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Green corrosion inhibitors - A sustainable approach to metal protection in aggressive environment

ABSTRACT

Corrosion is the process by which a metal degrades because of chemical or electrochemical interactions with its environment. Acids, alkalis, humidity, pH, temperature, and salts are some of the main elements that contribute to this deterioration of metal and cause corrosion. Acids are frequently used to clean, descale, and pickle metal surfaces. Corrosion is a significant industrial problem since metallic parts lose mass during this process. Plant extracts, polymeric compounds, nanomaterials, inorganic and organic materials, and pharmaceutical molecules have all been used as corrosion inhibitors to inhibit metals from corroding in acidic environments. The study focuses on the adsorption of bioactive compounds such as tannins, flavonoids, and alkaloids onto metal surfaces to provide barriers against corrosive substances; however, a significant barrier still remains that their commercial use is limited by the fact that their inhibitory efficiency decreases with increasing temperature, and their uniformity and effectiveness are also affected by environmental factors that result in plant composition diversity.

Keywords: *Green corrosion inhibitors, Carbon steel, Adsorption mechanism, Acidic environment, Thermal stability*

1. INTRODUCTION

Carbon steel's low cost and suitable mechanical qualities make it a widely utilized engineering material in a variety of sectors, including petrochemical, maritime, and construction. Never theless, a highly acidic environment can cause corrosion [1]. However, in acidic conditions, steel is extremely prone to corrosion, causing enterprises to suffer significant financial losses estimated at billions of dollars each year [2]. Acids are often employed in industrial processes that expose metals to corrosive substances, such as petrochemical operations, acid pickling, industrial cleaning, and petroleum refining [3]. A common industrial acid that corrodes metals through chemical or electrochemical processes is hydrochloric acid [4]. Additionally, The alloy's composition [5] , the harsh environment [6], the product's operating conditions [7], the level of environmental aggression [8], the presence of protective measures [9], and the product's service life [10] as illustrated in figure 1, are just a few of the numerous variables that affect how quickly metals and alloys corrode and also changes in temperature have a significant impact on the development of corrosion processes in metal structures, equipment, and products because they alter the pace of all chemical reactions [11].

The World Corrosion Organization (WCO) estimates that corrosion costs $2.5 trillion USD a year, which it claims, "[reflects] in part many decision-makers in industry and government not fully understanding the consequences of corrosion and how critical it is to control it." The direct as well as the indirect costs have been estimated as part of the many methodologies used to estimate the cost of corrosion. Corrosion-resistant alloys, cathodic protection, corrosion inhibitors, chemical treatments, etc. are all included in the direct cost. However, indirect costs include labor and equipment costs for operations connected to corrosion, revenue loss from production losses [12]. Therefore, a strong drive to reduce/prevent the losses due to corrosion have been widely encouraged over decades ago.

There are essentially two ways to prevent corrosion in materials, particularly steel structures: cathodic protection and the use of corrosion inhibitors. A sacrificial anode or impressed current

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can be used to provide cathodic protection for a metal structure. Making a metal function as a cathode in an electrochemical cell provides cathodic protection. In comparison to the other metals, they are designed to protect, sacrificial anodes are extremely active metals with a greater negative electrochemical potential.

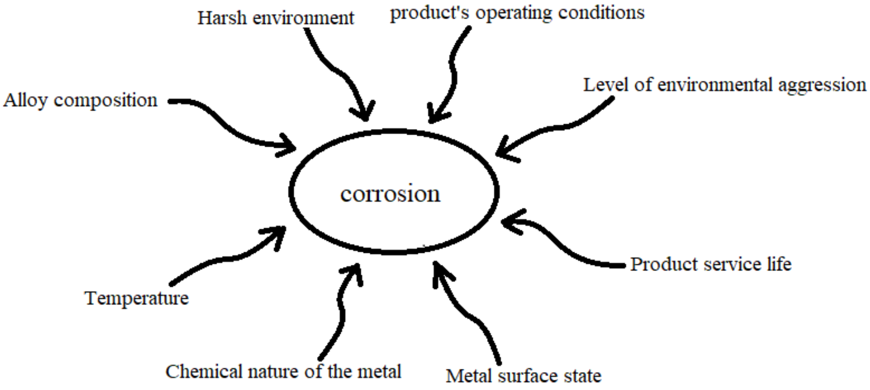

Fig. 1. Some factors affecting the rate of aqueous corrosion of metallic materials

Thus, instead of protecting the metal, sacrificial anodes are consumed. By polarizing the open circuit potential of cathodic sites into the same circuit potential of anodic sites, the entire circuit becomes cathodic in nature, preventing corrosion. This stage results in impressed cathodic protection [13]. Nevertheless, one of the most economical ways to prevent corrosion in carbon steel is to use chemical substances called corrosion inhibitors. In essence, a corrosion inhibitor reduces the rate of corrosion by preventing the metal from reacting with its surroundings. It is usually employed in modest amounts. Inhibitors therefore make it possible to employ inexpensive carbon steel as a structural material. Corrosion inhibitors work by (i) creating a layer that adheres to the metal surface, (ii) generating corrosion products, such as passivator iron sulfide (FeS), and (iii) generating precipitates that can remove or deactivate an aggressive component [14]. These film-forming or interface inhibitors fall into one of three categories: anodic, cathodic, or mixed-type, depending on the electrochemical processes they are blocking [15]. Anodic inhibitors, often referred to as passivation inhibitors, reduce the pace of anodic reactions by forming salts, hydroxides, or oxides that are sparingly soluble at nearly neutral circumstances. Conversely, cathodic inhibitors work by creating a barrier that shields cathodic regions from oxygen in alkaline settings and hydrogen in acidic ones, therefore slowing down cathodic or reduction processes. Mixed inhibitors influence both the anodic and cathodic reaction sites by forming an adsorptive film on the metal surface. About 80% of organic inhibitors fall into this category.Based on the chemical nature of the inhibitors, they can be divided into organic and inorganic [16]. Many of the well-known inhibitors are organic substances with conjugated double or triple bonds that include π-electrons and electronegative functional groups. By donating electrons through their π-orbitals, they consequently exhibit good inhibitory characteristics [17]. Additionally, the metal surface and functional groups with heteroatoms like oxygen, sulfur, nitrogen, and other elements with unbound lone pairs of electrons interact in a certain way that is crucial for inhibition. Increased inhibition is seen when these two characteristics coexist [18]. On the other hand, Inorganic inhibitors are those inhibitors in which the active substance is an inorganic compound. Furthermore, many inorganic inhibitors, particularly those containing phosphate, chromate, and other heavy metals, are currently being gradually restricted or outrightly banned by several environmental regulations due to their toxicity and disposal issues, especially in the marine industry where aquatic life is at risk [19]. Duethese drawbacks which are applicable to both organic and inorganic corrosion inhibitor, researchers have pushed towards finding or using non-toxic or organicgreen corrosion inhibitors that would impart maximum protection to the metallic structures but have least impact on mankind and nature [20, 21]. Green corrosion inhibitors are compounds that are ecologically biocompatible, biodegradable, inexpensive and nontoxic. Typically, they are composed of waste or extracts from natural plants, which are widely accessible in many nations. Most green inhibitor molecules typically have polar functional groups, aromatic rings, multiple bonds, and electronegative atoms like P, N, S, or O. These atoms can work with metal cations to create protective layers on the metallic surface that prevent the development (initiation and/or propagation) of the corrosion process [22][23]. Green inhibitors may be divided into two classes based on their chemical composition: inorganic and organic. Several types of organic green inhibitors include plants (extracts, oil), ionic liquids, amino acids, drugs, and natural polymers. Regarding inorganic inhibitors, the majorities in this group are toxic and cannot be considered as green inhibitors. Nevertheless, there are also some exceptions. Lanthanide salts, for instance, are inorganic rare-based elements with high biodegradability and low toxicity. While inorganic inhibitors work better in neutral environments, organic inhibitors are often better suited to acidic media. Furthermore, organic inhibitors may exhibit both cathodic and anodic behaviour, whereas inorganic inhibitors may only exhibit one of these. In general, the inhibition efficiency of organic green inhibitors is higher than that of inorganic ones. However, the focus is on environmentally friendly corrosion inhibitors that are thermally stable and have a greater variety of uses. Nevertheless, the full use of these environmentally friendly corrosion inhibitors is frequently restricted by the harsh, acidic, and hot conditions [24][25]. i.e. as observed in majority of the reviewed literature there is a consistent trend that the corrosion inhibition efficiency exhibits a negative correlation with temperature, indicating a decline in efficiency as temperature increases.

This review article provides a comprehensive evaluation of various green corrosion inhibitors, focusing on their inherent properties, adsorption mechanisms, and thermal stability at elevated temperatures. This will support environmentally friendly and sustainable production. Additionally addressed are the techniques for investigating corrosion and the inferred adsorption isotherms.

2. CORROSION

Corrosion is defined as the environmental degradation of a material and its intended qualities [26,27,28]. Many industries, including transportation, infrastructure, manufacturing, utilities, and production, may be impacted by this natural process. It can impact a wide range of materials, including wood, aggregates (concrete and composite materials), covalent and ionic compounds, and technical materials (metals, alloys, plastics, paint, and rubber) [29][30]. These corrosive environments include saltwater, saline solutions, alkaline (sodium hydroxide and potassium hydroxide), and acidic (hydrochloride acid, sulfuric acid, and nitric acid) media [31, 32].

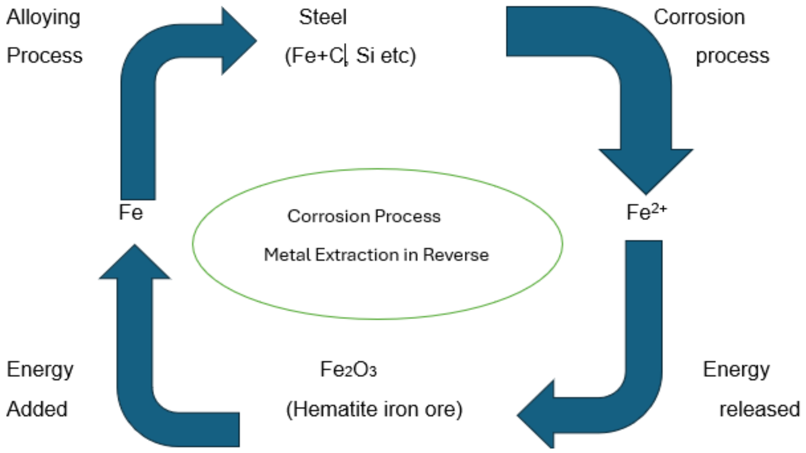

Figure 2. Metal Refining and Corrosion Cycle

In terms of metallic materials such as Fe, [33]&[34] describes corrosion as a spontaneous process that allows a substance to revert to its stable thermodynamic condition as shown in figure 2 above. Majority of these corrosion processes include electrochemical reactions, such as oxidation-reduction processes, and are significantly impacted by operational circumstances.

2.1. Corrosion Mechanism

Corrosion processes are electrochemical in origin and have many characteristics with batteries. Metal atoms can give up electrons to become positively charged ions when they encounter water molecules, so long as an electric circuit can be properly stretched across a large region to create widespread waste. All metals have the propensity to oxidize, albeit some do so more readily than others [35]. These electrochemical cells comprise of five parts namely.

a) Anodic zones
b) Cathodic zones
c) Electrical contacts between these zones
d) An electrolyte
e) A cathodic reactant

Electrons move from the anodic to the cathodic sites inside these electrochemical cells. To balance the flow of electrons, the charged particles, known as ions, travel across the conducting solution. Cathodic reactions cause anions to flow toward the anode, whereas the anode itself causes cations to drift in the opposite direction. The cathode does not corrode as a result, but the anode does. There also exist a voltage/potential difference amidst the anode and cathode. Many distinct micro cells form on the metal surfaces because of the constitutional phase difference caused by differences in stress, coatings, and defect levels (such as dislocations, grain boundaries, and kink sites), or by changes in ionic conductivity or composition in the conducting solution. Every comparable cell reaction is the consequence of two simultaneous anodic and cathodic reactions occurring on the metal's surface at the same speeds. Corrosion is the chemically spontaneous oxidation of the metal upon contact with the cathodic reactant [36].

2.1.1. Anodic Reactions

The metal undergoes corrosion at the anode, and in the presence of an electrical charge differential, the metal oxidizes to its ionic form at the solid-liquid interface. Generally, the anodic reaction is often the ionization of a metal that enters a conducting solution.

$$M \rightarrow M^{n+} + ne^- \quad (2.1)$$

Where ‘n’ is the metallic valence, e^- is the electron, M is the metal and M^{n+} its metal ion.

2.1.2. Cathodic Reactions

The cathodic reactions involve the environment and can represented by the following reactions:

$$R^+ + e^- \rightarrow R^0 \quad (2.2)$$

Where R^+ is the positive ion present in the electrolyte, e^- is the metallic electron and R^0 is the reduced species. Based on the environment, many cathodic reactions and electron consuming reactions are possible. The main reactions are as follows:

I. The anaerobic acidic aqueous environment

$$2H^+ + 2e^- \rightarrow H \quad (2.3)$$

II. The anaerobic alkaline aqueous environment

$$2H_20 + 2e^- \rightarrow H_2 + 2OH^- \quad (2.4)$$

III. The aerobic acidic aqueous environment

$$2O_2 + 4e^- + 4H^+ \rightarrow 4H_20 \quad (2.5)$$

IV. The aerobic alkaline aqueous environment

$$2O_2 + 4e^- + 2H_20 \rightarrow 4OH^- \quad (2.6)$$

2.2. Nature of Plant Extract

The effectiveness of plant extracts in literature suggests that they could potentially replace synthetic organic and inorganic inhibitors. Since the active ingredient's composition determines how green inhibitors work, several researchers have developed a number of theories to explain this phenomenon [37]. Organic molecules that include nitrogen, sulphur, and oxygen have been researched for their ability to lessen corrosion attack. Current research indicates that most organic inhibitors are absorbed by the metal surface by dislodging water molecules and creating a dense barrier layer [38].

The effectiveness of these organic corrosion inhibitors is directly related to the existence of polar functions with heterocyclic compounds, π electrons, and O, S, or N atoms in the molecule. For the adsorption process to be established, the polar function is typically thought of as the reaction center. The necessity to create inexpensive, non-toxic, and environmentally acceptable natural materials as corrosion inhibitors has drawn attention from researchers due to the multiple harmful consequences of the majority of synthetic organic inhibitors and stringent environmental restrictions [39]. However, well-known botanicals, medicinal and culinary herbs, and seed extracts are primarily utilized as potential inhibitors. The presence of the alkaloids erysotrine, erysodine, erythraline, hyponine, erythroidine, and erbydine in the seeds is primarily responsible for the high inhibitory efficacy of these extracts. Because they are readily available and inexpensive, extracts and oils from wild shrubs and plants that contain these alkaloids prove to be superior inhibitors [40]. Although the active ingredients of natural inhibitors differ among plant species, their structures are quite like those of their organic counterparts. Carrots have pyrrolidine in many locations, castor seeds contain the alkaloid ricinine, garlic has allyl propyl disulphide, and mustard seeds contain the alkaloid berberine, which has a lengthy chain of aromatic rings and a N atom in the ring. There is monomtrene-1,8-cineole in eucalyptus oil. Gallic acid, coumarin, sterols, and 2-hydroxy-1,4-naphthoquinone resin and tannin are all present in lawsonia extract. Gum exudate comprises reducing and nonreducing sugars, volatile monoterpenes, canaric and related triterpene acids, neutral sugar residues, and hexuronic acid. Bioflavonoids, unsaturated fatty acids, and primary and secondary amines are all present in garcinia kola seed. Carotene, flavonoids, amino acids, ascorbic acid, and pigments are all present in calyx extract [41].

Furthermore, A vast array of detailed publications about plant extracts as efficient corrosion inhibitors of iron or steel in acidic media have been published as we enter the twenty-first century as illustrated in figure 3, which shows an increasing number of publications which coincide with people's increased awareness of the need to protect the atmosphere.

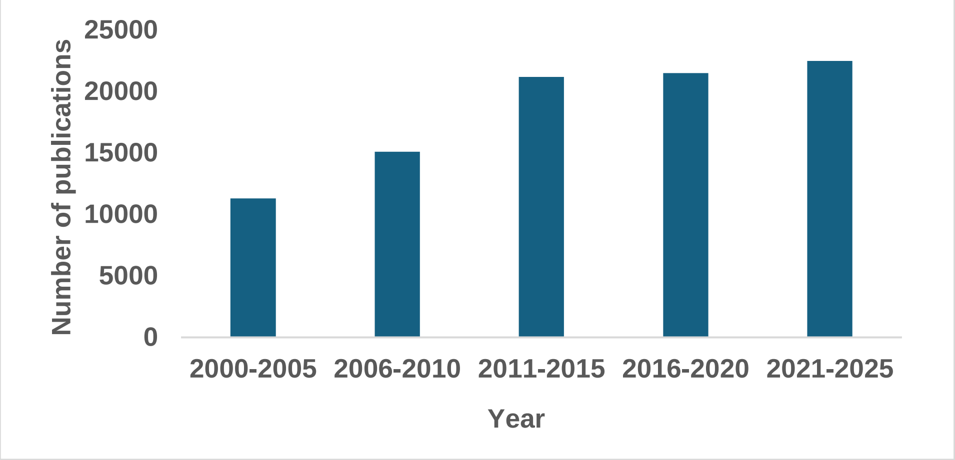

Figure 3 Publications on green corrosion inhibitors

Salvia officinalis, for example, has been shown to exhibit 96% inhibition efficiency at 2500 mg/L [42], At 340 mg/L, Osmanthusfragran exhibited 94% inhibitory efficiency [43], At 300 mg/L, Musa paradisica exhibits 90% inhibitory efficiency [44], At 6000 mg/L, mangrove tannins possess a 89% inhibitory efficiency [45], At 1000mg/L *Jasminum nudiflorum* exhibited 92% inhibitory efficiency [46], At 1200 mg/L, Lawsoniainermis has a 92% inhibitory efficiency [47], At 1000 mg/L, Dendrocalamusbrandisii exhibits 90% inhibitory efficiency [48], At 400 mg/L, aqueous coffee grounds exhibit an 83% inhibitory efficiency [49], At 4000 mg/L, Phyllanthusamarus has an 81% inhibitory efficiency [50], At 1000 mg/L, black radish has a 92% inhibitory efficiency [51] and At 200 mg/L, bamboo leaf has an 89% inhibitory efficiency [52]. All these investigations concur that the existence of complex organic species in plant extracts is frequently attributed to their inhibitory effectiveness.

2.3. Plant Extract Preparation

The preparation of plant extract, following the selection of plant parts, typically commences with the drying process, succeeded by grinding and sieving to convert it into powder form. Except for fruit juice extraction, the first drying step is often required for all plant components. The traditional drying method usually takes a long time to complete and is carried out at room temperature. For example, the bark takes around 20 to 30 days to dry [53][54], and it can be done either in the sun or in the shade. The required extract from plants can be isolated and extracted using a variety of techniques following the drying process. In accordance with the extraction principle, extraction techniques include solvent extraction, distillation, pressing, and sublimation[55]. In brief, extraction methods are based on heating, cooling and separating the active compounds in the presence of the solvent [56]. The most used technique for extracting plants among them is solvent extraction. Figure 4 illustrates the typical solvent extraction techniques that may be applied to the extraction of various plant elements, including the bark, wood, and leaves. The solvent extraction process requires the solvent to permeate plant tissue, dissolve the compounds, and then extract the phytochemicals [57].

Through solvent extraction, variables including temperature, extraction duration, solvent-to-solid ratio, and types of solvents with different polarity may affect the properties of the extract as well as the chemical composition and physical characteristics of the samples [58].For instance, it was discovered that raising the solvent-to-solid ratio improved the phenol yields from the extraction of black currants [59], or from grape pomace [60]. However, optimise value must be considered to balance the cost, solvent wastes and avoidance of saturation effects [61]. In addition, smaller particle sample size also can increase yield for extract [60].

Additional techniques for plant extraction include ultrasound-assisted, enzyme-assisted, and microwave-assisted approaches [62]. In addition, techniques that use compressed fluids as extracting agents, such as subcritical water, supercritical fluids, and pressurized fluids, have also been employed to prepare plant extracts [63]. Supercritical fluids are a novel family of substitute solvents that enable the selective extraction of phytochemicals from plant extracts at optimal processing times and mild temperatures [64]. Table 1 summarises the benefit of these plant extraction methods.

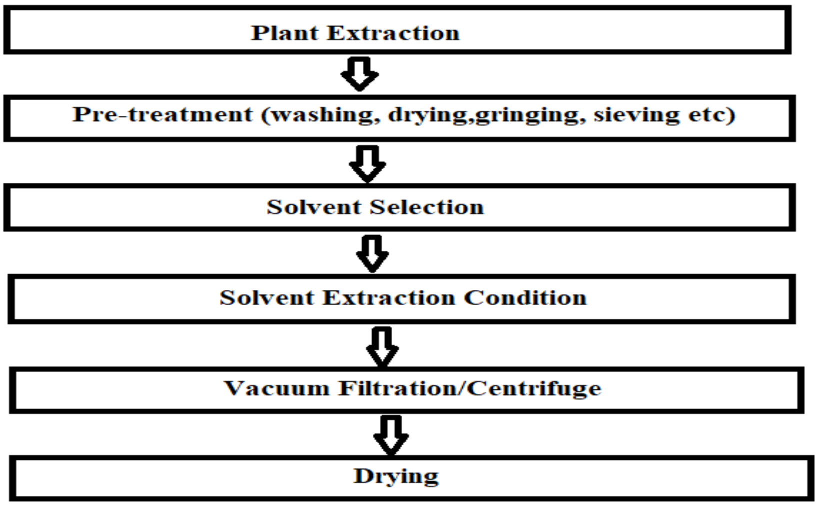

Fig. 4: Flowchart showing the solvent extraction method for plant extraction

Table 1. Advantages of different plant extraction techniques

Methods	Benefits	Ref.
Solvent extraction	Low energy consumption, large production capacity, fast action, easy continuous operation and ease of automation	[65].
Microwave-assisted extraction	By breaking and heating in concert, microwave irradiation can speed up reactions, decrease reaction times, and prevent harm to active ingredients that can happen with conventional, high-temperature heating techniques	[66].
Enzyme-assisted extraction	A new and efficient method for releasing bound chemicals that enables the utilization of the virtually whole plant matrix and, in turn, increases total yield.	[67][68].
Ultrasound-assisted extraction	Ultrasound energy facilitates organic and inorganic compounds leaching from plant matrix.	[69].
Compressed fluids as extracting agents	Permit the phytochemicals to be separated from the extracts selectively at the ideal processing period and at mild temperatures.	[64].

The other factor that needs to be considered for the plant extract preparation is the temperature during the extraction process. According to reports, the temperature range between 60°C and 80°C is ideal for the extraction process since it may produce the best extraction results [70]. The ideal extraction temperature is chosen to ensure the phytochemicals' efficient solubility. Higher temperatures increase solubility and mass transfer rates, which in turn reduce viscosity and enhance solvent mobility in matrices[71]. Furthermore, the ideal temperature for the extraction process must be maintained without breaking down the active ingredients (phytochemicals). The oxidation of phenolic chemicals brought on by high temperatures and extended extraction times might reduce the yield (extract)[71].

2.4. Characterization Techniques

Various characterization techniques have been employed in several research studies to aid in the proper understanding of the nature of leaf extracts as corrosion inhibitors. To determine the functional group present in an extract, the Fourier Transform Infrared Spectroscopy (FTIR) is employed for this purpose. The FTIR is a widely used analytical technique that measures the infrared spectrum of absorption or emission of a solid, liquid, or gas. It provides valuable information about the molecular composition and structure of materials by analyzing their vibrational energy levels[72].

FTIR spectroscopy operates on the principle that molecules absorb specific frequencies of infrared light, which correspond to the vibrational modes of the bonds within the molecules. When infrared radiation passes through a sample, some of the radiation is absorbed while the rest is transmitted. The absorbed radiation causes transitions in the molecular vibrations, leading to a spectrum that serves as a unique “fingerprint” for each substance[73].

Another important characterization technique is the Gas Chromatography-Mass Spectrometry (GC-MS). This is a powerful analytical technique that combines the capabilities of gas chromatography (GC) and mass spectrometry (MS) to identify and quantify active compounds within complex mixtures. This method is widely utilized across various fields, including environmental analysis, forensic science, pharmaceuticals, food safety etc [74]. In the first phase of GC-MS, gas chromatography separates volatile compounds in a sample.

2.5. Green Inhibitor Mechanism

To separate the metallic structure from its surrounding medium and halt the oxidation-reduction process, corrosion inhibition may involve the use of either organic or inorganic chemicals that adsorb on it. Organic inhibitors work by forming a protective layer on the metal or alloy surface as shown in figure 5, by adsorbing their molecules [75]. As an alternative, inorganic inhibitors function as anodic inhibitors and enhance corrosion resistance by enclosing their metallic atoms in a coating.

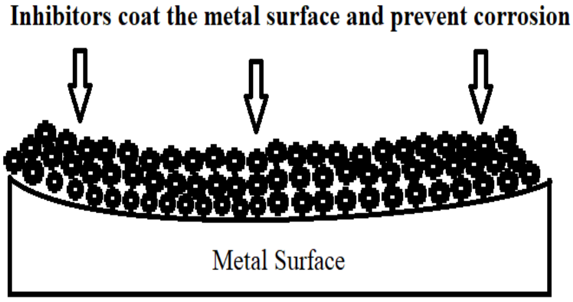

Figure 5. Corrosion inhibitor mechanism

Most corrosion resistance inhibitors that have been studied are hazardous and pose a serious risk to the environment when disposed of. As a result, environmental laws have restricted their usage. Inhibitors are still essential for preventing corrosion, though. Inhibitors are categorized based on:

1. Three types of electrode process, namely, anodic, cathodic and mixed

2. The chemical nature of the environment to acid inhibitors (organic or inorganic), neutral inhibitors, alkaline inhibitors and vapour-phase inhibitors.

To treat the surface of metals or alloys in a corrosive environment, green inhibitors are applied in extremely small amounts. When used into several industrial systems, plant extracts are believed to be a rich, naturally occurring chemical component [75] that influences the rate of corrosion by adsorbing beneficial species on metal surfaces when added to many industrial system through:

1. Changing the rate of anodic and/or cathodic reactions

2. Effecting the diffusion rate of aggressive ions interacting with metallic structures

3. Increasing electrical resistance of the metal surface by forming a film (coat) on it.

By delaying or halting the anodic or cathodic processes, or both, corrosion can be reduced. To lessen the oxidation and/or reduction of corrosive processes, inhibitors engage with anodic and/or cathodic reaction sites after being adsorbed on the metal surface to create a protective barrier [75]. It has been postulated that the inhibitors are adsorbed into the metal surface either by physical (electrostatic) adsorption or chemosorption [76].

However, in determining the corrosion inhibition efficiency of corrosion inhibitors, the corrosion rate of the metal in the corrosive environment can be determined by employing various electrochemical techniques. Electrical Impedance Spectroscopy (EIS) is an electrochemical technique that measures the impedance of a system over a range of frequencies. It provides insights into the electrochemical processes occurring at the electrode interface, making it a valuable tool for studying corrosion, battery performance, and bio-sensing applications [77]. EIS simply operates on the principle of applying a small sinusoidal voltage or current perturbation to an electrochemical system and measuring the resulting current or voltage response. The Nyquist plot is a plot of the imaginary part of impedance (-Im(Z)) versus the real part (Re(Z)). This plot provides information about different resistive and capacitive elements in the system. The impedance (Z) is defined as the ratio of the applied voltage (V) to the measured current (I) [78].

$$z = \frac{V}{I} \tag{2.7}$$

Potentiodynamic polarization (PDP) is a widely used electrochemical technique for studying the corrosion behavior of metals and alloys. This method involves varying the potential of an electrode in a controlled manner while measuring the resulting current, providing valuable insights into corrosion mechanisms and rates[79]. In PDP, the potential of a working electrode is systematically altered over a specified range, typically from a cathodic (negative) to an anodic (positive) direction. This is achieved using a potentiostat, which controls the potential and records the current response[80].

2.6. Adsorption Isotherm Model

In the presence of hostile media, adsorption is the initial stage of creating a corrosion-protective coating or coat on metallic surfaces at active spots. The adsorption process is really the most effective, promising, and frequently utilized basic method for treating wastewater, and corrosion inhibitors work through it to prevent metals from corroding. The quantity adsorbed as a function of temperature and pressure (or concentration) is typically used to empirically characterize the adsorption process. The relationship between the amount of the adsorbed material and its bulk concentration during the adsorption process at a specific pH and temperature is known as the adsorption isotherms. Typically, an isotherm equation that describes the adsorption isotherm and whose parameters represent the adsorbate's surface characteristics and affinity may also be used to infer the mechanism of the adsorption process [81].

2.6.1. Numerous isotherms have been used in adsorption systems and there including:

1. Brunauer, Emmett, and Teller (BET) model

The Brunauer–Emmett–Teller (BET) hypothesis is the foundation for a crucial analytical method for determining a material's specific surface area and attempts to explain the physical adsorption of gas molecules on a solid surface [82]and can be expressed as in Eq (2.8)

$$\frac{P/P^0}{n\left(1-\frac{P}{P^0}\right)} = \frac{1}{n_m c} + \frac{c-1}{n_m c}\left(\frac{P}{P^0}\right) \qquad (2.8)$$

P/P^0 is the relative pressure, n is the amount adsorbed (per unit mass of adsorbent, say), nm is the BET monolayer capacity, and c is usually related to the net heat of adsorption by c = exp (q^{st}-q_L)/RT, where q^{st} is the isosteric heat of adsorption in the monolayer and q_L is the heat of condensation. Conventionally, adsorption data are plotted as $(P/P^0)/n(1 - P/P^0)$ vs P/P^0, and the nm and c parameters are determined from the slope and intercept of the resulting line [83].

2. Freundlich model

Adsorption activities that take place on heterogonous surfaces can be described by the Freundlich isotherm [84]. The surface heterogeneity, the exponential distribution of active sites, and their energy are defined by the expression provided by this isotherm [85]. The Freundlich isotherm has the following linear shape [86].

$$log\, q_e = log\, K_f + \frac{1}{n} log\, C_g \qquad (2.9)$$

where K_F is adsorption capacity (L/mg) and $1/n$ is adsorption intensity; it also indicates the relative distribution of the energy and the heterogeneity of the adsorbate sites.

3. Sips model

Sips isotherm is a combination of the Langmuir and Freundlich isotherms. This model may be used to forecast adsorption on heterogeneous surfaces without the drawback of the Freundlich model's tendency to increase adsorbate concentration [87]. Consequently, this model predicts the Langmuir model (monolayer adsorption) at high adsorbate concentrations while reducing to the Freundlich model at low adsorbate concentrations [88] and it is given the following general expression [89].

$$q_e = \frac{k_S C_e^{\beta s}}{1 - a_S C_e^{\beta s}} \qquad (2.10)$$

where K_S is Sips isotherm model constant (Lg^{-1}), β_S is Sips isotherm exponent, and a_S is Sips isotherm model constant (Lg^{-1}).

4. Temkin model

The Temkin isotherm model assumes that the heat of adsorption (ΔH_{ads}) of all molecules in the layer decreases linearly as surface coverage increases, and it also considers the impact of indirect adsorbate/adsorbate interactions on the adsorption process [90]. The Temkin isotherm is valid only for an intermediate range of ion concentrations [91]. The linear form of Temkin isotherm model is given by the following [92]:

$$q_e = \frac{Rt}{b} In\, K_T + \frac{RT}{b} In\, C_e \qquad (2.11)$$

where b is Temkin constant which is related to the heat of sorption (Jmol−1) and K_T is Temkin isotherm constant (Lg−1) [93].

5. Langmuir model

The adsorptive capacity of different adsorbents is measured and compared using Langmuir adsorption, which was initially developed to characterize gas-solid phase adsorption [94]. The Langmuir isotherm balances the relative rates of adsorption and desorption (dynamic equilibrium) to account for surface coverage. The percentage of the adsorbent surface that is open determines adsorption, whereas the percentage of the adsorbent surface that is covered determines desorption [95]. The Langmuir equation can be written in the following linear form [96]:

$$\frac{C_e}{q_e} = \frac{1}{q_m k_L} + \frac{C_e}{q_m} \qquad (2.12)$$

where C_e is concentration of adsorbate at equilibrium ($mg\,g^{-1}$). K_L is Langmuir constant related to adsorption capacity ($mg\,g^{-1}$), which can be correlated with the variation of the suitable area and porosity of the adsorbent which implies that large surface area and pore volume will result in higher adsorption capacity

The adsorption mechanism, inhibitor's chemical and electrical properties, temperature, the kind of electrolyte used, steric effects, and the composition and surface charge of metals are some of the variables that influence the inhibitor's adsorption on and isolation from the metallic surface [97]. The most advantageous method for elucidating the interaction between the inhibitor and the metal surface is the Langmuir adsorption isotherm [98]. Adsorption—which can be either physical (physisorption), chemical (chemisorption), or a combined adsorption mechanism—approaches steady-state adsorption on the eroded surfaces and is thought to be the most efficient way to prevent corrosion.

Physical adsorption is related to the standard free energy of adsorption ΔG^0_{ads} in aqueous solution. if its value is -20 kJ.mol-1 an electrostatic interaction between the charged centers of molecules and the charged metal surface are linked to a dipole interaction between the molecules and the metal surface[99]. On the other hand, chemical adsorption is a process that creates a coordinating covalent bond by transferring or exchanging electrons from the inhibitor to the metallic surface. Compared to physical adsorption, the bonding strength is substantially higher where the value of ΔG^0_{ads} substantially higher around -40kJ.mol^{-1}or lower [100][101]. Green corrosion inhibitor adsorption slows down corrosion by reducing the amount of active metallic surface area, exposing inactive areas to corrosive substances. In most situations, the efficiency of inhibition decreases as the temperature rises, whereas green inhibitors are more effective at room temperature or low temperatures.

3.1. Overview of Previous Studies on Green Inhibitors for Mild Steel Corrosion in Acidic Environment

The use of green corrosion inhibitors to preserve mild steel in acidic environments has been the subject of a substantial amount of study throughout the past two decades. These research mostly assess how well different plant extracts, algae, biopolymers, and ionic liquids block corrosive environments like phosphoric acid (H_3PO_4), sulfuric acid (H_2SO_4), and hydrochloric acid (HCl) [102][13]. The bioactive components of these green inhibitors such as tannins, flavonoids, alkaloids, and polyphenols adsorb onto the metal surface and lower corrosion rates, which is primarily responsible for their efficacy. Researchers have also looked at how corrosion inhibition performance is affected by factors such inhibitor concentration, temperature, exposure time, and acid molarity[103]. To provide a comparative insight, Table 2 summarizes notable studies in this field, highlighting the type of green inhibitor used, experimental conditions, maximum inhibition efficiencies achieved, and the key findings derived from each study.

Table 2. Overview of previous studies on green corrosion inhibitors for mild steel in acidic environments

Green Inhibitor Used	The corrosive environment the material is exposed to	Max. Inhibition Efficiency/Conc. Of Extract	Temperature of Exposure and Duration	What to Deduce from the Study	Ref.
Pomegranate Arils Extract (PAE)	1Mol. HCL solution	74%/400mgl^{-1} 52.13%/400mgl^{-1}	298K 313K 333K	The negative impact of temperature on the inhibition efficiency of pomegranate Arils Extract (PAE) limits its use as a satisfactory inhibitor for mild steel in HCL media at elevated temperatures	[104]
MondialWhittei roots Extract(MWE)	0.5Mol. HCL solution	89.47%/1.0g/L	60°C/9h	The thermodynamics and kinetic studies disclosed that the inhibition was exothermic with pseudo-second order kinetics.	[105]
Lemon Balm (LB) extract	1M HCL	95%/ 800 ppm	280-320K/ 0.5,2,4,6,12 and 24h	The LB extract showed mixed corrosion inhibition and significantly retarded the mild steel dissolution rate in HCl solution	[106]
Chinese gooseberry fruit shell extract	1M HCL	94%/ 1000ppm	25ºC/ 2.5 and 5h	The Chinese gooseberry fruit shell extract behaved as a mixed type of inhibitor and controlled the corrosion rate of both anodic and cathodic reactions with a little cathodic prevalence	[107]
Sunflower seed hull extract	1M HCL	98.46%/ 400ppm	298-333K /72h	Physical adsorption was discovered to occur close to the chemical on the metal surface. Since this process is exothermic, additional adsorption does not occur when the temperature rises.	[108]
Sidaacuta (Malvaceae)	0.7M, 1.2M and 2.2M HCL	71.16%/ 15g/l, 30g/l and 45g/l	298K 318K 338K 358K 8hr	The crushed Sidaacuta leaves reduce mild steel corrosion in hydrochloric acid according to all four evaluated adsorption isotherm models, with the Langmuir isotherm retaining the best match.	[109]

Voacanga Africana	0.7M, 1.2M and 2.2M	88.69%/ 15g/l, 30g/l and 45g/l	298K 318K 338K 358K 8hr	The corrosion of mild steel was decreased by adding finely crushed Voacanga Africana leaves; the rate of corrosion was shown to rise as the acid concentration rose, while the effectiveness of inhibition increased with time.	[110]
Dacryodisedulis (DE)	1 M HCl and 0.5M H_2SO_4	79.1%/ 800mg/l	303 – 333 k/3h	The extract was found to inhibit corrosion of low carbon steel more in 0.5M H2SO4 at 800mg/l	[111]
Coriandrum sativum *L.*	1M H_3PO_4	72.75%/ 500ppm	30 -50°C/ 3h	CSE acts as mixed inhibitor by inhibiting both cathodic and anodic reactions to same extent	[112]
Laurus nobilis L. oil	3% NaCl solution	For pure aluminium 90.2% and 80.4% for AA5754/ 10-50 ppm	25°C/24hrs	Laurus nobilis L. oil inhibits the pitting corrosion of pure aluminium more effectively than the corrosion process observed on AA5754 alloy in the same range of concentrations	[113]
Neolamarckia cadamba crude extract (bark, leaves) and pure alkaloid (3β-isodihydro-cadambine)	1M HCl	Bark extract- 91% in 5mg/L Leave extract – 88% in 5mg/L Pure alkaloid – 89% in 5mg/L	25°c/30 min	As corrosion inhibitors, the results demonstrated that all the green inhibitors performed exceptionally well (greater than 80% at 5 mg L1).	[114]
Thyme leaves extract	2M HCl	84%/ 400mg/L	25°C-50°C/ 7 days	The inhibition efficiency at higher temperatures increased with higher inhibitor concentrations and the Thyme leaves extract acted as a mixed type of inhibitor on the metal surface	[115]
Task-specific ionic liquid	1 M HCl solution	78.7%/100 mg/L of TSIL	298k/2hrs	TSIL demonstrated comparatively high corrosion prevention capabilities for low carbon steel in a 1 M HCl solution. As the concentration of TSIL rose, so did the efficacy of inhibition.	[116], [117]
Spirulina platensis	1 M HCl and 1 M H_2SO_4	82.65%/500ppm	303 K /2hrs	The inhibition efficiency is better in H2SO4 medium than in HCl medium	[118]
Salvia aucherimesatlantica	0.5M H_2SO_4	86.12%/ 2 g/L	303 to 343 K/6hrs	Salvia aucherimesatlantica oil mainly acts as good inhibitor for the corrosion of steel in 0.5 M H_2SO_4	[119]
Cassia bark extract	0.5 M H_2SO_4 solution	88.02%/500ppm	298k/8hrs	In a 0.5 M H2SO4 solution, curcumin extracts prevent C-steel from corroding. As the concentration of the extract grows, so does the extracts' inhibitory effectiveness.	[120]
Chitosan	0.1 M HCl	92.1 %/ 1.8 mM	298K/24hrs	It was discovered that chitosan effectively inhibited mild steel corrosion in a 0.1 M hydrochloric acid solution. The findings showed that mild steel's weight loss and corrosion rate were successfully reduced by raising the chitosan content from 0.3 to 1.8 mM.	[121]
Dryopteriscochleata leaves extracts	1M H_2SO_4	95.09%/ 2400ppm	298k/6hrs	In a 1 M H2SO4 solution, methanol and water extracts of D. cochleata leaves suppress the corrosion of aluminum. As inhibitor concentration rises, inhibition effectiveness rises as well, but as temperature rises, it falls.	[122]

Shahsavari et al., (2022)[104] reported the effect of concentration and temperature on pomegranate Arils Extract (PAE) as a green corrosion inhibitor for mild steel in hydrochloric acid (HCL). During the experiment they discovered that the increase in temperature from 298-333K with $400mgl^{-1}$ of PAE increases the corrosion current density from 0.06 to $1.28mAcm^{-2}$ resulting to a negative inhibition efficiency at 333K. Synthesis of green corrosion inhibitor for mild steel in acidic environment as investigated by Ogunleye et al., (2020) [105]proved that at a fixed concentration of inhibitor of 0.5g/L, the corrosion rate (CR) increases from 1.6881-5.3187g/m^2h with corresponding temperature of 30°C and 45°C respectively. However, it was detected that the inhibitor efficiency increases as temperature increases from 303-333K and maximum inhibitor efficiency of 89.47% was achieved using 1g/L of Mondia Whittie roots Extracts (MWE). Asadi et al. (2019)[106] investigated the effect of Utilizing Lemon Balm extract as an effective green corrosion inhibitor for mild steel in 1M HCl solution: A detailed experimental, molecular dynamics, Monte Carlo and quantum mechanics' study. During the investigation it was discovered that that the LB extract had a maximum inhibition of 95% even after 24hr immersion.

Furthermore, it is interesting to note that comparing the results from the experiment with other published works on using synthetic and/or green corrosion inhibitors for carbon steel protection, the LB extract provided higher inhibition efficiency even at long exposure times. In examining A detailed electrochemical/theoretical exploration of the aqueous Chinese gooseberry fruit shell extract as a green and cheap corrosion inhibitor for mild steel in acidic solution, Dehghani et al., (2019) [107]discovered that from the SEM and AFM analyses that an increase in the concentration of the inhibitor led to the formation of a protective layer on the mild steel surface, which inhibited the surface from corrosive attacks. Sunflower seed hull extract as a novel green corrosion inhibitor for mild steel in HCl solution as examined by Hassannejad&Nouri et al., (2018), It was determined that the inhibitory property existed because of the reduction in charge and ion transfer on the metal surface brought about by the complex formation between the inhibitor and the metal surface ions. In Modelling of Corrosion Inhibition of Mild Steel in Hydrochloric Acid by Crushed Leaves of SidaAcuta (Malvaceae), Ndukwe et al., (2017a) discovered that the surface deterioration of mild steel in an uncontrolled 0.7M HCl solution is confined, as the SEM picture demonstrates. On the other hand, the addition of crushed SidaAcuta leaves notably stopped the steel's surface from corroding. Furthermore, the corrosion rate was observed to increase with increase in acid concentration. Ndukwe et al., (2017) reported the Modelling of Corrosion Inhibition of Mild Steel in Sulphuric Acid by thoroughly Crushed Leaves of Voacanga Africana (Apocynaceae). During the investigation it was discovered that Alkanoid, flavonoid, phytate, tannin, and saponin were all present in Voacanga Africana leaves, according to a phytochemical investigation which contributed to the inhibition efficiency. Furthermore, the mild steel corrosion was decreased by adding thoroughly crushed Voacanga Africana leaves. While the inhibition efficiency increased over time, the corrosion rate was found to increase as the acid concentration rose. In the Adsorption and corrosion-inhibiting effect of Dacryodisedulis extract on low-carbon-steel corrosion in acidic media as studied by Oguzie et al., (2010). They unraveled that according to the impedance results, the extract species' adsorption on the carbon steel surface produced the mixed-inhibition mechanism suggested by polarization measurements. However, between low and high DE concentrations in 1 M HCl, the adsorption behavior, as estimated by the Langmuir isotherm, exhibits clear variations, which we have linked to a transition from physisorption at low concentrations to chemisorption at high enough concentrations. Prabhu et al., (2013) researched on Coriandrum sativum L.—A novel green inhibitor for the corrosion inhibition of aluminum in 1.0 M phosphoric acid solution. During the experiment they discovered that the energy of activation value indicates that the CSE physically adsorbs onto the aluminum surface and the adsorption of CSE on the surface of aluminum follows Langmuir adsorption isotherm. During the examination of Laurus nobilis L. oil as green corrosion inhibitor for aluminum and AA5754 aluminum alloy in 3% NaCl solution by Halambek et al., (2013). They identified that due to the small proportion of iron (0.08%) in aluminum, the weight loss and potentiodynamic polarization measurement reveals that the AA5754 alloy resists corrosion better in a 3% NaCl solution than pure aluminum. Furthermore, they unraveled that these properties were most likely caused by various intermetallic particles, some of which are cathodic in character, especially Al (Mn, Fe, Cr) found on the surface of aluminum alloys. Raja et al., (2013) reported on Neolamarckiacadamba alkaloids as eco-friendly corrosion inhibitors for mild steel in 1 M HCl media. During their experiment they uncovered that the inhibitors inhibit corrosion through adsorption process and were found to follow Langmuir adsorption isotherm. Furthermore, by raising the system's resistance, the inhibitors decreased the rate of corrosion, according to impedance experiments, and the corresponding circuit was, shown to work well with CPE. In studying the effect of Thyme leaves extract on of mild steel in HCl by Ibrahim et al., (2012). They reviewed that in 2 M HCl, corrosion thyme extract is proven to be an effective corrosion inhibitor for mild steel. As concentrations rose, so did its inhibitory efficiency. The corrosion rate of mild steel rises with increasing temperature, according to test findings from several corrosion procedures. This suggests that the extract's adsorption on the metal surface is of the physisorption type. During the investigation on the Task-specific ionic liquid as a new green inhibitor of mild steel corrosion by Kowsari et al., (2014). They discovered that the TSIL worked as a mixed-type inhibitor in 1 M HCl solution and inhibited both anodic metal dissolution and cathodic hydrogen evolution processes, according to the potentio dynamic polarization curves. Additionally, Lower surface damage was observed on the sample immersed in HCl solution containing 100 mg/L TSIL than other samples and higher contact angle and a lower surface free energy were obtained when the steel sample immersed in HCl solution containing TSIL than the one without TSIL. Kamal et al.,(2012) investigated on Spirulina platensis – A novel green inhibitor for acid corrosion of mild steel. They uncovered that the inhibitor changed the anodic and cathodic Tafel slopes, demonstrating the inhibitor molecules' multimodal mode of action. Additionally, the presence of phytoconstituents such proteins, peptides, fatty acids (like c-linolenic acid), and amino acids (like methionine) is linked to the corrosion-inhibiting activity that S. platensis exhibits. In the Essential oil of Salvia aucherimesatlantica as a green inhibitor for the corrosion of steel in 0.5 M H_2SO_4 as reported by Znini et al., (2012). They discovered that according to chemical study, the main constituent of S. aucherimesatlantica oil may be camphor and the Inhibition efficiency on the steel may occur by action of camphor. As examined by Abdallah et al., (2018) on Some natural aqueous extracts of plants as green inhibitor for carbon steel corrosion in 0.5M sulfuric acid. They identified that the Adsorption proceeds according to Temkin's isotherm and additionally the molecular size of the main constituent of the three natural extracts employed determines the order of the inhibitory effectiveness. By raising the pitting potential to higher values, the natural extracts prevent C-steel from pitting and corroding. Rabizadeh et al., (2019) reported on Chitosan as a green inhibitor for mild steel corrosion: Thermodynamic and electrochemical evaluations. They uncovered that, the corrosion-inhibiting properties of this biodegradable addition (Chitosan) were diminished when the temperature was raised from 298 to 328 K. Furthermore, thermodynamic simulations show that chitosan was adsorbed onto substrates by both physical and chemical adsorption, which raised the enthalpy of activation and the activation energy for the corrosion process. In studying the evaluation of dryopteriscochleata leaf extracts as green inhibitor for corrosion of aluminum in 1 M H_2SO_4. They discovered that within the range of the concentrations examined, the methanol extract of dryopteriscochleata leaves is a more effective inhibitor than water extracts, according to all the methods used, which are in good agreement.

3. CONCLUSION

The need for eco-friendly and sustainable solutions throughout the world has made green corrosion inhibitors a competitive substitute for traditional chemical inhibitors. Mostly derived from plant extracts, these inhibitors provide a practical way to mitigate corrosion while lowering environmental toxicity and regulatory issues. Their efficacy is ascribed to the presence of bioactive substances including tannins, alkaloids, and flavonoids that aid in adsorption onto metal surfaces and provide protective coatings against corrosive chemicals. Despite their encouraging promise, there are several obstacles to overcome before green inhibitors may be used in practice. Their decreased inhibitory efficacy at high temperatures is a major drawback that limits their application in high-temperature industrial operations. Furthermore, seasonal and regional variations in plant composition might affect inhibitor efficacy, underscoring the necessity of consistent extraction and testing procedures. However, the increasing amount of research emphasizes how crucial green inhibitors are to developing long-term corrosion prevention techniques. Researchers can improve these inhibitors for wider industrial application by utilizing developments in materials science, electrochemical investigations, and computational modelling.

3.1. Future perspectives

The decrease in inhibitory effectiveness at high temperatures is one of the most urgent issues. Future research should explore methods to improve the thermal stability of plant-based inhibitors, such as incorporating nanoparticles, chemically altering bioactive components, or creating hybrid formulations with synthetic inhibitors. Secondly, as one method to increase the synergistic effects and improve overall performance, research should be done on integrating green inhibitors or combining other plant extracts with already available commercial inhibitors. Moreover, new, high-performance formulations can be produced by examining these interactions using computational and experimental methods.

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IZVOD

ZELENI INHIBITORI KOROZIJE – ODRŽIVI PRISTUP ZAŠTITI METALA OD AGRESIVNOSTI

Korozija je proces kojim se metal razgrađuje zbog hemijskih ili elektrohemijskih interakcija sa okolinom. Kiseline, alkalije, vlažnost, pH vrednost, temperatura i soli su neki od glavnih elemenata koji doprinose ovom propadanju metala i izazivaju koroziju. Kiseline se često koriste za čišćenje, uklanjanje kamenca i dekapiranje metalnih površina. Korozija je značajan industrijski problem, jer metalni delovi gube masu tokom ovog procesa. Biljni ekstrakti, polimerna jedinjenja, nanomaterijali, neorganski i organski materijali if armaceutski molekuli su svi korišćeni kao inhibitori korozije kako bi se sprečila korozija metala u kiselim sredinama. Studija se fokusira na adsorpciju bioaktivnih jedinjenja kao što su tanini, flavonoidi i alkaloidi nametalne površine kako bi se obezbedile barijere protiv korozivnih supstanci; međutim, značajna prepreka i dalje ostaje da je njihova komercijalna upotreba ograničena činjenicom da se njihova inhibitorna efikasnost smanjuje sa povećanjem temperature, a na njihovu uniformnost i efikasnost utiču ifaktori okoline koji rezultiraju raznolikošću sastava biljaka.

Ključne reči: *Zeleni inhibitori korozije, Ugljenični čelik, Mehanizam adsorpcije, Kisela sredina, Termička stabilnost*

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