REVIEW

Improvement corrosion properties of reinforcement concrete by corrosion inhibitors: A brief review

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The penetration and diffusion of chloride ions into reinforced concrete structures is the primary cause of steel reinforcement corrosion; thus, this work focused on demonstrating and reviewing the beneficial effects of green inhibitors and organic inhibitors on chloride-induced corrosion. Another motivation for selecting and testing these types of inhibitors was to investigate their effectiveness in greater depth using a variety of various scientific methodologies and a variety of very advanced laboratory and materials testing procedures. In this method, the chosen eco-friendly inhibitor (so-called green inhibitor) might be compared with at least a less hazardous inorganic inhibitor, which is also cheap and usually effective in mitigating iron corrosion in the given environment.

INTRODUCTION

Corrosion of reinforcing steel in civil infrastructure such as bridges, offshore constructions, and buildings has a substantial impact on their service life. The availability of chloride ions above the critical level in the vicinity of the reinforcement is the primary cause of reinforcing steel corrosion. These hazardous ions come from a variety of sources, including road deicing and airborne sea salts [1]. The strong alkalinity (pH > 12.5) of the concrete pore solution maintains its longevity. This shields the steel bar with a thin film of passive and stable oxide composed of hydrated cement paste products that are strongly adhered to the steel's surface [2]. Corrosion of concrete reinforcement is a significant issue. This process, which is common in coastal areas and harsh settings, is the most common cause of reinforced concrete structure degradation. This hazardous damage is caused by a reduction in the reinforcement section, a loss of adhesion between the metallic material and the concrete, or the formation of fractures in the cement coating that due to the corrosion of the concrete reinforcement the bond strength with concrete is reduced. This has the effect of reducing the load-bearing capacity of the engineering structure. The strong effect of chloride ions in the environment is a significant component that impedes and complicates civil engineers' work and restricts the longevity of concrete constructions. The chloride ions catalyze the dissolution reaction of the protective properties of the passive layer carry by inner layer of Fe_3O_4, resulting in fast disintegration and chipping and cracking [3]. The corrosion process not only reduces the cross-section of rebars but also produces expansive products, which initiate cracking in concrete as show in Figure 1 [4, 5].

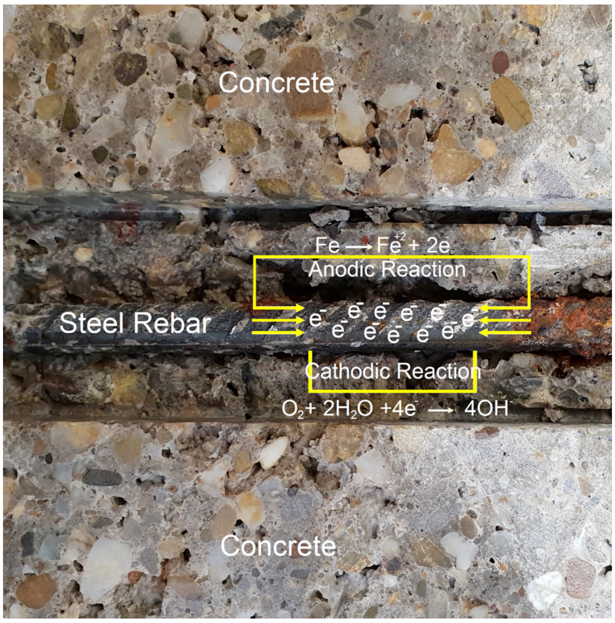


Fig. 1. The corrosion process in reinforced concrete [5]

Corrosion inhibitors have been used to prevent corrosion in reinforced concrete for many decades. They offer adequate corrosion protection even in the presence of large quantities of hostile chloride ions. These active groups either physically/chemically adsorb chloride ions to prevent them from directly accessing the reinforcing steel or chemically interact with the steel surface to generate a protective coating. Nonetheless, the vast majority of conventional corrosion inhibitors on the market are synthetic, which has a severe environmental impact as well as health and safety issues. As a result, numerous researchers have studied the performance and usefulness of green corrosion inhibitors, as well as various methods and types of chemicals [6]. The inhibitors on the market are highly expensive and have an impact on the overall cost of the building. Because of their carcinogenic nature, inorganic inhibitors based on nitrite have been prohibited. It is therefore vital to identify an alternative that may be supplied at low quantities to reduce the cost of concrete structures while also inhibiting chloride-induced corrosion. Organic inhibitors work well. The most effective inhibitors are those with mostly nitrogen, sulfur, or oxygen atoms in their structure. The typical functional groups that allow their attachment to the metal or alloy are amines ($-NH_2$), hydroxyls ($-OH$), or carboxyls ($-COOH$). Organic corrosion inhibitors have been used in the field of concrete materials since the early 1990s. They exhibit the capacity to stop mild steel corrosion in a media that resembles concrete interstitial water in the presence of chloride. The majority of organic commercially available corrosion inhibitors in concrete are amino alcohols, carboxylic acid salts, and amines carboxylates [7]. The addition of corrosion inhibitors to concrete as chemical admixtures during the mixing process would be a simple and cost-effective way to increase the durability of concrete structures. However, calcium nitrite is currently the sole inhibitor that has been shown to be effective in preventing chloride corrosion on real-world structures [6], the mechanism by which this protection occurs can be described as anodic inhibition. Calcium nitrite will prevent pitting corrosion of steel reinforcement in concrete where the Cl^-/NO_2^- concentration ratio in the pore fluid is as high as 1.0-1.5, and Cl^-/OH^- ratios are in the range 0.27-0.57 [8].

CORROSION MECHANISM IN CONCRETE

Corrosion can be characterized by electrochemical reactions, where the anodic (electron-producing) reaction is, for example, iron oxidation. The electrolyte's characteristics determine the cathodic (electron-consuming) reaction. The cathodic reaction for corrosion in concrete involves oxygen reduction. The mentioned reactions for corrosion of steel in concrete are:

$2Fe \rightarrow 2Fe^{2+} + 4e^-$ anodic reaction, oxidation of iron

$O_2 + 2H_2O + 4e^- \rightarrow 4OH^-$ cathodic reaction, oxygen reduction

The reactions take place at separate locations on the steel surface, known as anodic and cathodic regions. The corrosion system is characterized as an electrochemical cell established on a steel surface exposed in concrete. An electrochemical cell is made up of an anodic and cathodic area, an electron conductor (rebar), and an ion conductor (electrolyte). Concrete pore water serves as an electrolyte [9]. The potential pH diagram, also known as a Pourbaix diagram, provides the first indication of corrosion for carbon steel in an aqueous solution with a pH of 0-14. In the absence of dissolved chloride species, the pH level established in the concrete structure (about 13) allows for the formation of a stable passive layer on the surface of the steel [10]. The corrosion rate of steel in concrete is determined by the steel's characteristics, the concrete's qualities, and the environment's properties [9].

Carbonation-induced corrosion

Carbonation is a type of corrosion of reinforcing bars in concrete structures that occurs when CO_2, as a natural gas in the air, penetrates the surface of concrete through the dry portions of interconnected pores in concrete microstructures and reacts with $Ca(OH)_2$ in the moist portions of pores to form $CaCO_3$ and water (H_2O) [11]:

$$CO_2 + Ca(OH)_2 + \rightarrow CaCO_3 + H_2O$$

The remaining CO_2 reacts with C–S–H to produce more $CaCO_3$. Water is required for the carbonation process because CO_2 dissolves in water to generate carbonic acid H_2CO_3 [12]. The adsorption of water and CO_2

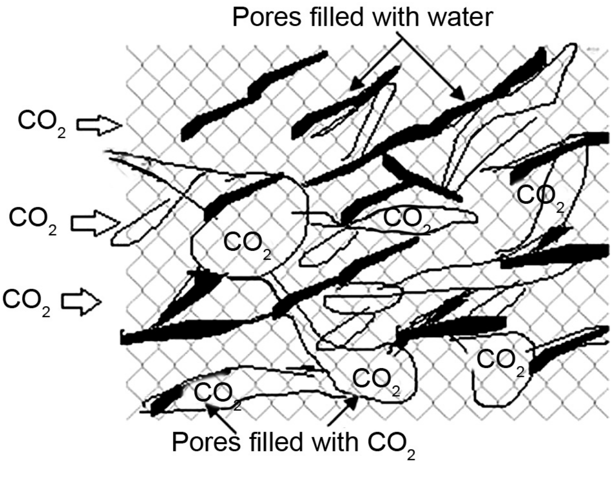


Fig. 2. Carbonation assisted by water and CO_2 adsorption [13]

by concrete is depicted schematically in Figure 2. Water infiltrates through small, linked pores, whereas CO_2 collects in big pores. The linked pores combine to generate huge pores that deliver the water needed to form carbonic acid:

$$CO_2 + H_2O \rightarrow H_2CO_3$$

Because of $Ca(OH)_2$ reduction, the carbonation process lowers the pH of the pore solution to less than 9, resulting in the breakdown of the passive oxide layer that protects the steel bars from corrosion [14]. An oxide film of corrosion products coats metal surfaces, acting as a barrier to further chemical processes. This is the passive layer's primary purpose in reducing corrosion damage [15]. This corrosion is a multistep process that begins at the interface between the steel bars and mortar. Corrosion spreads owing to oxide production, which is mostly dependent on the availability of oxygen and water at the concrete-steel bar infiltration interface [16].

Chloride ingress into concrete

The most common cause of reinforced concrete deterioration is the ingress of chloride-contaminated water, either from deicing salt used for snow and ice control on roads for winter maintenance purposes or from the marine environment, where bridges, for example, span tidal estuaries [17]. Although chlorides have a minor effect on pH, their presence causes depassivation of protective coatings, which leads to pitting corrosion [18]. The diffusion of chlorides from the environment caused by direct exposure to a marine environment or the use of deicing salts and chemicals is the primary source of chloride-induced corrosion in most structures. Chloride transmission into crack-free concrete occurs via four main methods. Capillary action, diffusion due to high concentration on the surface, permeation under pressure, and migration due to electrical potential gradients are some of them [19]. Although the chloride attack mechanism does not directly corrode steel reinforcement, it does breakdown the protective iron oxide film and encourage corrosion. Chlorides do function as corrosion catalysts. The process of chloride diffusion through concrete, however, differs from that of carbonation in that it affects the passive layer without the need for pH lowering [20]. Fig. 3 illustrates the corrosion process brought on by chloride ingress.

CORROSION INHIBITORS

Corrosion inhibitors can be categorized into different classes based on how they prevent corrosion, the environment they are exposed to, the protective layer they are covered in, the sort of structure and composition they have, or even their physicochemical characteristics. The application of inhibitors allows for the speed and intensity of the corrosion process to be limited, while total prevention of corrosion is practically unattainable. Because of ecological limitations, chemicals less hazardous to the environment are routinely used to replace inorganic inhibitors [22]. Corrosion inhibitors' effectiveness is influenced by both their active groups and corrosion mechanisms. For the purpose of preventing chloride ions from directly accessing the reinforcing steel, these active groups either physically or chemically adsorb chloride ions, or they chemically interact with the steel's surface to create a barrier or protective coating [6]. The half-cell reactions that occur during corrosion are anodic and cathodic. Anode and cathode electrodes

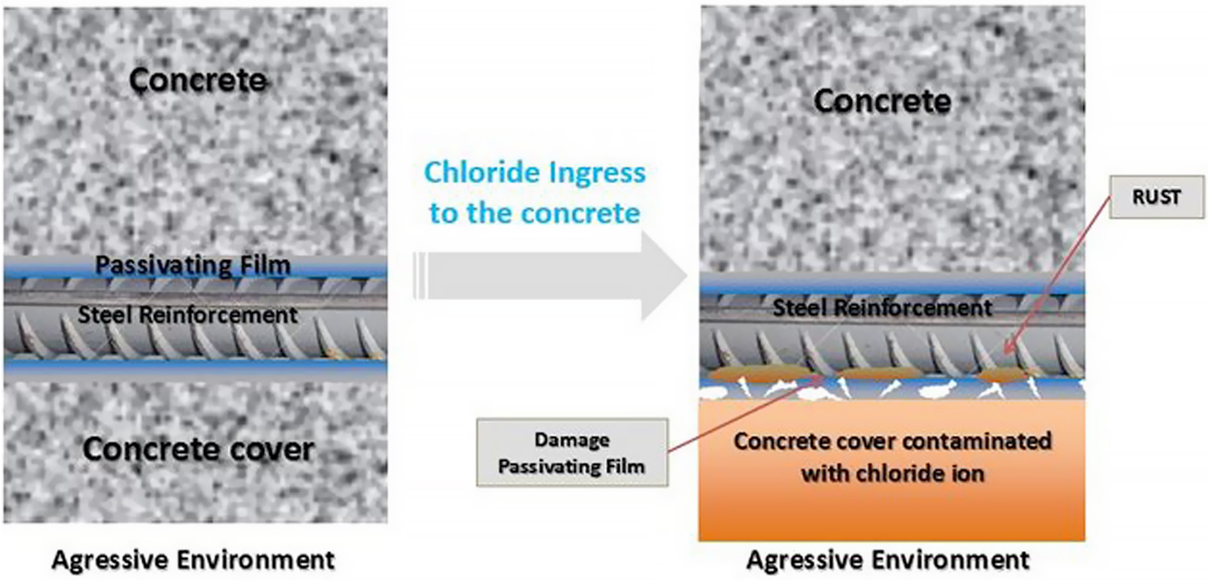


Fig. 3. Corrosion process induced by chloride ingress [21]

connected to steel act as a system during corrosion. as an electrolyte solution in pore-filled concrete sculptures, water [22]. The development of a passive coating or a decrease in corrosion rate following depassivation can both postpone those electrochemical reactions. Organic inhibitors can postpone cathodic, anodic, or combined reactions [22]. For the purpose of defending the integrated steel rebar/concrete construction, corrosion inhibitor technology was created. The chemicals used by these inhibitors create a monomolecular layer between the metal and the water in order to function. The molecule of a film-forming amine has one hydrophilic end and one hydrophobic end. The most efficient forms of inhibitor interact at the anode and cathode simultaneously, and the majority of existing admixtures for migrating inhibitors are based on amino carboxylate chemistry [23]. These molecules will organize themselves perpendicular to the reinforcement and parallel to one another, establishing a barrier [24]. According to laboratory testing, even in the presence of up to 2.20% chlorides, these corrosion inhibitors can move through the concrete to shield the rebar from corrosion [25].

Organic inhibitors

The use of organic inhibitors is one of the many effective corrosion prevention techniques for rebar in concrete that is appealing because of their characteristics, low price, and market accessibility. Organic inhibitors are available in two forms: admixture corrosion inhibitors (ACI), which are added directly to freshly mixed concrete, and migrating corrosion inhibitors (MCI), which are applied to the concrete's surface and then allow for penetration into the concrete. Combining amine carboxylates and amino alcohols, migrating corrosion inhibitors (MCIs) exhibit adaptability as admixtures, surface treatments (coatings), and in rehabilitation initiatives. Following corrosion testing, inspection of the embedded steel rebar revealed no corrosion attack for the MCI-treated concrete samples, whereas untreated concrete had localized corrosion. According to depth profiling and X-ray photoelectron spectroscopy, the inhibitor had arrived at the rebar surface in around 150 days [26]. Even in the presence of chloride ions, the amine-rich compound on the rebar surface enhanced corrosion protection for the MCI-treated steel rebar and stopped the growth of red rust [26].

Migrating corrosion inhibitors (MCI) placed to the surface are useful for mending the chloride-contaminated concrete. Chloride and inhibitor ions in concrete are transported via three mechanisms: natural diffusion, electrical migration, and movement of pore solution due to capillary suction or pressure gradient. Typically, MCIs are based on commercial organic molecules. Aminoalcohols permeate the pores of the concrete quickly and protect the steel rebar by generating a hydrophobic coating by physical or chemical adsorption. This inhibitor also reacts with the cement particles, forming an insoluble substance that blocks the pores of the concrete. Because of their ease of use, low cost, and safety, these inhibitors are an appropriate technique for rehabilitating reinforced concrete structures [27].

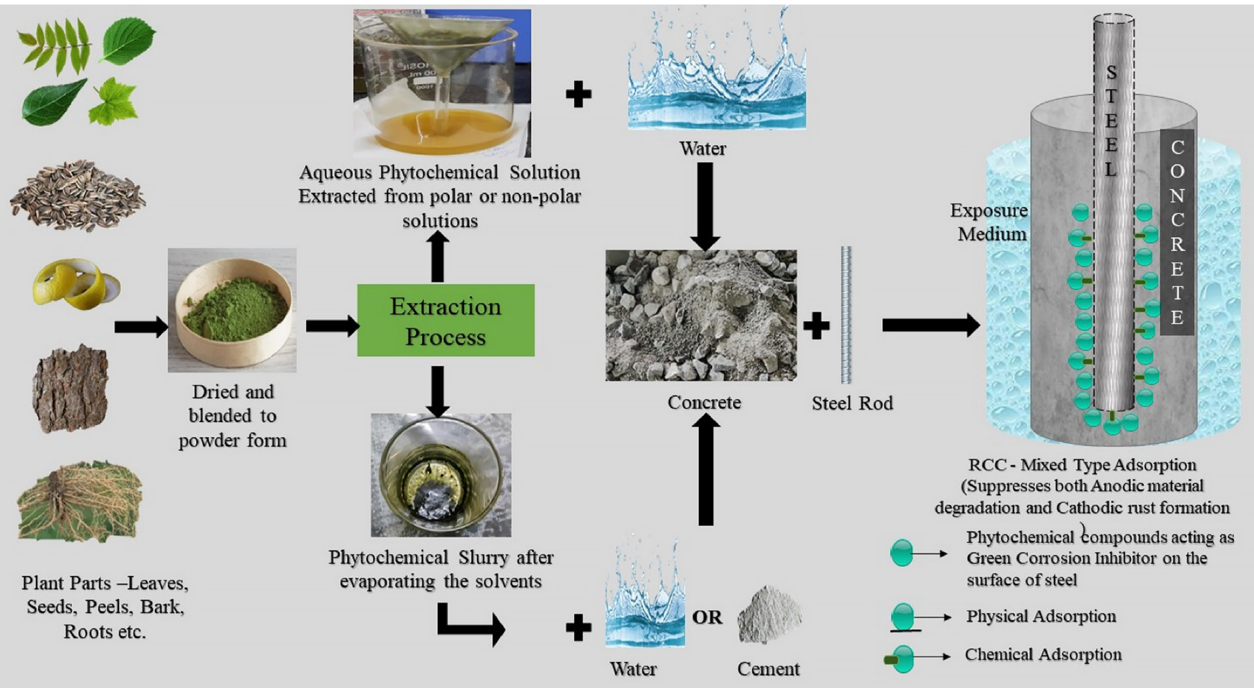


Fig. 4. Concise overview of green corrosion inhibitors in RCC [6]

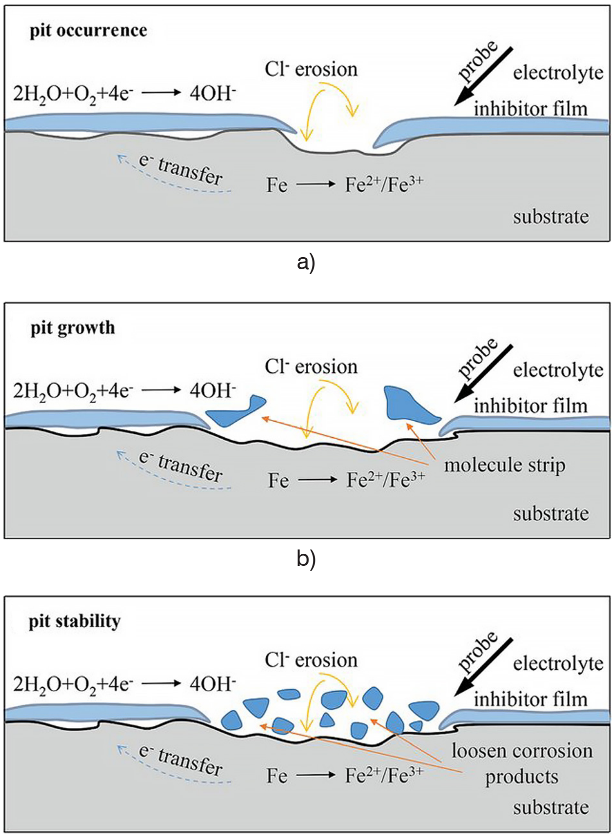
Green corrosion inhibitors

Due to their renewability, accessibility, and relatively low cost, natural materials and plant extracts are frequently employed as corrosion inhibitors since they are environmentally friendly. There is a wide range of green corrosion inhibitors, and many of them might be used in the field as effective and useful corrosion inhibitors. Numerous variables, including the pH of the corrosive environment, the types of aggressive ions present, and the molecular structure of the corrosion inhibitors, all affect how well these materials stop corrosion. The main challenge is determining which active natural product components are responsible for a decrease in the corrosion activity in metals [28]. Chemisorption inhibitors are those that make chemical bonds with the surface of the metal or corroding metal, whether covalent or coordinate. At increasing temperatures, the inhibitor efficiency of this type of inhibitor rises by promoting the formation of chemical bonds. On the surface of metals, these two mechanisms may occur concurrently. Plant extraction is one of the methods often utilized to create green corrosion inhibitors [29]. Because of their easy availability, eco-friendliness, biodegradability, low toxicity, and renewable properties, green corrosion inhibitors such as biopolymers, surface active agents, pharmaceutical compounds, chitosan, honey, yeast, plant extracts, and amino acids, among others, are gaining traction for reducing corrosion in reinforced concrete structures. When these eco-based inhibitors are added as an admixture to the mixing water in the reinforced concrete specimen, the organic compounds present in these inhibitor sources are adsorbed on the metal surface, forming a passivation layer that protects the active area from corrosion action, as illustrated in Fig. 4 [30].

Mechanism of green corrosion inhibitors

Various researchers are studying the green corrosion inhibitor (natural plant extracts) inhibitory mechanism in depth in order to determine its efficiency. Adsorption is a process that occurs on the surface of metals to impede the inhibitory mechanism of plant extracts [31]. Several aspects of the chemicals contained in a certain extract are known to influence adsorption and, thus, inhibitory qualities. According to Ben Harb et al, the inhibition efficiency is expected to be affected by molecular size, carbon chain length, conjugated bonding, aromaticity, the ability of the film to be dense or reticulated, the resistance of the bond to the metal substrate, the number and nature of bonding groups and atoms within a molecule, and an appropriate solubility of phenolic compounds in solvent extraction [32]. Figure 5 depicts a schematic illustration of pit corrosion on carbon steel. Under the influence of Cl ions in Figure 5a, corrosion begins at the exposed area with the loss of an electron from the metal Fe. As a result of the corrosion reaction, the inhibitor molecules in the area were gradually eliminated, as seen in Figure 5b. Although the corrosion reaction was regulated by the “firmly” molecule, which is located distant from the unprotective area, loose corrosion products assemble on the concentrated region. As a result, it showed how well the inhibitor protected against the aggressive solution [33].

In summary, inhibitor efficiency is proportional to inhibitor concentration, whereas corrosion rate is inversely related to inhibitor concentration. The majority of the literature reports that plant extracts obey mixed-type inhibition based on potentiodynamic polarisation studies. SEM photos confirm the formation of a protective coating on the surface of metal. We infer that corrosion prevention is dependent on inhibitor molecule absorption on the metal surface and the use of green inhibitors in an alkaline medium containing chloride ions. Because green corrosion inhibitors are generated from plants, they are both cost effective and environmentally friendly. Green inhibitors, on the other hand, will undoubtedly be employed for corrosion prevention in the future [34]. Plant extraction is one of the most frequent methods for developing green corrosion inhibitors. Liu et al. investigated the effect of ginger



a)

b)

c)

Fig. 5. Illustration mechanism under protection of inhibitor molecule by SVET analysis: a) pit occurrence; b) pit growth; c) pit stability [33]

extract on chloride-induced carbon steel corrosion in a simulated concrete pore solution. They discovered that 2% ginger extract delayed corrosion commencement by producing a carbonaceous organic coating on both the anodic and cathodic surfaces of the steel. Nonetheless, the inhibition efficacy of ginger extract is much lower when compared to standard corrosion inhibitors such as sodium nitrites [29]. Another plant-based substance that provides exceptionally excellent corrosion prevention in basic chloride solutions is methanol extracted from olive leaves. The inhibitor efficacy of olive leaves has been reported to be as high as 91%, which can be attributed to the presence of N, O, and p-electrons in their structure [29]. According to researchers, extracts of many sections of plants have the potential to be used as corrosion inhibitors, and this property is not limited to their leaves. Plant extraction of esfand seed, for example, revealed the production of protective complexes and chelates films on the surface of carbon steel. This generated layer was hydrophilic, which was related to the presence of organic chemicals in the extract such as mercapto, amine, hydroxyl, phosphate, and carboxylic components. After 219 hours of immersion in 3.5% chloride solution, the inhibitor efficiency of esfand at 1000 ppm was more than 93%, which is comparable to other synthetic organic inhibitors [35].

Amines

Amines such as alkanolamine, monoethanolamine (MEA), diethanolamine (DEA), and triethanolamine (TEA) are widely used as corrosion inhibitors because they (for example, alkanolamine) move through the pore structure of the concrete and form a protective film on the surface of the reinforcing steel. Furthermore, it has been observed that these inhibitors can diminish chloride ion migration in concrete and have been classed as mixed corrosion inhibitors by inhibiting anodic and cathodic reactions [36]. Amines and alkanolamines, like ammonia (NH_3), have basic nitrogen atoms in their molecules with a lone electron pair (N:). Nitrogen's lone electrons have an effect on the molecule's form. Because of the lone electron pair, amines have a trigonal pyramide geometrical shape, with electrons filling the fourth corner, similar to ammonia. Amines are organic ammonia (NH_3) derivatives with the generic formula NR3, where R might be hydrogen (H) or an alkyl or aryl group. They, like ammonia, have a basic nitrogen atom with a lone electron pair (N:) in their molecules, which makes amines organic bases and polar compounds. Because of their lone electron pair and polar nature, such compounds are regarded as adsorption reaction centers and excellent inhibitors of active corrosion sites on metal surfaces. Amines are commonly used to prevent rusting [37]. Organic amino inhibitors' inhibition efficacy is still insufficient, and the development of amino-based inhibitors with improved inhibition performance is widely needed. In general, adopting hybrid inhibitors, which are a combination of two or more types of inhibitors, can be an effective way to improve corrosion protection efficiency. Amino alcohols, for example, can be synthesized by combining alkanolamines with amines or, alternatively, with organic acids [38]. The amino-alcohols are frequent inhibitors that are based on alkanolamines and amines or on organic acids [39]. It has been reported that mixed inhibitors based on amino alcohols can be utilized as concrete admixtures or in repair products for existing constructions [40]. However, such compounds are additionally functionalized by various functional groups to provide more effective corrosion protection for steel. Alkanolamines (aminoalcohols) include both amine and alcohol functional groups. Aminoalcohols, like amines, have been claimed to be effective corrosion inhibitors for rebar [41]. Tritthart has investigated the transfer mechanism via the concrete cap. Who established that the amino-alcohol molecule is not bound by cement and remains mostly dissolved in the pore fluids, creating ideal conditions for great mobility. However, according to a separate study conducted by Tritthart. In the case of surface application, he discovered that only a very small amount of aminoalcohol penetrated from mixed inhibitors, implying that penetration is inhibited by some mechanism, such as clogging of the pores by solidification of other compounds of the mixed inhibitor [42].

Carboxylate

When the organic inhibitor is added to the system, the polar compound's hydrophobic group orients itself perpendicular to the metal surface and inhibits aggressive corrosive fluids. Aggregated hydrophobic groups produce a tight barrier that prevents chloride penetration into the concrete. In addition to amines and alcoholamines, compounds containing O, S, or P in their structure are effective corrosion inhibitors in alkali media. Carboxylic acids have polar groups, whereas oxygen has lone electron pairs capable of chemical and physical bonding. They produce carboxylate anions, which can adsorb on the surface of steel. Carboxylates

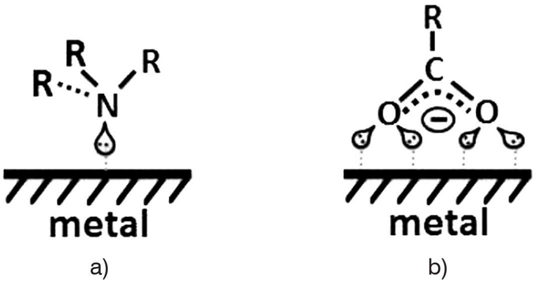


Fig. 6. Scheme of functional groups a) amines, b) carboxylates on the metal surface [43]

adsorb on the metal surface via the ionic form of the carboxylic group ─COO─ group, which is delocalized on the oxygen atoms in the ─COOH group. Figure 6 shows how inductive and resonance effects on the electron density of the molecule influence the adsorption of organic inhibitors on steel [43].

Aminoacids, which have amine and carboxylic groups in their structure, are another class of chemical inhibitors [44]. The ─COOH─ group interacting with the metal surface exhibits adsorption. If the steric effect is large, alkyl chains and other substituents make a good physical barrier against chloride ions. The presence of two or more relatively near carboxylate groups in the molecule improves inhibitive effects. This allows for the complexing of not only ferrous, but also ferric species that compete with chloride ions [45].

CONCLUSIONS

This review provides a complete analysis to establish the efficacy of these green and organic inhibitors in reinforcing steel corrosion protection. It categorizes and ranks a wide range of corrosion inhibitors. Furthermore, it suggests a set of criteria for selecting effective inhibitors based on their properties and sustainability needs. They may operate in locations with heavy chloride contamination. Many factors influence the inhibition efficiency of these materials, including the pH of the corrosive environment, the type of aggressive ions present, and the molecular state of the corrosion inhibitors. Their corrosion protection mechanism is based on the presence of active groups such as carboxylates, amines, and hydroxide in the structure of these green materials. These active groups are physically/chemically or physico-chemically adsorbed on reinforcing steel, forming a protective layer that inhibits cathodic, anodic, or both processes. The new protective coating may repel chloride ions and provide additional corrosion prevention. However, the concentration of these active groups determines the mechanism of protection as well as the inhibitor efficiency. In general, some forms of green corrosion inhibitors have a corrosion protection efficacy of up to 94%, which is comparable to popular synthetic inhibitors. Extracting the functional group from such green organic compounds can increase corrosion protection efficacy by up to 99%. More research is needed, however, to evaluate their technical qualities, such as modeling functional groups for corrosion prevention and their effectiveness against the combined attack of corrosive ions.

CONFLICTS OF INTEREST

The authors certify that they NO affiliations with or involvement in any organization or entity with any financial interest (such as honoraria; educational grants; participation in speakers' bureaus; membership, employment, consultancies, stock ownership, or other equity interest; and expert testimony or patent-licensing arrangements), or non-financial interest (such as personal or professional relationships, affiliations, knowledge or beliefs) in the subject matter or materials discussed in this manuscript.

Data Availability Statement

Statement for Data available on request from the authors: The data that support the findings of this study are available from the corresponding author upon reasonable request."

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