

Review

# The Use of Plant Extracts as Green Corrosion Inhibitors: A Review

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**Abstract:** The corrosion of metals is very important, both economically and environmentally, and is a serious concern. Since the past decades, traditional (chemical) corrosion inhibitors to prevent corrosion have been and are still being used. Although these inhibitors can be said to be a good choice among other protection techniques because of their good efficiency, the toxicity of many of them causes environmental problems, and, due to the change in the laws on the use of chemicals, many of them are no longer allowed. Hence, during the past years, research on green corrosion inhibitors (GCIs) increased and very favorable results were obtained, and now they are very popular. It can be said that biodegradability and easy preparation are their most important factors. Meanwhile, the use of plants, especially their extracts, has been studied a lot. Plant extracts contain compounds that have anti-corrosion properties. In this review, the use of plants as GCIs is investigated, focusing on recent advances in their use. Also, the phenomenon of corrosion, corrosion protection (including coatings, nanoparticles, and chemical inhibitors), and other GCIs are briefly reviewed.

**Keywords:** plants; corrosive environment; corrosion protection; green chemistry

## 1. Introduction

It can be said that the development of the economy causes the advancement of technology and the rapid development of society, although it is difficult to ignore the negative consequences it has for the environment [1]. Pollution is directly related to the increase in human population density, in fact, the increase in population causes an increase in human activities, which affects the ecosystem [2]. It can be said, simply, that any human activity that causes the reduction of the quality of the environment is pollution [3]. Environmental pollution is the most important problem in the world, which was created by humans, and continuing knowledge of the source of pollution and its consequences is important because unawareness causes activities that may increase pollution [4]. In general, pollution can be classified into three types: activities that cause air, water, or soil pollution [5]. Deforestation, toxic pollutants, plastics, industrial effluents, agricultural and household waste, greenhouse gas, and improper waste disposal can be considered examples of pollution [6–10]. Meanwhile, corrosion can be considered a phenomenon that causes huge losses and has negative environmental consequences [11]. In fact, corrosion is a spontaneous process, although it can be caused by man-made activities, and many factors play a role in the creation and progress of corrosion (see Figure 1) [12]. In fact, corrosion is an electrochemical process [13]. The destruction of metal structures is very high all over the world, and this problem not only causes economic damage but also poses a serious threat to safety [14]. Corrosion can be considered an important issue for many industries such as oil and gas, construction, marine, aerospace, petrochemical, military, and ceramic industries [15]. High strength and corrosion resistance are always two main and important factors for the use of metallic materials [16]. Corrosion reduction is always a priority in a project where metallic and metal-based alloys are used [17].



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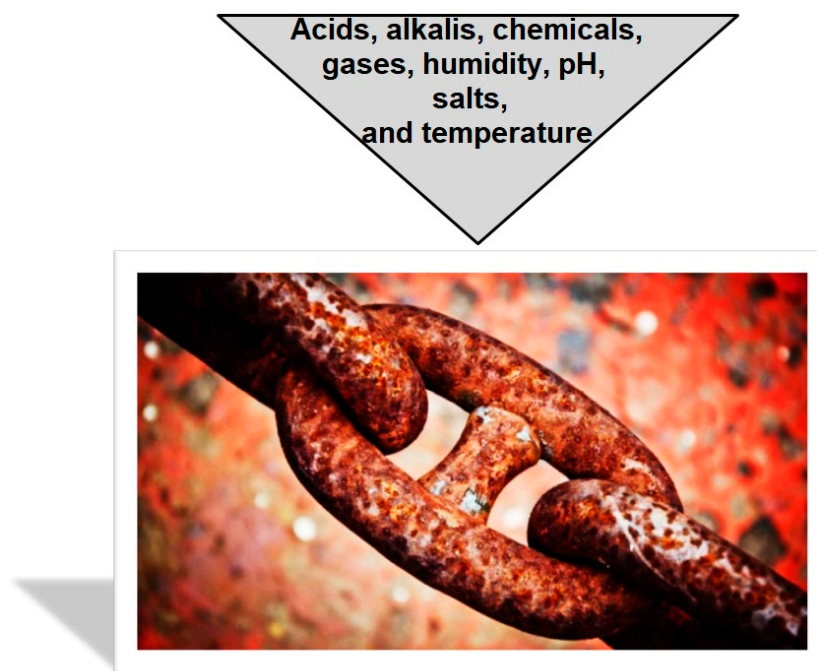
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**Figure 1.** Effective factors in the creation and progression of corrosion.

Researchers are always looking for low-cost and good-performance solutions to prevent and reduce corrosion [18]. Today, corrosion inhibitors are widely used [19]. In general, corrosion inhibitors can be divided into inorganic and organic categories based on their chemical composition [20,21]. Although these inhibitors are used, they have problems such as toxicity, short sustainability, and rapid decomposition [22]. For example, chromates and benzimidazole derivatives that can prevent corrosion are very toxic and cause great harm to the environment [23]. The use of nanomaterials such as hollow mesoporous silica, clay, carbon nanotubes, and graphene oxide is another way to prevent corrosion [24,25]. Also, these nanomaterials can be considered as hosts, and corrosion inhibitors can be placed in them [26–28]. Coating technology is another solution to retard corrosion. In fact, the coating is an outer layer that prevents the penetration of corrosive species like a shield. Among a wide variety of materials, polymers are known as a unique coating due to their properties such as hydrophobicity, anti-fouling, self-healing, and mechanical, chemical, and thermal resistance [29]. Considering the pollution of corrosion inhibitors, one of the main goals of industries is to use non-toxic and environmentally friendly materials, and of course, cost-effective materials. For several years, GCIs (including plant extracts and fruit waste) have received much attention [30]. Many of them have antimicrobial properties in addition to anti-corrosion properties due to being rich in bioactive compounds [31]. It can be said that almost all parts of the plant such as leaves, roots, bark, flowers, fruits, wood, and seeds are used to prepare the inhibitor, but the leaves are considered more often because they have the highest abundance of phytochemicals, although this does not apply to all plants [32,33]. Usually, GCIs are used in very low concentrations, and, due to their adsorptive properties, they can be absorbed on exposed metal surfaces [34]. One of the most important issues in their preparation is the solvents used to extract plant extracts, for this reason, it is important to choose the right solvent (in terms of availability, cost-effectiveness, and safety) [35,36]. Also, the extraction temperature is important, in the recommended literature the ideal temperature is between 60 and 80 °C [37,38].

In this review, the use of plants as GCIs is investigated as well as the phenomenon of corrosion, corrosion protection (including coatings, nanoparticles, and chemical inhibitors), and other members of the category of GCIs are briefly reviewed.

## 2. Corrosion and Factors Affecting It

Corrosion is always lurking in metal structures; this phenomenon has a slow but continuous behavior and causes major failures [31]. Although corrosion is mostly slow, in some environments, such as the sea and industrial towns, it is very rapid and severe [14,31]. In general, metals and metal-based alloys are affected by many factors such as moisture, chemicals, pollutants, fuels, and mineral raw materials, and thus, the conditions for corrosion are created [14]. As mentioned earlier, corrosion is mainly electrochemical in nature and flows in layers of moisture on the metal surface [14]. The mechanism of corrosion flow can be briefly explained. After the initial contact of the metal with the atmosphere (the duration of contact can be from several hours to several days), a surface layer (consisting of oxides and hydroxides) and an electrolyte solution (due to the absorption of water and precipitation of aggressive gases), which can be a very thin layer moisture or an aqueous thick layer, forms on the metal [14,39]. As the thickness of the aqueous layer increases, first the corrosion rate increases (because the electroconductivity of the electrolyte increases) and then decreases (because the rate of oxygen diffusion to the metal surface reduces) [14,15]. Next, the  $\text{Fe}^{2+}$  ions that enter the solution undergo hydrolysis and oxidation processes, and finally, this process continues with the absorption of oxygen or hydrogen release and leads to the formation of iron corrosion products [14]. Corrosion products, which are called rust, are a barrier between the metal and the environment and include many oxides, hydroxides, and oxyhydroxides [14,40]. Three factors have an effect on corrosion, which are environmental factors, metallurgical factors, and structural factors [14,41]. Environmental factors can be considered temperature fluctuations, humidity, and pollutants [14]. As mentioned previously, temperature is an important factor in the corrosion rate. Briefly, in constant humidity, the increase in temperature causes an increase in corrosion, but in closed-air spaces, decreasing the temperature without extra dehumidification increases the rate of corrosion [15]. In general, it can be said that the effect of temperature is partly dependent on the humidity of the environment. One of the pollutants is sulfur dioxide, and there is a high content of it in industrial areas and cities with high density [14]. Sulfur dioxide is the result of the combustion of sulfur-containing fuels and has the ability to accelerate the cathodic reaction of hydrogen evolution, in fact, it plays the role of an oxidizing agent [15,42,43]. Rain has both a positive and a negative role in the corrosion process, in fact, rain has a positive effect by removing chlorine ions and some pollutants from the metal surface, but on the other hand, as the metal surface becomes wet, the possibility of transferring active corrosive compounds (such as sulfur dioxide) increases [15,44]. In fact, sulfur dioxide can acidify the electrolyte on the surface, resulting in the formation of soluble corrosion products [15]. It can be said that the positive or negative effect of rain depends on the presence of pollutants in the atmosphere, although the direction and speed of the wind are also important during rain [14,15]. Also, pH is an important factor, in general, when it decreases, the intensity and rate of corrosion increase [45]. Chloride ions are one of the most aggressive species that can cause corrosion and there is a high content of them in the marine atmosphere [14,15]. In coastal areas, chlorine ions dissolve in the moisture layer and cause the solubility of corrosion products to increase and the passive layers to be destroyed [14,31]. Also, a high salinity atmosphere can lead to the formation of thick layers of rust on the wet surface of carbon steel, causing it to easily separate from the metal substrate, as a result, the surface becomes unprotected and this means accelerating the corrosion process [14,34]. Moreover, an increase in temperature and intensity of ultraviolet (UV) radiation causes the release of chlorine ions [45,46]. Processing and manufacturing method, type, and concentration of alloying elements can be considered metallurgical factors that are directly related to corrosion [14]. For example, in steel, different alloying elements can cause corrosive behavior [47]. The structural factors are different for each structure, but in general, it can be said that the design of the structure, the geometric features of the structure, the spatial orientation of the structure relative to the earth's surface or sky, and the orientation of the surface relative to the main points are of great importance [14,48]. Finally, it can be said that corrosion is the cause of stress concentration and loss of material strength [49,50].

### 3. Corrosion Protection

Protective coatings have the most important effect in preventing corrosion. In this regard, many coatings are used such as paint, polymer, metallic coating, and phosphating coating [15,31]. Meanwhile, the use of nanoparticles to strengthen the coating is common [51]. It can be said that paint and varnish are used more than other coatings to protect structures, and, usually, these coatings consist of several layers (primer and topcoat) [52]. First, it is necessary to prepare the metal surface, which is done manually and mechanized (such as brushes, sand, and shot blast tools), and organic solvents or alkaline solutions (such as sodium hydroxide and potassium hydroxide) are used for degreasing [14]. Then, it is applied on the surface of the primer to improve adhesion for the next layer (paint) [15]. To improve the properties of the primer layer, pigments, and corrosion inhibitors (which will be explained in Section 4) can be added to it [14,31]. Various resins such as epoxy, vinyl ester, acrylic, and polyurethane are used for corrosion protection [12,23,28]. Epoxy has properties such as chemical stability, high mechanical properties, and good adhesion to surfaces, and, due to hydroxyl and amino groups, they can coordinate with metal d-orbitals [28,53]. But, if the epoxy is exposed to UV radiation for a long time, due to the photodegradation reactions in the matrix, it causes the loss of mechanical properties and yellowing of the epoxy [14]. However, nanoparticles such as clay, carbon black, graphene oxide, and graphene can be used to improve UV resistance and other properties [28,54–56]. Another resin used is vinyl ester, the reasons for which are elasticity, adhesion, and UV resistance [12]. Acrylic coatings have high resistance to acids and alkalis, but their properties decrease in long-term contact with water, also they have a relatively high cost [14]. Their properties depend on factors such as functional groups, side chain length, and the presence of a methyl or hydrogen group on  $\alpha$ -carbon [14,53]. Polyurethane has high flexibility, abrasion resistance, high adhesion, and UV resistance, but low thermal stability and resistance to mechanical stress are its weak points [23,30]. The disadvantages mentioned in the coatings can be improved to a great extent with additives such as clay, metal oxide, carbide, carbon nanomaterials (such as carbon nanotubes, graphene, and graphene oxide), silica, and nitride [14]. However, in the field of corrosion protection, conductive polymers (such as polyethylene, polypyrrole, polyaniline, and polythiophene) have also been used [15]. These polymers are not toxic, also they exhibit two behaviors (physical and electronic barrier) [57]. Metal coating (or cathodic protection) is another method used. In short, zinc, aluminum, magnesium, and their alloys are used as coatings because their electrode potential is more negative than iron (or steel) [15]. For phosphating, the steel is immersed in an acidic solution (metallic phosphate salts), and a crystalline layer of phosphate forms on the surface of the steel [14,15]. In fact, an uneven surface is created. These structures are only used in dry indoor environments because their service life will be greatly reduced by getting wet [15].

### 4. Corrosion Inhibitors

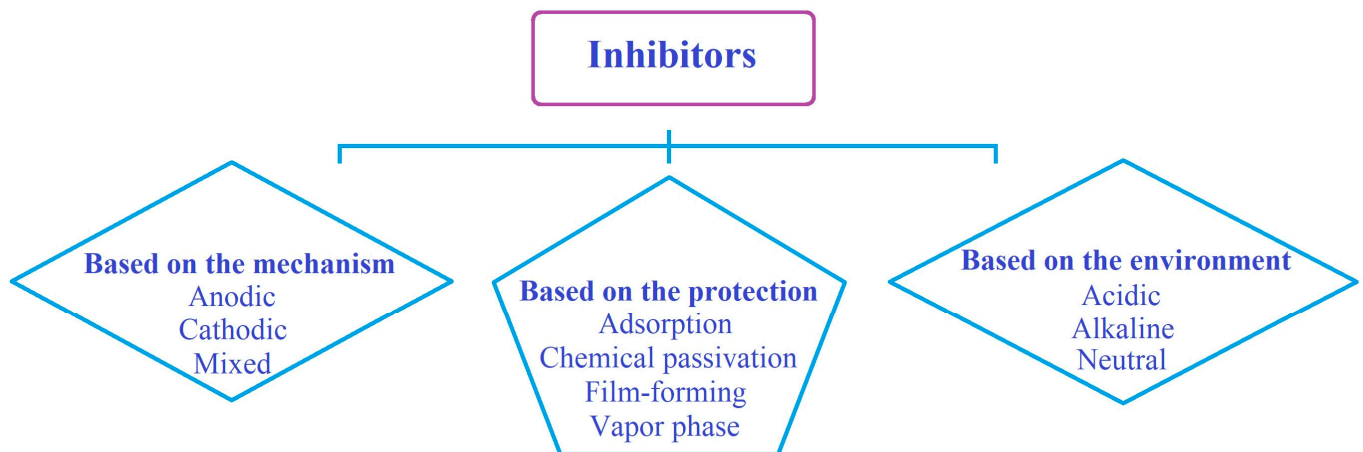
An inhibitor is a substance (chemical or natural) that, when added in low concentrations (1 to 15,000 ppm) in a corrosive environment, will reduce or prevent corrosion [58]. This explanation can be considered the simplest interpretation for an inhibitor. Before using them, several important factors such as toxicity, environmental compatibility, cost, availability, and evaluating and identifying corrosion problems should be considered [23,30,31]. To reduce corrosion, the inhibitor can affect the components of a corrosion cell (which includes the anode, cathode, electrolyte, and electronic conductor), so the inhibitor may cause anodic inhibition, cathodic inhibition, resistance inhibition, and diffusion restriction [58]. In general, an ideal inhibitor should have factors such as no toxicity and pollution problems, long-term effectiveness, good protection at very low concentrations, uniform and localized corrosion suppression, protection of all materials exposed to corrosion species, efficiency under all conditions (temperature and velocity), as well as not creating deposits on the metal surface [31,58]. Many inhibitors are composed of organic and inorganic compounds (usually composed of elements of groups V B and, VI B) [14,58]. The effectiveness of organic

inhibitors can be increased with halogen ions [59]. Briefly, it can be said that the highest and lowest effects are related to  $I^-$  and  $Cl^-$ , respectively, and no effects have been reported for fluoride [58]. In fact, this efficiency improvement is related to the synergism of halogen ions, as metal adsorbs halogen ions, whose charge shifts the surface in a negative direction, thus increasing the adsorption of cationic organic inhibitors because organic inhibitors are adsorbed on the surface of metals [58,59]. Inhibitors generally behave in two ways, either they form a protective shield on anodes or cathodes or they are directly adsorbed on the metal surface [58,60]. Based on the mechanism, the inhibitors are divided into three groups: anodic, cathodic, and mixed, which are briefly discussed in Table 1. Additionally, Figure 2 shows the classification of corrosion inhibitors based on mechanism, environment, and protection [58,61].

**Table 1.** A brief description of inhibitors based on mechanism.

Inhibitor	Corrosion Inhibition Method	The Most Common Inhibitor
Anodic	By forming a protective film along the anode, they increase their potential and reduce the corrosion reaction.	Chromates and tungstates
Cathodic	By blocking the cathode sites, they reduce the rate of the reduction reaction of the electrochemical corrosion cell, and as a result, the corrosion is reduced.	Elements arsenic and antimony
Mixed	By forming a film that causes deposits on the surface, they block the anodic and cathodic sites and delay corrosion.	Silicates and phosphates

Note: Data from [58,59].



**Figure 2.** General classification of inhibitors.

Usually, the mechanism of the inhibition process is due to absorption [60,61]. In general, the chemical structure of the inhibitor and the surface charge of the metal affect the absorption [58]. Important factors play a role in inhibition, and inhibition always results from one or more mechanisms (see Figure 3) [58,60].



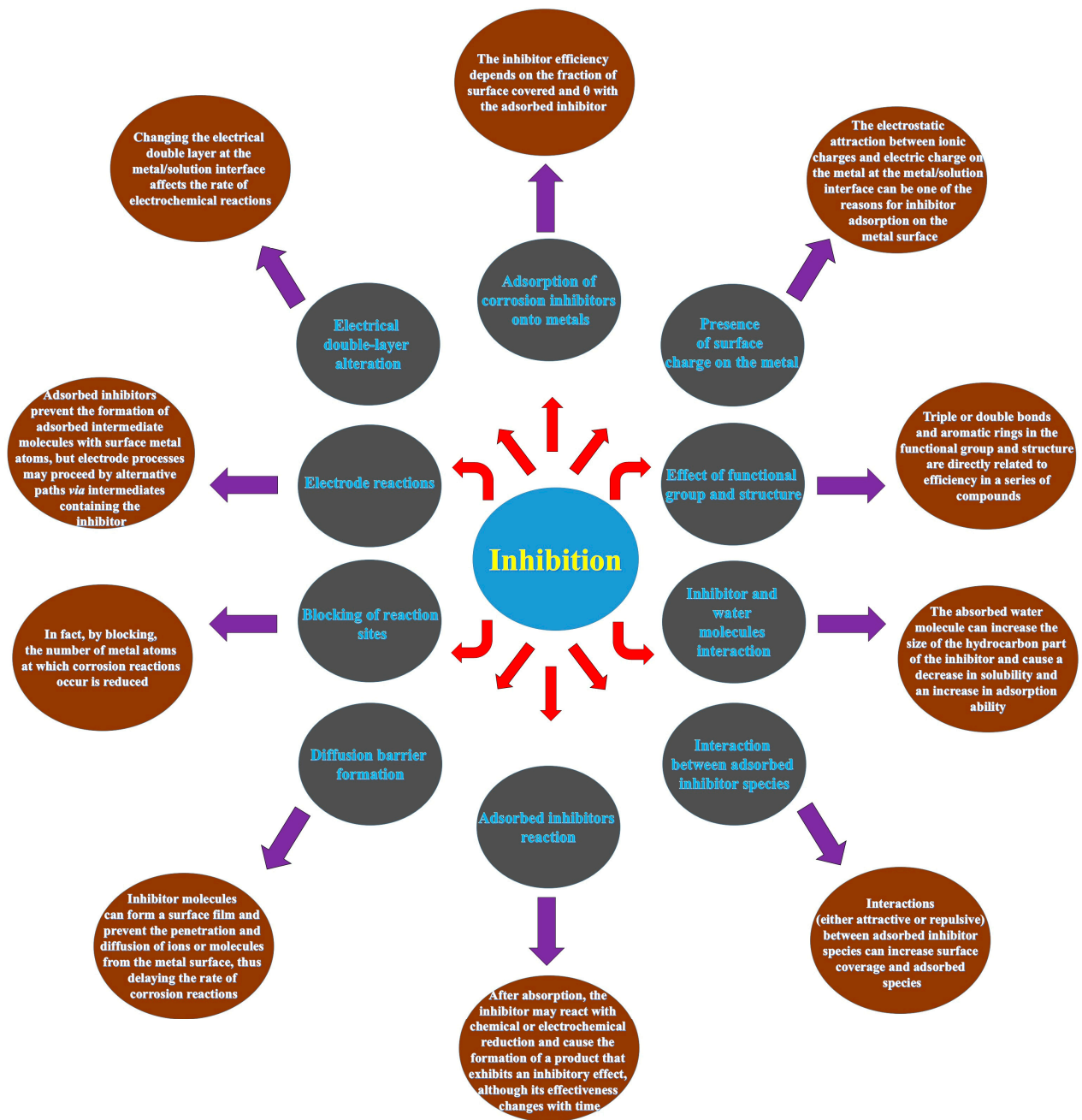


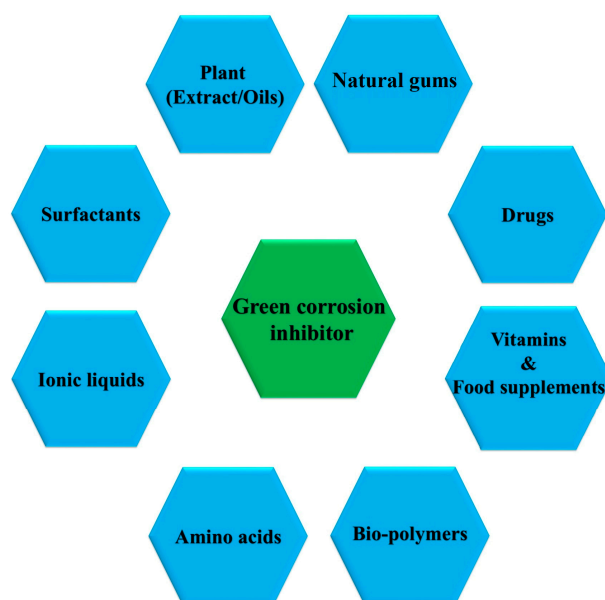
Figure 3. Important factors in inhibition.

## 5. GCIs

### 5.1. Background

As mentioned earlier, there are various methods to prevent corrosion, but it can be said that the use of corrosion inhibitors is one of the most effective and practical methods [62–64]. Corrosion inhibitors contain compounds based on whether they are inorganic or organic [65]. For inorganic inhibitors, nitrate, nitrite, chromate, dichromate, and phosphate can be mentioned, and on the other hand, organic inhibitors contain functional groups, heteroatoms (such as O, N, S, P),  $\pi$ -bonds, and aromatic heterocyclic rings [65–68]. Many corrosion inhibitors have a high protective effect, but recently, their use has been limited due to high cost, toxicity, non-biodegradability, and health hazards [63,69,70]. They have a significant effect on ecosystems and living beings, so they have important environmental concerns [63,70]. Some of them contain heavy elements that cause water and soil pollution due to the accumulation and non-biodegradation of these elements

and, eventually, cause their accumulation in plants and animals, and in this way, enter the food chain and have irreparable effects on human health [71,72]. Therefore, a less expensive, available, and non-toxic alternative to common corrosion inhibitors is essential. Hence, research was conducted on the development of corrosion inhibitors based on natural resources, and in fact, a new concept called green corrosion inhibitors emerged [64,73]. According to new legislation, chemicals must be safe for the environment and humans, so high efficiency is not a sufficient condition for using an inhibitor [74,75]. In this regard, green corrosion inhibitors have gained a lot of popularity. Their inhibition results are comparable to conventional organic and anodic inhibitors, and in some cases, they have been more efficient, although it has been reported in the literature that the efficiency of some of them decreases greatly at high temperatures [62,76–78]. In fact, the development of green corrosion inhibitors still needs to be improved. It can be said that most natural products can be considered green corrosion inhibitors because they are rich in compounds that are effective for corrosion inhibition [73,79,80]. When talking about green corrosion inhibitors, plants are always unconsciously considered, while a wide range of sources can be considered (see Figure 4) [81–84].



**Figure 4.** Types of green corrosion inhibitors.

As seen in Figure 4, the use of various sources can be considered for use, and many of them, in addition to anti-corrosion properties, can reduce microbial aggression due to the presence of compounds such as polyphenols, alkaloids, and terpenes [81,82,85].

## 5.2. Classification of GCIs

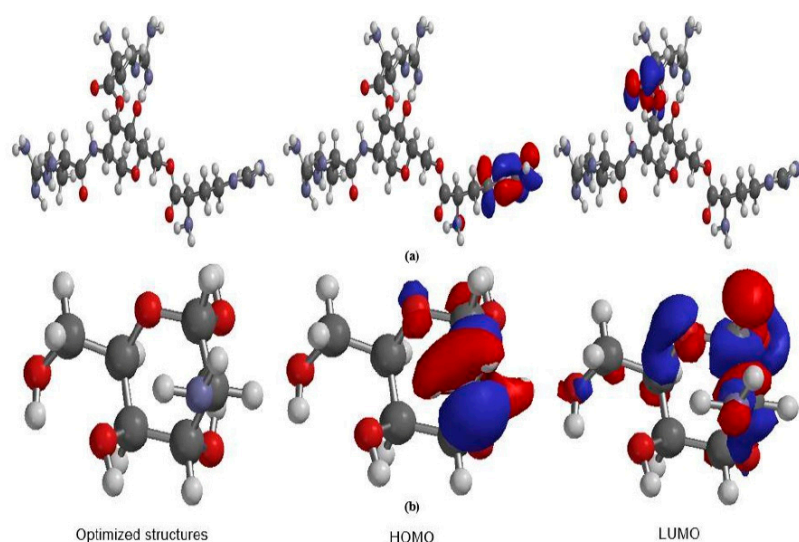
### 5.2.1. Amino Acids

Amino acids are one of the most popular inhibitors and have been used to inhibit corrosion in many metals such as iron, steel, aluminum, copper, and nickel [62]. Their popularity is due to their reversibility, non-toxicity, easy to make, biodegradability, environmentally friendly, solubility in water, and low cost [62,74,86]. The presence of heteroatoms such as N and O (because they contain carboxyl groups and amino groups) as well as conjugated  $\pi$ -bonds-electron systems can be considered as the main reasons for their anti-corrosion properties [62]. However, the molecular structure of the side chain is also very important. In fact, the adsorption energy increases with the lengthening of the side chain, and its maximum value on the iron surface (based on the side chain) is in the following order: acidic < polar < nonpolar < basic [87]. Also, having a negative or positive charge depends on the pH and they usually have a positive charge in an acidic environment [74]. Usually, their corrosion inhibition mechanism is based on the absorption and formation of a film

on the metal surface [74,88]. Methionine, cysteine, lysine, arginine, glycine, and valine can be mentioned as some of the amino acids that have been investigated for corrosion inhibition [62,74]. Yeganeh et al. [62] investigated the effect of L-methionine on the corrosion inhibition of stainless steel in the sulfuric acid ( $H_2SO_4$ ) solution. The results showed that methionine increases the charge transfer resistance and decreases the corrosion current density, so the electrochemical impedance spectroscopy results showed a maximum efficiency of 97%. Methionine has two groups,  $-NH_2$  and  $-S-CH_3$ , and corrosion inhibition of methionine was reported due to the chemisorption process (chemical bond between sulfur and iron) and the physisorption process (due to the interaction of functional groups and the surface).

### 5.2.2. Bio-Polymers

Polymers extracted from natural resources (renewable resources) can be considered as a new and attractive approach to inhibit corrosion. Their mechanical and thermal properties, non-toxicity, and compatibility with the environment can be considered to be their strengths [89,90]. They are widely used in various applications such as cosmetics, food preservation, medicine, and textiles [89,91]. The use of a polymer as a host for a corrosion inhibitor can be considered a successful combination because, in addition to the low use (concentration) of the inhibitor, the polymer can be a long-term protector [74]. As described in Section 3 (corrosion protection), polymers can resist solvents and many aggressive species. One of these polymers that is used to inhibit corrosion is chitosan because it has many adsorption sites [92]. Chitosan is used for many applications such as cosmetics, medicine, water treatment, and also the protection of bronze objects [93,94]. The poor solubility of chitosan in water is perhaps considered its only weakness (as an inhibitor), although this problem can be solved by using chitosan derivatives such as carboxymethyl chitosan (while retaining the number of adsorption sites) [92]. Dalhatu et al. [95] investigated the effect of L-arginine grafted onto chitosan on the corrosion inhibition of mild steel in 0.5M HCl. The evaluation showed that the inhibition efficiency at optimum concentration rose to 91.4%. The optimized structures are presented in Figure 5. It can be said that the absorption performance of a corrosion inhibitor molecule depends on the highest occupied molecular orbital (HOMO) and the lowest unoccupied molecular orbital (LUMO), in fact, HOMO and LUMO are related to the electron donation characteristics and electron accepting behaviors, respectively. HOMO is mainly localized over the L-arginine while, LUMO density is localized exclusively on the chitosan ring, indicating that these regions are mainly involved in electron donation and acceptance, respectively, during the metal inhibitor interactions.

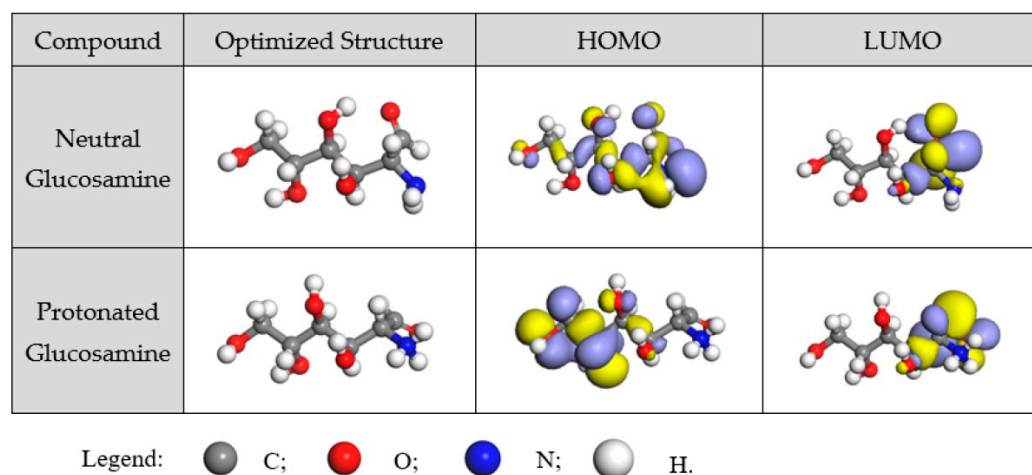


**Figure 5.** Optimized structures and frontier molecular orbital electron density distribution (HOMO and LUMO) of (a) Chitosan-L-arginine and (b) chitosan. Reprinted/adapted with permission from Ref. [95]. 2024, MDPI.



### 5.2.3. Drugs

Pharmaceutical products are one of the candidates for corrosion inhibition due to having heteroatoms and various compounds [96]. In fact, expired drugs are considered waste, but many of them can be used as inhibitors, thereby preventing the waste of resources [96]. Although the drugs are designed for therapeutic use and no corrosion protection application is reported for them by the manufacturer, the use of other compounds along with them can increase the anti-corrosion effect [97,98]. To date, the effectiveness of many drugs to inhibit corrosion has been evaluated, such as naproxen, glucosamine, clotrimazole, amoxicillin, ciprofloxacin, doxycycline, and streptomycin [74,99,100]. Chaoui et al. [66] investigated the effect of two hydrazone derivatives based on Naproxen ((E)-N'-(2,4-dimethoxybenzylidene)-2-(6-methoxynaphthalen-2-yl) propanehydrazide (HYD-1), and N'-cyclohexylidene-2-(6-methoxynaphthalen-2-yl) propanehydrazide (HYD-2)) on corrosion inhibition of mild steel in a hydrochloric acid (HCl) solution. Naproxen is a medication used to treat pain and joint swelling. The results showed that both compounds have high inhibitory performance and this effect increases with increasing their concentration, and they are also in the category of mixed inhibitors (they affect both anodic and cathodic reactions). Moreover, the results of electrochemical evaluations showed maximum inhibition efficiency for HYD-1 and HYD-2 of 96% and 84%, respectively. Both compounds prevent corrosion via adsorption on the surface, and also, increasing the temperature reduces the protection of the inhibitors so that, for HYD-1, the maximum inhibition efficiency decreases from 96% (at a temperature of 303 K) to 81% (at a temperature of 333 K). Feng et al. [101] investigated the effect of two types of expired glucosamine drugs (glucosamine sulfate (GS) and glucosamine hydrochloride (GH)) on the corrosion inhibition of carbon steel in an H<sub>2</sub>SO<sub>4</sub> solution. Also, in this study, iodide ions were used to increase the inhibitory effect. The evaluation showed that GH has better inhibition compared to GS, although the performance of GS improved with the addition of iodide ions. Also, they were in the category of mixed inhibitors and their inhibitory properties were due to the covering effect. Figure 6 shows that, for the neutral glucosamine molecule, the HOMO is mainly distributed around the amino and carbonyl groups, so these groups easily interact with metals by creating negative charges from heteroatoms of N and O. Also, the LUMO is distributed in the carbonyl group, so it is an active site for accepting electrons. This study shows that glucosamine is not only useful for the treatment of osteoarthritis but it can also be used to inhibit corrosion.



**Figure 6.** Optimal structures of neutral and protonated glucosamine molecules and their HOMO and LUMO distributions. Reprinted/adapted with permission from Ref. [101]. 2024, MDPI.

### 5.2.4. Ionic Liquids

The use of ionic liquids as corrosion inhibitors has been widely evaluated [102]. In fact, they are salts composed of cations and anions that are in the liquid phase at (or close to)

room temperature [103,104]. It can be said that their unique properties such as high thermal and chemical stability, non-flammability, very low toxicity, low volatility, and the ability to absorb metal surfaces have made them popular [74,105,106]. The efficiency of ionic liquids depends on factors such as temperature, pH, specific interactions, and chemical structures, and they also prevent corrosion by forming a protective film on the surface [106,107]. Many ionic liquids that contain cations with nitrogen atoms have been investigated as corrosion inhibitors; in fact, the corrosion properties of ionic liquids derived from quaternary ammonium are much better [104,106]. These properties include higher purity products, cheaper synthesis methods, tensioactive properties, and less toxicity (compared to ionic liquids derived from heterocyclic groups) [106,107]. Arellanes-Lozada et al. [106] investigated the effect of methyltrioctylammonium methyl sulfate and trimethyltetradecylammonium methyl sulfate on the corrosion inhibition of API-X52 steel in an HCl solution. The results showed that the inhibitors are very effective at a temperature of 40 °C, which is related to the high thermal stability of methyl sulfate anions. Also, they were in the category of mixed inhibitors.

#### 5.2.5. Plants

Plants can be considered an extremely rich source of compounds with anti-corrosion properties. Today, the use of plant extracts and oils to inhibit corrosion is widely investigated [28,30,31]. Unlike synthetic (chemical) inhibitors, they are non-toxic in nature [74]. Also, they are very popular due to their cheap price, high abundance, easy preparation method, environmental friendliness, and biodegradability [83,108]. However, their long-term use and preservation are partly affected by biodegradability [74]. It can be said that the first use of a plant extract as a corrosion inhibitor dates back to 1930, which was the extract of *Chelidonium majus* and some other plants in a pickling bath of H<sub>2</sub>SO<sub>4</sub> [108,109]. Plant oils and extracts include a wide range of phytochemical compounds such as tannins, flavonoids, and polyphenols, so they have a high potential for corrosion inhibition [83,85,109]. Many plant compounds have been evaluated for corrosion inhibition, but given that there are about 300,000 different types of plants in the world, there are still numerous types of plants remaining [83,109]. In Section 5.3, plants will be thoroughly investigated.

#### 5.2.6. Surfactants

Typically, surfactants are used in applications such as shampoos, detergents, paints, coatings, and concrete, but they also have other uses, in fact, they are also used as corrosion inhibitors [110]. Their advantages include cheapness, easy preparation methods, non-toxicity, and high performance [74]. Surfactants have hydrophilic and hydrophobic parts [74,111]. The hydrophilic part interacts with polar entities and the hydrophobic part interacts with hydrophobic entities. In fact, this dual nature of their interaction with surfaces and interfaces is very influential and decisive [112,113]. Generally, surfactants are classified as barrier-type inhibitors because they reduce corrosion by creating a physical barrier, in fact, they can act as anodic or cathodic inhibitors [112,114]. In fact, it is the absorption of surfactant on the surface that can prevent corrosion, and this absorption depends on factors such as the electrochemical potential of the surface, temperature, the concentration of the adsorbent, and the composition of the solution [112]. Still, many studies in the field of surfactants are about their ability to inhibit corrosion in acidic environments [115–118].

#### 5.2.7. Vitamins and Food Supplements

Vitamins and food supplements are also in the category of green corrosion inhibitors, although priority is usually given to substances with expired dates [84]. Vitamins are of great interest due to factors such as good solubility, non-toxicity, low price, and suitable functional groups [84,119]. Much research has shown that vitamins such as B, C, D, and E can be used as corrosion inhibitors [84,120–125]. They are soluble in water (directly added to the corrosion medium) or fat (used to make superhydrophobic self-assembling protective layers), so the first factor for their selection is the medium to be used [84,122,125].

Argiz et al. [120] investigated the effect of ascorbic acid (vitamin C) on the corrosion inhibition of steel in cement mortars contaminated by chlorides. The results showed that ascorbic acid can inhibit corrosion and this ability is stable for 8 months. Also, its highest efficiency, obtained by adding a  $10^{-3}$  mol/L concentration in the mixing water, is 97.3%. Uzoma et al. [126] investigated the effect of aspartame on the corrosion inhibition of T95 carbon steel in HCl. The results showed that aspartame is a mixed inhibitor and the inhibition efficiency increases with increasing temperature (60 to 90 °C). Also, at 90 °C using 2000 ppm of aspartame, an inhibition efficiency of 86% was obtained after 4 h of immersion.

#### 5.2.8. Natural Gums

The last member of the category of green corrosion inhibitors is natural gums. Gums are generally classified as a pathological response to plant damage and are exudated from the stem or root of the tree [127]. They are a mixture of long-chain polysaccharides, whose hydrolysis often produces arabinose, galactose, mannose, and glucuronic acid [128,129]. Natural gums are used in the food processing, cosmetic, and pharmaceutical industries [127,128]. In the past years, many studies have been conducted on natural gums as corrosion inhibitors, in fact, gums have compounds containing oxygen atoms, nitrogen, and COOH functional groups and can form complexes with metal ions on metal surfaces [127,129]. However, it is possible to improve the stability and properties of gums through the addition of some agents such as surfactants or salt of halides, and self-healing agents [128]. Palumbo et al. [130] investigated the effect of guar gum on the corrosion inhibition of N80 carbon steel under a sweet environment in a saline solution. The performance of guar gum increased with increasing concentration and immersion time but showed a decreasing trend with increasing temperature so that the inhibition efficiency with  $0.4 \text{ g L}^{-1}$  of guar gum at 25 °C was 76.16%, but by increasing the temperature to 50 °C, the inhibition efficiency decreased to 63.19%. Timothy et al. [131] investigated the effect of *Berlinia grandiflora* and cashew on the corrosion inhibition of mild steel in HCl. The results showed that the inhibition efficiency (at 20 °C) for *Berlinia grandiflora* and cashew (using 1000 ppm of each) is 92% and 75%, respectively. Also, the inhibition efficiency decreased to 76% and 67% for *Berlinia grandiflora* and cashew after 48 h of immersion, respectively. Moreover, after 24 h of immersion at 60 °C, the inhibition efficiency for *Berlinia grandiflora* and cashew was 80% and 57%, respectively. Table 2 shows some of the natural gums and a brief description of them.

**Table 2.** A brief description of some natural gums.

Natural Gums	Brief Description
Gum arabic	Its other name is acacia gum, which is obtained from the <i>Acacia Senegal</i> tree and is used in drug delivery, adhesive, anti-corrosion agents, and food packaging.
Guar gum	It is obtained from the <i>Cyamopsis tetragonoloba</i> plant and is used in agriculture, drug delivery, biomedical, pharmaceuticals, coatings, and food industries.
Xanthan gum	It is secreted by the <i>Xanthomonas campestris</i> bacteria and is used in cosmetics, the oil industry, tissue engineering, and food industries.
<i>Albizia</i> gum	It is obtained from the <i>Albizia</i> tree and is used in the cosmetics, pharmaceutical, and food industries.
<i>Dacryodes edulis</i> gum	It is obtained from the <i>Dacryodes edulis</i> tree and is used in cosmetics, pharmaceutical, adhesive, and anti-corrosion agents.
Cashew tree gum	It is obtained from the <i>Anacardium occidentale</i> tree and is used in medicine, anti-corrosive agents, emulsifying agents, and coating.
<i>Raphia hookeri</i> gum	It is obtained from the <i>Raphia hookeri</i> tree and is used in pharmaceuticals, emulsifiers, and anti-corrosive agents.

Note: Data from [127,128,132,133].

### 5.3. Using Plants as GCIs

In Section 5.2.5, the use of plants to inhibit corrosion was briefly explained. In general, the result of using plants with an inhibition efficiency of more than 80% can be considered a good inhibitor. Different parts of plants can be used, including leaves, roots, fruits, flowers, oil, woody parts, whole plants, and even waste [30–33]. In fact, these inhibitory properties are due to their rich natural compounds. Usually, plant extracts are more often used to investigate corrosion inhibition [32,33,74]. In short, the solution that consists of the active ingredients of the plant and a certain medium (solvent) is called an extract [82]. Therefore, the method of preparation of plant extract is very important. Figure 7 shows the general process of preparation of plant extract.

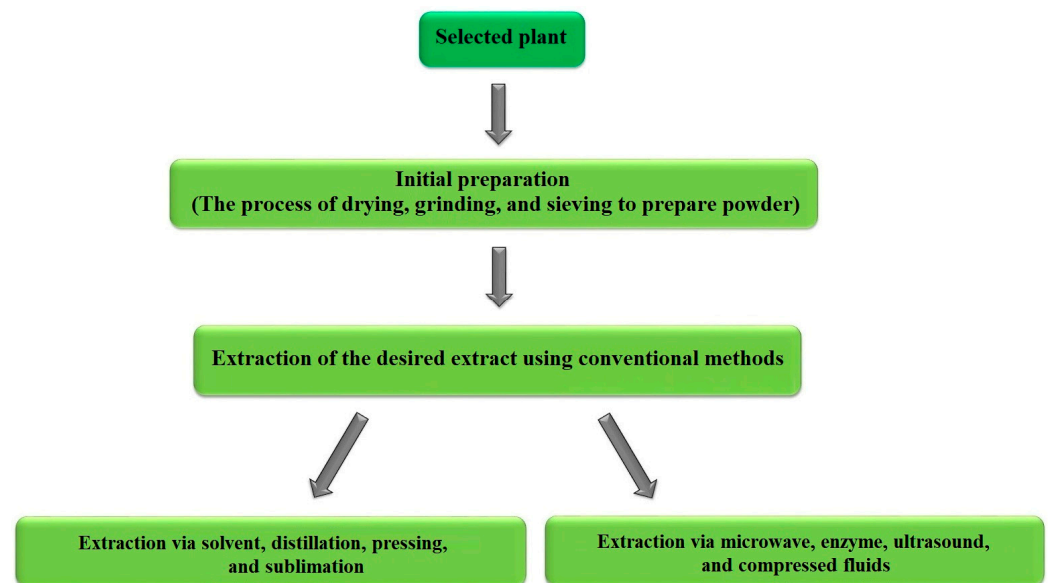


Figure 7. The process of preparing plant extracts.

Solvent extraction is usually more popular and is the most widely used because it is easier, faster, and has a higher production capacity than other methods [134]. In this method, the solvent penetrates into the plant tissue and causes the required compounds to be extracted [74,134]. Many factors affect the quality of the extract, including the type of solvent (polar or non-polar), the ratio of solvent to solid, and temperature (usually between 60 and 80 °C is the best) [134]. But, in general, the solvent has the most key role, because it is responsible for dissolving the active compounds from the plant tissue, also, the solvents affect the properties of the extracts [82,135]. Table 3 summarizes some of the most commonly used solvents for extraction and the active extracts (phytochemicals) commonly obtained from them.

Table 3. Types of phytochemicals extracted from common solvents.

Solvent	Acetone	Ethanol	Methanol	Water
Phytochemicals	Flavonols and Tannins	Alkaloids, Flavonols, Polyacetylenes, Polyphenols, Propolis, Sterols, Tannins, and Terpenoids	Anthocyanins, Flavonols, Lactones, Quassinoids, Phenones, Polyphenols, Saponins, Tannins, Terpenoids, Tatarol, and Xanthoxylines	Anthocyanins, Lectins, Polypeptides, Saponins, Starches, Tannins, and Terpenoids

Note: Data from [136].

The inhibition mechanism of GCIs has been widely investigated. In general, the inhibition mechanism and inhibition efficiency are directly dependent on the active phytochemical compounds of the plant extract [65,134,137]. The absorption mechanism of GCIs is classified into three ways: chemisorption, physisorption, or a combination of the two (mixed) [76]. In general, the interaction between the charged metal surface and the inhibitor determines the type of absorption, in fact, the weak polar interaction causes physisorption, and the molecule with strong electrostatic forces on the metal surface causes chemisorption [64,138]. But, in general, a mixed inhibitor provides the best protection because it affects both cathodic and anodic reactions [64,134,139]. As mentioned before (Section 5.1), temperature has an important effect on the performance of GCIs. Evaluating efficiency at different temperatures is essential because many extracts can have very different performances with increasing and decreasing temperatures [82]. In fact, with increasing temperature, the amount of absorption in physisorption and chemisorption decreases and increases, respectively [76,138]. Also, the immersion time can have a great effect on the performance of the inhibitor [82]. Sivakumar and Srikanth [83], in investigating the inhibitory effect of *Mimusops Elengi* extract on the corrosion of mild steel in HCl medium, reported a decrease in inhibition efficiency with increasing temperature. Also, inhibition efficiency increased with increasing immersion time (from 1 to 12 h), which was related to increased absorption of extract compounds on the substrate surface. Abdallah et al. [63], in the study of the use of natural nutmeg oil to prevent the corrosion of carbon steel in HCl, reported that, as the temperature increases (30 to 60 °C), the corrosion rate values increase while inhibition efficiency values decrease. Begum et al. [109] also reported, in the study of the use of *Spilanthes acmella* leaves extract for corrosion inhibition of mild steel in HCl, that, with increasing temperature (303 to 333 K), efficiency and corrosion rate decrease and increase, respectively. Barbouchi et al. [79], in the study of the use of *Terebinth* extract to prevent iron corrosion in NaCl, reported that, with the increase of immersion time (up to 72 h), the thickness of the created protective layer increases. Although there are many studies that report a decrease in efficiency with increasing temperature, some researchers report an increase in efficiency, such as Méndez et al. [140], who used *Ilex paraguariensis* extract to inhibit the corrosion of aluminum in HCl and the results showed that the maximum value of efficiency at 315 and 323 K is 69 and 71%, respectively, and these results were obtained using a concentration of 0.248 g L<sup>-1</sup> of the extract. Regardless of the adsorption mechanism, temperature, and immersion time, other parameters such as the density of functional groups, polarizability, and electronegativity are also very important [73]. However, in addition to the nature of the inhibitor and its chemical composition and surface charge of the substrate, the type of corrosive medium is also important [65,134,141–143]. For example, extracts that contain aromatic rings, multiple bonds, and heteroatoms can be easily protonated in an acidic medium, which creates positively charged inhibitory species that can adsorb through chloride ions on the positively charged steel surface [134]. Dehghani et al. [69] in investigating the use of *Ziziphora* leaf extract molecules to prevent corrosion of mild steel in HCl, reported that protonated forms of acacetin, chrysin, and thymonin (leaf extract molecules) electrostatically interact with adsorbed chloride ions. Ginger has been extensively studied for its anti-corrosion activity. The compounds in it such as gingerols, shogaols, and curcumin play an important role, in fact these compounds accumulate on the surface of the metal and block the anode and cathode sites, which is due to  $\pi$ -electrons and heteroatom oxygen atoms in the phenolic compounds [12,23]. Also, gingerol is hydrophobic, so the penetration of corrosive species is delayed [23]. *Ganoderma lucidum* mushroom is suitable for anti-corrosion activity due to its diverse compounds. In fact, amine and hydroxyl groups in *Ganoderma lucidum* compounds (such as proline and triterpenoids) can transfer electrons with Fe<sup>2+</sup> in anodic regions and physically absorb on the mild steel surface and form a protective film, and *Ganoderma lucidum* contains hydrophobic compounds (such as quercetin, proline, and gallic acid) [27]. Matcha is another plant that has important compounds. The main compounds of matcha (mainly catechins) can be adsorbed on the steel surface, which is due to the  $\pi$ -electron transfer



between the oxygen atoms and the aromatic rings of these compounds with the vacant *d*-orbitals of the steel surface [30]. Table 4 reports a summary of the performance of some corrosion inhibitors (plant-based) in different media.

**Table 4.** Corrosion inhibition performance of some plants in different corrosion media.

Plant	Substrate	Corrosive Medium	Maximum Inhibition Efficiency (%)	Reference
<i>Artemisia vulgaris</i> — <i>Solanum tuberosum</i>	Mild steel	H <sub>2</sub> SO <sub>4</sub>	88.06–83.22	[73]
<i>Coriaria nepalensis</i>	Mild steel	H <sub>2</sub> SO <sub>4</sub>	97.03	[77]
Terebinth ( <i>Pistacia terebinthus</i> L.)	Iron	Sodium chloride (NaCl)	86.4	[79]
<i>Ixora coccinea</i>	Mild steel	H <sub>2</sub> SO <sub>4</sub> -HCl	77.96–89.38	[144]
<i>Justicia secunda</i>	Aluminum	HCl	94.30	[145]
<i>Gongronema latifolium</i>	Mild steel	HCl	81.69	[146]
<i>Clinopodium acinos</i>	Mild steel	HCl	89.90	[147]
White tea	Mild steel	HCl	96.00	[148]
<i>Momordica charantia</i>	Carbon steel	H <sub>2</sub> SO <sub>4</sub>	93.51	[149]
<i>Malva sylvestris</i>	Mild steel	NaCl	91.00	[150]
<i>Lilium brownii</i>	X70 steel	HCl	85.00	[151]
<i>Pueraria lobata</i>	10# steel	HCl	94.37	[152]
<i>Allamanda cathartica</i>	Mild steel	H <sub>2</sub> SO <sub>4</sub>	72.75	[153]
Cauliflower	Copper	H <sub>2</sub> SO <sub>4</sub>	99.00	[154]
Peach pomace	Mild steel	NaCl	88.00	[155]
Binda rind	Mild steel	HCl	97.33	[156]
Betel	Carbon steel	HCl	94.90	[157]
<i>Platanus acerifolia</i>	Carbon steel	NaCl	99.86	[158]
<i>Catharanthus roseus</i>	Mild steel	NaCl	84.00	[159]
Mutiti	Low—carbon steel	H <sub>2</sub> SO <sub>4</sub>	86.23	[160]
<i>Chamaerops humilis</i>	Mild steel	HCl	88.00	[161]
<i>Elaeoselinum thapsioides</i>	Carbon steel	HCl	82.00	[162]
<i>Punica granatum</i> L.	Mild steel	HCl	87.30	[163]
<i>Gentiana olivieri</i>	Mild steel	HCl	89.70	[164]
<i>Tithonia diversifolia</i>	Mild steel	HCl	79.99	[165]
<i>Matricaria aurea</i>	Mild steel	HCl	93.56	[166]
<i>Pogostemon quadrifolius</i> floral	Mild steel	HCl	95.79	[167]
<i>Taxus baccata</i>	Carbon steel	HCl	74.26	[168]
<i>Prunus dulcis</i>	Mild steel	HCl	88.00	[169]
<i>Artemisia herba-alba</i>	Stainless steel	Phosphoric acid (H <sub>3</sub> PO <sub>4</sub> )	85.00	[170]
Artemisia oil	Steel	H <sub>3</sub> PO <sub>4</sub>	74.00	[171]
<i>Psidium Guajava</i>	Mild steel	H <sub>3</sub> PO <sub>4</sub>	82.00	[172]
Guar gum	Carbon steel	H <sub>3</sub> PO <sub>4</sub>	95.00	[173]

Table 4. Cont.

Plant	Substrate	Corrosive Medium	Maximum Inhibition Efficiency (%)	Reference
<i>Rhynchosytilis retusa</i>	Mild steel	H <sub>2</sub> SO <sub>4</sub>	93.24	[174]
<i>Acacia catechu</i>	Mild steel	H <sub>2</sub> SO <sub>4</sub>	98.54	[175]
<i>Solanum xanthocarpum</i>	Mild steel	H <sub>2</sub> SO <sub>4</sub>	98.14	[176]
<i>Veratrum</i>	Copper	H <sub>2</sub> SO <sub>4</sub>	97.00	[177]
<i>Brassica oleracea</i> L.	Q235 steel	H <sub>2</sub> SO <sub>4</sub> -HCl	92.30–93.80	[178]
<i>Thymus satureioides</i>	316L stainless steel	NaCl	82.00	[179]
<i>Pinus nigra</i>	Carbon steel	HCl	97.00	[180]
Palm seed	Carbon steel	HCl	95.00	[181]
<i>Cucumis sativus</i> L.	Carbon steel	H <sub>2</sub> SO <sub>4</sub>	92.80	[182]
Harmal roots	Carbon steel	H <sub>2</sub> SO <sub>4</sub>	94.10	[183]
Palm leaves	Carbon steel	HCl	96.80	[184]
<i>Aerva lanata</i>	Aluminum	NaCl	88.00	[185]
<i>Ziziphus joazeiro</i>	Mild steel	HCl	94.70	[186]
<i>Trochodendron Aralioides</i>	Mild steel	HCl	96.42	[187]
Aquatic Artichoke	Mild steel	HCl	98.70	[188]
<i>Cynara scolymus</i> L.	Mild steel	NaCl	96.10	[189]
<i>Juglans regia</i> L.	Mild steel	HCl	95.00	[190]
<i>Rosa damascene</i>	Carbon steel	H <sub>2</sub> SO <sub>4</sub>	75–96.1	[191]
<i>Cinnamomum camphora</i>	Low carbon steel	H <sub>2</sub> SO <sub>4</sub>	89–97	[192]
Olive leaf	Copper	KOH	90.68	[193]
<i>Justicia brandegeana</i>	Carbon steel	H <sub>2</sub> SO <sub>4</sub>	82.41	[194]
Coffee waste	Carbon steel	HCl	76–96	[195]
<i>Chamaerops humilis</i>	Carbon steel	HCl	94	[196]
Grape seed oil	Carbon steel	HCl	79	[197]
<i>Glebionis coronaria</i> L. flower	Mild steel	HCl	95	[198]
Fig leaf	Mild steel	HCl	94	[199]
<i>Cyclotrichium niveum</i>	Mild steel	HCl	97.3	[200]
Pumpkin leaf	Copper	H <sub>2</sub> SO <sub>4</sub>	89.98	[201]
<i>Pipper cubeba</i>	Carbon steel	Sulfamic acid	96	[202]
<i>Bagassa guianensis</i>	Zinc	NaCl	97	[203]
<i>Aloe vera</i> (L.) Burm. F.	Bronze B66	NaCl	89	[204]
Asafoetida	Mild steel	NaCl	90	[205]
<i>Skytanthus acutus</i>	Carbon Steel	NaCl	90	[206]
Orange peel	Magnesium alloy	NaCl	85.7	[207]
Nettle leaves	Mild steel	NaCl	95	[208]
<i>Santolina chamaecyparissus</i>	304 stainless steel	NaCl	86.9	[209]
<i>Myrmecodia Pendans</i>	Carbon steel	NaCl	91.41	[210]
<i>Ficus pumila</i> Linn.	XC38 steel	NaCl	91.3	[211]

Normally, HCl, H<sub>2</sub>SO<sub>4</sub>, and H<sub>3</sub>PO<sub>4</sub> are used as acidic corrosion media. These acids are used in the industry in various processes such as descaling, chemical coloring, acid pickling, phosphating, and drilling operations, and metal equipment comes into contact with them [81]. Therefore, more of these environments are used to evaluate corrosion inhibition. In addition to the mentioned media, plant extracts are also used in corrosive media saturated with CO<sub>2</sub> [212]. In this regard, the extracts obtained from many plants such as olive leaf, *Jatropha curcas*, *Coptis chinensis*, *Dimocarpus longan*, tobacco leaf, dandelion, licorice, *Sida acuta*, Kuding, and Pomelo have been investigated and some of them have had acceptable results [135]. In recent years, many studies have been conducted to use plant extracts as GCIs for reinforced concrete structures. Normally, in reinforced concrete, steel undergoes pitting corrosion through contamination with chlorides or carbonation [70]. Hence, it is very important to use a corrosion inhibitor. Kola nut extract has been reported to prevent chloride ions from reaching the surface of the armature [213]. Also, *Bambusa arundinacea* extract prevents the conversion of hydroxide from calcium to calcite and has a pore-blocking effect [214]. Valdez-Salas et al. [215] investigated the effect of *Azadirachta indica* leaf extract on the corrosion inhibition of steel in concrete mortars contaminated by aggressive chloride. The results showed that *Azadirachta indica* does not change the integrity and physicochemical parameters of concrete, and after 182 days of investigation, it showed a long-term protection against corrosion of 95%, which is a similar result to commercial inhibitors. Naderi et al. [216] investigated the effect of licorice plant extract on the corrosion inhibition of steel rebar in concrete contaminated by aggressive chloride. The results showed that the extract has an inhibition efficiency higher than 80%. Polyphenolic compounds such as tannins and flavonoids are abundant in licorice extract, which causes corrosion inhibition. In general, we can say that GCIs are an exciting and partly cost-effective solution for protecting metals. On the other hand, GCIs, especially plant inhibitors, are very popular due to their non-toxicity (sometimes very low toxicity) and eco-friendliness. There are still many species of plants that have not been investigated, but it can be said that, in the not-too-distant future, GCIs will find their real place in the industry.

## 6. Conclusions

To sum up, the use of plants as GCIs was reviewed, and corrosion, corrosion protection, and other members of the GCI category were briefly reviewed. It can be said that the main reason for using GCIs is their availability, and non-toxicity. Although they have become surprisingly popular, there are still some problems associated with their use. Perhaps the most important issues are the inhibitory effect, compatibility with other materials, and cost. As mentioned earlier in this review, the effectiveness of GCIs depends on several factors such as substrate type, corrosive media, temperature, and inhibitor concentration. Hence, it can be said that a GCI can be used depending on the existing conditions. Also, there is a possibility of the incompatibility of GCIs with some coatings or paints. Moreover, for large-scale applications, there is a possibility of high cost to prepare some GCIs. So, it can be said that research efforts should be focused on cost-effectiveness and performance improvement in different conditions. In general, in the near future, GCIs can be considered a green solution to protect metals.

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