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The Impact of Drugs as Corrosion Inhibitors on Aluminum Alloy in Coastal-Acidified Medium

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<http://dx.doi.org/10.5772/intechopen.72942>

Abstract

The use of corrosion inhibitors has proven to be one of the effective methods of corrosion protection of metals and alloys. Aluminum alloys are good candidate materials for structural components in major industrial application owing to their excellent corrosion resistance. However in high intensity coastal and acidified medium the challenge of micro porosity and hydrogen embrittlement distributed along the interface cannot be neglected as suitable corrosion inhibitive compounds is needful without significantly reacts with the environmental components. The challenges of most effective organic and inorganic Inhibitors when dissolved in aqueous environments are increasingly due to their toxicity. Drug as inhibitive compound have been seen as suitable replacement for this high toxic organic inhibitors since both react by adsorption on a metallic surface. They are usually compounds that form film and cause the formation of precipitates on the metal surface, thereby blocking both cathodic and anodic and sites. Therefore, this study covers the general overview of impact of some drugs as corrosion inhibitive compound on aluminum for industrial applications and their environmental impact.

Keywords: corrosion inhibitors, protection, coastal, acidified, drug and impact

1. Introduction

Aluminum is a silvery white material and a member of boron group. It is one of the most abundant metal in the Earth's crust, and the third most abundant element within, after oxygen and silicon. It is yielding, long-lasting, lightweight, malleable metal with physical appearance varying from silvery coloration to approximate gray, which depend on the roughness of the surface. Aluminum is not a Ferro-magnetic material. It is also not soluble in alkanols, despite the fact that, in certain form, it can be water-soluble. It has an appreciable yield strength of

S/N	Research work	Result	Reference
1	Meclizine (Antihistaminic Drug) as Corrosion Inhibitors of Aluminum in HCl Environment	The result showed that the drug excellently inhibited the aluminum in the acid medium at 303 K	[2]
2	Clotrimazole and Fluconazole (Antifungal drugs) as Corrosion Inhibitors of Aluminum in HCl Environment	The results pointed out that both drug server as excellent inhibitors in the Medium	[3]
3	Amine Compound as Corrosion Inhibitors of Aluminum in HCl Environment	The result indicated 92% inhibitive efficiency at 30°C and 83% at 60°C using 100 ppm inhibitor concentration	[4]
4	Voltaren (expired) Drug to inhibit Aluminum in HCl Environment	Corrosion potential (Ecorr) was shifted to the direction that is more noble by the Inhibitor molecules	[5]
5	Domiana Extract as inhibitor of Aluminum in Acidic medium	From the result, the inhibition efficiency displayed direct variation with inhibitor’s concentration but demonstrated inverse relation with temperature	[6]
6	Corrosion Inhibitor of Aluminum using Chloroquine in HCl Environment Media	It was deduced that chloroquine excellently inhibited the aluminum in the acidic environment and the efficiency of the inhibitor increased with concentration of inhibitor but not favored by temperature	[7]
7	Corrosion Inhibitor of Aluminum using <i>Aloe Vera</i> in NaCl medium	The result showed that <i>Aloe Vera</i> is potential green corrosion inhibitor in saline environment	[8]
8	Corrosion inhibition of Aluminum Alloy in NaOH Medium using Anti-inflammatory drug	The result showed the potential of the Omeprazole drug in inhibiting corrosion in the basic environment	[9]
9	Corrosion inhibition of Aluminum using Fluconazole & Clotrimazole in HCl Medium	The result displayed the antifungal drug as potential inhibitor of aluminum in the acidic environment	[10]
10	Corrosion inhibition of Aluminum using green corrosion inhibitor in acidic medium	The result showed the extract as eco-friendly inhibitor in the acidic environment	[11]

Table 1. Analysis of inhibitive drugs and their performance.

about 10 MPa for the pure metal, while that of its alloys can range from about 150 to 650 MPa. Aluminum is about 33% dense and stiff in comparing to steel.

The machinability and formability of aluminum is excellent with outstanding corrosion resistance due to its ability to form an effective and tenacious thin surface layer of its oxide in atmosphere. It is also noteworthy that the strongest alloys aluminum are less corrosion resistant because of the possibility internal galvanic cell that may be developed in service [1]. This corrosion resistance is also often greatly reduced in the presence of various salts or dissimilar metals and often corrosion of aluminum can be mitigated using corrosion inhibitors, even the use of inhibitive drugs (Table 1).

2. Passivity of pure aluminum and its alloys

2.1. Corrosion of aluminum

Al has a reactive metal, can react with acidic, basic or salty foods to release itself into the food product. Also, an aluminum container that is worn out or pitted may contaminate the food by

releasing excess aluminum into it [12]. Therefore, temperature does have an effect on the corrosion of aluminum. The aluminum subjected to the higher temperature does corrode because of the increase in particles' collision [13]. Corrosion of aluminum depends on environment, design, alloy and preventive measures put into considerations. A dirt-free aluminum surface is highly reactive and will spontaneously react with air or water to form aluminum oxide. This oxide is usually stable and possess excellent adhesion to the surface of metal therefore protects the material (aluminum) from further corrosion or oxidation. Thus aluminum has excellent corrosion resistance in stable oxide layer environments [14]. This oxide layer will be weaken in environments where there is low or high pH particularly, in the presence of aggressive ions. That is, at a pH below 4 and beyond a pH of about 8.5, there will generally be an increase in the aluminum corrosion rate, though this depend on the ions that are present in that particular environment. The oxide layer will be broken down by the aggressive ions locally and localized corrosion attacks start. Of all the ions that display high aggression, chloride ion (Cl^-) is of the most practical importance [15]. From thermodynamically point of consideration aluminum is a metal with high activity. Nevertheless, in aerated environment, the metal is rapidly covered with an intense oxide layer [16]. This oxide layer is basically inert, and prevents further oxidation. The thickness of this layer depends largely on the temperature, environment and alloy elements. The oxide films that are formed in air at ordinary room temperature are about 2.5 nm of thickness on pure aluminum. Heating to about 400°C may results in films up to about 25 nm (this is about 10 times that at room temperature [17]. If the oxide film is destroyed, e.g. by a scratch, new oxide will spontaneously and immediately form on the uncovered metal [18]. Thus pure and even many of its alloys display excellent performance in corrosion protection.

3. Aluminum alloy

Major reason for alloying aluminum is to increase hardness, strength and resistance to fatigue, stress relaxation, wear or creep. The Effects of different alloying elements and combinations of them on these properties are specific and related to the phase diagrams of constituent alloy elements.

Figure 1 shows the relationships between some of the more commonly used alloys in the 6xxx series. The elements that are most frequently used in commercial aluminum alloys to provide better strength especially, when related to strain hardening via cold working or heat treatment, or both are; zinc, copper, silicon, magnesium and manganese (**Figure 2**). All these elements have high solid solubility in aluminum, and usually, the solubility increases as temperature increases.

Aluminum and its alloys possess significant industrial and economic value due to its light weight, good thermal and electrical conductivity as well as low cost [20]. The most important property of aluminum is its ability to effectively resist atmospheric and several aqueous solution because of the quick formation of protective oxide of thin film which adhere tenaciously to the surface, that serves as barrier between the corrosive environment and the metal.

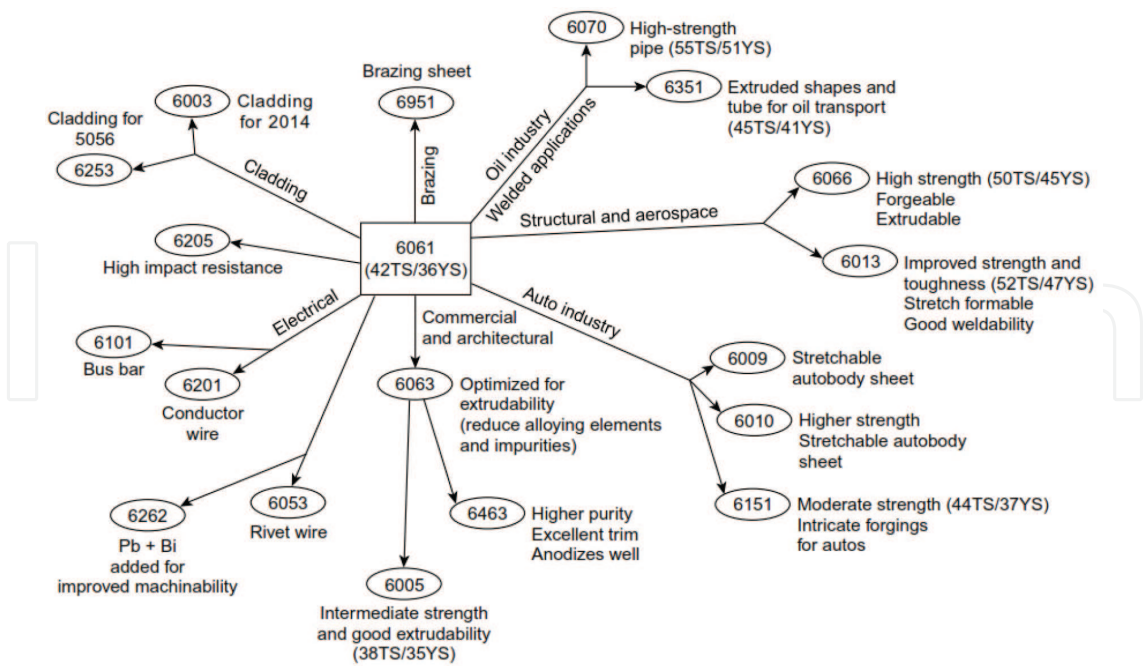


Figure 1. Relationships that exist among commonly used alloys in the 6xxx series (Al-Si-Mg). Their yield strength (YS) and tensile strength (TS) in ksi units [19].

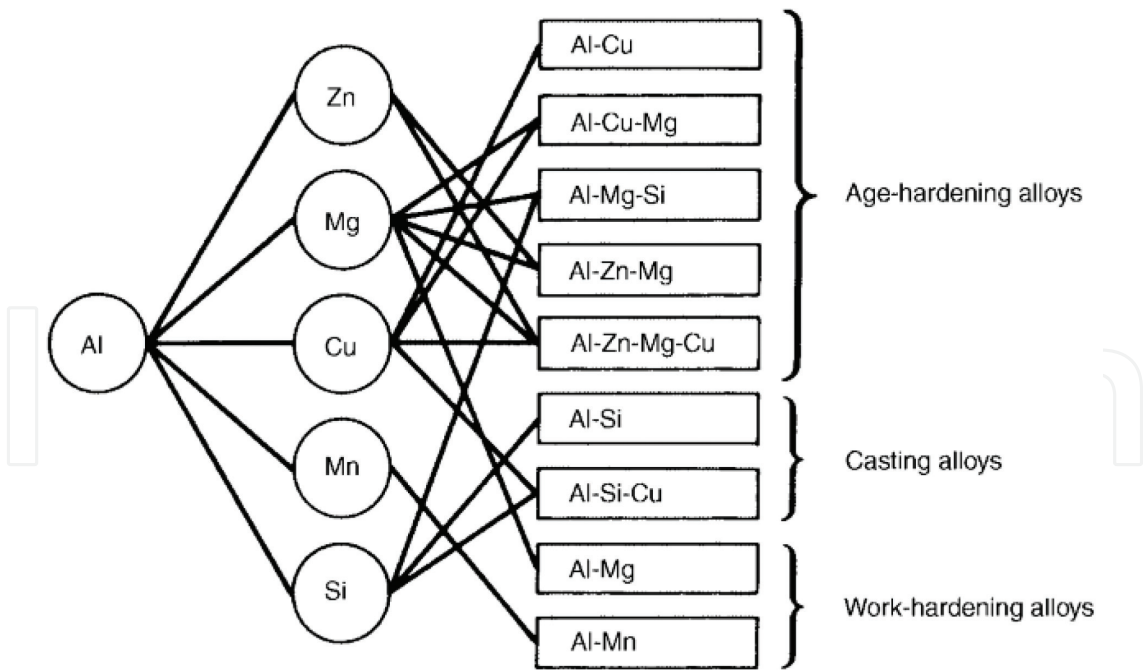


Figure 2. The principal aluminum alloys [19].

Element	Half Reaction	E°(V)
Gold	$\text{Au}^{3+}_{(\text{aq})} + 3\text{e}^- \longrightarrow \text{Au}_{(\text{s})}$	+1.50
Platinum	$\text{Pt}^{2+}_{(\text{aq})} + 2\text{e}^- \longrightarrow \text{Pt}_{(\text{s})}$	+1.18
Silver	$\text{Ag}^{+}_{(\text{aq})} + \text{e}^- \longrightarrow \text{Ag}_{(\text{s})}$	+0.80
Iron	$\text{Fe}^{3+}_{(\text{aq})} + \text{e}^- \longrightarrow \text{Fe}^{2+}$	+0.77
Copper	$\text{Cu}^{2+}_{(\text{aq})} + 2\text{e}^- \longrightarrow \text{Cu}_{(\text{s})}$	+0.34
Tin	$\text{Sn}^{4+}_{(\text{aq})} + 2\text{e}^- \longrightarrow \text{Sn}^{2+}_{(\text{aq})}$	+0.13
Nickel	$\text{Ni}^{2+}_{(\text{aq})} + 2\text{e}^- \longrightarrow \text{Ni}_{(\text{s})}$	-0.25
Iron	$\text{Fe}^{2+}_{(\text{aq})} + 2\text{e}^- \longrightarrow \text{Fe}_{(\text{s})}$	-0.44
Chromium	$\text{Cr}^{3+}_{(\text{aq})} + 3\text{e}^- \longrightarrow \text{Cr}_{(\text{s})}$	-0.74
Zinc	$\text{Zn}^{2+}_{(\text{aq})} + 2\text{e}^- \longrightarrow \text{Zn}_{(\text{s})}$	-0.76
Manganese	$\text{Mn}^{2+}_{(\text{aq})} + 2\text{e}^- \longrightarrow \text{Mn}_{(\text{s})}$	-1.18
Titanium	$\text{Ti}^{2+}_{(\text{aq})} + 2\text{e}^- \longrightarrow \text{Ti}_{(\text{s})}$	-1.37
Aluminium	$\text{Al}^{3+}_{(\text{aq})} + 3\text{e}^- \longrightarrow \text{Al}_{(\text{s})}$	-1.66
Magnesium	$\text{Mg}^{2+}_{(\text{aq})} + 2\text{e}^- \longrightarrow \text{Mg}_{(\text{s})}$	-2.27
Cerium	$\text{Ce}^{3+}_{(\text{aq})} + 3\text{e}^- \longrightarrow \text{Ce}_{(\text{s})}$	-2.34
Praseodymium	$\text{Pr}^{3+}_{(\text{aq})} + 3\text{e}^- \longrightarrow \text{Pr}_{(\text{s})}$	-2.35
Sodium	$\text{Na}^{+}_{(\text{aq})} + \text{e}^- \longrightarrow \text{Na}_{(\text{s})}$	-2.71
Calcium	$\text{Ca}^{2+}_{(\text{aq})} + 2\text{e}^- \longrightarrow \text{Ca}_{(\text{s})}$	-2.87

Figure 3. Aluminum in the redox strength of metals.

The reaction is in line with the position of aluminum in the redox strength of metals (**Figure 3**). It has been observed that this film (oxide) is stable at a range of about 5–9 range of pH [21].

4. Pitting corrosion of aluminum

Aluminum usually suffers pitting corrosion in the presence of chloride ions. Pitting corrosion is a dangerous form of localized corrosion that selectively attacks areas of a metal surface where there is; (i) a surface scratch or a mechanically induced break in an otherwise protective film, (ii) an emerging dislocation or slip step caused by applied or residual stresses and (iii) a compositional heterogeneity such as an inclusion, segregate or precipitate [22]. Pitting is one of the most insidious forms of corrosion. It is very destructive to many engineering materials since it causes perforation of the metal. As a result of its localized nature, it often causes sudden unexpected failures. Pitting is often difficult to detect because corrosion products may cover small pits. General illustration of pitting corrosion on aluminum alloys is shown in **Figure 4**.

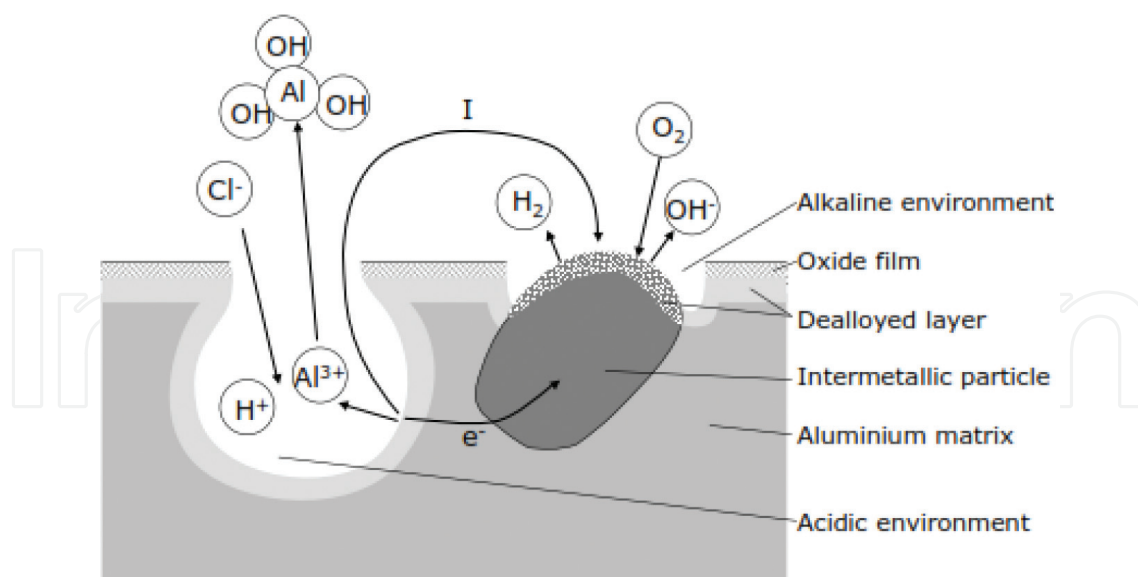


Figure 4. General illustration of pitting corrosion on aluminum alloys [18].

5. Corrosion inhibitors

Corrosion inhibitors have been established to be effective and easier means of corrosion control. Using chemical inhibitors to reduce the rate of corrosion processes is somewhat amenable. Corrosion inhibitors found applications in various industries, examples are; oil and gas, chemical industries, production, refineries, heavy manufacturing, water treatment and etc. [23]. In extraction the oil, chemical and processing industries, the use of corrosion inhibitors are usually considered to be the primary line of protection. Therefore, a large numbers of scientific research have been concentrated to the issue of corrosion inhibitors [24–29]. **Table 1** shows application of corrosion inhibitors in some specific system environment. An inhibitor is a chemical substance or mixture of substances which when added in very little concentrations to a corrosive environment, it effectively retards or prevents corrosion without significantly reacts with the environmental components.

Inhibitors can be classified as either organic or inorganic compounds and they are usually dissolved in aqueous environments. Some of the most effective inorganic inhibitors are carbonates, chromates, nitrites, silicates and phosphates. The organic inhibitors comprise amines, nitrogen compounds, sulfur compounds such as thioethers, thioalcohols, thioamides, thiourea and hydrazine. Chromates and zinc salts are used increasingly less due to their toxicity and are currently largely replaced by organic inhibitors.

Also, with reference to their influence on the electrochemical reaction relating to metal, and their environment corrosion inhibitors (the inorganic inhibitors) can also be classified as anodic, cathodic or mixed corrosion inhibitors (**Figure 5**). **Figure 6** displays the adsorption mechanism of an organic inhibitors on aluminum. These have been in application for quite some years and the most common examples of their uses are in films and coatings on metals where chromate, nitrate, borates, phosphate, benzoates, and oxides are incorporated, Nitrites

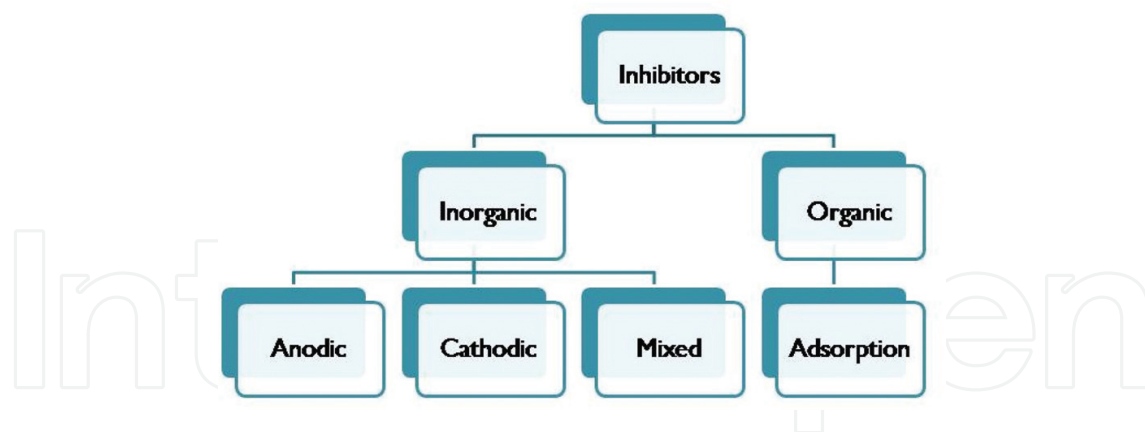


Figure 5. Classification of corrosion inhibition.

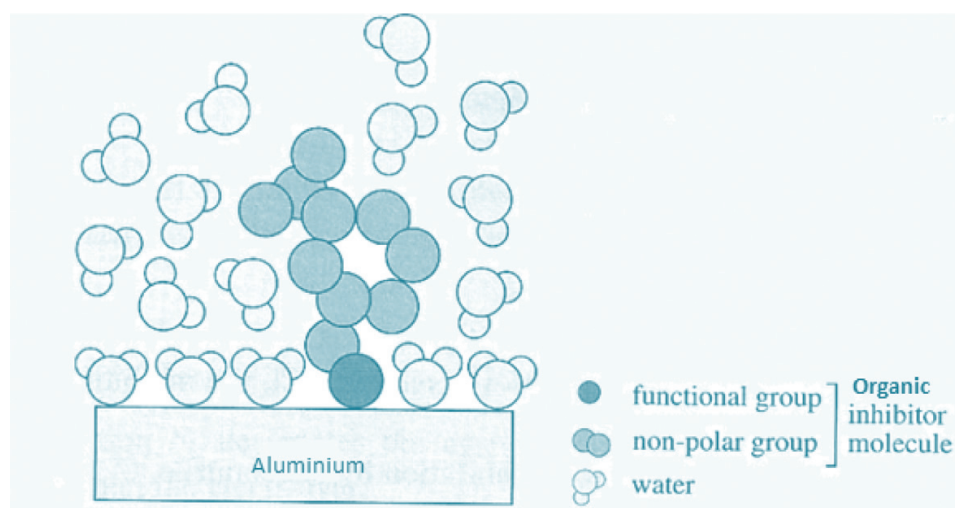


Figure 6. Adsorption mechanism of an organic inhibitor on an aluminum surface in an aqueous environment.

are being used as inhibitor in concrete reinforcement. While the organic inhibitors react by adsorption on a metallic surface.

Anodic inhibitors generally act by forming a protective oxide film on the surface of the metal to be protected resulting in a large anodic shift of the E_{corr} (corrosion potential). This shift usually forces the surface of metal into the region of passivation. That is, they slow down the anodic dissolution reaction rate of and produce products of reaction which form a thin film over anode. Examples are; Chromates, nitrates, tungstate and molybdates.

Cathodic inhibitors usually work by either slowing down the cathodic reaction or precipitating (selectively) on cathodic regions to inhibit the diffusion of reducing parts to the surface. That is, they interrupt the flow of electrons from the anode to the cathode and produce products of reaction which precipitate selectively at cathodic sites. Examples are zinc sulfate, nickel sulfate, aminoethylene and polyphosphate.

Mixed inhibitors act by reducing both the anodic and cathodic reactions. They are usually compounds that form film and cause the formation of precipitates on the metal surface, thereby blocking both cathodic and anodic sites. Common examples are the phosphates and silicates.

Therefore, this study deals with general overview of aluminum, corrosion inhibitors, their classifications, Inhibitive drugs, considerations in employing them, their efficiency industrial applications and their environmental impact.

6. Properties of a good inhibitor

In order to prevent or mitigate the corrosion of materials (metallic), inhibitors in question must be able to satisfy these following measures [26]:

- i. It must be effective in corrosion protection at a very little concentration of the inhibitor.
- ii. It must offer protection to all the part of the component that are exposed from the corrosion attack of corrosion.
- iii. It must maintain its effectiveness at extreme operating conditions (high velocity and temperature).
- iv. During application, over or under dosage of inhibitor, the rate of corrosion must not drastically increased.
- v. The inhibitor or inhibitor's products must not form any deposit on the surface metal especially at regions of heat transfer.
- vi. It must mitigate both localized and uniform corrosion.
- vii. The range of effectiveness should be long.
- viii. It must not pose health hazard or pollution potential.

7. Drug inhibitors

Typical examples of organic eco-friendly inhibitors are fast green and carmine. The molecules of fast green have aromatic ring with electroactive nitrogen and oxygen atoms while molecules of carmine have aromatic rings (electron rich) with electroactive oxygen atoms. These molecular structures are shown in **Figure 7**. These compounds are adsorbed at the metal surface and they block the active site and consequently, decreasing the corrosion rate and consequently increase the inhibition efficiency [30]. Nevertheless, Most of organic inhibitors are poisonous and harmful to the environment. The increasing health and ecological awareness have drawn the attentions toward research and development of very effective and efficient eco-friendly inhibitors.

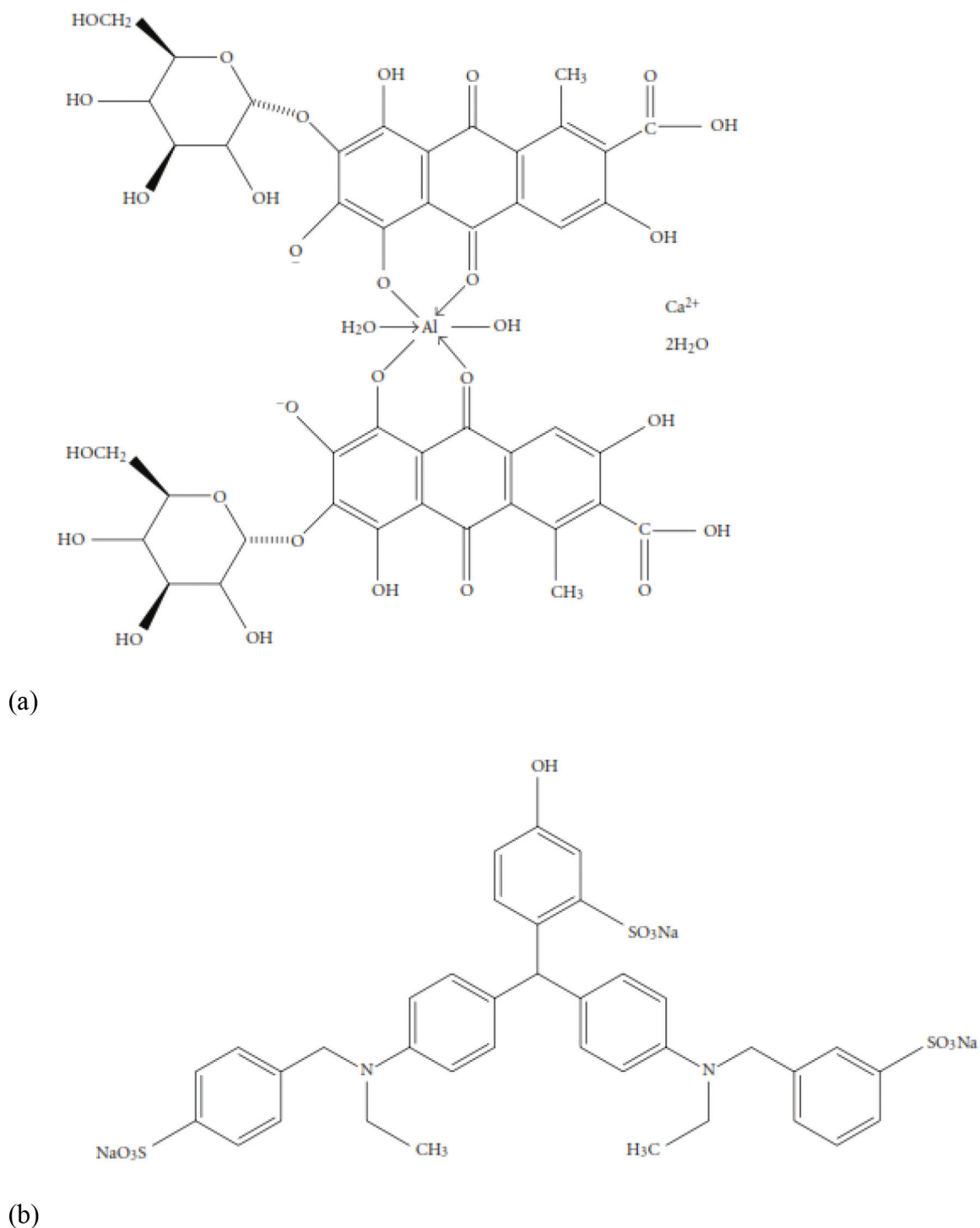


Figure 7. Structure of (a) carmine and (b) fast green [31].

Therefore it is vital and essential to develop cheap and environmentally secure corrosion inhibitors [32, 33]. Recently drugs have now been used as corrosion inhibitors. According to Gece [34], the use of drugs as corrosion inhibitors of metals corrosion possesses some merits over using some inorganic/organic inhibitors because of their little or insignificant impact on the environment. Drugs are nontoxic, cheap, negligible negative effects on environment, so it

suggested replacing the traditional toxic corrosion inhibitors [35]. Many Researchers in this field generally agree that drugs are corrosion inhibitors that can compete favorably with discovered green corrosion inhibitors and that most of these drugs can be synthesized from natural products. The choice of some drugs used as corrosion inhibitors is based on the following: (a) drug molecules contain oxygen, nitrogen and sulfur as the main active centers, (b) drugs are reported to be environmental friendly and important in biological reactions and (c) some drugs are easy to produced. Therefore, it has been established from research findings that, since majority of these drugs are natural product or plants extracts have been discovered to accomplish the aim of employing a cheap, environmentally suitable, abundantly available and effective high inhibition efficiency and little or insignificant environmental impact. **Tables 2 and 3** reveal application of corrosion in specific system environment and examples of drugs (quinolones) commonly used as corrosion inhibitors in our common environment respectively [34]. Nowadays, studies are now focusing on using pharmaceutical drugs nontoxic corrosion inhibitors to slow down the corrosion of metals and their alloys. For instance, Abdallah [36] used rhodanine azosulpha drugs to inhibit the corrosion of 304 stainless steel in HCl solution, [37] also anti-bacterial drug to control the deterioration of aluminum in HCl solution. Similarly, Obot and Obi-Egbedi [38] used antihypertensive drugs to slow down the corrosion of aluminum and aluminum alloys in aqueous solutions.

The presence of benzene ring in the molecules and heterocycles like pyridine, thiophenes, isoxazoles, others that present in drugs has sensitized researchers all over the world to intensifying their investigation of drug compounds as green corrosion inhibitors as save alternative to the conventional toxic corrosion inhibitors [39].

A number of studies have reported the use of drugs particularly antibacterial as corrosion inhibitors [40]. For instance, Eddy and Ebenso [40] investigated on Corrosion Inhibition and Adsorption Characteristics of Tarivid on Mild Steel in H_2SO_4 using thermometric and gasometric methods. Their finding showed that tarivid drug inhibits the deterioration of mild steel in the acid environment (H_2SO_4). The inhibition efficiency of tarivid was established to increase in values as its concentration increased however; it decreased as the temperature increases. The mechanism of physical adsorption was proposed from the acquired kinetic and thermodynamic factors. Also, Langmuir isotherm adsorption model was followed. Abdallah [36] studied the effect of Rhodanine azosulpha drugs as corrosion inhibitors on the corrosion of 304 stainless steel in HCl environment using gravimetry and potentiodynamic methods. Parallel adsorption technique was used for the inhibition process on the steel surface because of the presence of multiactive center within the molecules of the inhibitor.

Solmaz et al. [41] have shown that Rhodanine is a good corrosion inhibitor for low carbon steel in hydrochloric acid. They used electrochemical techniques for the corrosion findings.

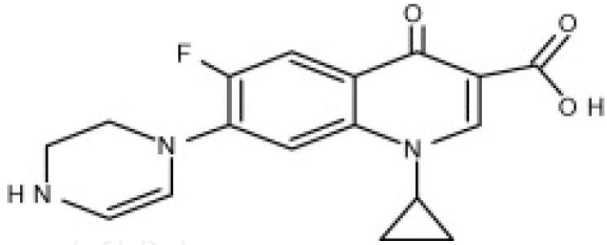
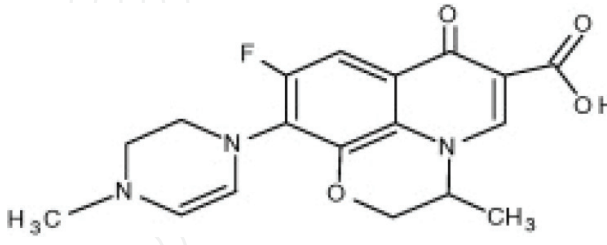
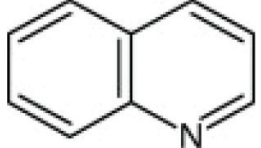
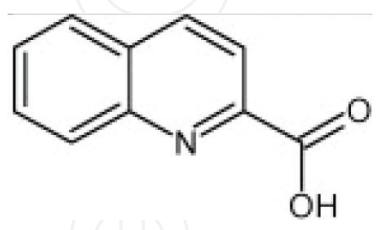
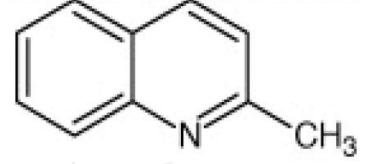
The effect of a number of antibiotic drugs, like; spectinomycin, paromomycin and streptomycin on the corrosion performance of zinc metal in hydrochloric acid environment was studied by [42], using some electrochemical and gravimetric techniques. The percentage of inhibition efficiency was discovered to be increasing as the concentration of drug increases but decreases with temperature. It has been discovered that majority of the drugs used display vital roles in biological reactions due to their antibacterial, anticonvulsant, antidiabetic, and other properties

[10, 11]. The criteria used in selection of these drugs as corrosion inhibitors depend on the following (i) whether the molecules contain sulfur, oxygen and nitrogen as active centers, (ii) the ease of production as well as purification. (iii) They are eco-friendly.

System	Inhibitor	Metals	Concentration
Acids			
HCl	Ethylaniline	Fe	0.5%
..	Mercaptobenzotriazole	..	1%
..	Pyridine + phenylhydrazine	..	0.5% + 0.5%
..	Rosin amine + ethylene oxide	..	0.2%
Sulfuric	Phenylacridine	..	0.5%
Phosphoric	Sodium iodide	..	200 ppm
Others	Thiourea	..	1%
..	Sulfonated castor oil	..	0.5-1.0%
..	Arsenic Oxide	..	0.5%
..	Sodium arsenate	..	0.5%
Water			
Potable	Calcium bicarbonate	Steel, cast iron	10 ppm
..	Polyphosphate	Fe, Zn, Cu, Al	5-10 ppm
..	Calcium hydroxide	Fe, Zn, Cu	10 ppm
..	Sodium silicate	..	10-20 ppm
Cooling	Calcium bicarbonate	Steel, cast iron	10 ppm
..	Sodium chromate	Fe, Zn, Cu	0.1%
..	Sodium nitrite	Fe	0.05%
..	Sodium phosphate monobasic	..	1%
..	Morpholine	..	0.2%
Boilers	Sodium phosphate monobasic	Fe, Zn, Cu	10 ppm
..	Polyphosphate	..	10 ppm
..	Morpholine	Fe	variable
..	Hydrazine	..	O ₂ scavenger
..	Ammonia	..	neutralizer
..	Octadecylamine	..	variable

Engine coolants	Sodium chromate	Fe, Pb, Cu, Zn	0.1-1%
..	Sodium nitrite	Fe	0.1-1%
..	Borax	..	1%
Glycol/water	Borax + MBT	All	1% + 0.1%
Oil field brines	Sodium silicate	Fe	0.01%
..	Quaternaries	..	10-25 ppm
..	Imidazoline	..	10-25ppm
Seawater	Sodium silicate	Zn	10 ppm
..	Sodium nitrite	Fe	0.5%
..	Calcium bicarbonate	All	pH dependent
..	Sodium phosphate monobasic + Sodium nitrite	Fe	10 ppm + 0.5%

Table 2. Application of corrosion inhibitors in specific system environment [24].

S/N	Drug (used as corrosion inhibitor)	The molecular structure	The clinical usage	Class of inhibitor	Type of environment (corrosive medium)
1.	Ciprofloxacin		Typhoid fever, Prostatitis, Diarrhea	Mixed	Saline (NaCl)
2.	Ofloxacin		Urethritis, Gonorrhea, Cervicitis	Mixed	Acidic (HCl)
3.	Quinoline		Antimalarial	Mixed	Acidic (HCl)
4.	Quinaldic acid		Antimalarial	Mixed	Acidic (HCl)
5.	Quinaldine		Antimalarial	Mixed	Acidic (HCl)

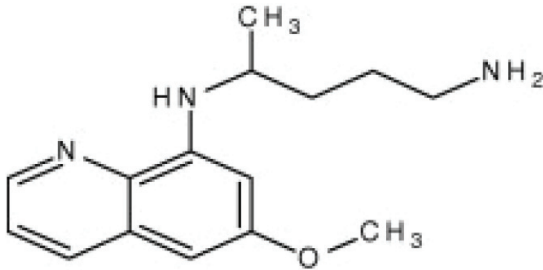
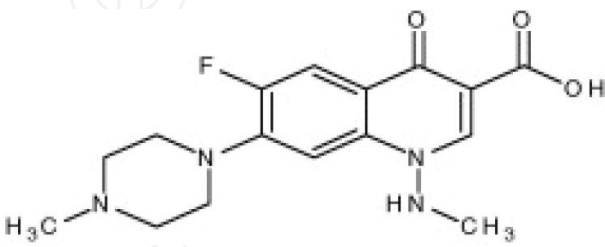
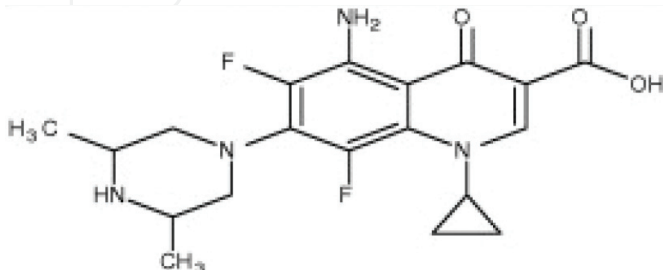
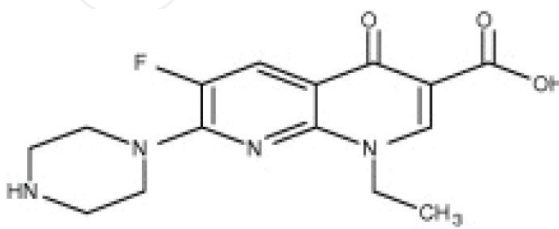
S/N	Drug (used as corrosion inhibitor)	The molecular structure	The clinical usage	Class of inhibitor	Type of environment (corrosive medium)
6			Antimalarial pneumonia	Mixed	Acidic (HCl)
7.	Amifloxacin		Typhoid fever, Diarrhea (infectious)	Mixed	Saline (NaCl)
8.	Sparfloxacin		Urinary tracts, Typhoid Fever, Bones	Adsorption	Acidic (HCl and H ₂ SO ₄)
9.	Enrofloxacin		Infections in urinary tracts	Mixed	Saline (NaCl)

Table 3. Examples of drugs (quinolones) commonly used as corrosion inhibitors [35].

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