Eco-friendly Green Corrosion inhibitors in Chloride Contaminated Natural Sea-Water: A review

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Abstract:
Generally, the metallic equipment is fabricated from copper metal due to its enormous application in various industries. Nevertheless, many applications of copper are confined just because it communicates with aggressive chloride ion present in seawater. The behavior of chloride ion accelerates corrosivity, for example, crevice and pitting corrosion initiation, and their circulation on metal surface and lessen the efficacy of the metal by reducing its life span. Notwithstanding, this review paper explicitly examines eco-friendly corrosion inhibitors (CI) in chloride contaminated Natural sea-water for the security of ‘copper metal’ from corrosion. In this review article, the emphasis has been given to green inhibitors owing to better protection ability, effectiveness, and eco-friendliness. The literature furnishes the recognition of the different behavior of green inhibitors and their execution in chloride contaminated marine environment with their polarization efficiencies.

Keywords: Natural Sea water, Copper metal, Green Corrosion Inhibitors, Corrosive Environment.

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1. Introduction

Copper is highly considerable metal in marine equipment due to its corrosion resistivity in neutral environment and high mechanical, electrical & thermal conductivity. However, when it approaches to high chloride contaminated medium (natural sea water) its surface starts getting degraded due to pitting corrosion, in such
conditions protection of copper is only the key. Copper metal has outstanding application in various industries as shown in Fig. 1. However, the natural sea-water is extremely competitive and aggressive medium to all metals including copper [1]. Such hostile medium can cause critical corrosion issues to the copper metal structures within a very short period of time and then the corrosion commodities (corrosion products) could pollute the neighboring atmosphere (air, seawater, living content, etc.) [2]. It is possible to identify those constituents, which describe the type and rate of corrosion attack on metal can be physical, chemical, and biological. Some notable parameters, where the metal is exposed i.e. water temperature, dissolved oxygen, salinity, flow rate, pH, and biological activity (bio-deposits) and environmental zone, establishes seawater corrosivity [2, 3]. The ‘dissolved oxygen’ acts as an oxidizing-agent, and is an incredibly necessary part of the corrosion process, and its transport takes place by diffusion to the outside of the metal, which is a real step in the kinetics of corrosion. The concentration of oxygen in very cold waters can be up to 11 ppm high [7]. The deposition of oxygen on the metal surface can create crevice corrosion at the localized places. The consequence of the amount of oxygen on metal depends on the nature of metal for example, on the copper surface, removal of dissolved oxygen may reduce the corrosion to a good extent [4]. Oxygen accumulation on the metal surface may produce corrosion of the crevice at localized locations [5, 6].

**pH of saline water and its effect on copper corrosion.** The pH of the seawater is noticed between 7.5 to 8.3 at the open surface, which generally depends upon the concentration of CO2 dissolved in seawater. The level of pH alters due to the photosynthesis process of plants and the respiration process of microorganisms. The biochemical oxidation process of microorganisms declines the level of pH and increases the acidity level which ultimately leads to an increase in the level of CO2 gas in seawater [7, 8]. Practically, periodic fluctuation in pH does not affect the behavior of copper and other metal corrosion. Although, the deposition of incrustations on metal surface can be affected by pH fluctuation, which directly or indirectly influence the rate of corrosion [1]. The seawater flow velocity degrades the quality of metal. In the case of copper metal, the flow velocity has a critical velocity beyond which the excessive corrosion of copper occurs. However, when the speed of natural seawater is equal or less one meter per second then copper performs aptly without any notable change in the corrosion rate [7]. Besides, the high flow rate of seawater causes severe corrosion such as cavitation on the metal surface. The zones of seawater to which metal surface is exposed can be divided into several parts i.e. a) atmospheric, b) splash/spray, c) tidal (low and high), d) full immersion or submerged (deep ocean and shallow water) and e) mud-zone as shown in Fig. 2. The exposure of copper metal to these areas determines the variety of corrosion and attack. At the time of tides, splash zone and tidal are those areas that affect copper corrosion severely [9].

The coastal areas of the seawater are accounting for the main corrosion issues in metallic bodies due to the excessive economic industrial and human movements. The main substances, such as nitrates, nitrites, sulfates, ammonia, phosphate, and sulfurs are released by industries and enhance the pollutant's concentration in the seawater which ultimately becomes the purpose of copper metal degradation [10].
Fig. 1. Presents a schematic illustration of a varieties of industries based on copper exhibiting high vulnerability towards corrosive attacks

Fig. 2 Various marine regions around the metallic body of the harbor structure.
1.1 Salinity of Seawater

The salinity of seawater frequently fluctuates from 32.1 to 37.1 g/kg (~ 3.5 %), and usually, this sort of alteration does not change the rate of metal corrosion [8]. The dissolution of sodium chloride salt (NaCl) is observed to be 90 % in the seawater along with Mg, K and Ca. The salinity alters according to the evaporation intensity of seawater or the addition of freshwater arriving from rivers. The behavior of chloride ion accelerates corrosivity, for example, crevice and pitting corrosion initiation, and their circulation in metals [11]. From the thermodynamic subject, the increase in the temperature of seawater normally stimulates the reaction of ‘electrochemical-corrosion’ on metals, this happens when some other parameters are constant. For example, the increase in temperature of seawater decreases the solubility of oxygen, and enhances the biological activity. The precipitation of magnesium hydroxide (Mg(OH)₂) and calcium carbonate (CaCO₃) accumulated on the exterior of the metal works as a shielding mechanical barrier [12]. The copper metal exhibits different natures of corrosion with respect to the change in temperature of the seawater, especially in the month of summer. In addition, it encourages the formation of a calcareous coating on the metal surface, which helps to mitigate the rate of corrosion. In this method, the general inference is that the rise in water-temperature upsurges the rate of copper corrosion, however, the rate of corrosion decreases at temperatures above 60˚ C due to the consequence of the drop in dissolved-oxygen levels. The corrosion of copper and its alloys in marine areas from north to south is particularly susceptible to the temperature of seawater. However, water heating contributes to the release of gases, especially oxygen (O), during this month [8].

1.2 Composition of Seawater

Seawater occupies over 70 % of the surface of the earth and known as the most available natural electrolyte. Corrosion phenomena destroy most of the metallic formations in the sea-water [13]. Sea-water is the predominant corrosive element in the case of underwater corrosion, since it is an aggressive solution of vast concentrations of ionic salts [14]. As an outcome, the extra existence of additional dissolved or suspended aggressive elements, diffused gases, existing organics, or solutes increases corrosivity. Some ions, such as Na⁺, Cl⁻, Ca²⁺, SO₄²⁻, Mg²⁺, K⁺, Br⁻, HCO₃⁻, F⁻, Sr²⁺, B³⁺ and dissolved gases, such as O₂ and CO₂, are typically the main component of the seawater solution, making it the metallic body's hostile medium [11, 15]. Compared to other ions, sodium and chloride ions are present at greater concentrations [16]. The salinity of one kg of natural saline water is defined as the mass (gm) of dehydrated salt [17]. The salinity of the seawater is around 35.15 gm/kg. To put it in a different way, sea-water contains about 3.5 percent of dissolved salt with respect to mass. The list of ions existing in seawater is extremely large. Notwithstanding, they exist in little concentrations. Table 1 lists out the 11 ionic species which are ample in natural sea-water (salinity 35.157 %, density 1.023 g / cm³ at 25 °C).
Table: 1 Composition of natural and sea water [18]

| S.N. | Ions     | Formula weight | Natural sea water (g/kg) |
|------|----------|----------------|-------------------------|
| 1    | [Cl⁻]   | 35.453         | [19.353]                |
| 2    | [Na⁺]   | 22.9898        | [10.76]                 |
|      | [Mg²⁺]  | 24.312         | [1.294]                 |
| 3    | [SO₄²⁻] | 96.06          | [2.712]                 |
| 4    | [K⁺]    | 39.102         | [0.386]                 |
| 5    | [Ca²⁺]  | 40.08          | [0.413]                 |
| 6    | [Br⁻]   | 79.91          | [0.067]                 |
| 7    | [Sr²⁺]  | 87.62          | [0.008]                 |
| 8    | [HCO₃⁻] | 61.01          | [0.142]                 |
| 9    | [F⁻]    | 19             | [0.001]                 |
| 10   | [H₃BO₃] | 61.83          | [0.026]                 |

[*a to j show formula weight of the corresponding ions]

1.3 Corrosion Inhibitors

Copper has been discovered to be more resistant to the effects of the humid atmosphere and other chemical compounds. [19, 20]. However, it is found that copper is more susceptible to corrosion in competitive media (chloride contaminated media). The presence of passive film on metal in such a harsh environment provides insignificant protection to copper. The mechanism of copper corrosion in chloride-contaminated sea water have been reported in countless literatures [21-23]. Besides, it is necessary to protect copper from insignificant causes which account for its physical deterioration. Since the use of inhibitors defends copper metal under harsh conditions. Inhibitors are widely used to avoid the degradation of the attractive appearance of copper metal in a hostile environment. The ambiguity of inhibiting copper corrosion has mesmerized numerous researchers. As a result, a large number of efficient inhibitors have been evaluated till date [24-27]. Organic inhibitors, as compared to inorganic inhibitors, are more effective among all evaluated inhibitors owing to extraordinary properties such as high-efficiency, cost-effectiveness, and strong adsorption on metal [28]. A Scheme of substantial organic inhibitors has been prepared to show some azoles and their derivatives, as well
as a few schiff bases, amines, and amino acid drug-compounds (Scheme 1) [29, 30]. Inhibitors are classified in three categories as follows:

1.31 Organic inhibitors & Inorganic corrosion inhibitors

The aromatic organic molecules contain electron clouds around the ring, which makes it electron-loaded compound [31, 32]. These inhibitors predominantly include compounds, such as amino acids [33], amines [34, 35], azoles [36] etc. The organic inhibitors contain sulfur (S), phosphorus (P), oxygen (O), nitrogen (N), heteroatom in the molecule enhances the action of inhibition via adsorption on copper surface and updates the organic-molecule against the corrosive attack [22, 32, 37-41]. Besides, In-organic Corrosion Inhibitors such as molyb-dates (MoO₄²⁻), tetra borates (B₄O₇²⁻), chromates (CrO₄) [42, 43] used to lessen the corrosive effect on copper surface, but their utilization has become restricted due to their less stability in hostile media and carcinogenic toxic effects [44]. However, the toxicity of organic inhibitors and inorganic inhibitors has sparked the hunt for green corrosion inhibitors. Nevertheless, organic inhibitors are less toxic as compare to inorganic inhibitors [45].

Scheme 1. Chemical structure of common N-containing heterocyclic inhibitors related to copper corrosion.
1.32 **Green corrosion Inhibitors (GCI)**

In addition, green inhibitors exhibit biodegradable nature and lack of toxicity of heavy metals or other harmful compounds. Moreover, green plant extracts are accessible, widely available, sustainable, eco-friendly and ecologically acceptable. That’s why green inhibitors are being used extensively in place of organic corrosion inhibitors [46-49]. Hazards created by the toxicity of organic and inorganic inhibitors have led to an increase usage of modern GCI [49]. There is various application of GCI in diverse fields as shown in Fig. 3. Most of these naturally occurring inhibitors serves as anti-corrosion agents, and are environmentally safe and harmless when disposed of. Green inhibitors act when used in a small volume to treat the surface of metals or alloys in a corrosive climate. Plant-extracts are considered to be rich-naturally synthesized chemical-compounds. The best source of green inhibitors are natural products since they contain the plurality of ‘heteroatoms’ such as ‘N, O, S, and P’ similar to organic inhibitors. Therefore, these heteroatoms present in plant extract behaves as an active center, assist adsorption on metal surface and adsorb either via physical adsorption process or chemical adsorption process by creating a film that defends the surface and restrain from hostile anions, such as chloride and sulfate etc [50-52]. However, studies on the precise mechanisms of the adsorption process and the recognition of the main substance are still scant. During corrosion, metal ions migrate to the solution in active areas (anodic site) and transfer electrons from the metal. Corrosion can be minimized by slowing down or totally blocking anodic or cathodic reactions, or both. Inhibitors are adsorbed on the metal surface, provide a shield and predominantly interfere with anodic reactions to reduce oxidation or corrosive reactions.[53].

![Fig. 3 Application of green corrosion inhibitors in multiple industries and fields.](image)
1.4 A detailed description on green Inhibitors used in Natural sea-water for copper corrosion

All the green corrosion inhibitors used in natural sea water have been listed in Table 2. Al Kharafi et al [54] studied the performance of ‘nitrogen-containing azolic inhibitor’ for the hindrance of copper corrosion (CC) in contaminated seawater (thio-sulfate and sulfide). The Gibbs-free adsorption energy was estimated to be 49.15 kJ/mole. The inhibition efficiency (ƞ %) of this inhibitor was recorded to be 97% at 20 °C. Furthermore, several green-inhibitors were inscribed for the restrain of copper-corrosion in natural sea-water.

R. Oukhrib et al [55] extensively examined the effectiveness of green "Ziziphus lotus essence" and mentioned the notable reduction in the kinetic process of copper corrosion. The inhibition efficiency (93%) was achieved at the highest conc. of 5 g/L of green inhibitor. He further determined the thermodynamic studies by altering temperature from 20 to 50 °C. Khadija El Mouaden et al [56] declared 89% corrosion efficiency of chitosan (green inhibitor) in sulfide co ing seawater at 8000 ppm follows a physical adsorption process on copper metal. They also found very negligible or zero concentration of the corrosion products after 90 days on the copper surface when chitosan was used as an inhibitor, further, confirmed by FESEM-EDS analysis. R. Oukhrib et al [57] showed the use of flowers extract (stamen and petals) of crocus sativus., L commonly known as Saffron flower’s extract in natural sea water against copper corrosion and found 84% inhibition efficiency at 2000 ppm of inhibitor. The green inhibitor (saffron extract) behaved as cathodic type inhibitor according to the outputs of corrosion tests. B. Sakthi Swaroop [58] reported the extract of neem leaf as green inhibitor against microbial corrosion on copper in natural seawater. He used 3 vol % of inhibitor and stated that the inhibitor’s organic molecules may adsorb on the copper surface and decontaminate the surface of copper through its bactericidal effect. He also observed the stability of inhibitors film up to 3 weeks. In his experiment, he immersed coppers samples with and without neem extract in the seawater solution and carried out corrosion test and morphological test. The presence biofilm and colonies of aerobic bacteria (A. sulfureus) were investigated on copper surface confirmed by FESEM analysis, which leads to the formation of green-blue color water. Rani and Selvara [59] reported about the inhibition efficiency of grape (Vitis vinifera) extract against copper corrosion in sea water. The effect of immersion time, temperature, seed concentration and skin extract have been examined adopting different corrosion test. The adsorption of grape extract on copper obeyed Langmuir adsorption isotherm. S Sulaiman [60] reported the use of ‘Allium cepa’ and showed 46 % inhibition efficiency against copper corrosion inhibitor at 0.6 g/L concentration. The inhibition efficiency of Allium cepa is found to be less effective in case of copper metal in natural sea water. C. Wrubl and E. D. Mor [61] investigated the effectiveness of ‘zinc gluconate’ (ZnGO) inhibitor for the corrosion of copper in natural sea-water by adopting weight loss tests and potentiostat polarization. It was revealed that, in aerated conditions (naturally), 60 % efficiency of inhibition of copper corrosion is attained in the presence of ZnGI inhibitor between 6.10^4 to 8.10^3 mol/L concentrations. The whole action of ZnGI mechanism is cathodic.
The mass loss of copper sample was determined after 28 days of immersion in sea-water with a variety of concentrations of ZnGI inhibitor.
Table 2. shows the detailed information of corrosion inhibitors used in natural sea-water.

| S.N. | Inhibitor group | Inhibitor | Inhibitor Concentration | Solution-Condition | Medium | Adsorption isotherm | Efficiency (%) | Ref |
|------|-----------------|-----------|-------------------------|--------------------|--------|--------------------|----------------|-----|
| 1    | Azolic          | [5-phenyl-1-H tetrazole (PTAH)] | 0.01 mol L⁻¹ | Sulphide and thio sulphate contaminated sea water at 293 K | Natural seawater | Langmuir | 2.17×10⁻⁷ | [54] |
| 2    | Green (Natural inhibitor) | [Ziziphus lotus] | 5 g/L | 293 K and 323 K | Natural seawater | - | 93 | [55] |
| 3    | Green           | [Chitosan] | 800 ppm | 298 K | Sulfide Synthetic seawater | - | 89 | [56] |
| 4    | Green           | [Saffron extract (Crocus sativus, L)] | 2000 ppm | 292 K | Natural seawater | - | 84 | [57] |
| 5    | Green           | [Zadirachta indica leaf extract] | 3 vol % | 298 k | Neutral seawater | - | - | [58] |
| 6    | Green           | [Emblica officinalis (Amla leaves extract)] | 1000 ppm | 303 K | Natural seawater | Langmuir | 79.99 | [62] |
| No. | Green Plant | Concentration | Temperature | Time | Solution | Adsorbent | pH | Immobilization Method | Reference |
|-----|-------------|---------------|-------------|------|----------|-----------|----|----------------------|-----------|
| 7   | *Vitis vinifera* Seed and skin extract | 1000 ppm | 24 h, 298 K | 76.08 | Natural seawater | - | - | Langmuir | [59] |
| 8   | *Allium cepa* | 14 g/L | 39.5 °C | 60 | Natural seawater | - | - | - | [60] |
| 9   | Zinc Gluconate (ZnGl) | 4x10^{-1} to 10^{-2} mol/L | 25 °C | 60 | Natural seawater | - | - | (Weight loss test) | [61] |
2. Conclusions

The study of the corrosion inhibitors under different conditions such as environment form, temperature, inhibitor concentration, contamination, etc., provides an opportunity to select an efficient eco-friendly, and economic green corrosion inhibitor for a particular environment. Most of these naturally occurring inhibitors serve as anti-corrosion agents, and are environmentally safe and harmless when disposed of. Green inhibitors work when applied at extremely low concentrations to treat the surface of metals or alloys in a corrosive environment. Plant extracts are considered to be rich, naturally synthesized chemical-compounds and a good adsorbent due to the presence of plurality of heteroatoms such as N, S, O and P. From the extensive literature review, it is notable that rising the inhibitor concentration in the chloride contaminated solution advances the efficiency of corrosion inhibition. Now a days, the progress of green chemistry and green chemical technology has provided innovative synthetic methods for the synthesis of modern corrosion green inhibitors with increased corrosion inhibition ability.

3. Conflict of interest

The authors have no conflict of interest in this review article.
4. References

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