriethoxysilane (PFDTES) modification, which effectively inhibited the adhesion of corrosion pollutants and other organisms. The obtained coating can be used for the surface modification of ships serving in seawater. Rodič et al. [111] prepared a superhydrophobic film on the surface of aluminum using a one-step ultrasonic process. They found that the resulted hydrophobic coating had good corrosion resistance, anti-fouling performance, and a certain degree of anti-icing. The hydrophobicity of the coating increased with the alkyl chain length in the alkyl-silane precursor, and the surface hydrophobicity of fluoroalkyl silane modification was better than that of alkyl-silane. The anti-corrosion and anti-fouling durability of coatings, which significantly impact the practical application of metals, has rarely been mentioned in the above studies.

Aluminum alloy products will inevitably be subjected to external wear, such as the friction, erosion, or wetting of seawater, rain, and other media during their service time. These external forces will decrease the protective durability of the superhydrophobic coating, thereby shortening the service life of the metal substrate. Gupta [107] reported that some superhydrophobic layers have poor adhesion to the substrate, resulting in lower-strength mechanical forces (such as finger touch) that can destroy the micro/nanostructure of the coating, causing an irreversible loss of the superhydrophobicity of the coating. Eventually, the coating loses its protective effect on the substrate. In addition, Table 4 details the durability and mechanical wear resistance test results of a series of aluminum superhydrophobic coatings. In recent years, how to construct superhydrophobic coatings on aluminum alloys with chemical stability and mechanical durability has become a research hotspot.

Table 4. Stability test results of superhydrophobic coating on the aluminum alloy surface.

The Primary Preparation Process of the Coating	Chemical Stability	Mechanical Durability	References
Sodium hydroxide etching; Lauric acid impregnation modification	WCA dropped to 125.3° after immersion in 5% acetic acid solution for 6 days.	The peel test was carried out with 100 N/m insulation tape, and the coating lost its superhydrophobicity after 15 viscous peels.	[55]
Hydrochloric acid etching; Deposition of Zn-Al LDH film; Stearic acid impregnation modification.	Lost superhydrophobicity after 7 days of immersion in 0.6 mol/L NaCl solution; Lost superhydrophobicity after 14 days of immersion in distilled water.	Using 1500 mesh SiC abrasive paper, the coating lost its superhydrophobicity after applying 2 N pressure friction for 250 cm.	[83]
Anodizing; electrodeposited TiO ₂ nanoparticles; Octadecyl trimethoxy-silane impregnation modification	WCA dropped to 141.8° and lost superhydrophobicity after 7 days of immersion in seawater.		[112]

Table 4. Cont.

The Primary Preparation Process of the Coating	Chemical Stability	Mechanical Durability	References
Anodizing; Lauric acid impregnation modification. Scrub treatment; Hydrochloric acid etching; Stearic acid impregnation modification.	Using 2000 mesh SiC sandpaper load 100 g for uniform friction, lost superhydrophobicity when the wear distance is 300 cm.		[58]
High-speed wire discharge machining; Hydrochloric acid etching; Perfluorooctanoic acid impregnation modification	Lost superhydrophobicity after 14 days of immersion in NaCl solution (0.6 mol/L). Lost its superhydrophobicity after rubbing the sample for 260 cm at 20 kPa using 1000 sand-grained sandpaper. Using a 200 g weight drop from a height of 12 cm and hitting the coatings directly at approximately 15.50 cm/s, it lost superhydrophobicity after 35 times.		[113] [114]
Hydrochloric acid etching; Stearic acid impregnation modification. Laser etching; 1H, 1H, 2H, 2H- Perfluoro-octyl-triethoxy-silane impregnation modification.	After 1 day of immersion in 3.5 wt% NaCl solution, WCA < 150°. Lost superhydrophobicity after a 14-h salt spray test.		[115]
Spray nano-SiO ₂ particles and 1H, 1H, 2H, 2H-perfluorodialkyltriethoxysilane modified epoxy resins.	The coating lost superhydrophobicity after rubbing 150 cm with a 1000 mesh sandpaper load of 200 g.		[84]
Spray polystyrene microspheres and polydimethylsiloxane.	A peel test was performed using 3 M VHB tape, and the coating lost superhydrophobicity at the 40 th peel. The coating lost its superhydrophobicity after wearing 900 cm with a 2000 mesh sandpaper load of 100 g.		[116]
Spray F-SiO ₂ nanoparticles, epoxy resin, fluoro-silicone paint, and fluorinated polyurethane.	Heated at 260 °C for 1 h, the coating lost its superhydrophobicity.		[56]
Spray SiO ₂ nanoparticles and epoxy resins modified with dodecyl-trimethoxy-silane. Plasma oxidation;	50 µL of water is used for dripping onto the surface of the coating at a rate of 160 drops per minute from a height of 40 cm, and the coating lost its superhydrophobicity after 2 h.		[57]
Chemical vapor deposition of fluorinated SiO ₂ nanoparticles.	After 40 h of irradiation under ultraviolet light, the WCA of the coating drops to 149.3°, while after 12 h of irradiation, the SA of the coating increases to 15.7°.		[85]
One-step electrodeposition of Iron (III) chloride hexahydrate and myristic acid.	After 9 days of immersion in 3.5 wt% NaCl solution, the coating lost its superhydrophobicity.	After peeling off 20 cycles with 3 M tape, the coating lost its superhydrophobicity.	[62]
Thermomechanical and microwave-assisted hydrothermal treatment; Chemical vapor deposition perfluoro-octyl-triethoxy-silane.	After rubbing at a constant speed of 60 cm with a load of 40 g using 240 mesh sandpaper, the coating lost its superhydrophobicity.		[69]
Electrodeposition of SiC particles, trimethyl cerium palmitate, and palmitic acid first; then electrodeposition of palmitic acid.	After 6 days of immersion in 3.5 wt% NaCl solution, the coating lost its superhydrophobicity.		[77]
	After 9 days of immersion in 3.5 wt% NaCl solution, the WCA of the coating decreases to 147.5°, while on day 8, the SA of the coating increases to 10.5°.	After rubbing 300 cm with a 50 g weight load of 1000 mesh sandpaper, the WCA of the coating is reduced to 135.4°. A 2 kg steel rod is rolled over the specimen surface, and the coating loses its superhydrophobicity after 18 cycles.	[81]

4.2. Methods for Improving the Performance of Superhydrophobic Coating on the Surface of Aluminum Alloy for Anti-Corrosion and Anti-Fouling

4.2.1. Improve the Preparation Process of Superhydrophobic Coating on the Surface of Aluminum Alloy

When the superhydrophobic surface is continuously exposed to the corrosive medium, the external friction and wear and the pressure of the corrosive liquid medium will damage the composition and structure of the hydrophobic coating, resulting in the coating losing its hydrophobicity and eventually losing its anti-corrosion and anti-fouling effect. Therefore, studying how to enhance the hydrophobic durability of superhydrophobic coatings in different environments is essential for their practical applications. According to the formation mechanism of existing superhydrophobic coatings, the two main factors affecting their performance are the rough structure of the coating surface and the low surface energy substances' properties on the coating surface. For this reason, many existing studies have focused on adjusting the construction of rough surface structures and the selection of low surface energy substances. Commonly used methods for constructing the hierarchical structure on an aluminum alloy surface mainly include direct mechanical grinding, chemical etching, anodizing, chemical conversion, etc. Alternatively, rough structures can also be obtained by electroplating or electrodepositing nanoparticles (e.g., SiO₂, TiO₂, CeO₂) on the surface of aluminum alloys [117]. In addition to the use of low molecular weight

organic substances, there are also studies on the use of polymer organic compounds or polymers, with higher hardness and better stability to improve the wear resistance of the coating. However, most of the optimization of the existing coating preparation process is to modify and optimize the coating's rough structure and low surface energy substances separately. Separately optimizing the composition and structure of the coating cannot combine the composition's structure and the optimization effect well, i.e., they cannot exert their synergistic influence. Moreover, the uniformity of dispersion of low surface energy substances on the rough structure, the type of bonding with the rough structure, and the degree of adhesion between the coating and the substrate all have essential effects on the durability of the coating. Therefore, one of the current trends in modifying superhydrophobic coatings is preparing functional nanoparticles and polymer–polymer combinations of inorganic–organic composite coating by one-step modification optimization.

Li et al. [118] prepared an Al_2O_3 /poly-siloxane composite superhydrophobic coating on the surface of 2024 aluminum alloy by the one-step electrodeposition method. The chemical stability and durability of Al_2O_3 /poly-siloxane composite superhydrophobic coating are shown in Figure 6. There was no significant change in WCA, SA, and morphology after seven days of immersion in 3.5 wt.% NaCl solution. When exposed to the atmospheric environment for up to 90 days, the changes in WCA and SA were still small, and there was no apparent change in morphology, indicating that a superhydrophobic coating with good durability could be obtained on the surface of aluminum alloy by one-step electrodeposition. Fan et al. [119] first applied a polydimethylsiloxane (PDMS) layer on the surface of 6082 aluminum alloy by spraying method. Then, they sprayed a mixed solution of hydrophobic SiO_2 particles and PDMS on the PDMS coating prepared in the previous step to form a superhydrophobic composite coating PDMS/SH. The results of the composite coating's mechanical durability and chemical stability are shown in Figure 7. The coating surface is rubbed at a constant speed with 80 g of sandpaper under 1000 mesh sandpaper, and it is still superhydrophobic when the wear distance reaches 1600 cm. Meanwhile, the coating was rinsed in an aqueous solution with pH = 1–14, placed in an environment of 1–100 °C, or irradiated with ultraviolet light of 0.51 W/m² for 0–7 days. At the end, the WCA of the coating was larger than 150°, indicating that the siloxane superhydrophobic coating prepared by the two-step spraying method had good chemical stability.

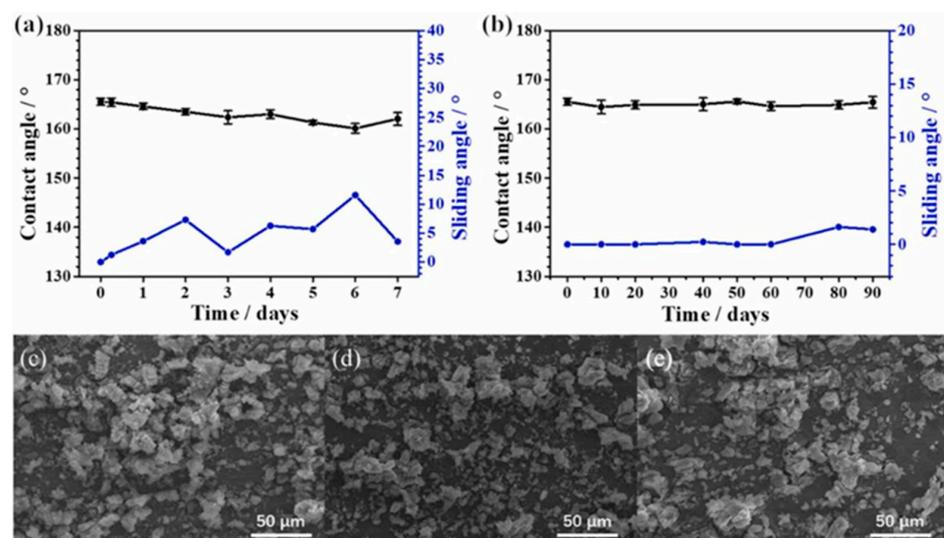


Figure 6. (a) Variation of the water contact angle and sliding angle of the Al_2O_3 /poly-siloxane hybrids coated aluminum alloy with the immersion time in 3.5 wt.% NaCl solution and (b) storage time in the atmospheric environment at room temperature. (c) SEM image of the pristine Al_2O_3 /poly-siloxane hybrids. (d) SEM images of the Al_2O_3 /poly-siloxane hybrids coated aluminum alloy stored in 3.5 wt.% NaCl solution for seven days and (e) in the atmospheric environment at room temperature for 90 days [118].

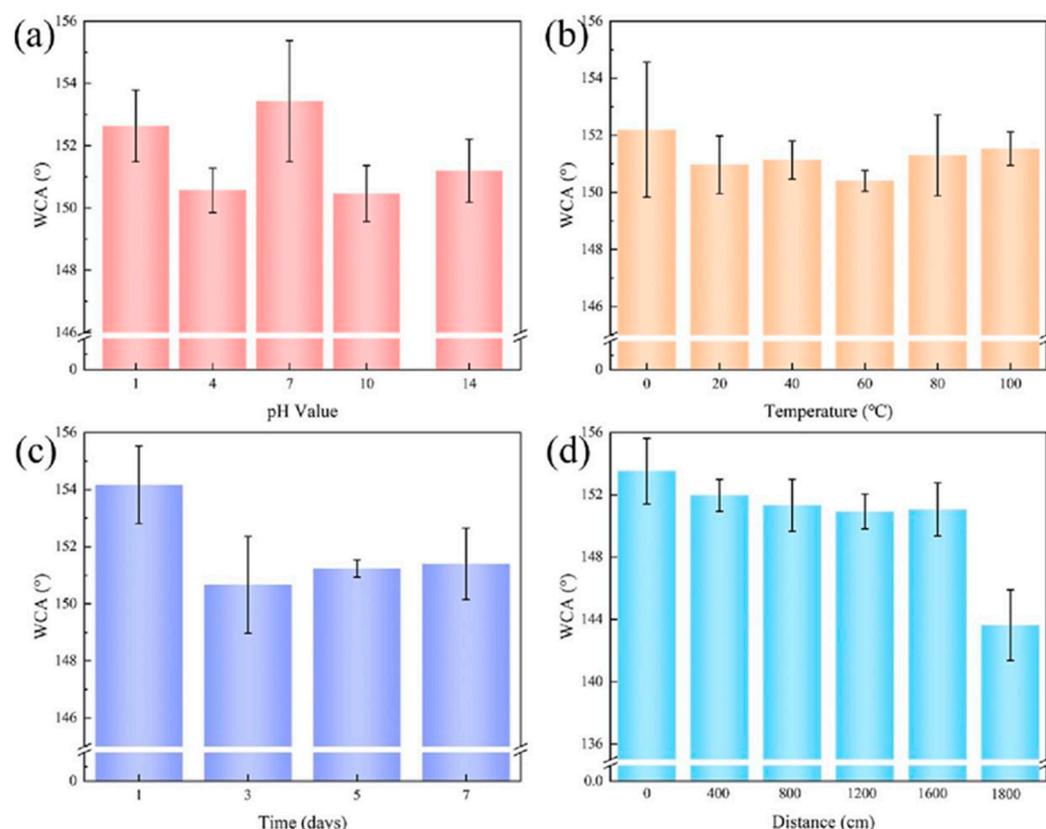


Figure 7. (a) The WCA value of PDMS/SH composite coating under different pH values, (b) different temperatures, (c) different UV irradiation times and (d) different abrasion distances [119].

4.2.2. Preparation of Superhydrophobic Coatings with Self-Healing Properties

In the actual application of superhydrophobic coatings, local wear and scratches will inevitably occur, and this minor local damage will also make the structure of the coating change and evolve into failure of the entire coating. Therefore, based on optimizing the preparation process of the superhydrophobic coating, making the superhydrophobic coating have a self-healing function through a suitable strategy is another effective method to improve its anti-fouling durability. Compared with traditional superhydrophobic coatings, the main advantage of superhydrophobic surfaces with self-healing performance is that the coating can restore its original structure, i.e., local damage in practical applications can be restored through its repair function without causing the entire coating structure to change or fail. So, the self-healing coating can extend its service life, improve its protective durability, and reduce equipment maintenance costs. There are two main methods for preparing self-healing superhydrophobic coatings [120]. One is the storage and release of healing agents. That is, the corrosion inhibition ions or superhydrophobic components are initially stored in the micro-nanostructure of the coating. They will move to the local damaged area to avoid the expansion of the damaged area over the entire coating, and the coating will return to its original properties after some time. The other method is rough micro-nanostructure regeneration, which generally requires secondary coating treatment, which limits practical applications. Compared with the second method, the current research mainly uses the storage and release of healing agents to obtain self-healing superhydrophobic coatings. Among these, nanomaterials not only serve as corrosion inhibitors but also enhance the adhesion between coatings and substrates, which can fundamentally solve the problem of coating durability [121,122].

Liu et al. [123] prepared a durable superhydrophobic coating (SHS) with a self-healing ability by plasma anodizing and impregnating the healing agent fluoroalkyl silane (FAS-17). The results showed that the superhydrophobic coating became superhydrophilic after

oxygen plasma oxidation for 2 min, and it can recover its superhydrophobicity after heating at 70 °C for 30 min. It maintains its superhydrophobicity after eight cycles (Figure 8a,b), indicating that the resulting coating has a specific self-healing function. The self-healing process of the coating is also analyzed in Figure 8c. After the surface FAS-17 is damaged by external wear, heating the coating allows the FAS-17 in the pores to diffuse to the outermost layer driven by surface tension, thus forming a new layer of FAS-17 and making the coating superhydrophobic again. The investigation of the chemical stability of the coating, as shown in Figure 8b, indicated that the oxygen plasma-treated coating could maintain its superhydrophobicity after being heated at 80 °C for 30 days. At the same time, a load of 500 g of 1000 mesh sandpaper is placed on the surface of the coating, and the horizontal friction is 12 cm at a speed of 5 cm/s. After repeating this process 30 times, the WCA of the coating can still be maintained at about 160°, indicating that the coating has good mechanical wear resistance.

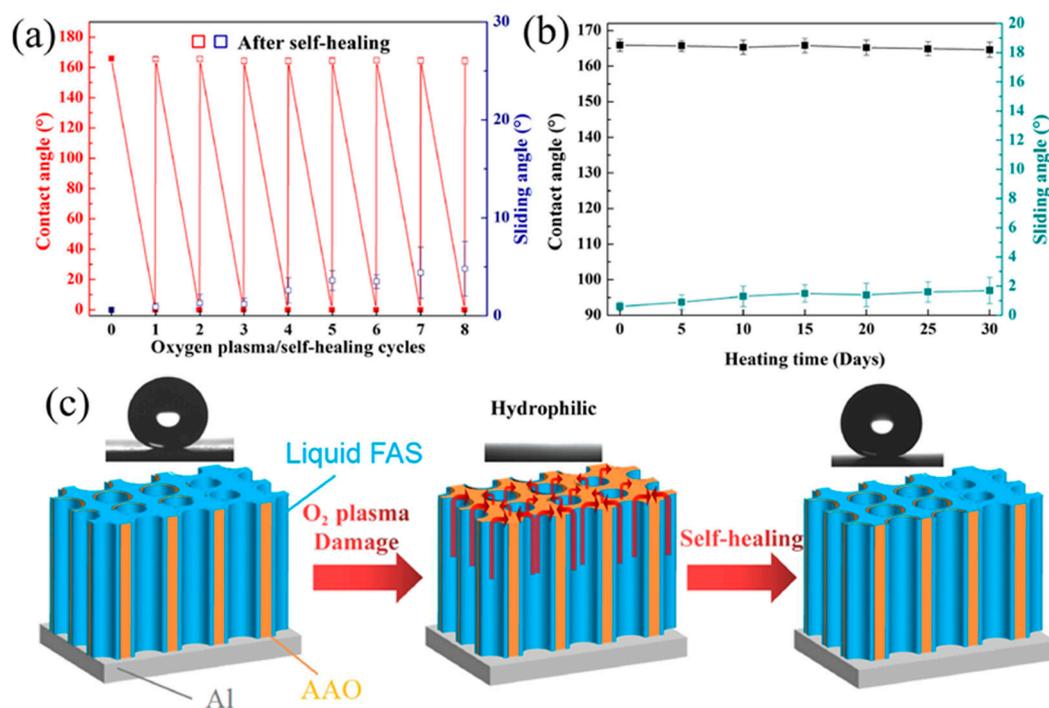


Figure 8. (a) CA and SA for water at room temperature (25 ± 3 °C) on the SHS after several oxygen plasma treatments and after being heated up at 70 °C for 30 min; (b) CA and SA of SHS after oxygen plasma treatments and after storing at 80 °C for different times and (c) schematic illustration of self-healing mechanism. [123].

Furthermore, layered double hydroxide (LDH) materials are ideal for self-healing superhydrophobic coatings due to their unique chemical composition and microstructure. Pan et al. [124] obtained a lithium aluminum LDH film (SLDH) modified by secondary alkane sulfonate (SAS) with high corrosion resistance by in situ growth on 6N01 aluminum alloy. The neutral salt spray (NSS) test found that the 6N01 aluminum alloy covered with Li-Al LDH film had severe pitting corrosion, but covered with SLDH film was almost undamaged after 360 h of exposure to NSS. This study also preliminarily proposes the anti-corrosion process of SLDH in corrosive media, as shown in Figure 9. When corrosion occurs, the inserted SAS anions are released and exchanged with corrosive Cl^- . Simultaneously, Cl^- is fixed in the interlayer space to balance the charge, slowing down the corrosion rate. Liu et al. [70] prepared a Ni-Al LDH superhydrophobic coating with self-healing properties on aluminum alloy 6061 by hydrothermal reaction and low surface energy substance stearic acid (SA) modification. The superhydrophobic coating has excellent self-cleaning properties and low fouling adhesion, which can effectively improve the corrosion

resistance of aluminum alloys. In addition, after the coating loses its superhydrophobicity in different harsh environments, external thermal stimulation can quickly restore its superhydrophobicity, showing good hydrophobic stability. The self-healing process of the Ni-Al LDH superhydrophobic coating is shown in Figure 10 [70]. When the coating is corrupted and becomes hydrophilic, the SA molecules stored between the LDH layers migrate to the coating surface. The coating can return to its original superhydrophobicity after the hydrophobic group returns to a certain amount. This self-healing process of SA migration can occur at room temperature, and external thermal stimulation can accelerate this self-healing process.

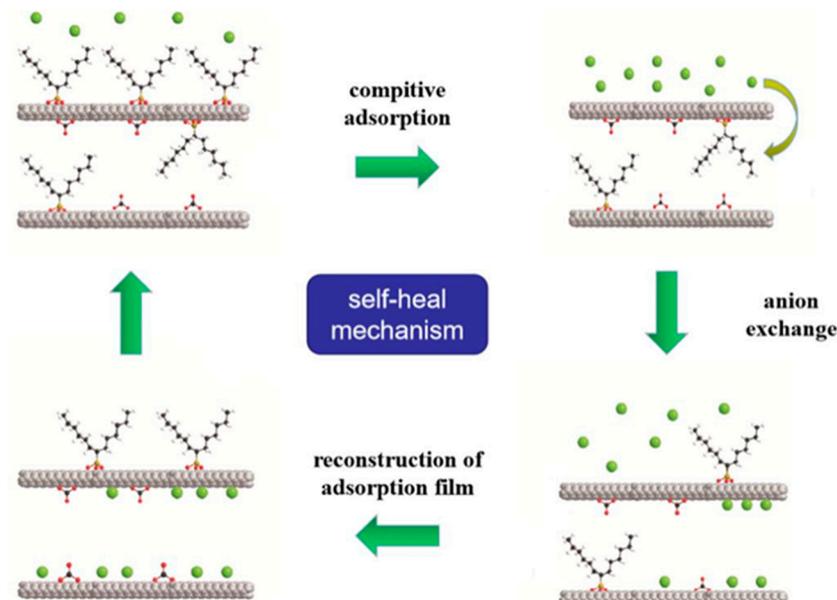


Figure 9. Schematic illustration of inhibition mechanism of SLDH [124].

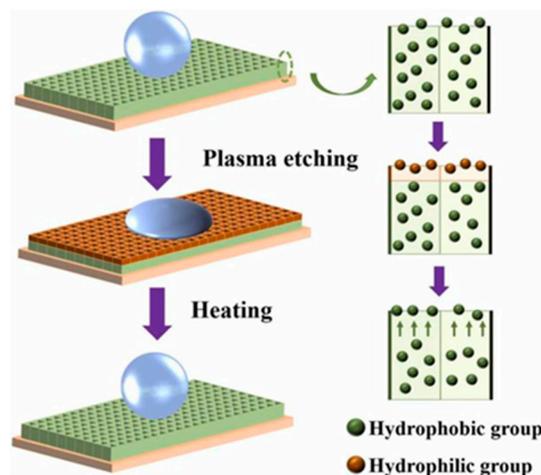


Figure 10. Schematic diagram of the self-healing process of superhydrophobic coating [70].

4.2.3. Introduction of Buffer or Sacrificial Layer into a Superhydrophobic Coating

Constructing the multi-layer superhydrophobic structure of the “superhydrophobic coating–buffer layer/sacrifice layer–super hydrophobic coating” on the aluminum alloy surface means introducing a buffer layer or sacrifice layer between the superhydrophobic surface at the bottom of the base and the outermost superhydrophobic layer to improve the anti-fouling durability of the entire coating. When the outer coating is subjected to external wear and loses its superhydrophobic properties, the buffer layer in the middle can be used as a buffer to protect the nanostructure of the underlying layer. When the outermost layer

is destroyed, its superhydrophobicity will temporarily disappear. At that time, the liquid corrosion medium (such as an aqueous solution) will directly contact and dissolve the sacrificial layer. After the sacrificial layer is completely dissolved, the superhydrophobic surface of the lowest layer will be exposed so that the coating will regain superhydrophobic properties. The process by which the coating peels off the damaged surface and reveals the new superhydrophobic character is shown in Figure 11. Wu et al. [125] used water-soluble polyvinyl alcohol (PVA) polymer binder as buffer layer and sacrifice layer to prepare a long-term anticorrosive superhydrophobic coating on the superhydrophobic aluminum surface coated with polytetrafluoroethylene (PTFE). The results show that, when the outermost superhydrophobic surface is scratched, the water-soluble sacrificial layer can be dissolved in rainwater or other aqueous solutions and detached from the coating. The exposed coating is a PTFE superhydrophobic coating, which can extend the service life of the superhydrophobic surface. Also, after the preparation of a double-layer superhydrophobic coating, the corrosion resistance of aluminum is improved, and even the repaired surface has better corrosion resistance than a coating sprayed only with PTFE. Lv et al. [126] added potassium titanate whiskers and SiO₂ particles (PTW-SiO₂) modified by amino silicone oil (ASO) into the poly(phenyl sulfide) PPS matrix by sol-gel method and then sprayed the prepared solution on the surface of aluminum plate to prepare a solid superhydrophobic PPS/ASO/PTW-SiO₂ composite coating. After immersing the superhydrophobic coating in 3.5 wt% NaCl solution for 28 days, the corrosion current density (I_{corr}) increased from 3.32×10^{-11} A/cm² (the I_{corr} of immersion for 1 h) to 1.289×10^{-10} A/cm². However, the I_{corr} was still five orders of magnitude lower than the I_{corr} of the blank aluminum plate, indicating that the PPS/ASO/PTW-SiO₂ superhydrophobic composite coating with a multi-layer structure has good corrosion durability.

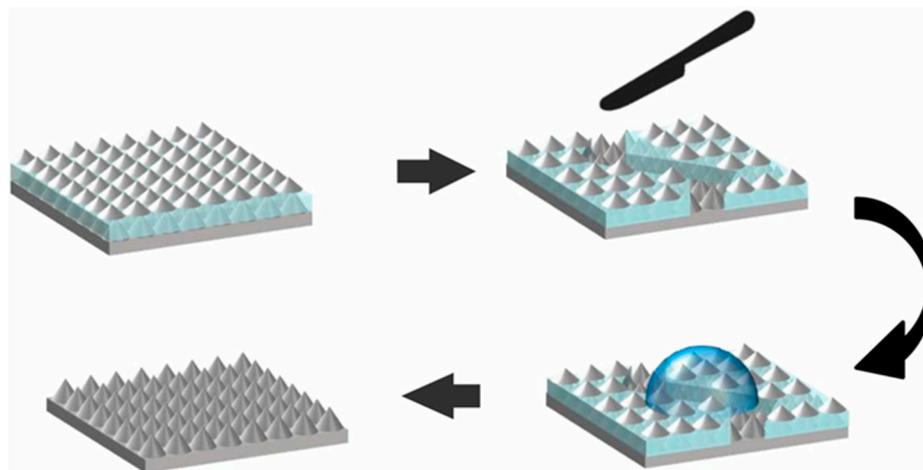


Figure 11. Shedding damaged surface and exposing new superhydrophobic surface [125].

5. Conclusions

The corrosion and fouling of aluminum alloy have become a focus problem that limits its wide application. The preparation of functional coatings on aluminum alloys' surfaces can improve their corrosion resistance, effectively improve the utilization rate of aluminum alloys, and thus save energy in response to the call of the new era of "carbon peak" and "carbon neutrality". Compared with the single-functional coating, various fields favor the multi-functional coating, so the superhydrophobic coating is applied to the aluminum alloy surface for its advantage of versatility. Most existing processes for preparing superhydrophobic coatings on the surface of aluminum alloys use a two-step method. First, build a micro-nano structure on the surface of the aluminum alloy, then modify the rough structure with low surface energy substances to obtain a superhydrophobic coating. Or prepare/formulate a superhydrophobic paint at first, then apply spray and spin to the surface of the aluminum substrate to get a superhydrophobic film. Therefore, if the superhydrophobic coating can be prepared on the surface of aluminum alloy by the one-step

method, it can not only optimize and improve the preparation process of the existing superhydrophobic coating but also provide a fast, simple, and environmentally friendly new method/technology for the preparation of superhydrophobic coating.

Furthermore, superhydrophobic coatings can effectively improve the corrosion resistance and anti-fouling performance of aluminum alloys and can even be applied to some extreme environments, such as the needs of an aerospace fuselage to be resistant to ultraviolet rays, nuclear reactors need to be resistant to radiation, marine ships need to be resistant to seawater corrosion, functional devices need to resist strong acid and alkali corrosion, etc. It is worth noting that superhydrophobic coatings have not yet been widely used and industrialized in aluminum alloy surface treatment, mainly for two reasons. One is that superhydrophobic coatings' durability and mechanical wear resistance are not outstanding. The other is that fluorine-containing low surface energy substances harm the environment. Based on this, this paper summarizes some new ideas to improve the durability of superhydrophobic coatings on the surface of aluminum alloys. (i) Increase the adhesion between the coating and the substrate by optimizing the coating preparation process, such as using a one-step in situ method to construct a rough structure composed of low surface energy substances on the substrate surface. (ii) A self-repairing superhydrophobic coating is prepared, for example, by introducing a low surface energy substance into self-healing LDH films and forming the two organic mosaics into a composite coating. (iii) A sacrificial layer with a similar structure and composition is prepared on the surface to enhance the protective durability of the multi-layer coating, etc. Nowadays, improving the durability and mechanical wear resistance of anti-corrosion and anti-fouling coatings on the surface of aluminum alloys has become one of the most critical challenges for commercializing superhydrophobic coatings on aluminum alloy surfaces.

Author Contributions: Q.Z.: Original draft preparation, writing, and revision; X.D.: Conceptualization, draft revision, review, and editing; Y.L. and X.F.: Part of the literature reading and knowledge summary; D.C.: Supervision and funding acquisition; Z.Z.: Supervision and draft review and revision. All authors have read and agreed to the published version of the manuscript.

Funding: This study was funded by the National Natural Science Foundation of China (52101079), Guangdong Basic and Applied Basic Research Foundation (2020A1515110825, 2020B1515120093), the Key-Area Research and Development Program of Guangdong Province (2020B010186001) and Science and Technology Project of Guangdong Province (2020B121202002).

Institutional Review Board Statement: Not Applicable.

Informed Consent Statement: Not Applicable.

Data Availability Statement: Not Applicable.

Conflicts of Interest: The authors declare no conflict of interest.

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