

# Inhibition of Microbial Corrosion by Green Inhibitors: An Overview

**Ghosal, Joydeep**

*Department of Biotechnology, Manipal Institute of Technology, Manipal Academy of Higher Education,  
Karnataka, INDIA*

**Lavanya M.\*<sup>+</sup>**

*Department of Chemical Engineering, Manipal Institute of Technology, Manipal Academy of Higher Education,  
Karnataka, INDIA*

**ABSTRACT:** Corrosion is a spontaneous process that affects valuable metal products, is dangerous, and causes expensive damage to many industries. There are many kinds of corrosion; among them is microbial corrosion. The effect of Microbiologically Induced Corrosion (MIC) can be explained in three steps: development of biofilm, alterations of the environment of the metal surface, and metal degradation (or corrosion). Common bacteria responsible for inducing MIC are sulfate-reducing bacteria, iron-reducing bacteria, and acid-producing bacteria. Metals and alloys have been treated with corrosion inhibitors to prevent microbiological corrosion. Expensiveness, toxicity, and in certain instances, ineffectiveness of these inorganic inhibitors have shifted the attention towards the organic green inhibitors. These green inhibitors used against corrosion are derived from plant extracts and organic substances. In addition to being ecologically acceptable and environmentally friendly, plant extracts are readily available, inexpensive, and renewable. This review paper explains microbial corrosion in metals and the mechanisms of MIC. In addition, this paper also reviews metal corrosion inhibition, green inhibitors, their types, and the impact of molecular structures on corrosion inhibition.

**KEYWORDS:** Metal corrosion; Microbial corrosion; Corrosion inhibition; Green inhibitors.

## INTRODUCTION

The gradual decaying of materials by a chemical or an electrochemical reaction(s) with their immediate surroundings is corrosion. Corrosion is a naturally occurring phenomenon through which a material can transform into its more chemically stable form. The same term, corrosion, has also been accepted to degrade non-metallic materials, but it is still widely used to refer to metals' decaying [1,2].

Corrosion of materials has led to problems that have taken a toll on those we consider assets. The annual cost for rectifying corrosion globally is approximated at USD 2.5 trillion. This cost, in turn, has affected the economies of countries adversely. Statistics suggest that these costs will rise shortly as the world moves towards an era dominated by machines [3].

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\* To whom correspondence should be addressed.

+ E-mail: lavanya.m@manipal.edu

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Corrosion comes in many forms. In recent history, these were grouped into eight forms familiar to us. Uniform or general attack, galvanic or two-metal corrosion, crevice corrosion, pitting corrosion, intergranular, selective leaching/parting, erosion, and stress corrosion are only a few of them [3].

A type of corrosion, which can be defined as accelerating the forms mentioned earlier, is microbiologically induced corrosion, also known as microbial corrosion. Microbial corrosion is sketched out as the degradation of metals brought about by microorganisms' presence and activities [4]. This form of corrosion can be harmful to numerous engineering materials [5]. The existence of water (moisture) and bacterial biofilm development on the metal surface is held accountable for the erosion of metals in this microbiologically induced corrosion (MIC) process [6].

The critical among the microorganisms concerned in such types of corrosion is bacteria. The responsible bacteria are usually unicellular without a true nucleus. The existence of Sulfate-Reducing Bacteria (SRB) and different sorts like acid-producing bacteria, iron-oxidizing bacteria, and slime forming bacteria, and on different occasions, supplementary microorganisms such as yeasts, algae, sulfur bacteria, and molds can be held responsible for MIC. MIC typically targets the external surface of metals in an aqueous environment. Microorganisms tend to colonize on surfaces of metals in patterns that are in the shape of detached colonies or spots instead of continuous films [7].

Microbial corrosion can affect a wide range of industries or structures, affecting the environment. A few of the examples are oil and gas industries, sewer systems, power plants, metal bridges, and as well as in aircraft fuel tanks and marine reservoirs [8–13].

Fundamentally, the anticipation of MIC would intend to forestall the formation and advancement of biofilm on the metal surface. To prevent microbial corrosion, various methods are exploited. Countless operating procedures have been proposed to tackle this anomaly. Some of them include a choice of corrosion-resisting materials, material design, coating the surface with an anti-corrosive protective coating, anodic and cathodic conservation (electrical defense). One of these techniques is the utilization of inhibitors for corrosion. The technique involving inhibitors is the most perceptive and proper method to forestall MIC. [2,3,5,14].

Efforts have been undertaken over the years to find suitable organic corrosion inhibitors for usage in different corrosive conditions. Thioaldehydes, nitrogen-based materials and their derivatives, aldehydes, sulfur-containing substances, acetylenic compounds, and different alkaloids such as strychnine, quinine, nicotine, and papaverine are all utilized as inhibitors in acidic medium. Potent inhibitors in neutral media include phosphate, nitrite, benzoate, and chromate. Inhibitors reduce or avoid the reaction of metal with different mediums. They help in reducing the corrosion rate by:

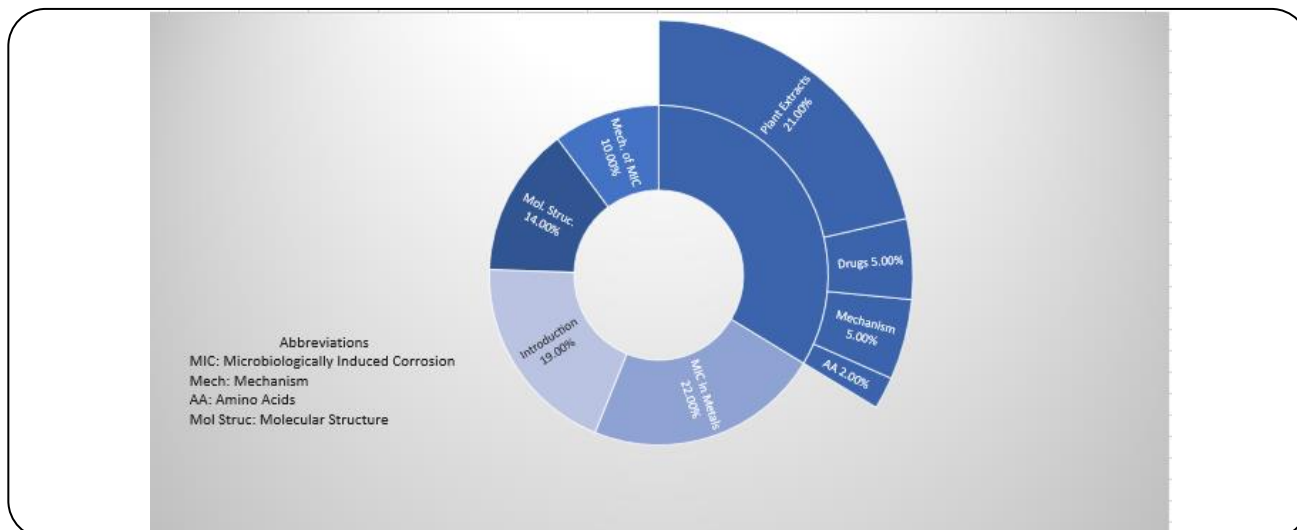
- Ion/molecule adsorption on metal surfaces.
- Altering the anodic and/or cathodic reactions.
- Reducing the rate at which reactants diffuse to surface of the metal.
- Reducing the metal surface's electrical resistance.
- Inhibitors that are mostly simple to use and can be used *in situ* [15].

In recent years the use of harmful inhibitors has been reduced due to the awareness of the safety towards the environment. Green corrosion inhibitors are employed to reduce toxicity and increase the usage of more environment-friendly products. Green inhibitors do not contain any heavy metals or potentially harmful components and are biodegradable. Green inhibitors are utilized to avoid corrosion in metal in acidic and alkaline environments. Because of the dangers posed by the poisonousness of traditional inorganic and synthetic inhibitors, the use of green inhibitors has grown. Natural products are the source of most green inhibitors, which act as anti-corrosion agents and are environmentally friendly and harmless when discarded. When green inhibitors are present in insufficient amounts, they can be used to treat the surface of metals in erosive situations [15,16].

Fig. 1 represents the total number of papers researched and reviewed to write and complete the review of this article.

## MICROBIAL CORROSION IN METALS

In 1891, Garrett's hypothesis that microbes play a role in metal corrosion was perhaps the first to be published. By demonstrating the presence of substantial quantities of sulfur, he proved that iron and sulfur bacteria were responsible for the corrosion of the inside and outside of water pipes [4].



**Fig.1: The percentage distribution of total review and research articles cited in this manuscript**

Both aerobic and anaerobic conditions can cause microbial corrosion. In aerobic corrosion, oxygen acts as an electron acceptor at the cathodic regions, allowing to delivery of hydroxides. Compared to lightly colonized areas, the typical non-consistency of a microbial film on an outer metal surface causes oxygen reduction underneath a thick layer. An area of oxygen reduction becomes anodic to several oxygenated regions during this reaction. Following that, a vulnerable metal travels into the solution at the anode, while at the cathode, electrons condense with water and oxygen. Gaines' work has linked the oxidation of metallic materials in anaerobic conditions to the behavior of microorganisms. The importance of sulfate-reducing bacteria has been consistently emphasized [17]. Metals such as copper amalgams and structural steels, for example, disintegrate across the entire surface without crevices or galvanic impacts [7].

The summary of different investigations on microbial corrosion affecting different metals is represented in a tabular form at the end of the following matter.

#### **Microbial Corrosion in Aluminum**

*Aspergillus niger* is a prevalent fungus in natural habitats, especially in warm and humid regions.

The rapid development and wide temperature and pH tolerance range of *A. niger* may explain its widespread distribution in many habitats. Previous research has shown that *A. niger* can grow in the temperature range 6–47 °C and remain active over an extensive wide pH range of 1.4–9.8 [18].

In a humid and clean environment, the corrosion of aluminum amalgam 2024 (AA 2024-T3) coupons exposed to *Aspergillus niger* fungus cultivated on glucose-comprising potato dextrose agar was investigated. Corrosion rate value in the systems containing only NaCl was  $37.2 \pm 3.5 \mu\text{m}$ , whereas the corrosion rate value for coupons exposed to *A. niger* with NaCl was  $149 \pm 3.5 \mu\text{m}$ . In addition, the rate of corrosion of aluminum alloy 2024-T3 by *Aspergillus niger* was four times faster than that of sodium chloride. Furthermore, intense pitting corrosion was observed, similar to that seen in aluminum alloy 2024-T3 when exposed to sodium chloride, and Al reduction correlated with copper enhancement [19]. From this experiment, we can conclude that *Aspergillus niger* increased the corrosion rate more than the coupons subjected to only NaCl solution. The comparison of results in these two conditions implied that the weight loss of the coupons caused by *A. niger* was much more significant than that caused by exposure to only NaCl.

In the presence of the *Aspergillus niger* fungus, corrosion responses of non-anodized and anodized aluminum amalgam 6061 were examined. The corrosion rate of anodized aluminum amalgam was slower than that of non-anodized aluminum amalgam. Anodized AA 6061 had reduced microbe adherence than non-anodized AA 6061, and microorganism growth was lower on anodized AA 6061 than non-anodized AA 6061. As a result, the corrosion rate of non-anodized AA 6061 was higher than that of anodized AA 6061. Localized corrosion was visible using optical microscopy. Localized corrosion was shown

to be more likely on anodized AA 6061 than on non-anodized AA 6061 [20].

Extracellular Polymeric Substances (EPS), which are made up of various macromolecules, mediate early cell attachment to the material surface and constitute form a biofilm matrix [21]. Secondary metabolites are produced inside the biofilm [22]. Microorganism attachment begins reversibly and then becomes irreversible. Microbe attachment is mediated by electrostatic contact and van der Waals forces, which are the same factors that hold the biofilm structure together. Physical adhesion between the microbe and the substrate regulates the initial attachment of cells. Adsorption occurs when the first colonists form weak, reversible bonds (slightly more strong than electrostatic repulsive forces) and hydrogen bonds with a surface. Irreversible attachment is achieved by secreting more EPS into a sponge-like matrix. This adhesive substance links the microbes to one other and the surface collectively indefinitely [23].

The majority of MIC reports on aluminum alloys 2024 and 7075 emerge from aircraft or underground fuel storage tanks. The United States Air Force (USAF) aviation fuel tanks (building with AA 2024-T3) were investigated for microbial contamination. There were 12 genera discovered, with four *Bacillus* species and two *Staphylococcus* species among them. Microbial adaptability to modifications in fuel content, notably additives and biocides, may be demonstrated by changes in the microbial community constitution, allowing bacteria to dominate the material. Microbial proliferation in jet fuel storage tanks causes fuel quality to deteriorate and aluminum alloys to corrode. The corrosion of AA 7075, on the other hand, has been studied in the presence of the fungus *Cladosporium resinae* and bacteria [24].

The role of microorganisms in aircraft corrosion damage was investigated in collaborative research including many European aircraft manufacturers. A total of 208 microorganisms were gathered from corroded locations of 7 separate aircrafts, including 158 bacteria, 36 yeasts, and 14 fungi. The results demonstrate that isolated bacteria from the genera *Penicillium*, *Enterococcus*, *Aerococcus*, *Micrococcus*, *Aspergillus*, *Bacillus*, and *Staphylococcus* corroded AA 7075 in a powerful way [25].

The conditions for the breakdown of passivity of aluminum alloys in the presence of microbiological contaminants in jet fuels were investigated in this study.

*Cladosporium resinae*, *Candida sp.*, and two strains of *Pseudomonas aeruginosa* were employed as microorganisms isolated from fuel storage tanks. *C. resinae* contributes to the corrosion process by altering the passive film/electrolyte characteristics, which results in metal pitting. In these conditions, the metabolic products of *C. resinae* are unable to trigger the breakdown of passivity of aluminum in the potential region where pitting occurs. Fuel water systems with oxidizing properties (particularly those with *C. resinae* and some strains of *Pseudomonas aeruginosa* as contaminants) provide ideal conditions for pitting. The elevated redox potential values reported in the presence of jet fuel pollutants are linked to these conditions [26].

### **Microbial corrosion in Zinc**

The effect of the biofilm of sulfate-reducing bacteria (SRB), which is associated with the 70/30 copper-zinc amalgam corrosion in culture medium under anaerobic conditions, was investigated in the laboratory. The open-circuit potential measurements displayed that the free corrosion potential was much nobler in the sterile culture medium than in the presence of SRB. The corrosion rate of the alloy initially decreased then stabilized upon exposure to the sterile culture medium. Inactive SRB, the corrosion rate initially decreased rapidly than gradually increased. These results indicate that the presence of SRB increases the corrosion rate of the alloy by three times. The presence of SRB decreased the linear polarization resistance of the alloy. With increasing the exposure time to culture medium inoculated with SRB, the impedance first increased and then decreased and presented two-time constants. This behavior was attributed to the formation of an adherent corrosion product layer, and it was correlative with the structure of the SRB biofilm. When there existed the acidic substance produced by SRB, the primary biofilm became loose, porous, and lost its protection, and enhanced the corrosion of the alloy [27]. From this investigation, we can conclude that the corrosion rate was higher in the medium containing SRB than in the sterile culture medium.

Because of their practical value, SRB is the most investigated bacteria in the MIC literature. The valence of the sulfur element ranges from -2 to +6. SRB can utilize various sulfur compounds with a valence greater than -2 as terminal electron acceptors in addition to sulfate.

Bisulfite ( $HSO_3^-$ ), thiosulfate ( $S_2O_3^{2-}$ ), and elemental sulfur are among them. SRB are strict anaerobes. However, they can survive for a short time in the presence of oxygen without growing [28].

*Aspergillus niger* was identified in metal samples exposed to maritime, rural, and urban environments in Lithuania. Under laboratory circumstances in a humid atmosphere, Al and Zn samples were exposed to *A. niger* for two years. The wild strain *A. niger* worked as a corrosion activator or inhibitor depending on the metal it colonized. Zinc showed microbially caused corrosion acceleration, while aluminum showed signs of corrosion inhibition. According to EIS data, under the effect of *A. niger*, two-layer structures grew on Zn and Al. The primary cause of Zn corrosion acceleration is a reduction in the thickness of the inner layer (close to the metal), which has the highest passivating capacity. The research showed that microorganisms might be utilized as corrosion shields instead of hazardous chemicals, whose use is becoming increasingly limited [29].

The erosion activity of nickel-zinc amalgam layers was investigated in a nutrient broth medium, and the bacteria *Aeromonas eucrenophila* detached from a corroded water treatment arrangement. Polarization curves indicated that the corrosion density and corrosion current displayed a cathodic shift after inoculation that confirmed an enhancement in corrosion rates for Ni-Zn amalgam layers. The QCM tests indicated the mass depletion and impact of corrosion of *Aeromonas eucrenophila*. With increased incubation time, the bacterial biofilm that developed on the amalgam plate surface grew in coverage and heterogeneity, causing the Ni-Zn compound layers underneath the biofilm to deteriorate in the pattern of pits and crevices erosion [30].

#### **Microbial corrosion in stainless steel**

An experiment was carried out in the laboratory on the corrosion activity of stainless steels submerged in sterile and natural seawater to see if the observed enhancement of free corrosion capabilities in sea-submerged stainless steels could be explained by the colonization of biotic microbiological slime on the surface of the metal. The investigation outcomes affirmed that all stainless steel samples remained undamaged from local attacks. Some cases of crevice corrosion were observed on two steel types, AISI 316 and 304, in the loops fed with natural

seawater. The results confirmed that a microbiological component protruding to metal drenched in standard seawater surfaces might modify the oxygen reduction kinetics. The most apparent explanation appears to be enzymatic catalysis on oxygen release, triggered by enzymes generated by the benthonic population and confined in the next instantaneous region of the stainless steel by similar biopolymers that held the biofilm together. This mechanism speeds up the initiation of localized attacks and their extension and progression, explaining why typical situations have more aggression than sterile situations with the same amount of salt. We can conclude from this study that a microbiological component adhering to the surface of metals in natural seawater can change the oxygen reduction kinetics [31].

Many authors have observed that natural seawater tends to be more corrosive than artificial seawater to various structural materials, including stainless alloys. According to various authors, this observation is caused by the action of marine bacteria on surfaces soaked in natural seawater. In the current study, AISI 316 SS pitted within three days in natural seawater, but equivalent samples remained pit-free in filtered seawater for at least seven days. The sole difference between these two sets of testing was that the natural water had a standard concentration of biofilm-forming bacteria, whereas the filtered water had far lower quantities [32].

2205 Duplex Stainless Steel (2205 DSS) microbial corrosion in oilfield-created water containing SRB (sulfate-reducing bacteria) was examined. The four phases of SRB growth in the oilfield water injection system were viscous growth, logarithmic growth, stable growth, and decline. Sulfate-reducing bacteria metabolism instigated surrounding parameter changes. pH enhanced during viscous, logarithmic, and stable sulfate-reducing bacteria development, diminishing in the decay stage. Alternatively, the conductivity of a solution is enhanced during logarithmic growth and viscous phases, diminishing during steady development and decay phases. The sulfide concentration diminished during steady sulfate-reducing bacteria development, expanded in viscous and logarithmic growth stages and stayed steady in the decay phase. Sulfate-reducing bacteria metabolism enhanced stainless steel corrosion during logarithmic growth and viscous phases. Nonetheless, during stable development, the close-packed microorganism film attached

to the surface of the metal and repressed erosion. The depletion of the microorganism biofilm, combined with erratic delamination of metabolite layers and corrosion products, led to the formation of local corrosion galvanic cell [33].

#### **Microbial corrosion in mild steel**

Due to its low cost, mild steel is a standard metal in the construction industry. In either case, as compared to other metallic materials, it has poor resistance towards corrosion in both aqueous and non-aqueous environments [34].

An anaerobic bio-digester of wastewater mild steel sample was exposed to aqueous solutions as part of an experiment. The material showed bacteria adhesion, especially sulfate-reducing bacteria (SRB), which produced insufficiently plentiful biofilms accompanied by corrosion products on the mild steel surface. Experiments were performed in the presence of a gaseous phase containing and at 1 and 7 atmospheres pressure. The corrosion potential at 1 atm rapidly decayed from -0.3V and remained at low values (~-0.7V). On the surface, a black, poorly adhering film formed. The dark film was still present at 7 atm, but it did not entirely cover the surface. Mild steel endures generalized corrosion at both pressure circumstances. Microorganisms have attached themselves to the material examined. It forms a corrosion product layer in the solution that is non-protective and contains oxides and sulfides [35].

The pressure on corrosion is influenced by partial pressures of acid gases (e.g., polysulphide and CO<sub>2</sub>), which may impact the dissolution of corrosive species into solution. Higher pressures can hasten corrosion by hastening the breakdown of the metal's protective surface layer. On the other hand, higher pressures may aid in creating an adherent and dense surface layer, reducing corrosion [36].

In a continuous flow system under completely anaerobic circumstances, mild steel corrosion was explored in the presence of a diverse colonized biofilm and sulfate-reducing bacteria (SRB). The biofilm formation was influenced by reinforcing the closed channel flow reactor with the low-concentration substrate at varied dilution rates. In the absence of ferrous iron, there was no direct association between corrosion and SRB activity. During a 21-day experiment (SRB at  $2.6 \times 10^9/\text{cm}^2$ ), the metal surface preserved its scratch lines when the growth of iron sulfide layer on mild steel was prevented

before the biofilm formed. Localized corrosion became obvious after 14 days and continued for 21 days when the iron sulfide layer formed before the biofilm grew. In the localized corrosion area, an intergranular and pitting attack was observed. All biogenic sulfide precipitated at high iron concentrations (about 60 mg/L in bulk water), and corrosion was significantly increased [37].

#### **Microbial corrosion in copper**

Using revolving cylinder samples in seawater containing sulfide, oxygen, polysulfide, and sulfur, corrosion rates of copper amalgams 715 (70:30 Cu: Ni) and 706 (90:10 Cu: Ni) were calculated. A control test was also carried out using uncontaminated deaerated seawater. These specimens can transform predominantly non-corrosive deaerated seawater into a similarly corrosive circumstance. Rates of corrosion were checked utilizing linear polarization and potential step strategies. In any case, corrosion rates in the polluted conditions were not significantly different from those in uncontaminated aerated seawater. Theoretically, the porous surface film (most likely cuprous sulfide) produced in all polluted environments could cause accelerated attack by inhibiting the formation of the protective cuprous oxide layer on subsequent exposure to uncontaminated aerated water. From this investigation, it was inferred that Cu- Ni amalgams could erode quickly in de-aerated seawater if these typically passive alloys were created functional by the existence of sulfur or if sulfide was accessible as an oxidant [38].

The corrosion behavior of carbon steel, pure aluminum, and copper in seawater was studied. Corrosion potential for carbon steel and aluminum had shifted negatively over time. Corrosion shifts the corrosion potential from positive to negative. In seawater, certain metals corrode quickly. Biofilms are difficult to generate on corroding metal substrates. Bacteria rarely adhere to copper surfaces. Hence the corrosion potential is rarely altered [39].

#### **Microbial Corrosion in Titanium**

In this study, the function of titanium (Ti) corrosion in a nutrient-rich media was investigated in the presence and non-appearance of *Actinomyces naeslundii*. *Actinomyces naeslundii* had the option to colonize the exterior of Ti and then create a thick biofilm. Pictures of SEM unconcealed

**Table 1: Microbial Corrosion in Metals.**

Metal	Medium	Microorganism Involved	Type of Corrosion	References
Aluminum/Al Alloy	Dextrose agar, culture medium	<i>Aspergillus niger</i> (fungus)	Pitting.	[18-22]
Zinc/Zn Alloy	Seawater, culture medium	Sulfate-reducing bacteria (SRB)	Pitting, Micro-pits.	[23,24]
Stainless Steel	Artificial and natural seawater, oilfield water	SRB	Crevice, local attack.	[25-27]
Mild Steel	Aqueous solutions, continuous flow sys.	SRB	Localized corrosion, Intergranular attack	[28-31]
Copper	Seawater, artificial groundwater	Microbes in seawater	Uniform corrosion, Pitting	[32,33]
Titanium	Nutrient rich medium, semi-continuous media	<i>Actinomyces naeslundii</i> (bacteria), <i>Desulfovibrio vulgaris</i> (SRB type)	Localized corrosion, pitting	[34,35]

the micro-pitting corrosion rate on the surface of the metal after eradication of the biofilm. From EIS, the electrochemical corrosion conclusions demonstrated a vital depletion in the corrosion-resistant ( $R_p$ ) value after submerging the titanium in *Actinomyces naeslundii* culture for three days. Similarly, XPS disclosed a decrease in the comparative extents of oxygen and titanium. After submersion of the metal in *Actinomyces naeslundii* culture, a comprehensible decrease of ascendant titanium dioxide ( $TiO_2$ ) was observed in the surface oxides. The outcomes implied that *Actinomyces naeslundii* created metabolites could diminish the stability and strength of the protective  $TiO_2$ , which is present in the surface oxides, that progressively reduces the resistance for corrosion of Ti, bringing about enhanced titanium corrosion when submerged in *Actinomyces naeslundii* solution as a function of time [40].

Titanium (ASTM Grade 2) pitting corrosion was examined by disclosing pieces to a culture, which is semi-continuous, consisting of SRB (sulfate-reducing bacteria) type, *Desulfovibrio vulgaris*. The metal pieces were disclosed to a semi-continuous culture for ninety days alongside a control set in a media, not inoculated. Epifluorescence microscopy revealed substantial habitation by the sulfate-reducing bacteria. The existence of two kinds of pits, enormous pits of diameter 2mm, which is hemispherical in shape and numerous micro-pits, were exposed by Optical microscopy. In control pieces disclosed to sulfate-reducing bacteria-free medium, no pitting was detected. Confocal Scanning Laser Microscope (CSLM) and Scanning Electron Microscopy (SEM) pictures demonstrated the eroded Ti exterior with various micro-pits, alongside conventional SRB cells, which were

rod-shaped in regions of the pits. Energy-Dispersive X-ray Analysis (EDAX) demonstrated peaks for Fe, C, P, O, Ti, and N in the corroded area. X-ray Photoelectron Spectroscopy (XPS) information revealed clear phosphorous and titanium sulfur peaks. From this investigation, it was inferred that within the existence of 0.2- 0.4 mM of sulfide and at room temperature, in addition, the putative phosphine, sulfate-reducing bacteria could advance pitting erosion of titanium by the development of titanium sulfide [41].

Table 1 consists of information regarding the microbial corrosion effecting different metals.

### MECHANISM OF MICROBIAL CORROSION

The mechanisms by which microbial corrosion principally include the fundamental electrochemical mechanisms of corrosion, specifically, the eradication of electrons employing oxygen or hydrogen ions. One case is the corrosion mechanism of iron without oxygen and at neutral or near-neutral pH values. Several hypotheses have been presented to explain this mechanism, the premier one being the typical cathodic depolarization hypothesis [7].

Fig. 2 displays the classification of mechanisms of microbial corrosion.

#### *Cathodic Depolarization*

In 1934, *Van der Vlugt* and *Von Wolzogen Kuehr* recommended the corrosion process instigated by SRB (sulfate-reducing bacteria) using the oxidation of the cathodic hydrogen as detailed in the cathodic depolarization hypothesis, which is a depolarization. Once metallic material is disclosed to water, it is polarized by an anodic reaction (dropping positive metal ions).

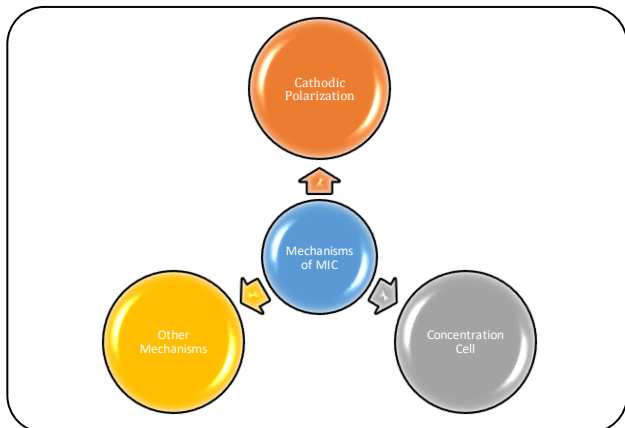


Fig. 2: Different Mechanisms of Microbial Corrosion.

In the absence of oxygen, the free electrons create hydrogen that colonizes on the surface of the metal, which will form a dynamic equilibrium that minimizes cathodic reaction (water-acquired protons). SRB is anticipated to deplete the created hydrogen. Therefore, oxidation of iron occurs. The mentioned mechanism enhances the dissolution of anodic metal and simultaneously ferrous sulfate (FeS) and ferrous hydroxide (Fe) as products of corrosion are generated [42].

The cathodic depolarization process is explained in the form of equations which are given below [4,42].

Anodic reaction:



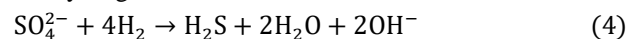
Water dissociation:



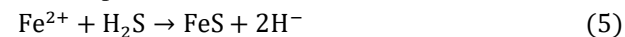
Cathodic reaction:



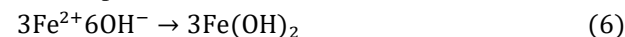
Hydrogen oxidation:



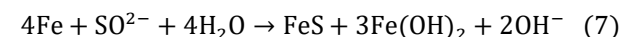
Precipitation:



Precipitation:



Final Reaction:



### Concentration cell formation

The adhesion to a metal surface (biofilm development) by aerobic creatures reduces the oxygen concentration at the metal surface, leading to an oxygen differential and

establishing corrosive, oxygen-differential cells [4]. Adsorbed cells develop, replicate, and make communities, which are physical irregularities on the surface of a metal, bringing about in local cathodes and anodes and developing differential aeration cells [43]. Because of the developed oxygen concentration cell, the colony that acts as an anode has a low oxygen concentration. The formation of the corresponding cathode demonstrates a higher oxygen concentration in the surrounding environment [44]. The non-uniform biofilm layer is one reason that promotes the production of differential aeration cells. The biofilm works as a barrier, preventing oxygen diffusion to the metal surface. However, since biofilms are deposited in a patchy and heterogeneous manner, oxygen concentrations vary between coated and uncoated metal areas. Bacterial aerobic respiration is another factor that may lead to differential aeration cells. Areas of metal under respiratory colonies serve as anodes during aerobic respiration, while areas away from these colonies, with higher oxygen concentrations, serve as cathodes [45].

### Other mechanisms

#### Mechanism inside biofilms

It is conventional to discuss microbes inside biofilms as anaerobic or aerobic and as individual species. In any case, microbes create synergistic colonies that regulate joined processes, which is not possible by individual species. Moreover, a solitary microbe can, at the same time, influence corrosion through various mechanisms. Cell lysis or death inside a complete-grown biofilm does not necessarily mean eliminating the impact on electrochemical techniques. It had been affirmed that pitting corrosion carries on underneath accumulations of iron-oxidizing bacteria free of biochemical action of the bacteria. Likewise, ferrous sulfate generated microbiologically boosts reactions of corrosion in the non-existence of viable cells [43].

#### Production of oxidizing agents

Organic and inorganic acids are among the most microorganism-generated corrosive products. The sulfuric acid generation by several species in the genus *Thiobacillus* causes the most spectacular metal erosion and degradation of rubber, concrete, and stone. In unadulterated cultures comprising *Thiobacillus thiooxidans*, essential sulfur might generate up to 10% sulfuric acid. A current



case of extreme underground erosion of tubing made of copper in the south of Maryland was most likely because of the mentioned mechanism. A marsh was set up for a building site by excavating and heaped to permit the water to drain. The marsh soil was then spread over an enormous region and combined with the primary sandy clay soil. The soil pH was 2.0, and chemical tests suggested significant free sulfuric acid [46].

Besides imposing corrosion issues, hydrogen sulfide is foul-smelling and venomous, causing a problem to employees of sewers when the concentration is high. Under anaerobic conditions, hydrogen sulfide is generated fundamentally inside the biofilms, wrapping the immersed surface of the pipes. Although, the corrosion of concrete happens when hydrogen sulfide is discharged from the wastewater to the sewer environment, where it eventually absorbs to the surface of the concrete higher than the wastewater. On the concrete surface, hydrogen sulfide is oxidized to sulfuric acid. Gypsum is produced as the sulfuric acid reacts with the alkaline components of the concrete. Since gypsum has minimal structural strength, this operation leads to exceptional structural deterioration of concrete pipes. Temperature is a vital parameter in the production of hydrogen sulfide. Rising temperatures lead to enhanced oxygen consumption rates, resulting in anaerobic circumstances and enhancing the rate of hydrogen sulfide formation [47].

In marine resources and industry, anaerobic Sulfate-Reducing Bacteria (SRB) and aerobic Iron-Reducing Bacteria (IOB) frequently induce microbial corrosion. IOB absorbs oxygen in the medium to make a proper development habitat for anaerobic sulfate-reducing bacteria and then advance the erosion of the matrix by sulfate-reducing bacteria. IOB and SRB collaborate to create biofilms on metal surfaces during this procedure. Corrosion materials, sessile cells, and Extracellular Polymeric Substances (EPS) from these two bacteria make up these coatings. As time passes, the stability of biofilms reduces, and afterward, some portion of them will tumble off to make heterogeneous films. Localized corrosion of products produced by these heterogeneous films to quicken corrosion rate. Heterogeneous biofilms produce corrosion in localized form because when these films happen at the surface of a material, the regions with thick biofilms forestall diffusion of oxygen to them, and the aerobic bacteria in biofilms likewise preclude the oxygen

beneath the biofilms. Each of them succeeds in creating areas with low oxygen concentrations. As a result, these areas act as anodic sites, causing the product to deteriorate. Simultaneously, regions with thinner biofilms or no biofilms have higher oxygen concentrations, which operate as cathodic sites for electron consumption and oxygen reduction reactions [13].

#### **MECHANISM OF CORROSION INHIBITORS**

At low concentrations, inhibitors are chemicals or mixtures that inhibit, avoid, or mitigate corrosion in hostile situations. The inhibitor's mechanism is usually one or more of the three mentioned below:

- The inhibitor is chemically adsorbed (chemisorption) on the metal surface and resulting in a protective thin film with inhibitory qualities, or a mixture of inhibitor ions and metallic surface produces a protective thin layer with inhibitory properties.
- The inhibitor promotes the production of a film by preventing the base metal from oxidizing.
- The inhibitor reacts with a potentially corrosive component in aqueous media, resulting in a complex product [48].

#### **GREEN INHIBITORS**

As there has been an ascent in environmental consciousness and a modification in guidelines that prohibit inorganic and synthetic inhibitors of corrosion because of their noxiousness, green inhibitors are of attraction. Natural or organic materials are a quality supply of green inhibitors, where their extracts consisting a large portion of the essential elements, for example, N, S, O, and C, which are effective in organic compounds, aid in adsorption of these components on amalgams or metals to create a layer that blocks corrosion and safeguards the surface [16].

#### ***Mechanism of green corrosion inhibitors***

The corrosion inhibitory property of natural products has been attributed to a variety of mechanisms of action:

Capsicum frutescens (CF) fruit extracts' antimicrobial activity was examined. The anti-corrosion effect of the ethanol extract on low carbon steel in acidic media was investigated experimentally. Using the agar disc diffusion method, the antibacterial efficacy of ethanol, methanol, water, and petroleum spirit extract against

corrosion-associated Sulfate-Reducing Bacteria (SRB), *Desulfotomaculum* species, was examined. Because of the phytochemical elements included in CF extract, such as alkaloids (8.8%), tannins (0.4%), and saponins (0.4%), it successfully reduced both corrosion and SRB growth (39.2 %). Because of the phytochemical constituents present in CF extracts, such as alkaloids (8.8%), tannins (0.4%), and saponins, it effectively inhibited both corrosion and SRB development (39.2 %). Molecular dynamics (MD) simulations were used to show the electronic structure and adsorption behavior of the active alkaloidal ingredients of CF extract, capsaicin and dihydrocapsaicin, which gave molecular-level insights into their contributions to the extract's corrosion inhibitory function [49].

In a 1 M HCl medium with and without various concentrations of a Hydro-alcoholic Extract of used Coffee Grounds (HECG), the temperature on corrosion of C38 steel was investigated. Temperatures between 293.15 to 323.15 K were used for the potentiodynamic polarization technique and electrochemical impedance spectroscopy. It was discovered that as the temperature and inhibitor concentration was increased, the inhibition efficiency decreased. At all temperatures examined, the adsorption of HECG on the surface of C38 steel was spontaneous and followed the Langmuir isotherm. The physical adsorption of HECG compounds on the surface of C38 steel was demonstrated by the thermodynamic adsorption values [50].

Allura Red (AR), Fast Green (FG), Amaranth (AM), Sunset Yellow (SS), and Tartrazine (TZ) were investigated for their corrosion of mild steel inhibition properties in a solution of 0.5 M HCl with the help of quantum chemical calculations, gravimetric, and potentiodynamic polarization. The findings demonstrated that the dyes under investigation were excellent corrosion inhibitors with significant inhibition efficiency. The inhibitory efficiency of all colors studied increases with concentration and decreases with temperature. In the presence of KI, the inhibition efficiency of dyes improves because of synergistic interactions between iodide (I) ions and dye molecules. According to potentiodynamic polarization results, the dyes examined were mixed-type inhibitors in the presence and the absence of KI. The adsorption of these investigated dyes on mild steel surfaces, in the presence and absence of KI, requires a physical adsorption process and follows the Langmuir adsorption isotherm. The S, O,

and N heteroatoms in the dye molecules are the most likely sites for encounters with mild steel, according to quantum chemical calculations [51].

Microorganisms produce secondary metabolites under constant stress conditions. A single microbial type can produce various secondary metabolites, for example, antibiotics. Antibiotics can be taken orally, digested, and absorbed into the bloodstream. In a study, an expired antibiotic (cefdinir) was used as a corrosion inhibitor for mild steel in 1M HCl medium, providing an 81.4% of inhibition efficiency [52–54].

### **Types of green inhibitors**

#### *Plant extracts*

Plant extracts have become a common source of inhibitors. They are an intriguing alternative to synthetic inhibitors because they are readily available, cost-effective, organic, environmentally friendly, health-friendly, and sustainable. They are abundant in ingredients that have a high inhibition quality [55,56]. Saponins, anthraquinones, proteins, amino acids, alkaloids, polyphenols, glycosides, tannins, flavonoids, and other heterocyclic compounds are among the phytochemical constituents found in plant extracts. These phytochemicals are regarded as corrosion inhibitors [57,58]. Because the inhibitory effect is due to the adsorption of inhibitor molecules on the metal surface, forming a protective coating by blocking the active sites, phytochemicals found in plant extracts are utilized as corrosion inhibitors [59]. The adsorption functioning of organic inhibitors for corrosion is influenced by factors such as the inhibitor molecule's planarity, temperature, solubility, electronic structure, corrosive conditions, and the nature of metal. The utilization of plant extracts as another option for organic corrosion inhibitors has numerous advantages like biodegradable reduced environmental risk as most plant extracts are durable and non-toxic and are related with a negligible adverse effect of the encompassing region [60].

#### *Drugs*

Over the last two decades, extensive research and development have resulted in discovering new classes of corrosion inhibitors. The value of using a variety of drugs as corrosion inhibitors has increased. Drugs (chemical medicines) appear to be suitable candidates to substitute conventional toxic corrosion inhibitors due to their natural

origins, non-toxic properties, and minimal negative impacts on the aquatic environment [61]. The existence of heterocycles, heteroatoms, and the benzene ring, as isoxazoles, pyridine, thiophenes, have produced drug products as a reassuring supply of green inhibitors. The inhibitory effect of Candesartan drug on Carbon Steel (CS) in a 1 M HCl acidic medium was investigated. EDX, AMF, and SEM techniques were used to examine the surface morphology of the inhibited CS. The inhibitor had an Inhibition Efficiency (IE) of 79.8% at a concentration of 300 ppm [62].

Abdallah and Jahdaly examined the effectiveness of three amino-glycoside, amikacin, gentamicin, and kanamycin, as inhibitors for corrosion for aluminum with a purity of 99.99 % in a solution of 1 M HCl at 30°C. The inhibition effect was observed in the given order amikacin > kanamycin > gentamicin. According to the authors, the change in molecular weight and the number of heteroatoms present in these compounds were the reasons behind the effectiveness of inhibition order among the three antibacterial drugs. The authors attributed that these drugs are adsorbed through the oxygen atoms present in the heterocyclic rings and the amino groups on the aluminum surface. Coordination bonds could have developed between the nitrogen and oxygen atoms' unshared electron pairs and the empty p-orbitals of aluminum [63].

*Obot et al.* used the gravimetric approach to demonstrate the inhibition efficiency of two antifungal drugs, Clotrimazole (CTM) and Fluconazole (FLC), on aluminum corrosion in 0.1 M HCl. on the metallic surface, both drugs followed the Langmuir adsorption isotherm. The CTM and FLC drugs displayed maximum efficiencies of 88% and 82%, respectively, at  $1 \times 10^{-4}$  M concentration [64].

#### *Amino acids*

The amino acids exhibited a decent capacity to inhibit erosion of various metals in several habitats. Like many organic corrosion inhibitors, the inhibitive impact of those materials was ascribed to the aggregation of the inhibitor substances by “indirect” or/and “direct” adsorption onto the metal surface, which lessens the metal interaction with the erosive sources in solution. The key elements that affect the means of adsorption are the protonation state and chemical nature of the surface, as well as the magnitude and nature (charge) and inhibitor nature. Usually, two

adsorption methods could be regarded. In one method, the neutral inhibitor might be adsorbed through a chemisorption method on the metal surface, and in the second method, the charged inhibitor can collaborate electrostatically with a charged metal surface [65].

Utilizing Density Functional Theory (DFT) technique in gas aqueous states, three amino acids inhibition efficiencies (serine (S), tryptophan (W), and tyrosine (Y)) had been examined as green inhibitors on carbon steel erosion. Both the experimental and theoretical outcomes demonstrated that the sequence of efficiency of inhibition is W (tryptophan) > Y (tyrosine) > S (serine). A correlation of the outcomes in aqueous media and gaseous stage displayed that the trends in the molecular properties are comparative. Comparing tryptophan with the remaining inhibitors shows an exemplary electrophilic character. Therefore, highlighting its good capability as an electro-donor. The tryptophan has the biggest hardness and hence a convenient back-donation charge [66].

Table 2 is summarized on green corrosion inhibitors produced by natural products that have been used as in several aggressive media on different metals and alloys to protect them.

### **ROLE OF MOLECULAR STRUCTURE ON CORROSION INHIBITION**

The goal for corrosion prevention is to create inhibitors for corrosion with a better wetting behavior, better adsorption efficiency, and higher surface coverage. An organic molecule that meets all the above requirements is described as an effective corrosion inhibitor. The structure and temperature of operation and the composition of the target metal surface are significant factors of inhibitor adsorption. The hydrocarbon chain length, stereochemical influences, heteroatoms' presence, the size and presence of aromatic rings, and other structural features of an inhibitor molecule all influence inhibition adsorption [81].

#### ***Effect of carbon chain length***

The hydrocarbon chain length attached to a corrosion inhibitor molecule can significantly impact the corrosion inhibitor's hydrophobicity and film-forming behavior. Large alkyl chains can increase hydrophobicity while crowding the metallic surface, preventing inhibitor adsorption [81]. Marco investigated the carbon-chain length effect for poly-carboxylates of the series

Table 2: Different natural sources for green inhibitor used in different mediums.

Source of Natural Product	Microbe Involved	Alloy/metal studied	Studied Media	Inhibition Efficiency (%)	References
<i>Allium sativum</i> (garlic extract)	<i>Bacillus subtilis</i> A1, <i>Streptomyces parvus</i> B7	Carbon steel, Stainless steel	NaCl	81-CS 72-SS	[67]
<i>Catharanthus roseus</i> and <i>Lawsonia inermis</i> leaf extract	<i>Bacillus oleronius</i> EN9	Mild steel	Re-circulating water environment	<i>L. inermis</i> -75 <i>C. roseus</i> -44	[68]
<i>Azadirachta indica</i> (neem)	<i>Bacillus subtilis</i> A1, <i>Streptomyces parvus</i> B7, <i>Pseudomonas stutzeri</i> NA3 and <i>Acinetobacter baumannii</i> MN3	Carbon steel	NaCl	81- abiotic system 72- biotic system	[69]
<i>Citrus reticulata</i>	<i>Desulfovibrio sp.</i>	copper	1 M HCl	79.8	[70]
Methanolic extract of <i>Artemisia pallens</i> (MEAP)	<i>Bacillus megaterium</i> SKR7	Mild steel	1.5% NaCl	83	[71]
<i>Turbinaria ornata</i> (TO) extract	<i>Bacillus megaterium</i>	Mild steel	Conc. HCl	92.4	[72]
Glycolipid biosurfactant	<i>Bacillus subtilis</i> A1, <i>Streptomyces parvus</i> B7, <i>Pseudomonas stutzeri</i> NA3 and <i>Acinetobacter baumannii</i> MN3	Carbon steel	Abiotic and biotic systems	80- abiotic 87- biotic	[73]
<i>Citrus reticulata</i> reduced silver nanoparticles	<i>Desulfovibrio sp.</i>	X80 steel	1 M HCl	76.2	[74]
<i>Polyalthia longifolia</i>	SRB	Mild Steel	Postgate medium B (KH <sub>2</sub> PO <sub>4</sub> , NH <sub>4</sub> Cl, CaSO <sub>4</sub> , MgSO <sub>4</sub> .7H <sub>2</sub> O, sodium lactate, yeast extract, ascorbic acid, thioglycolic acid, sodium chloride, and FeSO <sub>4</sub> .7H <sub>2</sub> )	87.23	[75]
Aloe vera ( <i>Aloe barbadensis</i> ) extract	SRB, IRB	Steel	SRB- postgate medium IRB- Winogradsky medium	SRB-66 IRB-89	[76]
<i>Salvia officinalis</i>	<i>Pseudomonas aeruginosa</i>	304L Stainless Steel	2216E medium: NaH <sub>2</sub> PO <sub>4</sub> , NH <sub>4</sub> NO <sub>3</sub> , NaF, Na <sub>2</sub> SiO <sub>3</sub> , H <sub>3</sub> BO <sub>3</sub> , SrBr <sub>2</sub> , SrCl <sub>2</sub> , KBr, Na <sub>2</sub> CO <sub>3</sub> , KCl, CaCl <sub>2</sub> , Na <sub>2</sub> SO <sub>4</sub> , MgCl <sub>2</sub> , NaCl, ferric citrate, yeast extract and peptone	97.5	[77]
Pyocyanin extracted from <i>Pseudomonas aeruginosa</i>	<i>B. thuringiensis</i> EN2, <i>T. aaidingensis</i> EN3 and <i>B. oleronius</i> EN9	Copper	0.2M HCl	72	[78]
Catechin hydrate (CH)	<i>Pseudomonas aeruginosa</i>	304L Stainless Steel	2216E medium	99.8	[79]
Benzothiazole	<i>Desulfovibrio caledoniensis</i>	hot-dip galvanized steel	Modified Postgate medium	80	[80]

$^{-}\text{OOC}-(\text{CH}_2)_n-\text{COO}^{-}$  (n in the range of 0–7) and pointed out that for the inhibition effect, there is an optimal chain length. With increasing intermediate chain length, the pitting potential ( $E_{\text{pit}}$ ) increases at first, then decreases until it reaches its maximum for an intermediate chain length (n = 3) [82].

In 15% HCl solution, an imidazoline, 2-heptadecyl-1-[2-(octadecanoylamino) ethyl]-2- imidazoline (QSI) with -

$\text{C}_{17}\text{H}_{35}$  as the length of the tail chain was synthesized, described, and examined as an inhibitor for corrosion for low carbon steel. The influence of the length of the pendant hydrocarbon chain on inhibitory efficiency ( $\eta$ ) was studied. Although it effectively resisted low carbon steel corrosion in the investigated media, QSI had a diverse pattern of action. The highest concentration tested (400 mg/L) afforded  $\eta$  of <50%. As the length

Table 3: Effect of molecular structures on corrosion inhibition.

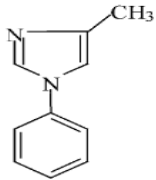
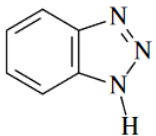
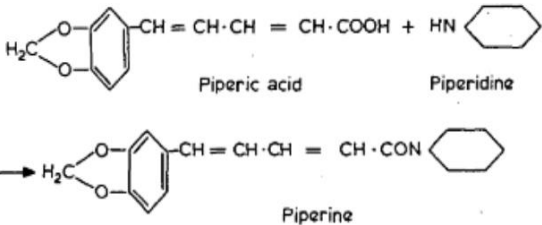
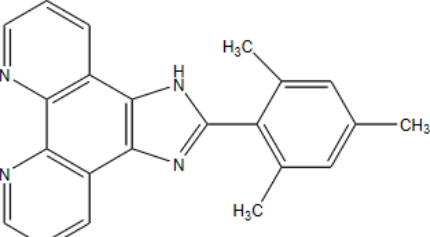
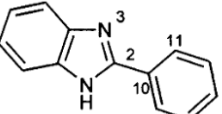
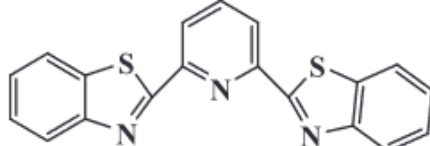
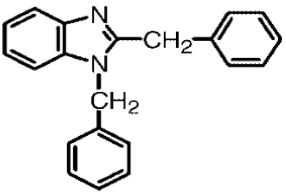
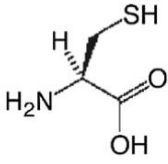
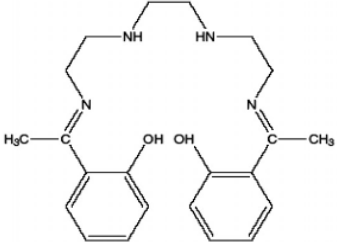
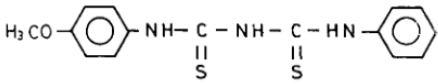
Inhibitor Used	Studied metal/medium	Inhibition Efficiency (%)	References
 1-phenyl-4methylimidazole	Copper/H <sub>2</sub> SO <sub>4</sub>	93	[86]
 1H-Benzotriazole	MS (Mild steel)/1 M HCl	92.6	[87]
 Piperic acid + Piperidine → Piperine	Aluminum/2 N HCl	65.8	[88]
 2-mesityl-1H-imidazo[4,5-f][1,10]phenanthroline	Mild steel/0.5 M H <sub>2</sub> SO <sub>4</sub>	52	[89]
 benzylbenzimidazole	Iron/HCl	95	[90]
 2,6-Bis-(2-benzothiazolyl)pyridine	Mild steel/ 1 M HCl	93-95	[91]
 1,2-Dibenzylbenzimidazole	Mild steel/ 1 M HCl	95-97	[92]

Table 3: Effect of molecular structures on corrosion inhibition. ( )

Inhibitor Used	Studied metal/medium	Inhibition Efficiency (%)	References
 L-Cysteine (Cys)	Copper/ 1 M HNO <sub>3</sub>	61	[93]
 2,2'-((2E,12E)-3,6,9,12-tetraazatetradeca-2,12-diene-2,13-diyl)diphenol	Iron/2 M HCl	94	[94]
 1-anisidyl-5-phenyl-2,4-dithiobiuret	MS/ 1,3,5 M HCl	98.6-1M 98.9-3M 98.1-5M	[95]

of the hydrocarbon pendant chain increases, the inhibition efficiency of imidazoline decreases. Imidazoline compounds with a pendant group hydrocarbon length of -C<sub>13</sub>H<sub>27</sub> and -C<sub>15</sub>H<sub>31</sub> outperform QSI with a pendant group hydrocarbon length of -C<sub>17</sub>H<sub>35</sub> [83].

#### Effect of heteroatoms and stereochemistry

Organic compounds with polar groups and heteroatoms like O-, N-, S-, and/or  $\pi$ -electrons are more effective inhibitors. Since complexes can form between the free or  $\pi$ -electrons of the organic inhibitor, typically nitrogen, sulfur, or oxygen, and an empty d-orbital of the metal, these compounds can be adsorbed on a surface [84]. As a result, a heteroatom with a lower electronegativity value can form strong bonds (charge sharing) with a metallic surface than a heteroatom with a higher electronegativity value. Almost all corrosion inhibitors feature one or more heteroatoms in their molecular structures, especially in polar functional groups, which improves their adsorption on metallic surfaces [60].

Furthermore, organic compounds with -CN and -NO<sub>2</sub> substituents are electron withdrawing in nature, and their -R- and/or -I-effects are likely to reduce the inhibitory efficacy of organic inhibitors. -CN and -NO<sub>2</sub> substituents, on the other

hand, boost the inhibitory efficiency of organic inhibitors, whether they are too big or too small [85].

Table 3 displays the impact of molecular structure of different inhibitors used on different metals and mediums.

#### CONCLUSIONS

Metals and their alloys are used in various industrial applications and daily life. Degradation or corrosion of metals leads to heavy industrial loss and economic loss. The prevention of metal corrosion is vital to avoid these losses. Corrosion inhibitors are one such method employed to prevent metal corrosion. The green corrosion inhibitors replace the synthetic inhibitors those are known to be toxic, harmful to the environment and expensive. Although the green inhibitors are biodegradable and eco-friendly, they also display some drawbacks including the time taken for the production of the extract, various stages involved and the cost. Other inhibitors, such as drugs and ionic liquids also involve limitations of high cost and multistage manufacturing. There is a good deal of study left on green corrosion inhibitors yet to be determined.

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