

Influence of Inhibitors on the Corrosion of Al and Al-composites in Chloride-containing Solutions - A Review

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Abstract Corrosion is a natural, inevitable process, and is one of the world's most serious problems. Losses incurred due to corrosion are extremely expensive for society. Several technological strategies have been explored and implemented to address these losses. The use of inhibitors to prevent corrosion is a common and efficient method to reduce corrosion losses. This review covers Al and Al-composite corrosion inhibitors in chloride-containing solutions, because of their popularity in a broad array of industrial applications. A vast number of studies in the literature detail the common tendency of Al and Al-composites with reinforcements to deteriorate. Accordingly, it is worthwhile to employ inhibitors to protect them, as discussed in the present work. The emphasis is on selecting the smartest corrosion inhibitor and evaluating its performance. According to the study, the most commonly used corrosion inhibitors are 1,4-naphthoquinone (NQ), 1,5-naphthalene diol, 3-amino-1,2,4-triazole-5-thiol (ATAT), ammonium tetrathiotungstate, clotrimazole, amoxicillin, antimicrobial and antifungal drugs. Electrochemical impedance spectroscopy (EIS), potentiodynamic (PDP), and weight loss were among the most commonly used modern electrochemical technologies to test inhibitors' efficacy under environmental conditions.

Key words inhibitors, aluminum, composites, drug.

1. Introduction

Corrosion-related expenses can account for ~3-4 % of GDP of the world's most developed countries, which is comparable to that spent on public education and healthcare.¹⁻³⁾ Thus, corrosion and its associated phenomena have always intrigued the scientific community. In this context, Al composite materials have become more prominent in nuclear and aerospace industries owing to their high strength ratio.³⁻⁶⁾ A variety of methods and combinations of techniques are utilized for corrosion control of aluminum and composites in various environmental conditions. Among the most commonly tested protection methods are anodizing,^{7,8)} protective coating,⁹⁻¹²⁾ Inhibitors including such as conversion, inorganic, and even Nobel metal,¹³⁻¹⁵⁾ organic,¹⁶⁻¹⁸⁾ & inorganic inhibitors.^{19,20)} Moreover, corrosion inhibitors investigated for aluminum and Al-composites have been published in

several research articles and patents.²¹⁻²³⁾

The focus of this short overview is solely on recent corrosion inhibitors for composite and Al-based materials in chloride-containing environments. In this review article, we initially address the basic corrosion of aluminum and its composites and then switch over to different corrosion inhibitors utilized for solutions containing chlorides.

2. Electro corrosion of Al and Al-composite in Chloride-containing Solution

The potential implies that Al is a thermodynamically powerful oxidizing agent, following only magnesium among important engineering materials.²⁴⁾ Similarly, Al composites are usually prone to corrosion in various electrolytes due to galvanic interactions occurring between both the reinforcement and the matrix, thus causing

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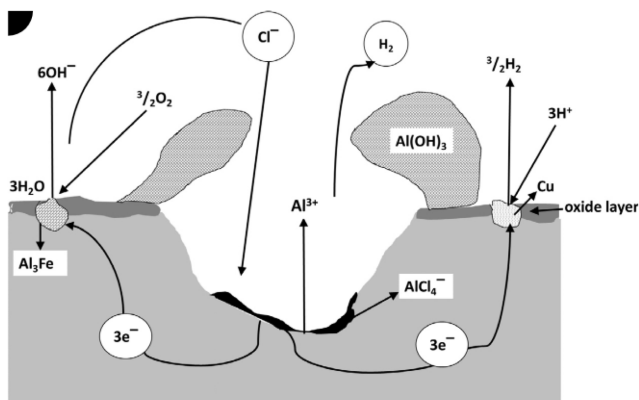


Fig. 1. Pitting corrosion of Al alloys in solutions containing chlorides.³²⁾

preferential corrosion at the interface due to the development of compounds, and defects just at the interface and affecting fissures that produce corrosion trajectories.²⁵⁾ The Al-based composites have inadequate corrosion resistance when compared to base alloys because the reinforcement phase leads to the breakdown of protective film and corrosion occurs. In reality, the reinforcement particles are indeed an impediment to protecting the surface film of aluminum alloys. The aluminum matrix has a natural thin oxide surface layer that protects against pitting propagation.^{24,26)} The Al corrosion process occurs frequently in oxygenated chloride-containing environmental conditions.^{5,27-29)} The chloride anions strike the oxide coating, weakening it in the most vulnerable areas. As shown in Fig. 1, due to the formation of local hydroxide the cathodic sites tend to be more alkaline.³⁰⁾ The dissolved oxygen is critical for a pitting attack. Because of precipitation of aluminum hydroxide outside the pits, a cone-shaped reserves of electrochemical compounds results near the pits' edges.^{31,32)}

3. Organic Compounds as Corrosion Inhibitors

Even though organic compounds consist of numerous hetero-atoms such as nitrogen, sulfur, oxygen, and phosphorous that offer adsorption zones, they have been widely utilized as inhibitors for Al and Al-composites.³⁰⁾ The following section will discuss the utilization of organic complexes as corrosion inhibitors for metallic samples in chloride-containing environments research cited,³³⁻³⁵⁾ in the last 2 decades. Fig. 2 depicts the tested corrosion inhibitors in the corrosive environment.

4. Organic Compounds as Corrosion Inhibitors in Chloride Environments

Potentiodynamic and Electrochemical Impedance Spectroscopy (EIS) techniques were used to investigate the inhibitor performance of 1,4-naphthoquinone (NQ) as an Al corrosion inhibitor in aerated and de-aerated chloride environments as reported.³⁶⁻³⁹⁾ The impact of inhibition increased with increasing inhibitor content in both aerated and deaerated solutions. The introduction of NQ (1 mM) resulted in the utmost inhibition effectiveness. With the inclusion of the inhibitor, the corrosion and disintegration potentials switched to more positive values. The transitions in deaerated solution are higher than in aerated electrolytes. The results revealed that NQ molecules form a tight bond on the metal surfaces, shielding against further anodic disintegration, based on SEM images and quartz crystal analyzer measurement systems. Sherif and Park also used various electrochemical approaches to examine 1,5-naphthalenediol's capabilities to safeguard aluminum and composite in 0.5 M NaCl electrolyte. Sherif and Park, 2005 et al.⁴⁰⁾ also found that the inhibitory performance grew as the compound level increased.

The compound significantly reduced anodic and cathodic currents, as well as corrosion current and concentration.

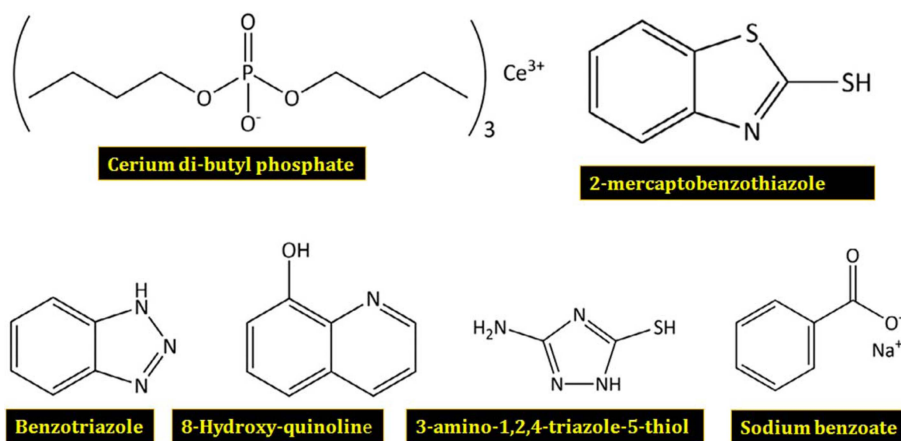


Fig. 2. The corrosion inhibitors for Al and its composites in chloride-containing solution.³⁰⁾

The inhibitor molecules are also boosting corrosion and disintegration potentials. According to the authors, 1,5 naphthalene diol upon adsorption on aluminum and its composite surface, layer up in the preformed pits and protect them from further anodic disintegration. Furthermore, it was proposed that corrosion inhibitor prevents the formation of a complex between aluminum hydroxides and chloride ions.⁴⁰⁾ Sherif used the EIS techniques to explore the impact of 3-amino-1,2,4-triazole-5-thiol known as ATAT on the electro-corrosion inhibitors of aluminum in spontaneously aerated 0.6 M NaCl electrolytes.⁴¹⁾ The effectiveness of inhibition increased as the concentrations of ATAT increased. The presence of the compound increased aluminum's polarization resistance. The addition of ATAT lowered the anodic and cathodic corrosion as well as passivates the current while shifting the electrochemical corrosion and pitting corrosion potentials to a significant level of negativity. The ATAT molecules, as per researchers, not only repaired the defects in the film of oxide formed on the metallic surface but also prohibited the formation of soluble chloride and oxy-chlorides complexes. Further, it was noticed that N-thiazolyl-2-cyanoacetamide was investigated for the corrosion of aluminum using weight loss and polarization techniques. They found that increasing the inhibitor concentration and decreasing the temperature increased inhibition efficiency. They also examined the efficacy of inhibitors with Ba^{2+} , Sr^{2+} , Ca^{2+} , and Mg^{2+} ions on N-thiazolyl-2-cyanoacetamide and found that the efficiency of inhibition increases when 1 mM MCl_2 ($\text{M} = \text{Mg}, \text{Ca}, \text{Sr}, \text{Ba}$) is added, owing to the synergistic and mixed kinds of inhibitors.⁴²⁾ Aluminum composites must be corrosion-resistant in hostile environments if they are to be used for an extended period. Aluminum is a highly reactive material with an oxide film on the surface that shields it against corrosion. However, aluminum composites are more susceptible to corrosion than bare alloys owing to reinforcements such as SiC and Al_2O_3 , which break down the protective oxide layer on the metallic surface. Furthermore, many studies^{11,43-47)} have been published on corrosion inhibitors such as polymer coating, anodizing, as well as chemical passivity. Amongst corrosion control methods, anodizing is most capable of protecting composites.^{8,9,48)} There are several coatings, but chromate conversion is one of them, which is a kind of chemical passivity, and acts as a corrosion inhibitor for aluminum composites. However, it is toxic and even environmentally hazardous. The conversion coating of rare-earth metals (Ce^{3+} , Y^{3+} , La^{3+} , Pr^{3+} , and Nd^{3+}) is used to protect aluminum composites from pitting corrosion.^{7,49,50)} The scientists^{51,52)} use cerium type conversion coating, which is a type of chemical passivity, to protect aluminum composites from corrosion. Because cerium coating develops

a hydrated oxide film of cerium at the cathode side on the surface of materials, it impedes corrosion behavior. This film functions as a cathodic inhibitor by impeding the kinetic model of oxygen reduction only at the cathodic site of the metal surface.⁵³⁾ Hydrogen peroxide and salt perchlorate bath would indeed be required to stimulate the growth of the cerium oxide as well as hydroxide film.⁵⁴⁾ In this context, Chen and Mansfeld⁴⁴⁾ revealed corrosion via pitting in Al6092 with SiC_p Al composite. They also used impedance spectra to assess the resistance to corrosion of Al 6092 with SiC_p composite (after one week, dipped in 0.5 N NaCl). For modification, the Ce-Mo process and sulfuric acid anodizing were used. Anodizing to hot water had no effect on resistance to corrosion of composites; but anodizing with dichromate seal showed significant improvements in corrosion resistance, however, it was abandoned due to environmental concerns. The effect of Cerium conversion treatment on corrosion prevention of Al5A06 with SiC_p aluminum composites at room temperature has been investigated.⁵¹⁾ The influence of different conversion solutions, temperature, and time of soaking on the Cerium conversion coating for corrosion protection of SiC_p with Al 5A06 aluminum composites was examined. The ideal corrosion control was indeed obtained in $\text{CeCl}_3 \cdot 7\text{H}_2\text{O}$ within NaCl (3.5 %) solution at 45 °C for 60 minutes, followed by curing at 100 °C for half an hour resulting in the development of cerium oxides as well as hydroxides just on cathodic region and an Aluminum oxide film here on metal matrix. Ahmad and Aleem⁵⁵⁾ used cerium chloride, sodium molybdate (Na_2MoO_4), and Potassium dichromate ($\text{K}_2\text{Cr}_2\text{O}_7$) as Al 6013 with SiC_p composite inhibitors. The localized corrosion was mentioned in an Al6013 with SiC_p -based composite in 3.5% NaCl electrolytes. It is induced by secondary phases of copper (Cu), iron (Fe), and chromium (Cr) just at the interface of the Al6013 with SiC_p matrix composite. A cathodic kinetic reaction is suppressed by cerium treatment at the Al6013 with SiC_p composite interface, which inhibits localized electrochemical corrosion on the matrix. A protective hydroxide or cerium oxide (CeO_2) film forms, which precludes the matrix alloy from corroding. In contrast to sodium molybdate and K2 treatments, cerium chloride action was perhaps the most effective inhibitor of Al6013 with SiC_p composite. Xue Wenbin et al.⁵⁶⁾ created an anti-corrosion film on an Al2024/ SiC_p composite created using the MAO strategy in a silicate environment. A potentiodynamic polarization test was used to inhibit corrosion in a 3.5 % NaCl solution. MAO film grows 2 layers on the composite surface that function as corrosion protection. The majority of the SiC_p fused to form SiO_2 and only a trace of SiC_p stayed in the film adjacent to the Al2024 with the SiC_p matrix composite interface. The inner layer is composed

of mullite (Al_2O_3), while the outside layer is composed primarily of silicon oxide. Tjong and Huo^{52,57}) recently found that oxidizing inhibitor as a cerium converting treatment in situ Al or Al_2 deposits on the surface of the composite while dipped in reduced CeCl_3O_3 , TiB_2 , and H_2 composite indicates patches of film O_2 concentrations. When dipped in 10 g and 100 mL CeCl_3 and hydrogen (H_2) at 30 °C, a dense film forms on the surface of the Al with Al_2O_3 , and TiB_2 reinforcement composite, improving corrosion resistance when subjected to NaCl electrolytes. Corrosion behavior and inhibition of AA6061 with 20 % Al_2O_3 and AA2014 with 10 % Al_2O_3 composites were dipped in 0.1M NaCl electrolytes with a variety of corrosion inhibitors such as including sodium molybdate dehydrate ($\text{Na}_2\text{MoO}_4 \cdot 2\text{H}_2\text{O}$), Sodium tungstate dehydrates (Na_2WO_4), ammonium tetrathiotungstate ($\text{H}_8\text{N}_2\text{S}_4\text{W}$), ammonium tetrathiomolybdate ($(\text{NH}_4)_2\text{MoS}_4$), $(\text{NH}_4)_6\text{Mo}_7\text{O}_{24}$ (ammonium (para)-tungstate), ammonium (meta)-tungstate hydrate ($(\text{NH}_4)_6\text{W}_{12}\text{O}_{39} \cdot x\text{H}_2\text{O}$), sodium poly Even $\text{H}_8\text{N}_2\text{S}_4\text{W}$ (ammonium tetrathiotungstate) was an effective inhibitor for the AA2014 with 10% $\text{Al}_2\text{O}_3\text{O}_2\text{Cr}_2$ composite.⁵⁸⁾

5. Drugs corrosion inhibitors

As recently found, drugs were also explored as corrosion inhibitors, particularly for steel corrosion inhibition. However, investigations are focusing on drugs used as oxidation inhibitors for aluminum and composites.

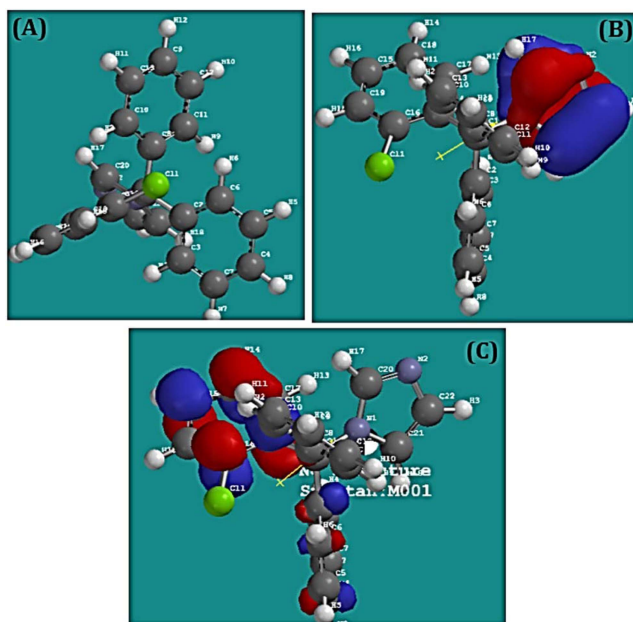


Fig. 3. The optimized structure (a), an optimized configuration of the clotrimazole molecule (b), the HOMO configuration of the clotrimazole molecule (c), and the LUMO configuration of the clotrimazole molecule produced by Density Functional Theory (DFT) by using the DMol3 technique.⁶²⁾

Amongst all the drugs used for corrosion protection, the chemical structures of anti-microbial and anti-fungal drugs were the most extensively researched compounds as good corrosion inhibitors for Al and composites in a variety of electrolytes.^{36,59-61)}

Furthermore, one research team recently found that Clotrimazole (CTM) is a specific inhibitor of aluminum corrosion in an acidic environment. The inhibition performance improves with inhibitor concentration, attaining 90.9 % (1×10^{-4} Molarity) at room temperature. Fig. 3 illustrates the optimized molecule of Clotrimazole, which itself is (1-[(2-chlorophenyl)-diphenyl-methyl] imidazole). Although, a few research works on the potential employment of anti-hypertensive & anti-emetic drugs just like corrosion inhibitors were also carried out. All the drugs revealed were examined as corrosion inhibitors in a variety of electrolytes.⁶²⁾

5.1. Antibacterial drugs

Anti-microbial drugs are amongst the most extensively used therapeutic drugs in the world. The terminology anti-microbial, anti-biotic, and anti-infective refer to a broad range of pharmaceutical medicines including anti-bacterial, anti-fungal, anti-viral, and anti-parasitic medications. Anti-bacterial compounds are the most often used among these, though similar rules will apply to compounds from some other groups too.

The mixture of antibacterial drugs and metals appears straightforward because metals and composites are used in many applications during which antibacterial activity is desired.⁶⁴⁾ The findings of S.M. Tamborim et al. using the Scanning Vibrating Electrode Technique (SVET) indicates a decrease in electrochemical action in the doped cellulose acetate (CA) feature film with amoxicillin as compared to the undoped ones and indicate some healing effects. Under the oxidation process, CA films doped with 2000 ppm amoxicillin illustrated the impact of amoxicillin as either a corrosion inhibitor via lower current densities after 3 days of exposure.⁶³⁾ This is depicted in Fig. 4-5.

5.1.1. Penicillin

Penicillin was one of the first anti-bacterial drugs to be searched owing to its wide use in modern medical science. The lead compound was produced naturally (obtained from *Penicillium* fungal species) and also qualifies to yield a few novel semi-synthetic derivative drugs with differing spectra of action and performance against a range of infections. Penicillin work by preventing the production of cross-links in the bacterial cell membrane, weakening it to the point where the osmotic pressure in the bacteria's inner surface would become progressively uncompensated, resulting in cell demise. Because their biological intention is indeed an enzyme concerned with

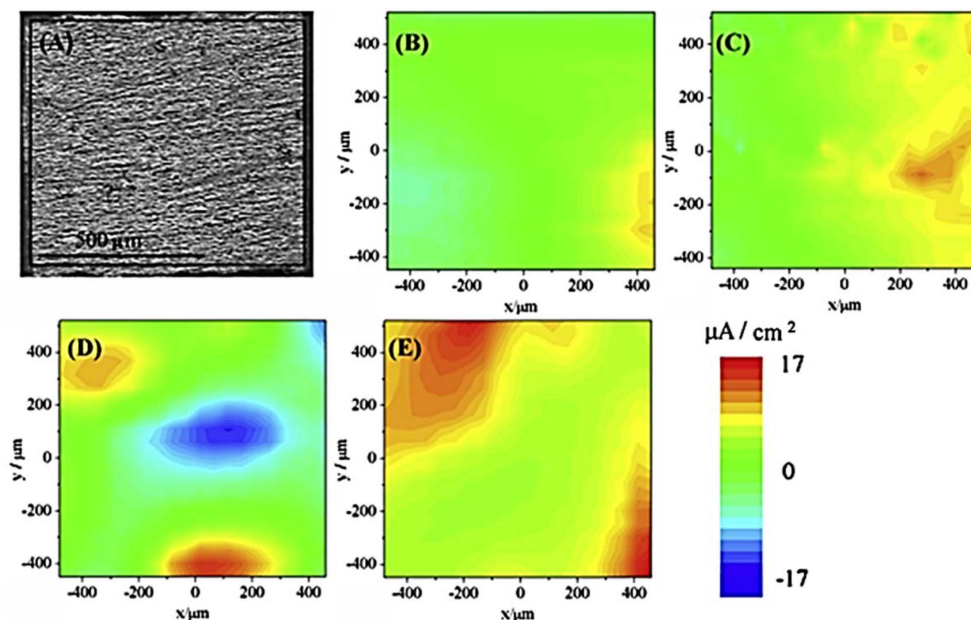


Fig. 4. (A) Optical image of the surface of an uncoated AA2024-T3 substrate (CA) depicting the analyzed area and SVET graphs of ionic currents analyzed 200 m above the surface while subjected to 0.05 M NaCl electrolytes which contain 2000 ppm amoxicillin for (B) 5 minutes, (C) 1 hour, (D) 1 day, and (E) three days.⁶³⁾

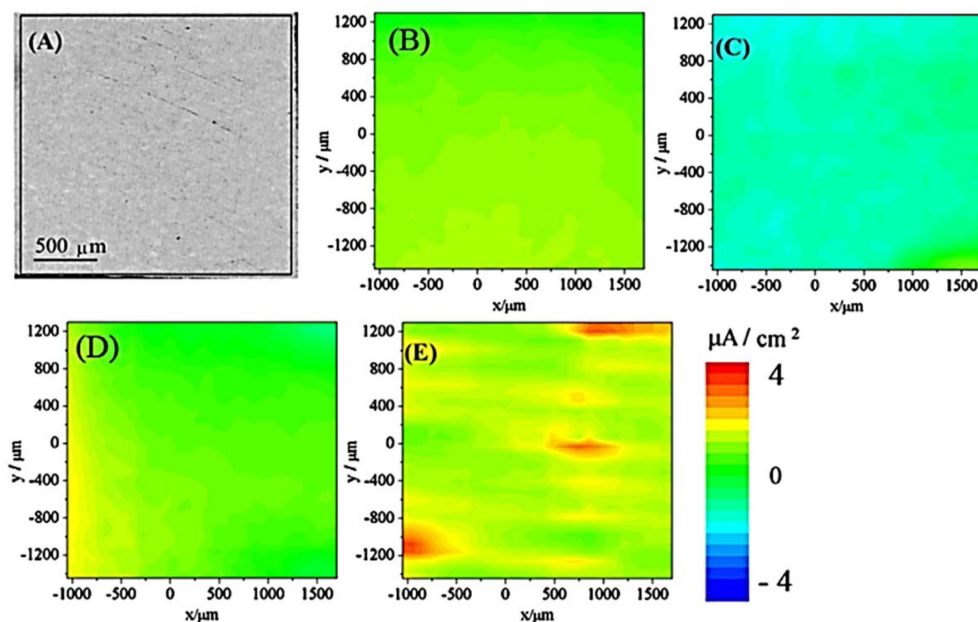


Fig. 5. (A) Optical image of AA2024-T3 substrate surface coated with CA film (doped with amoxicillin). The optical image showing the analyzed area and SVET maps of the ionic currents measured 200 lm above the surface, exposed to 0.05 M NaCl solution during (B) 5 minutes, (C) 1 hour, (D) 24 hours, and (E) 72 hours.⁶³⁾

the synthesis of bacterial cell walls, their corrosion inhibitors interaction is most probably related to their chemical structures, most usually due to the heteroatoms manifest in their so-called β -lactam heterocyclic framework. To the best of our knowledge, only two research have been carried out to analyze the corrosion inhibitors potency of penicillin for the prevention of metals, which

include two β -lactamase resilient penicillin, as well as dicloxacillin., and two compounds from an amino-penicillin group, namely amoxicillin, and ampicillin. Resistant penicillin, cloxacillin, and dicloxacillin, as well as two compounds from the amino-penicillin functional group, amoxicillin, and ampicillin, seem to be the most commonly utilized antibiotics.^{60,65,66)}

5.1.2. Cephalosporin

Cephalosporin was observed in the mid-twentieth century as a crude extract from the fungus *Cephalosporium*. Since they have an analogous spectral range of antimicrobial effect as penicillin, individuals quite often use it as a 2nd line treatment once penicillin was used or when their insolubility in aqueous solution would not be a constraint. Because they possess an analogous structure to penicillin, these are both vulnerable to β -lactamase, a common bacterial approach for gaining resilience to antimicrobial drugs. The anti-corrosion ability of penicillin is still most probably attributable to the π electrons, which may also improve their own adsorb to the metallic surface.⁶⁷⁾

5.1.3. Aminoglycosides

Amino-glycosides seem to be anti-bacterial medicine that binds specifically to a bacterial 30S subunit of the ribosome, causing a misreading of mRNA and avoiding the bacterium from synthesizing proteins required for growth. The amino-modified glycoside observed in their framework inspired their identity. The medicine involves gentamicin and kanamycin (popularly known as kanamycin A), and both are de-oxystreptamin-containing aminoglycosides, and amikacin, which would be typically used as a last resort against multidrug-resistant bacteria. Despite the note that the primary framework of aminoglycosides is still not centered on the β -lactam ring, some characteristics of the antibiotics discussed earlier in this section were seen. Several amino-glycosides contain heterocycles within their structures. Solitary saccharide O-containing monomers could have served as conditional centers in almost the same way.⁶⁸⁻⁷⁰⁾

6. Conclusions

Corrosion is inevitable for Al and its composites, and it has a similar impact to natural disasters. As a result, completely preventing these issues is impossible. Attempting to take preventive measures to inhibit corrosion of the metal surface using inhibitors is more economically efficient. The inhibitors offer a streamlined solution for environmental corrosion protection. It was seen that the corrosion resistance increased with increasing inhibitor content (1,4 naphthoquinone) in both aerated and deaerated sodium chloride solutions because it formed a tight bond on the metallic surface and shielded further oxidation reaction. Moreover, the 1,5 naphthalene diol adsorbent surface on the aluminum surface protects them from further anodic dissolution due to the formation of hydroxides (in chloride ion solutions). In this review, the ATAT molecules not only repaired the defects in the oxide film on the metal surfaces but also stopped the

forming of soluble chloride and oxy-chloride complexes in the electrolytes. Interestingly, antibacterial drugs including clotrimazole (CTM), amoxicillin, amino-penicillin, and cephalosporin play a very significant role as corrosion inhibitors, which most likely owe to π -electrons, also improve their adsorption on metallic surfaces in chloride environments. Finally, a study of inhibitors for aluminum composites is peculiar and would become more appealing immediate future.

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