Superhydrophobic Coating Fabrication for Metal Protection Based on Electrodeposition Application: A Review

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Abstract

In recent years, superhydrophobic media has attracted tremendous attention due to its industrial applicability value, especially in anti-corrosion performance. The superhydrophobic coating, which has a robust and water-repellent capacity, can catch the air to form several “airbags” on the substrate’s surface, isolating the corrosion media. Various superhydrophobic coating preparation technologies have been suggested, but each has its own set of flaws. On the other hand, electrodeposition, as a relatively mature industrial processing application, offers distinct advantages. However, until now, there have been few reviews on the electrodeposition preparation of anticorrosive superhydrophobic coatings. Therefore, the author has described several fabrication techniques based on superhydrophobic coatings in this review, including the advantages and disadvantages. Superhydrophobic coatings conventional concepts and wettability, as well as the model wetting concepts, have been reviewed. The coating processing status and the corrosion-resistant potential through the electrodeposition of metal and comparable composite are detailly encapsulated. Furthermore, electrodeposition parameters, including current density, crystal modifiers, and a deposition time of the coating morphology, are reported, following the ultrasonic-assisted, jet, pulse, and magnetic field-induced electrodeposition, respectively, as the recently developed technologies for preparing a coating. Finally, technology limitation is shown as well as the obstacles and prospects, and the improvement of the superhydrophobic coating’s durability as a prospects research focus has been recommended.
Keywords

Electrodeposition, Corrosion Resistance, Superhydrophobic Coating, Wettability

1. Introduction

The study of superhydrophobic technology got its inspiration from the lotus leaves self-cleaning effect since the past decades, based on various superhydrophobic surface preparations. Applications such as anti-corrosion [1], self-cleaning [2], anti-ice and anti-fog [3], and oil and water separation [4] are leading industrial interest. Corrosion of metal has been a hazardous phenomenon on resources, economic, and safety risks globally [5]. Past data shows that corrosion cost of damage is about 4% - 6% GDP annually [6]. The superhydrophobic coating is an alternative due to its high water resistance, reducing the corrosion rate, thereby isolating the metal from the corrosive media [7]. The term superhydrophobic surface is a medium with a water contact angle (WAC) equal to or above 150˚ [8] and sliding angle (SA) less than 10˚ [9]. The surface energy and surface morphology are factors that relate to the wettability of the superhydrophobic surface.

Consequently, the superhydrophobic surface can be prepared by any of the two methods: either by the “top-down” method in placing the micro-nano structure on the hydrophobic material surface or the “bottom-up” method, where micro-nano structure surface graded using low surface energy [10]. Based on these ideas, several technologies such as chemical etching [11], electrospinning [12], sol-gel [13], anodizing [14], and salt spraying [15] were developed to fabricate superhydrophobic coatings. The combining of these methods can raise the hydrophobicity capacity of the metal surface to a high magnitude. Research has combined shot peening, dislocation etching, and stearic acid treatment to coat copper [16]. Stearic acids are absorbed by shot peening to increase surface roughness, while etching is conducive to the micro-nano ordering structure. Also, the chemical treatment can potentially reduce surface energy. All these techniques improve the hydrophobicity of the media. However, these technologies are made of costly equipment and toxic raw materials, hazardous to the environment, and carcinogenic to human health.

Furthermore, some technology’s preparation process is complex and challenging to handle and contains low preparation efficiency, making it difficult during large-scale industrial applications. Based on other fabricating superhydrophobic coatings, the electrodeposition technology advantages are many [17]. These advantages are: 1) electrodeposition technology is widely used for composite coating and nanomaterial fabrication [18]; 2) the technology works under a suitable power supply, electrode, and electrolyte; 3) it has a low-cost efficiency, a simple application method, and is easier to be used and control; 4) the coating morphological surface can be easily controlled by adjusting the voltage, current...
density, and deposition time with repeatable sequences; 5) it can be applied on the surface of many complex metals because of its not limited by the substrate’s shape and size; 6) Both the preparation efficiency and batch preparation can be determined. The movement of metal ions in the electrolyte and reduction on the cathode surface under a suitable power supply is known as electrodeposition.

During this process, the superhydrophobic effect can be achieved by controlling the process condition with microstructure constructed on the surface [19]. The pulse electrodeposition [20], jet electrodeposition [21], and composite electrodeposition [22] are known techniques for industrial application nowadays. The ultrasonic [23] and magnetic field induced methods as combining electrodeposition techniques have developed many unexpected results. This review highlighted the current development of electrodeposition technology in preparing superhydrophobic coatings for corrosion-resistant. It points out current electrodeposition developments and various metal protection applications with superhydrophobic coatings, emphasizing the mechanism, corrosion-resistant performance, wettability, and wetting concepts. Secondly, electrodeposition parameters, electrodeposition on metals, and electro-composite material application are discussed. Finally, the prospect and challenges with research directions to upgrade anti-corrosion with its durability have been greatly focused on.

2. Concepts of Superhydrophobic Surfaces

2.1. Wettability

2.1.1. Surface Tension and Free Energy

Wettability reflects the spreading of water droplets potentially on a homogeneous flat surface. Some surfaces term to be hydrophilic, where droplets are spread rapidly, forming a liquid film. However, other natural surfaces like the lotus leaves show a strong hydrophobicity when encountered with water droplets, thereby remaining spherical and eventually rolling off quickly. Typically, a water contact angle equal to or less than 90˚ is regarded as hydrophilic or is hydrophobic if it is greater than 90˚ [24], which indicates the dividing line of wettability. Gravity and surface tension led to a solid surface with attraction and repulsion sources among water molecules that eventually cancel each other, forming a balanced state. Due to the attraction forces between molecules, droplets tend to shrink inward with a thin layer on the liquid surface.

On the contrary, the droplet tends to spread due to gravity, but the droplet remains spherical when the surface tension is assertive. With an increase in droplet size and a dominated gravity, the droplet will eventually spread by attracting the surrounding water molecules. Research shows that wettability has been affected by surface free energy on the solid surface [25]. In chemical bonding, molecules in the liquid have higher potential energy than those outsiders, causing the generation of the surface free energy [17]. Although in the field of thermodynamic both surface tension and surface energy are different concepts but somehow related. Studies show that hydrophobicity is enhanced by lower
surface energy [26]; these low surface energy materials are used for fabricating superhydrophobic coatings on thin films.

2.1.2. Morphology of the Surface
A hydrophilic substance can also achieve a hydrophobic effect based on constructing a favorable surface morphology, clarifying that wettability is affected by surface tension or surface energy and the morphology of the structure. For example, many researchers have fabricated superhydrophobic coating on nickel. However, nickel itself is hydrophilic.

It was proven in the scanning electron microscope (SEM) that micron and nanometer structures support superhydrophobicity [27]; airbags were trapped between these micro nanoparticles forming a bulge, making the superhydrophobic surface repellent to droplets achieving an excellent anti-corrosion effect. Besides the lotus leaf, there are also natural superhydrophobic phenomena, but the waxy papillae of diameter 5 - 9 micrometer are distributed on lotus surface, which further covered with villi of 120 nm in diameter [28], (Figure 1(a) and Figure 1(b)), respectively. As shown in (Figure 1(c)), the water spider is due to its micro-bristles and nano-grooves distributed on the legs. These natural facts clarify the wettability of surface properties and micro-morphology of solid surfaces.

2.2. Model of Wetting Concepts
A British Scientist, Thomas Young, relates the contact angle and the surface tension on a smooth surface under a theoretical study of surface wettability, giving Young’s equation [29]. Equation (1):

$$Y_{SG} = Y_{SL} + Y_{LG} \cos \theta$$

where $Y_{SG}$, $Y_{SL}$, and $Y_{LG}$ are solid-gas, solid-liquid, and liquid gas, respectively, and $\theta$ is the water droplets’ intrinsic contact angle on a homogeneous surface. This theory is only suitable on a completely smooth surface with uniform chemically and nonelastic properties, as shown in (Figure 2(a)), but this surface does not exist. This Wenzel model [30] was proposed, which implies that the droplets will

![Figure 1. Natural patterns of superhydrophobicity: (a) Lotus leaf self-cleaning pattern; (b) A Lotus leaf micro-nano mastoid structure, (c) A water spider on the water surface.](image-url)
immerse as tiny grooves when contacted the rough surface, expanding the contact area than the apparent area of contact (Figure 2(b)). Equation (2):

\[ \cos \theta_w = r \cos \theta \]  

The roughness \( r \) is the ratio between the contact area to the apparent area of the droplet and rough surface. When the value of \( r \) is greater than 1, the wettability effect was magnified, where an increase in \( r \) makes a surface more hydrophilic and helps the hydrophobic surface improve its hydrophilicity eventually. Wenzel's model's application is unique on a surface with uniform composition, which gave rise to the Cassie-Baxter model [31]. In the Cassie-Baxter model, the droplet immerses on a composite surface of gas and solid, as shown in (Figure 2) Equation (3):

\[ \cos \theta_{lb} = f_1 \cos \theta_1 + f_2 \cos \theta_2 \]  

\( f_1 \) and \( f_2 \) are represented by the ratio of the contact area of Section 1 and Section 2 to the total contact area summing to 1, \( \theta_1 \) and \( \theta_2 \) which are the apparent contact angle of Section 1 and Section 2, respectively. However, these three models have been the widely accepted theoretical basis for studying droplet wettability behavior, but they are somehow limited and cannot modify all the wetting behaviors.

Some newly developed wetting models have been proposed based on the understanding of superhydrophobic surface micro-nano structure and classic wetting models’ limitations. The combination of the Cassie-Baxter model and Wenzel model known as the Marmur’s model was proposed [32], where droplets were partially inserted on an air cushion (Figure 2(d)), and it was scientifically proven as a transition between the two states. Reference [33] list the 1) Wenzel’s state, 2) Cassie’s state, 3) Transitional of Wenzel’s and Cassie’s state, 4) Gacko’s state, and 5) Lotus state as the five cardinal wetting states model of liquid droplets on a solid surface. The Cassie-Cassie mode, Cassie-Wenzel mode, Wenzel-Cassie mode, and Wenzel-Wenzel mode were classified as the four kinds of wetting states with micro-nano dual 36-scale structure on the surface.
but did not take into consideration the distribution of the effect of nanostructure on the sidewall of microstructure during wetting. Wu et al. [35] proved that this would affect the wettability. Some research points out that when droplets are in contact with solid surfaces of micro-nano dual structure, nine wetting states are formed, but the calculation method and condition of their contact angle were not described [36]. These models will no longer be applicable until the droplet size reaches a micron level or less on the surface, and the contact angle is determined by the Neumann-Borovka Equation (4) [37] [38].

$$\cos \theta = \frac{Y_{SV} - Y_{SL}/Y_{LV}}{-\Gamma/a}$$  \hspace{1cm} (4)

where $\Gamma$ is the line tension and “a” is the contact radius; however, there is a difference between the line tension experimental values and the theoretical because of the limited measuring technology and the difficulty in sample preparation [39]. Electrodeposition method of superhydrophobic coatings preparation

Electrodeposition can be divided into various media due to particle deposition, composite electrodeposition, metal deposition, and non-metallic. Based on superhydrophobic coatings as a recent research flashpoint, these three methods have been successfully used by researchers to develop superhydrophobic coatings on several substrates achieving an anti-corrosion effect.

3. Electrodeposition Parameter and Processing

The electrodeposition process’s three main factors are crystal modifier/bath composition, current density, and electrodeposition time, potentially affecting the coating’s composition and surface morphology.

3.1. Modification of Crystals

The electrodeposition bath consists typically of components like: 1) The main salts that deposit salt ions on the cathode surface; 2) The conductive salt mechanized the bath conductivity; 3) The buffer; stabilize pH near the cathode of the bath; 4) The anode activator which reduces the anodic polarization and dissolution of the anode; 5) Additives are added substances such as brightener, leveling agent, wetting agent to the coating, and having a significant effect. In electrodeposition application of a superhydrophobic coating, it is found that reagents can control crystal growth and other formations of micron-nano dual-scale structure, mechanizing the morphology of the coatings. These are crystal modifiers that benefit superhydrophobicity.

Soleimangoli et al. [40] prepared a Ni-Zn superhydrophobic coating by two-steps electrodeposition process, added ammonium chloride modifier to a bath composed of nickel chloride, zinc chloride, and boric acid, respectively. The coating surface becomes polyhedral after the addition of ammonium chloride, which changes from polyhedral-shape structure to micro-nano conical structure. Without a surface modification, ammonium chloride content reaches 400 g/L with a water contact angle reaching 159.8°, and the best superhydrophobic effect was obtained with increased roughness. The Ni-Zn coating’s better corrosion resis-
tant potential was gain with corrosion current density increasing by three orders of magnitude lower, compared without NH$_4$Cl. Another experiment [41] fabricates Ni superhydrophobic coating on copper substrate by mixing NiCl$_2$, H$_3$BO$_3$, with NH$_4$Cl. During the preparation process, the medication effect of NH$_4$Cl on crystallization transformed a single-shape micro-cone to nano-cone structure, achieving a superhydrophobic effect, moreover, exploring NHCl$_2$ role in the process based on electrochemical impedance spectroscopy and voltammetry. They found that nucleation’s activation energy increases when NH$_4$Cl is added to reduce the solution resistance and increase the system resistance charge transfer. Based on NH$_4$Cl strong complexity [42], the reaction:

$$\text{Ni}^{2+} + x\text{NH}_4^+ = \left[\text{Ni}\left(\text{NH}_3\right)_x\right]^{2+} + x\text{H}^+$$

shows the dissolving of NH$_4^+$ in the water reacting with Ni$^{2+}$, therefore, Ni$^{2+}$ in the solution changes from [Ni(OH)]$^+$ and [Ni(Cl)]$^+$ + [Ni(NH$_3$)$_x$]$^{2+}$ partially. Ammonium complexes are greater than the other two, affecting the electrode reaction and changing the coating morphology, inhibiting cathode surface nucleation. The screw dislocation growth theory is one of the phenomena that elude about NH$_4$Cl effect on coating morphology. It has been proven that atom in crystal growth is deposited as growth mode in their early stages. Also, accumulated atoms release internal stress in crystal lattice shearing displacement and dislocations formation. Dislocations as a step source always appear at the crystal interface and progress the growth steps spirally around, forming a spiral shape. The screw dislocation step distance is calculated using Equation (6) [43].

$$d = 19\rho_Y = 19V_mY/kT\ln\left(1 + \sigma\right)$$  \hspace{1cm} (6)

with $\sigma$ being the relative supersaturation solution growth interface, which can be calculated by Equation (7):

$$\sigma = a/a_* - 1$$  \hspace{1cm} (7)

with $a$ and $a_*$ it was being the activity of the saturated solution and supersaturated solute. The smaller the step distance, the greater the supersaturation. Besides hydrolysates and chloride [41], it was analyzed above that the addition of crystal modifier limited crystal growth high energy plane and absorbed the nickel ion electrolyte on one side of the crystal.

Hashemzadeh et al. [41] eluded that the coating morphology, wetting ability, and anti-corrosion performance could be affected by the crystal modifier in changing the preferred crystal orientation. Besides, Ethylenediamine was added to NH$_4$Cl and H$_3$BO$_3$ as a crystal modifier to adjust the coating surface nano-structure [44]. Nevertheless, the preparation of superhydrophobic coating based on crystal modifier and micro-morphological structure formation mechanism is further studies.

### 3.2. Current Density

An experiment has found that morphology and wettability of coating have been influenced by current density at a lower voltage [45]. It was analyzed that at high
current density, crystal size decreases, and at low current density, it increases, as shown (Figure 3(a) and Figure 3(b)). The micro-nano dual-scale structure appeared on the coating surface at an increased current density with an increased hydrophobic effect (Figure 3(c) and Figure 3(d)). A specific range causes it with an increased current density; the grain sizes become refined. The increased current density increases the cathodic overpotential, thereby providing another nucleation force, and the cathode nucleation rate gets more significant than the growth of the crystal [46], decreasing the crystal size. The current density becomes broadly enough at the diffusion-limited value in that the material diffusion process controls the whole electrodeposition process. Some crystals became passivated, forming micro-nano dual-scale structure and deposited ions on the cathode surface in time by the process of discharge.

However, current density does not always relate positively in terms of coating superhydrophobicity and anti-corrosion performances. Liu et al. [47] prepared a coated steel where the current density was low. It was found that the deposited crystals were sparse and uneven as the nanostructure could not grow further, but at an increased current density, micro-nano dual-scale structures were formed due to the growth of bud structures and eventually increases the hydrophobicity. Nevertheless, the nanostructure’s disappearance is due to crystal growth continuity, which results in a change to a single microstructure at a continued increase current density with crystals growth. This single microstructure hinders the superhydrophobic effect, which is not conducive.

Liu et al. [48] found that an optimal current density exists during the preparation of ceria coating on the magnesium alloy surface. The anti-corrosion performance could be affected if there is an increase in the current density, increasing the number of cracks, and widening of cracks.

Consequently, an excellent current density determines an optimal hydrophobic and corrosion resistance potential of a superhydrophobic coating.

![Figure 3](image-url). An increased current density nickel coatings imagery was observed by SEM [45].
3.3. Deposition Time

An optimal deposition time value can be obtained considering the effect of electrodeposition time on coating performance. An increase in deposition time will eventually decrease the hydrophobic effect beyond the critical point, and before the critical point, the deposition time will increase with an increased hydrophobicity. Wettability and surface morphology determine the superhydrophobicity, thereby affecting the coating morphology, Li et al. [49]. During a superhydrophobic zinc coating preparation on a steel surface based on a two-step electrodeposition technique, the water contact angle increases slowly as a secondary deposition time from 1 min to 3 min. The surface morphology changes from flat to full texture with an increase in roughness based on SEM observation. Afterward, the water contact angle did not increase further but decreases as the deposition time increases continually, with SEM results showing new surfaces relatively flat with decreased roughness. Simultaneously, if there is a shorter deposition time, the coating thickness will be small and easily crack and warp. Liu et al. [48] found that at just 5 min of deposition time, a coating surface got full of cracks and bends, shown in (Figure 4(a1)). An excellent anti-corrosive effect may not be obtained because the droplet’s adhesion with the surface was still vast after surface modification. Nevertheless, as the deposition time was increased to 60 minutes, more nanosheet structures were formed on the coating surface with increased coating thickness. The superhydrophobic and anti-corrosion effect becomes reinforced in which the cracks vanished with dense coating surface, as shown in (Figure 4(b1) and Figure 4(c1)). In brief, the superhydrophobic anti-corrosive coating task is realized based on superhydrophobicity relative to the surface morphology, which makes it unique from other anti-corrosion coatings. Therefore, deposition time necessity is more rigorous.

4. Metals Electrodeposition

4.1. Mechanism

Electrochemistry is the fundamental of electron deposition on a metal. It has to do with the movement and deposition of metal ions between the electrode and electrolyte, yielding to a new phase under an electric field. Past studies have proven that metal electrodeposition and an accepted theory have been formed [50]. There are two metal deposition stages, ranging from the transportation of

![Figure 4](image-url). A coating deposited due to increasing deposition time was observed by SEM imagery [48].
ions from the electrolyte to the electrode surface and deposited, following the atom being placed into the crystal lattice and growth [51]. This concord with the law of cathodic reduction of aqueous solution and the fundamental law of crystallization processing, but somehow complicated to the kinetic law of cathodic reduction because the electrode attracts particles continuously. The influencing of each of these processes resulted in the complexity of the metal deposition process on an electrode, specifying the four steps [52]: 1) Reaction particles (hydrated ions and complexions) move to the electrode through the process of electromigration, convection, and diffusion, respectively; 2) Rearrangement of water molecules or coordination number of metal complexes known as a pre-conversion process; 3) Reaction particles absorbed at the interface of electrode and solution which is the charges transfer process; 4) Crystallization of electro-process, in which atom from cathodic reduction develop to form crystal nuclei and grow later to form crystals, or, newly atoms spread on electrode surface at a particular position and enter the lattice and grow on the main metal lattice [53]. However, based on these four steps, the highest resistance and slowest rate step become the electrodeposition rate-determining step with changes in the electrochemical system and conditions. Both observation technology and chemical measurement technology are tools use to study the metal deposition of a system. For example, Cu on Ru surface in a sulfuric acid solution was prepared in observing the electrodeposition process with materials such as cyclic voltammetry, current-time transient measurement, and atomic force microscope being used. The obtained data showed that the Cu deposited or crystallization process did not meet the two-dimensional and three-dimensional nucleation model [54] with the growth of Cu crystal appearing anisotropy. Studies show that Ni deposited and crystallized on the Cu surface satisfied the instantaneous nucleation mechanism, and the main kinetic parameters were calculated [55]. Wasekar et al. [56] analyzed the mass transfer process at the cathode and developed a crystal size mathematical model of Ni with boron, sulfur, and phosphorus being present. It shows that deposition rate, current efficiency, and deposition particles corresponded and reciprocated the metal in the coating and current density, respectively. Electrochemical systems are becoming more complex in many fields by applying the electrodeposition technology with similarities and differences that can be studied in different systems using various technical means.

4.2. Magnesium Substrate Coating

One of the most common elements of the planet is magnesium. Magnesium and its alloys are widely used in aerospace, electronic devices, automobiles, and other fields because of their high strength, low density, excellent electrical conductivity, and electromagnetic shields [57]. The negative potential and activities of magnesium, on the other hand, are higher than those of iron, copper, and aluminum. It leads to a greater risk of corrosion [58]. On a surface of an AZ31B magnesium alloy with a WCA at 150˚ electrodeposition of 0.65 mA/cm², Liu et
On a surface of an AZ31B magnesium alloy with a WCA at 150˚ electrodeposition of 0.65 mA/cm², Liu et al. [48] developed a superhydrophobic CeO₂ coating over 60 minutes. Following the improvement in stearic acid, the WCA of the coating increased to 158˚, and petal nanosheets covered the surface. The sample anti-corrosion efficiency remained at about 90% after 48 hours of soaking at NaCl solution, which indicates that the coating had strong corrosion resistance to the substratum. After 60 days of atmospheric exposure and water droplet interaction with pH 1 - 13, however, the hydrophobic effect began to deteriorate sharply after 100 hours of diving in NaCl solution. Moreover, the superhydrophobic effect was discovered, and the coating no longer offers corrosion protection after being dragged by 140 cm with a weight of 50 g on a 600-grain sandboard. Metal ions can react directly to the electrolyte’s solution during the electrodeposition with functional groups of low-surface energy compounds resulting in the forming directly on the cathode surface of low surface energy metal compounds. Without surface modification after electrodeposition, this method generates a powerful hydrophobic effect. For instance, Zheng et al. [59] electrodeposited magnesium alloys with a magnesium nitrate and stearic acid ethanol solution as an electrolyte for 30 minutes at a constant voltage (30V). According to X-ray, the cover was composed principally of magnesium stearate, which had a small quantity of magnesium oxide, and a maximum WCA 156.2, and SA 4.9˚ photoelectron spectroscopy, the Fourier Transform Infrared Spectrometer, and Energy Dispersive Spectrometer. The current corrosion density of the layer was significantly lower than that of the base substrate after seven days in a 3.5-percent NaCl solution, and the WCA remained approximately 150˚ so that the surface coating could have relative long-term resistance to corrosion.

4.3. Aluminum Substrate Coating

Their high strength, low density, strong electrical and thermal conductivity, good processability, aluminum, and its alloys are commonly used in industrial fields.

While Al readily forms an oxide film in the air, which provides some protection, this film has poor corrosion resistance. The film is quickly dissolved when it comes into contact with acidic or alkaline media, causing extreme pitting corrosion, crevice corrosion, and other local corrosion. Using an ethanol solution containing nickel nitrate hexahydrate and stearic acid as the electrolyte, Xu et al. [60] prepared nickel stearate/nickel hydroxide superhydrophobic films on chemically cleaned aluminum alloy substrates by one-step electrodeposition. The results revealed that when the Ni²⁺/SA molar ratio was 0.4, the film’s surface had the best hydrophobic efficiency, with a WCA of 160˚ and a contact angle hysteresis value of 2.1˚. According to SEM photos, the formation of micro-nano dual-scale cauliflower-like structures on the surface was the fundamental explanation for its superhydrophobicity. Figure 5 depicts the polarization curves and the resulting polarization resistance and current density of the coatings. According
Figure 5. Represent (a) Polarization curves of potentiodynamic; (b) Polarization resistance, (c) Current density of a various sample with nickel stearate molar proportion [65].

to polarization curves, the corrosion resistance of the superhydrophobic coating-coated substrate was about 280 times higher than that of the uncoated substrate. The prepared coating’s durability, however, had not been mentioned in this literature.

Cathodic electrodeposition is the most widely used form; however, anticorrosive superhydrophobic coatings can be obtained by anodic in some cases. For example, Zhang et al. [61] prepared a superhydrophobic layer on aluminum alloy’s surface by anodic electrodeposition in the mixed solution of ferric chloride and myristic acid with aluminum alloy as the anode. During electrodeposition, Al was firstly oxidized to Al$^{3+}$ and then combined with CH$_3$(CH$_2$)COO—from myristic acid to form aluminum myristate and deposited on the anode surface. Finally, the media was covered with the micro-nano structure to form Danxia-landform-like morphology with a WCA of 155.1°. Through electrochemical tests, the corrosion potential of the matrix changed from $-1.280$ V to $-0.869$ V, and the corrosion current density was reduced by 85 %, demonstrating that a good corrosion protection effect was achieved. The successful cases of preparing various superhydrophobic coatings on the substrate by metal electrodeposition in the past six years are listed in Table 1. In summary, various superhydrophobic layers have been trained on different substrates metal electrodeposition to achieve a good anti-corrosion effect. However, in most cases, surface modifica-
tion is required, and the hydrophobicity depends on the micro-nano dual-scale structure of the surface so that the wear resistance and durability of the coating need to be further improved.

4.4. Copper Substrate Coatings

The copper’s excellent electrical conductivity, thermal conductivity, and ductility [62] are used in electronic, electrical, and other fields worldwide. Though copper is used widely, studies show that copper is resistant to corrosion in the saline environment [63]. Henceforth, fabricating coating on copper will avoid the adsorption and penetration of the saline particles. An experiment was successfully conducted with copper as a substrate at 60°C and pH four composing of the electrolyte solution of NiCl₂, H₃BO₃, and C₂H₁₀Cl₂N₂ by electrodeposition [64]: the result shows that the nickel coating current density (20 mA/cm² for 600 s and 50 mA/cm² for the 60 s) reflect an excellent superhydrophobic property at a water contact angle of 155°, and SEM tells that the coating has micro-nano dual-scale structures distributed on the nickel cone. Moreover, the superhydrophobic coating had excellent corrosion resistance in a 3.5 wt% NaCl solution based on electrochemical impedance spectroscopy and polarization curves analysis.

4.5. Steel Substrate Coatings

Though steels are widely used metal in the construction and industrial sectors due to their low price; corrosion is inevitable, causing the researcher never to stop developing an anti-corrosion substance to combat the effect. Past work has prepared zinc coating on X60 Steel by Electrodeposition using ZnSO₄ solution as an electrolyte, and current density (4 A/dm² for 20 min and 9 A/dm² for 3 min), pH four at temperature 25°C [49], and finally added perfluorooctanoic acid to the ethanol solution. The water contact angle was 154.21° and the sliding angle less than 5°, respectively. Liu et al. [47] prepared a Cu-Ni superhydrophobic coating by immersing 316 l stainless steel in NiCl₂, H₃BO₃, and CuCl₂ electrolytic solution at a current density (8 A/dm² for 20 min) and found that dense branch-like structure was deposited on the matrix, shows the surface morphology in (Figure 6(a) and Figure 6(b)). Structure size was about 4 - 8 micrometer, and dendrite structure was 300 - 600 nm, with water contact angle 161.27° and sliding angle 7.8°. Electrochemical test in 3.5 wt% NaCl solution was done to study the coating’s anti-corrosion performance, seen in (Figure 6(c) and Figure 6(d)). It was noted that the Cu-Ni coating obtained a better corrosion resistance corresponding to the lower corrosion current density. The coating anti-corrosion effect was 99% based on a calculation done by Equation (5).

\[ n = \frac{I_{\text{corr sub}} - I_{\text{corr coated}}}{I_{\text{corr sub}}} \times 100\% \]  

The excellent result of the coating anti-corrosion effect shows tremendous significance. After six months of exposure to air and 12 hours immersion in a strong acid with pH = 1, the coating contact angle had no change, but at pH = 14 in a strongly alkaline solution in 4 hours, the coating effect dropped speedily.
Other metal, such as aluminum is somehow less selective due to its poor corrosion resistance in a thin film, which may cause pitting or crevice corrosion once it contacts acidic or alkaline media. Thou has a low density, strong electrical and thermal conductivity, excellent processability, and high strength. Xu et al. [65] prepared an ethanol solution containing nickel nitrate hexahydrate and stearic and develop a nickel stearate-hydroxide superhydrophobic coating on aluminum alloy an excellent hydrophobic performance with the water contact angle, 160˚ ± 1˚ and hysteresis value to be 2.1˚ ± 1˚. Formation of micro-nano dual-scale structure on the surface was observed by SEM factoring hydrophobicity. (Figure 5) shows the coating polarization curve, the resulting polarization resistance, and current density, respectively. The polarization curve was used to calculate the superhydrophobic coated substrate’s corrosion resistance, which was 280 times higher than the substrate without coating. Although electrodeposition is used widely to fabricate coating on metal, anodic electrodeposition can also be applied to develop such a result. For example, reference [66] uses anodic electrodeposition with an electrolyte of ferric chloride and myristic acid on aluminum alloy substrate as the anode and successfully developed a superhydrophobic coating.

Micro-nano particles form a landform-like morphology with a water contact angle of 155.1˚, and electrochemical tests show that the corrosion potential changes from −1.280 V to −0.869 V, with a reduced current density of 85%, displaying excellent corrosion protection. Furthermore, Table 1 shows superhydrophobic
coating cases on the substrate based on metal electrodeposition for the past five years. In summary, metal electrodeposition has been developed by various coating substrates to achieve a good anti-corrosion effect. Nevertheless, surface modification is required in most cases, and the hydrophobicity is based on those micro-nano dual-scale structures present on the surface, to further improved the wear resistance and durability of the coating.

Besides metals, other nonmetals can be electrodeposited, forming a superhydrophobic coating. Nonmetal with conductive polymers with low surface energy can produce this superhydrophobic effect by one-step electrodeposition, for example, glass. An experiment shows that polythiophene on indium tin oxide glass by electrodeposition method was fabricated and form a superhydrophobic coating [67].

In another exciting experiment [74], a precursor solution of dodecyltrimethoxysilane (DTMS), ethanol, and potassium nitrate were mixed by preparing superhydrophobic DTMS silane films on low carbon steel based on electrodeposition on a three-electrode system. Researchers further proved that the coating effect was the best, with deposition time to be 300 s, applied potential at −1.3 V, and water contact angle of 152.9˚ and 3˚ sliding angles, respectively. The SEM analysis reflects nanosheets that were entangled to form a walnut-like micro-bundle on the substrate. (Figure 7) shows the potentiodynamic polarization curves of the sample prepared in 3.5 wt% NaCl liquid. However, the coated substrate’s corrosion density was calculated, and from that, it was reduced by three orders of magnitude than the bare substrate with an anti-corrosion potential of 99.74%. Even though 3.7 micrometer was the thickness of the prepared coating, its protection potential exceeded greatly than 30 μm thickness of superhydrophobic epoxy coating [75]. The water contact angle decreases after immersing in a NaCl solution for three days but steadily rises at 134.9˚ after eight immersion days. Past work invented that DTMS through hydrolysis in precursor solution produces a silanol group, where water electrolysis and oxygen reduction form an OH- radicle during electrodeposition.

This catalyzed silanol groups condensation process on the metal surface, cross-linking structure. In the end, DTMS adhered to the substrate as shown in (Figure 8) based on the one-step electrodeposition, which resulted in a micro-nano

Table 1. Some metals electrodeposition of superhydrophobic coating, several examples.

| Matrix       | Coating   | Electrolyte composition                        | Process conditions | WCA        | SA         | Ref  |
|--------------|-----------|------------------------------------------------|-------------------|------------|------------|------|
| Be-Cu alloy  | Cu Stearate | CuCl2 + HCl + stearic acid                       | 5 V (10 min)      | 163˚ ± 4˚  | 1.7˚ ± 0.2˚ | [68] |
| X90 steel    | Cu-Zn     | ZnSO4 + CuSO4 + NaOH + C4O6H4 KNa               | 2 A/dm² (60 min)  | 154.73˚   | 6.5˚       | [69] |
| Mg alloy     | Ce myristate | Ce(NO3)3 + myristic acid                       | 20 V (10 min)     | 160.4˚ ± 0.7˚ | -          | [70] |
| Mg alloy     | Ce stearate | Ce(NO3)3 + stearic acid                        | 30 V (10 min)     | 158.4˚ ± 0.9˚ | 2˚         | [71] |
| C45 steel    | Fe        | FeCl3 + palmitic acid + ethanol                 | 30 V (10 min)     | 160.5˚ ± 0.5˚ | 2˚ ± 0.5˚  | [72] |
| Copper       | Al stearate | AlOx + stearic acid                           | 30 V (10 min)     | 161˚ ± 1˚  | 2˚ ± 1˚    | [73] |
Figure 7. Curve 1 is Tafel plots of bare; Curve two is salinization treated traditionally, and Curve three portray MS substrate coated with DTMS superhydrophobic coating [74].

rough structure on the coating surface; however, DTMS achieved lower surface energy which eventually produced an excellent superhydrophobic and anti-corrosion effect.

5. Nonmetal’s Electrodeposition

Electrodeposited superhydrophobic coatings can be made from a variety of materials, not just metals. Many nonmetals, especially low-surface-energy conductive polymers, may achieve superhydrophobic effect directly by one-step electrodeposition, avoiding secondary surface modification and expanding the preparation of coatings. Xu et al. [76] used electrodeposition to create a series of polythiophene films (PL-PEDOT) on indium tin oxide (ITO) glass, with wettability. They used cyclic voltammetry to render the first super hydrophilic layer of poly(3,4-ethylene dioxythiophene) (PEDOT) on an ITO electrode. The overlying hydrophobic poly (2,2'-bithiophene) (PLTH) moieties with various electrodeposition charges (EC) were electrodeposited on the as-prepared PEDOT substrate. The coating surface’s wettability changed as the EC of PLTH changed, with the WCA steadily increasing from 0˚ to 150.4˚. Many fluorine-containing carbon chains have low surface energy, and they can be combined with specific organic substances through chemical reactions to produce new hydrophobic subderivatives (EOTT-Fn) with semi-fluorinated chains of various lengths. These derivatives were deposited on the gold substrate’s surface after electrodeposition, and that when n was more significant than 4, the character was superhydrophobic, with a contact angle greater than 158.9˚. They then produced an EDOT derivative with a short, fluorinated chain (C4F9) and a highly rigid methoxybenzothioate spacer [77]. The polymer monomer was then mixed with different electrolytes to induce electrodeposition on a gold plate. Following that, nanofibrous structures of various sizes were observed on the substrate’s surface, with a WCA of around 160˚, indicating that the coating had achieved a very superhydrophobic impact. Wang et al. [70] used stances to make a precursor solution used in electrodeposition to make superhydrophobic films. Darmanin et al. [78] synthesized dodecyltrimethoxysilane (DTMS) from a mixed solution of ethanol and potassium nitrate using 3,4-ethylensynthetic thiophenes (EOTT) as platform mo-
Superhydrophobic DTMS silane films were prepared on a low carbon steel surface using a three-electrode method after the precursor solution had been wholly hydrolyzed. According to further research, the hydrophobic effect was better when the deposition time was 300 seconds and the applied potential was $-1.3 \, \text{V}$. The WCA was $152.9^\circ$ at the time, and the SA was just $3^\circ$. According to SEM observations, the nanosheets were intertwined with each other to form a walnut-like micro-bundle on the substrate’s surface. Three orders of magnitude reduced the substrate’s measured corrosion current density with superhydrophobic DTMS coating compared to the bare substrate. The anti-corrosion performance was as high as 99.74 percent based on the prepared samples’ potentiodynamic polarization curves in 3.5 wt% NaCl solutions Figure 7. The coating’s safety performance was much higher than that of the superhydrophobic epoxy coating with a thickness of 30 m [79], despite its thinness of just 3.7 m. The coating’s WCA started to decrease after three days of immersion in NaCl solution but remained relatively strong at $134.9^\circ$ after eight days of immersion and remained nearly unchanged in the following year. Further research revealed that DTMS first formed active silanol groups in the precursor solution by hydrolysis. Water electrolysis and oxygen reduction created OH during electrodeposition, which effectively catalyzed silanol groups’ condensation on the metal surface, resulting in cross-linking structure and covalent binding (Si-O-Si and Si-O-Metal). As a result, as shown in Figure 8. Diagram of (a) DTMS reaction of electrochemically catalyzed condensation; (b) MS substrate coated with DTMS electrodeposition superhydrophobic films [74].
structure due to the drying and curing process, and DTMS has a low evaporation rate.

### 6. Composite Electrodeposition

The composite coating is formed when particles with unique properties combined with the electrolyte and co-deposited on the substrate surface, having a characteristic of the primary phase and high advantages of co-deposited particles. Therefore, a unique self-lubrication, wear resistance, corrosion resistance, and electrical conductivity properties are achieved by the composite coating, possessing an excellent industrial application in general and automotive engineering, and other fields. With sizes in micron and submicron to the nanometer, co-deposition particles phase is observed to be spherical, layered, needle-shaped, and the deposition phases are metals, conductive polymers, alloys, and conductive ceramics; other oxides shapes, carbides, borides, and nitrides [80]. Nanoparticles and metals co-deposition is efficiently used in fabricating a superhydrophobic coating. Particle dispersion in the electrolyte and matrix metal ions particles co-deposition are the two cardinal processes in composite electrodeposition [81]. Based on these, co-deposition models have been developed in the past decades. Guglielmi [82] relates the concentration composition coatings particle to liquid suspended particles and overpotential electrode by modifying electrophoresis and adsorption roles during co-deposition and not considering model factors, like electro adsorption and electrokinetic particles entanglement, and mass transfer [83]. Two forces affect solid particle’s movement to the cathode surface during the composite electrodeposition process: solid particles are suspended due to the bath flow and attract on the cathode vicinity; electrophoresis controls solid particles reaching the cathodic surface under a favorable electric potential capacity [84]. Henceforth, the composite electrodeposition is affected by the electrolyte particle dispersion capability and cathodic current potential, respectively. As shown in (Figure 9), a proposed model [85] of particles co-deposition

![Figure 9. MWCNTs-Ni/a-C:H superhydrophobic film preparation process [98].](image-url)
and essential phases were analyzed through five steps. 1) Ionic clouds and produced during the wrapping of some nanoparticles by surfactants and metal cations. 2) The convection layer, usually 1 mm in diameter, moves particles and these ionic clouds move towards the cathode. 3) They are the potential to reach hundreds of microns diffusing through the boundary layer concentration. 4) These particles are transferred through an electric double layering and adsorbed on a cathode surface in a few nanometer thicknesses. 5) With the reduced metal cation, these particles are co-deposited. An example [86] based on liquid-phase mass transfer analyzed silicon carbide and cobalt mechanism and assumed that particles deposited rate was due to electrode reaction. Fransaer et al. [87] studies spherical particles and rotating disk electrode metal of a co-deposition process, analyzing the effects of fluid velocity, particle concentration, and current density, respectively, but ignoring Brownian motion. The correctness of the above models is limited, which are centered on a particular deposition system. Concurrently, the present models did not rely on the particle inclusion effect of the electro-crystallization process, but the cathode’s area and morphology will be changed by the deposition of a particle on the matrix surface. They will inexorably affect the electro-crystallization process [83]. To develop an applicable composite electrodeposition mode, the author acknowledged that hydrodynamic, electrochemical, and adsorption factors must be considered exhaustively.

6.1. Corrosion-Resistant Performance

Many coatings developed in the past decades have poor corrosion resistance due to the micro-nano structure frailty. Many of the coatings are toxic and have a low adhesion rate to the substrate’s surface. Such coating locks mechanical durability to substance the substrate life spends. However, the electrodeposition has solved these problems to some extent by preparing an efficient anti-resistant composite coating. Mainly with graphene and carbon nanotubes being a new material for exploring composite electrodeposition expansion in fabricating superhydrophobic coatings. (Figure 8)

6.2. Graphene-Based Composite Coating

Material science has been supported brilliantly through graphene since the 21st century [88]. Graphene is hydrophobic and has a high specific surface area with excellent electrical conductivity [89] and mechanical properties applied in many fields. This technology has been explored in recent years by researchers globally for superhydrophobic composite coating. A chemical method was used by Choi et al. [90] in preparing petal-shaped graphene/Nafion hybrid films with an excellent hydrophobic effect, the water contact angle is 161°. The complex pretreatment graphene oxide (GO) and Nafion were treated with hydrazine solution at 850°C for 24 h, which was needed and needed purifying by dialysis for a week proving the preparation complexity. On the other hand, graphene superhydrophobic coating preparation using composite electrodeposition is simple. An
electrodeposition composite coating was prepared by directly adding graphene multi-layer to an electrolyte composed of nickel at a current density of 20 mA/cm² for 5 min. After being modified with myristic acid, a water contact angle was obtained to be 160.4° ± 1.5° with a sliding angle low as 4.0° ± 0.9°. Micron graphene bulge had randomly nano-nickel conical arrays, which were observed under SEM. A significant anti-corrosion effect was shown depending on the polarization curves of extrapolation results. It implied that the corrosion current density (about 0.005% of bare media) and polarization resistance was about 1.88 × 10⁴ times greater than the bare substrate. It was proven that after 60 cycles of wear test at 1.0 kPa, the water contact angle did not decrease, indicating an excellent coating wear resistance. Nevertheless, the coating prepared by the above method had a low graphene content, which needs modification. Bai et al. [91] upgraded this process by combining a high current approached and nickel pre-deposition. A graphene oxide/nickel composite electrodeposition coating of pinecone-like micro/nanostructure was placed on stainless steel substrate at a water contact angle of 162.7° ± 0.8° sliding angle of 2.5° ± 1°. Based on results from polarization curves, 99.98% obtained as the anti-corrosion efficiency after immersing in a 3.5 wt% NaCl solution for 36 h. Interestingly, after 100 cycles of the mechanical abrasion test, the water contact angle and sliding angle slightly decreased, demonstrating excellent mechanical stability. In recent years, some cases in which superhydrophobic composite coatings have been successfully prepared based on electrodeposition are listed in Table 2.

### Table 2. Superhydrophobic composite coating prepared by electrodeposition.

| Matrix | Coating | Electrolyte Composition | Process conditions | WCA  | SA   | Ref |
|--------|---------|-------------------------|-------------------|------|------|-----|
| ITO. Terthiophene (3 T)/3 T-Initiator | Surface-initiated atom transfer radical polymerization (SI-ATRP) | 0 - 1.2 V, a scan rate of 10 mV/s, 20 Cycles | 153° ± 2° | 0° | [94] |
| ITO. Perfluoroalkyl alkyl pyrrole | - | 1.25 V (300 s) | 152.3° | No SA | [95] |
| Gold PEDOT derivatives | - | 100 mA/cm² | 159.4° | 3.5° | [96] |
| Gold Fluorinated polypyrrole | Bu 4NClO₄ + anhydrous acetonitrile + pyreneMonomers | −0.7 V to EW(1.4 - 1.9 V), a scan rate of 20 mV/s | 160° | 1.0° | [97] |

6.3. Carbon Nanotube-Based Composite

Carbon nanotube (CNTs) as a one-dimensional nanostructure material, since the 90s, has been widespread for composite technology [92]. This material is an excellent choice for composite coatings fabrication because of its chemical and physical characteristics, including high surface area, good lubricity, high hardness, and wear resistance. A ZnO multi-walled carbon nanotubes (MWCNTs) characteristic was prepared on the copper mesh using the Sol-gel method, but PDMS must modify the coating for an excellent superhydrophobic effect [93].

However, a one-step electrodeposition technology using nickel(II) acetylacetonate
Table 3. Electrodeposition superhydrophobic composite coatings fabrication examples.

| Matrix    | Coating   | Electrolyte composition                                                                 | Process conditions         | WCA    | SA    | Ref  |
|-----------|-----------|-----------------------------------------------------------------------------------------|----------------------------|--------|-------|------|
| Copper    | Ni/TiO2/TMPSi | Plating solution: NiCl2 + H3BO3 + NiSO4 + SDS + TiO2 Modification: TMPSi                | 80.7 A/dm² (19.17 min), 50.5˚C + Surface modification | 151.6˚ | 6.2˚  | [22] |
| mild steel | Ni/WS2    | NiSO4 + NiCl2 + H2BO3 + CTAB + WS2 + Saccharin                                            | 4 A/dm² (20 min), 40˚C    | 158.3˚ | 7.7˚  | [100]|
| Stainless steel | Cu/MoS2   | CuSO4 + H2SO4 + MoS2 + CTAB + thiourea                                                 | 50 mA/cm² (2 min)         | 158.2˚ | 1.8˚  | [101]|
| Carbon steel | Ni/WC     | NiSO4 + NiCl2 + H2BO3 + CTAB + WC                                                      | 6 A/dm² (30 min), 40˚C    | 164.3˚ | 0˚    | [102]|
| Copper    | Ni/PTFE   | NiSO4 + NiCl2 + H2BO3 + PTFE particles                                                  | 100 mA/cm² (15 min), 60˚C | ≈155˚  | -     | [103]|

and MWCNTs methanol as the solution was used to develop an MWCNTs-Ni composite coating silicon media [98]. The electrodeposition preparation process was facile and superficial with a water contact angle of 158.89 and a sliding angle of 1.99, respectively. After 20 wear cycles, the coating maintains a superhydrophobicity displaying good wear resistance. The sample coating had an excellent anti-corrosion effect, with the corrosion current density decreasing notably after immersing into 3.5 wt% of NaCl solution.

Another experiment [99] was done with silicon wafers coated with a prepared MWCNTs-CO/a-C: by composite electrodeposition method, which obtained WCA to be 158.1˚ and SA to be 2.98˚. In 3.5 wt% of NaCl solution, the coating’s anti-corrosion potential reached 98.02%, and excellent wear resistance has proven by the sandpaper wear test. Composite electrodeposition derived the shortcoming of metal electrodeposition, providing knowledge for multi-functional composite superhydrophobic coating preparation. Secondary surface modification can also be avoided by adding suitable particles to the electrolyte solution so that the coating wear resistance and stability can be improved with a significant and durable anti-corrosion effect of the substrate.

7. Current Electrodeposition Development

Besides the widely used traditional electrodeposition technology for preparing a superhydrophobic coating, the current development of electrodeposition technology has shown significant advantages for corrosion protection; these developments are 1) Ultrasound-assisted electrodeposition; 2) Jet electrodeposition; 3) Pulse electrodeposition, and 4) Magnetic field-induced electrodeposition.

7.1. Ultrasound-Assisted Electrodeposition

An acoustic wave of 20,000 Hz frequency and above is known as ultrasound. According to studies, ultrasound can improve nanoparticle dispersion in the electrolyte and, significantly, change the coating morphology [104]. The presence of ultrasonic cavitation plays an essential role in the ultrasonic electrodeposition process, propagating through the liquid medium as positive pressure and negative pressure, as shown in (Figure 10). The intermolecular force of expansion becomes greater, forming tiny bubbles in the electrolyte during the negative pressure circulation. These bubbles have some physical and chemical impacts on
the electrodeposition process by continually gaining energy and expanding in volume, eventually collapsing with a large amount of heat and shock wave being released and jets at the moment of destruction [105]. It has been believed that ultrasonic cavitation affects the electrodeposition process in three ways: 1) deposition process strengthening effect; ultrasonic cavitation micro-jet stirring solid effect on the electrolyte can enhance ions movement, thickness reduction, and diffusion layer concentration gradient. It also accelerates the electrodeposition process and reduces the concentration polarization. 2) Grains size refinement; high temperature and pressure produce by ultrasonic cavitation and even release nearby local instant low temperature, protect grain growth, critical nucleation radius reduction, and nucleation rate improvement. (iii) Dispersion and stirring of nanoparticles; nanoparticles agglomerates are sometimes crushed by ultrasonic cavitation, uniformly distributing the solution nanoparticles. In many past experiments, the ultrasonic wave has been used to fabricate superhydrophobic coating by electrodeposition process as an excellent dispersing of plating solutions [106] [107].

Tudela et al. [108] found that nanoparticle distribution in a nickel matrix under ultrasound of a WS2 electrodeposition process was more uniform with refined grain size and improved hardness, indicating that ultrasound could increase nanoparticle deposition in a superhydrophobic coating. Li et al. [23] fabricated Ni-Cu/TiN composite coating on copper using ultrasound-assisted electrodeposition technology. It shows that ultrasonic waves’ presence enhances the composite coating properties’ improvement, mainly, the anti-corrosion performance. Therefore, ultrasonic cavitation is essential in fabricating a superhydrophobic coating based on electrodeposition because of the grain size refinement and coatings morphological control.

7.2. Jet Electrodeposition

Jet electrodeposition implies a favorable voltage applied between the cathode and anode, thereby jetting the electrolyte only on the cathode at high speed. In the jet coverage area where electrodeposition usually occurs, current passes through, and substances are deposited compared to no electric current area where the substance cannot be deposited. Henceforth, with comparison, jet electrodeposition is reasonable compared to traditional electrodeposition technology.
These are few reasons jet electrodeposition is advantageous: 1) It is selective; metals are deposited in the desired area during anode nozzle and cathode movement, making it easier for electrodeposition complex part implementation to save raw materials. 2) Substantial mass transfer to the cathode by high-speed jet creates a state of increasing the limit of current density and diffusion layer reduction on the cathode surface. 3) High current density increases the cathodic overpotential, refining grains and inducing micro-nano scale appearance for hydrophobicity improvement. 4) High current density can improve the potential deposition effect. 5) High jet velocity reduces hydrogen bubbles adhesion on the cathode surface; this reduces hydrogen embrittlement, pinholes, pitting, and minor cracks.

Shen et al. [110] prepared a superhydrophobic nickel coating through jet electrodeposition process by spraying nickel ion from the nozzle to graphite media surface in 4 L/min. Cauliflower-like clusters covered the coating surface, which was projected by SEM, indicating an excellent superhydrophobicity. The water contact angle was 155.5˚ and the sliding angle 6.5˚. The as-prepared coating maintains its stability after being exposed 6 months to air and stored at 200°C for four hours.

7.3. Pulse Electrodeposition

During pulse electrodeposition, the current is turned on and off in a particular pattern compare to the traditional DC electrodeposition process. It makes it unique for superhydrophobic coating fabrication [111] [112]. Metals ions at the cathode are reduced and deposited while current is conducted in that concentrated ion decrease tremendously. Nevertheless, when the current is switched off, the metal ions diffuse in the solution producing a new concentration ion at the cathode surface and stopping the ion reduction process. The current deposit upper limit is increased in that the diffused layer thickness reduces and decreased the polarization concentration. The two-dimensional nucleation rate \( H \), crystal nucleus critical radius \( r_c \). Moreover, the cathodic overpotential \( n \) best agreed with the below Equation (8), and Equation (9).

\[
H = K_c \exp \left(-\frac{bS}{\alpha}ight) \exp \left(\frac{E_x}{kT}\right) n
\]

(8)

\[
r_c = \pi h^2 E/6\alpha n
\]

(9)

Based on these equations, the nucleation rate \( H \), and the cathodic overpotential \( n \) increase instantaneously but the crystal nucleus critical radius \( r_c \), decreases with cathodic overpotential \( n \) being increased. Henceforth, the nucleation process enhances and was improved due to the high current density, which increases the cathodic overpotential, and the epitaxial crystal growth being interrupted as the current switched-off terminating grain coarseness. Although micro-nano dual-scale structures are foundational to superhydrophobicity, pulse electrodeposition characteristics are conducive to these structures’ growth, reducing cathodic hydrogen evolution reaction to obtain an excellent fabrication.
However, adjustable components such as waveform, period, frequency, duty cycle, current density, and deposition time affect the coating’s performance, and with better morphology control will characterize the pulse electrodeposition to the traditional DC electrodeposition technique in comparison. Pulse electrodeposition can be understood by Equation (10) and Equation (11).

\[
T = T_{on} + T_{off}
\]

(10)

\[
f = 1/T
\]

(11)

Equation (12), and Equation (13), respectively

\[
\gamma = T_{on}/T = T_{on}/T_{on} + T_{off}
\]

(12)

\[
I_{\rho} = I_{m}/\gamma
\]

(13)

\(T\), \(T_{on}\) and \(T_{off}\) represent the period, pulse-on time, pulse-off time, frequency is \(f\) and duty cycle is \(\gamma\), and peak and the average current density are \(I_{\rho}\) and \(I_{m}\).

Chen et al. [113] investigated the morphology and wettability of a nickel coating on copper base on pulse electrodeposition processing and how it is affected by peak current density, pulse-on time, and duty cycle. The results prove that, at an increased peak current density, nickel cone density increases with decreasing crystal size. On the other hand, they found that as the duty cycle increases, the nickel cone side wall’s roughness increases with nanocrystals’ secondary growth appearing. Both the duty cycle and the coating morphology were the same base on the effect of pulse-on time.

As the pulse-on time was extensive, sharper cones covered with shapeless secondary nuclei were seen. These components discussed above effectiveness on coating’s morphology affected the cathodic overpotential and ion mass transfer system.

Sangeetha et al. [114] use pulse electrodeposition to prepare Ni-W/PTFE composite coating on carbon steel surface, achieving an excellent superhydrophobic effect self-lubricating characteristic. Results are shown in (Figure 11), which relates the electrochemical impedance spectroscopy and potentiodynamic

![Figure 11](image)

**Figure 11.** Ni-W alloy matrix and Ni-W/PTFE coatings showed changes of PTFE particle concentration by Nyquist plot [114].
polarization curves of the as-prepared coating. It is observed that the Ni-W/PTFE coating has a lower corrosion current density, more protective corrosion potential, and a large arc radius compared to the Ni-W alloy hydrophobic coating.

**7.4. Magnetic Field-Induced**

Through the electrodeposition process, the magnetic field can affect both the composition and morphology of a superhydrophobic coating, as being recorded by many recent researchers. Thiemig *et al.* [115] found that nickel-based metals with a static magnetic field contain nano-co particles and magnetic particles perpendicular to the cathode surface, increasing nanoparticle deposition within the nickel base. The parallel magnetic field would reduce the co-deposition amount. Bund *et al.* [116] found within the nickel electrodeposition process after applying the magnetic field that the cathode limiting current density increases while the crystal size decrease, but somehow constant in another experiment [117]. Magnetic field as an influence technique on electrodeposition process; derived from two features, they are: 1) Based on magnetohydrodynamic (MHD) effect; this is called the Lorentz force acting on moving charges that are perpendicular to the magnetic field in that the magnetic field will affect the mass transfer rate in the solution. The magnetic field effect on the material transport is equal to the gently stirring [118]. 2) The co-deposition particles will follow high magnetic flux density based on composite electrodeposition in a ferromagnetic state.

However, the two points signified that the electrodeposition process's magnetic field application is significant in controlling coating morphology. Shen *et al.* [119] added ferromagnetic nano-nickel particle (Nip) of 500 nm in diameter in a plating solution and generated a magnetic field that was parallel by introducing into the electromagnet a constant DC, as shown in (Figure 12). It was a Ni/Nip anti-corrosive superhydrophobic coating on copper by magnetic field-induced scanning electrodeposition. The presence of the magnetic field deposition on the cathode surface developed a uniform micron-nanometer layered structure, and

![Figure 12. A magnetic field induced electrodeposition technology imagery [119].](image-url)
without the magnetic field, the composite coating had a disorderly particle dispersed with some uneven particle sizes. The coating maximum contact angle reaches 155.4˚ with magnetic field strength to be 120 mT, changing from hydrophilic to a superhydrophobic state after five days of exposure to air. The oxidation of nickel exposure to air produces a hydrophobic NiO, causing some wettability changes [120].

Perhaps, hydrocarbons in the air may reduce the surface energy, contaminating the coating [121]. The coating showed a perfect anti-corrosion performance with corrosion potential and corrosion current density to be −0.237 V and 0.06 μA/cm², and measurements were done in 3.5% NaCl solution showing a potentiodynamic polarization curve.

8. Obstacles and Prospects

Superhydrophobic coating preparation has been boom using electrodeposition technology in recent years. Despite that, most coatings are fabricated on a smaller scale in the laboratory without being applied in industries at a larger scale due to: 1) Many as-prepared superhydrophobic coatings still maintaining an insolvent steadiness and durability. At one point, the coating and the media’s poor adherence cause the superhydrophobic coating to be repelled during the use application. Due to the macro-nano structure brittleness, the coating surface structure is damaged by minor collision. Furthermore, the coating characteristic will be lost as the attached material layer falls from the surface because low surface energy materials customize its surface. Wherefore, the corrosion resistance of the coating becomes unstable, not providing a long-term potential. For example, Huang et al. [122] evaluated a coating pH durability with HCl = 1 pH, NaOH = 14 pH and CH₃COOH = 5 pH electrolyte solution, respectively, on dissimilar media applying amended TiO₂; PDMS. The outcome shows that the superhydrophobicity vanished when the coating was submerged for five hours in a robust acid solution, which eventually reduces the coating static contact angle below 150˚. While in a weak acid solution after submerged for six hours, the coating superhydrophobicity was conserved but not standard for empirical approach. Tan et al. [123] also evaluated a coated stainless steel within an electrolyte solution containing sodium hydroxide, sulfuric acid, and 3.5wt.% NaCl and the durability were analyzed, but the superhydrophobicity was vandalized succeeding a 20days soaking with an exceeding water contact angle and a 15° mega sliding angle. 2) Moisture issue; although the mechanism of superhydrophobic coating is repellent of water droplets, yet its bond becomes loose to steam adhering to the coating surface. The coating micro-nano structures become seal with the condensed water vapor to lose the hydrophobic coating effect below the dewing level. 3) Oil moisture issue; petroleum industries use superhydrophobic coatings to protect oil and gas pipelines from corrosion, a significant corrosion issue. Nevertheless, oil on these surfaces limits the water droplet’s surface tension prompting superhydrophobicity disappearances. Wherefore, future research
projects will be essential in fabricating the oleophobic and hydrophobic coatings against corrosion. 4) Preparing a superhydrophobic coating issue: past experiments were productive in fabricating a superhydrophobic coating by electrodeposition process by analyzing the dissimilarity in an electrolyte solution, current density, deposition time, and others. Perhaps the composition may be similar, but the fabrication steps used are dissimilar. Thus, due to the deficiency in the entire technical process or field norms, the substantial industrial applications are hindering.

More than the above faults, current research directions have been released to mitigate these issues. Particularly superhydrophobic coating durability needs significant improvement, or else there will be no relevance for them in corrosion-resistant. Foremost, surface modifiers following the electrodeposition should be circumvented because they are poisonous yet also fall off easily. Rather, few low surface energy substances combined with the electrolyte and deposited on the substrate surface are environmentally friendly. For a coating to be a multi-functional composite substance or superhydrophobicity and excellent wear resistance, and so on, it is achievable to include two or more sort of hydrophobic particles and other exceptional effects to the electrolyte solution.

Moreover, coating strength is ameliorated by optimizing the electrolyte composition. For instance, mussels secret Mytilus edulis foot protein 5 (MefP-5), adhering onto several media quickly [124]. Another researcher evaluated that 3,4-levodopa (DOPA) is responsible for the powerful adhesion [125]. Wang et al. [126] was inspired based on these occurrences and added dopamine to the electrolyte solution, and prepared a zinc/polydopamine/n-dodecyl mercaptan (Zn/PDOP/NDM) multi-functional composite superhydrophobic coatings onto copper, and steel, respectively. The WAC was 167.6˚, and the SA was 1˚; simultaneously, the crosscut tape test and abrasion test shows that the coating had an outstanding adhesion and wear resistance potential.

One of the potential research focuses direction hereafter is fabricating a superhydrophobic coating with an inhibitor that will resist long-lasting performance. The inhibitor is put on the substrate surface so as the coating superhydrophobicity is disoriented; the inhibitor can be emancipated to the coating surface as long-lasting protection. For instance, Zhao et al. [127] fabricated a SiO2 superhydrophobic coating using benzotriazole (BTA) as an inhibitor on the aluminum alloy surface. It was found that the discharge of the inhibitor with a pH mechanism improved the long-lasting anti-corrosion property of the coating. One of the options to enhance a coating’s durability is to fabricate an anti-corrosion coating that contains a self-repairing characteristic, which has been a hotspot research direction currently. If the superhydrophobic coating could have the characteristic of self-healing peradventure, the coating’s durability shall be ameliorated. Based on two points of view, the coating surface energy reduction and the vandalized surface structure.
9. Conclusion

A superhydrophobic coating anti-corrosion potential is achievable due to its exceptional water resistance structure hence and inevitably estranging corrosive substances from encountering the substrate. Electrodeposition application is regarded as a potential technique used in fabricating a superhydrophobic coating for a wide range of constructional and industrial applications or approaches. A substantial edge is in the composite electrodeposition process as it ameliorated the coating steadiness and avoided contingent surface modification. Coatings are prepared based on the strength of electrodeposition application, and their superhydrophobicity and corrosion-resistant potential can be tremendously pretentious by the crystal modifiers, current density, as well as deposition time. The ultrasound-assisted Electrodeposition, jet electrodeposition, pulse electrodeposition, and magnetic field-induced Electrodeposition are applications for substantial research benefits. Even though steadiness and durability are the problems the fabricated coatings are still facing, through adjusting these technologies, the coatings may probably be used extensively, exceptionally, and as an eco-friendly corrosion-resistant application some days.

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Conflicts of Interest

The authors declare no conflicts of interest regarding the publication of this paper.

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