



## Review Article

# Eco-friendly inhibitors for corrosion protection of metallic surfaces—a mini review

Milad Edraki<sup>a,b</sup>, Mohammad Banimahd Keivani<sup>c,\*</sup> 

<sup>a</sup> Polymer Department, Technical Faculty, South Tehran Branch, Islamic Azad University, P.O. Box 11365-4435, Tehran, Iran

<sup>b</sup> Department of Industrial Chemistry, Faculty of Rajaie, Lahijan Branch, Technical and Vocational University (TVU), Guilan, Iran

<sup>c</sup> Department of Chemistry, Payame Noor University (PNU), P.O. Box: 19395-4697, Tehran, Iran

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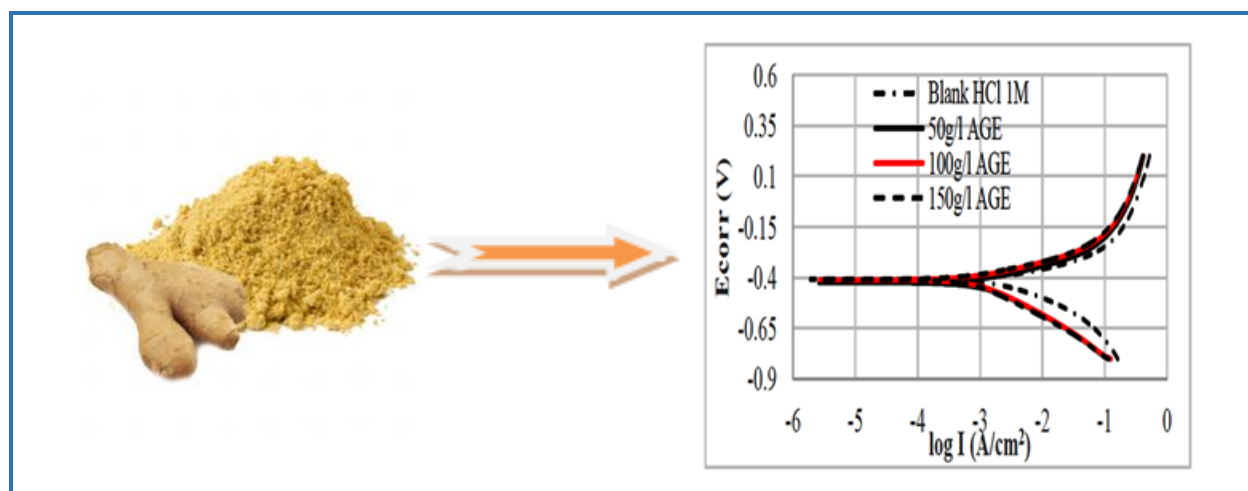
Electrochemical impedance spectroscopy (EIS)

### ABSTRACT

The corrosion of metals has been always one of the crucial issues in industries, causing pollution and waste of products, destruction, and devastation of equipment, leading to considerable human, economic, and environmental damages. Over the past years, several studies have been carried out on the recognition of various kinds of corrosion and their causing factors, and also prevention methods for corrosion and corrosion inhibitors. Currently, many prevention methods of corrosion and inhibitor chemicals are used; however, their side effects on the environment and human health are significant in a long-term exposure. Over the last decades, using natural inhibitors has been replaced by the chemical materials and attracted a great deal of attention from scientists. In the present research, a review was carried out to introduce natural inhibitors as green inhibitors and their uses and reaction mechanisms.

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## Graphical Abstract



## Biographies



2013 Onwards, Faculty member, Payame Nour University (PNU), Tehran center, Iran. 2000-2013, Faculty member, Iranian Academic Center for Education Culture & Research (ACECR), Guilan Branch, Rasht, Iran. The research fields in which he has interest and expertise include, Corrosion, Nanomaterial's synthesis, Formulation and Characterization of paints& surface coatings and polymer nanocomposites, conducting polymer, absorption & adsorption.



**Milad Edraki** currently, he works as a lecturer in the Department of Industrial Chemistry at Technical and Vocational University (Faculty of Rajaie, Lahijan Branch). He received his Ph.D. degree in Polymer Engineering from SouthTehran Branch – Islamic Azad University in January 2019 under the supervision of Associate Professor Davood Zaarei. The research fields in which he has interest and expertise include, Corrosion, Nanomaterial's synthesis, Formulation and Characterization of paints& surface coatings and polymer nanocomposites.

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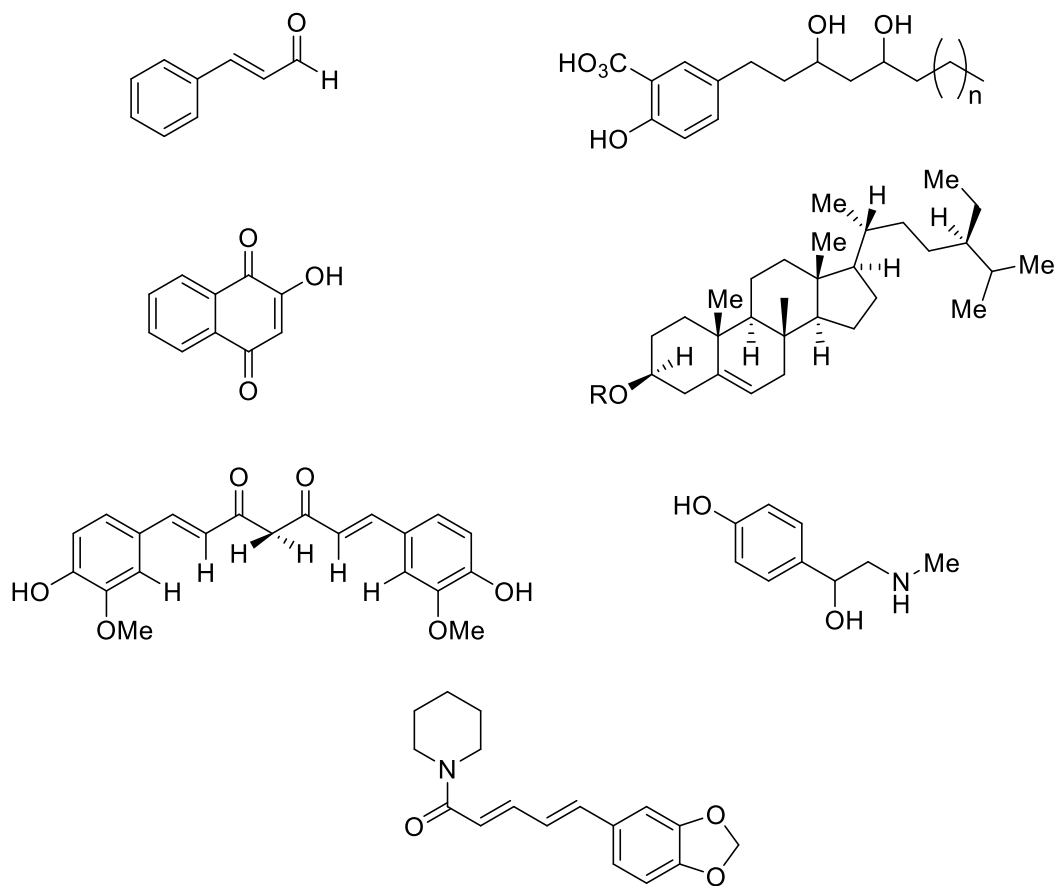
**Introduction**

Corrosion is a chemical interaction within the metal and its environment, causing lots of economic damages in the various industrial processes [1, 2]. Acidic solutions such as sulfuric, hydrochloric, and phosphoric are widely used for pickling steel surfaces to eliminate oxide layers in different industries [1, 3, 4]. In the pickling process, different types of corrosion inhibitors are used to control the dissolution rate of metals because of the high corrosion potential of acidic solutions. The available organic compounds within inhibitors contain sulfur, nitrogen, and oxygen, which have the ability to share their free electrons with empty orbitals of metallic ions on the metal surface. Additionally, the presence of polar groups including, -NH<sub>2</sub>, -CN, C=O, and -OH in the inhibitor molecular structure helps to adsorb inhibitor molecular on the metal surface [1, 5, 6]. Despite the proper function of organic inhibitors, the toxicity of these types of inhibitors has an undesirable environmental effect, so that green inhibitors are now replaced by organic compounds [7–14]. Green inhibitors have rich sources of electron releasing atoms and polar groups, which in addition to lower environmental risks, they can properly protect the metal surface against corrosion [15, 16].

Green inhibitors are biocompatible and do not have an undesirable effect on the environment [17–19]. As it has been mentioned in the previous studies, leaf and cortex of the plants, seed, and root of fruits are renewable sources and appropriate for green inhibitors [17–19]. Most green inhibitors have higher expenses than organic species. Furthermore, these types of inhibitors have rich sources of active groups, which can provide high inhibition efficiency [20–22].

Besides, most green inhibitors are extracted in non-aqueous solvents (such as methanol, ethanol, and hexane), making the extraction process more expensive and non-functional. On the other hand,

methanol, ethanol, and hexane have undesirable environmental effects on surroundings. Availability is another important issue for choosing a proper inhibitor [1, 23, 24]. Research demonstrates that some of the green inhibitors have a high ability to control the corrosion of steel; however, they are not completely available, making them ineffective and obsolete [1, 23, 24]. Nevertheless, in the present study, extracts such as *cinnamon*, *ginger*, *lawsonia inermis*, *nettle*, *turmeric*, *citrus aurantium*, and *black papper* are introduced as strong, available, cheap, and biocompatible inhibitors. Chemical structure of the green inhibitors is shown in Scheme 1.



**Scheme 1.** Schematic structure of a) *cinnamon*, b) *ginger*, c) *lawsonia inermis*, d) *nettle*, e) *turmeric*, f) *citrus aurantium* and g) *black pepper*

### Classification of Inhibitors

Based on the reaction type with active chemical surface areas of metal, inhibitors are divided into three classes, namely anodic, cathodic, and mixed [25]. Polarization method can determine how inhibitors interact with the surface. The basis of polarizing or potential change is as a line of one potential to another potential. During this change, viscosity of the current in accordance with applied voltage will be changed based on what factors control the corrosion process (activation,

concentration, and ohmic) [25]. Any change in viscosity of the charge of the electrochemical process of anodic or cathodic at the presence of an inhibitor is associated with the inhibitory polarizing behavior in the electrochemical process [25]. Therefore, the polarization method is the most essential one in classifying inhibitors into anodic, cathodic, and mixed [25].

### **Anodic Inhibitors**

This type of inhibitor material reduces corrosion by limiting anodic reaction and density of the corrosion current. In fact, what happens in terms of anodic organic substances is the significant polarization of anodic reaction with these compounds [25]. For instance, it has been shown that plant extract of mimosa has anodic inhibition action on the metallic surface of brass in sulfuric acid medium; because as shown in the [Figure 1](#), viscosity loss of current (the crossing of cathodic and anodic branches) was observed in the more positive potentials than corrosion potential [26].

### **Cathodic Inhibitor**

In this inhibitory mechanism, molecules reduce cathodic reaction speed. In other words, they are adsorbed on the cathodic areas, causing significant polarization of cathodic reaction [25]. For instance, it has been represented that the plant extract of giant bamboo has cathodic inhibition on aluminum in the sulfuric acid medium; because according to the [Figure 2](#), the current of viscosity loss is observed in the negative corrosion potential [27].

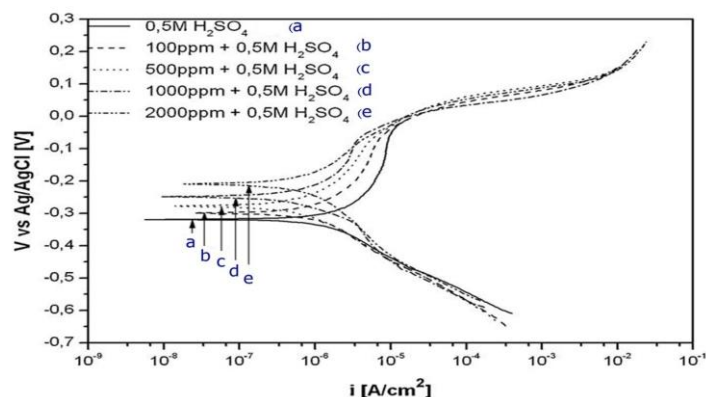
### **Mixed Inhibitor**

In inhibitor molecules, the propagation of electron density leads to molecules adsorption in both anodic and cathodic areas. In this context, electrochemical activities are affected in both anodic and cathodic and both of the reactions are polarized [25]. It has been shown that *Justicia gendarussa* has the mixed inhibition activity on the steel in the hydrochloric acid medium. This is due to the fact that, the viscosity loss of the current is observed in more positive and negative potentials than corrosion potential ([Figure 3](#)) [28].

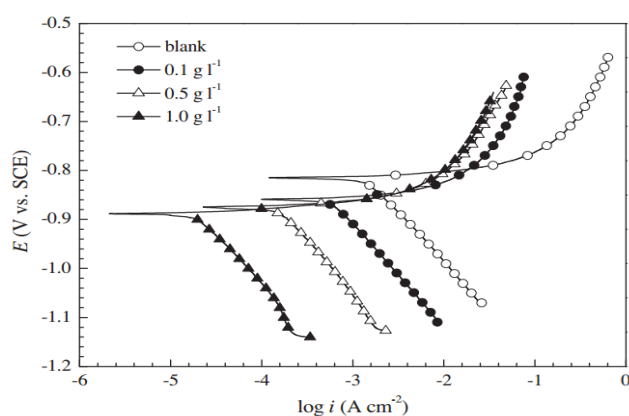
### **Green Inhibitors**

An inhibitor is a substance (or a combination of substances) added in a very low concentration to treat the surface of a metal that is exposed to a corrosive environment that terminates or diminishes the corrosion [29]. These are also known as site blocking elements, blocking species or adsorption site blockers, due to their adsorptive properties [29]. The term “green inhibitor” or “eco-friendly

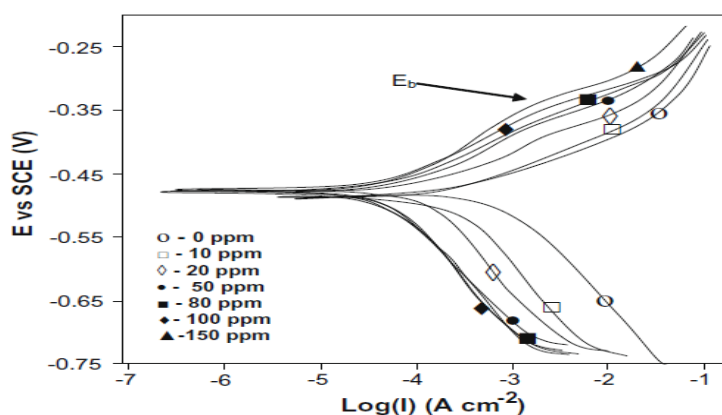
inhibitor” refers to the substances that have high biocompatibility in nature [29]. The inhibitors such as plant extracts presumably possess biocompatibility due to their biological origin.



**Figure 1.** The polarization curve of brass in sulfuric acid medium with and without plant extract of mimosa [26]



**Figure 2.** Tafel curve of aluminum polarization in sulfuric acid medium with or without giant bamboo [27]



**Figure 3.** Tafel curve of steel polarization in hydrochloric acid medium with and without Justicia Gendarussa [28]

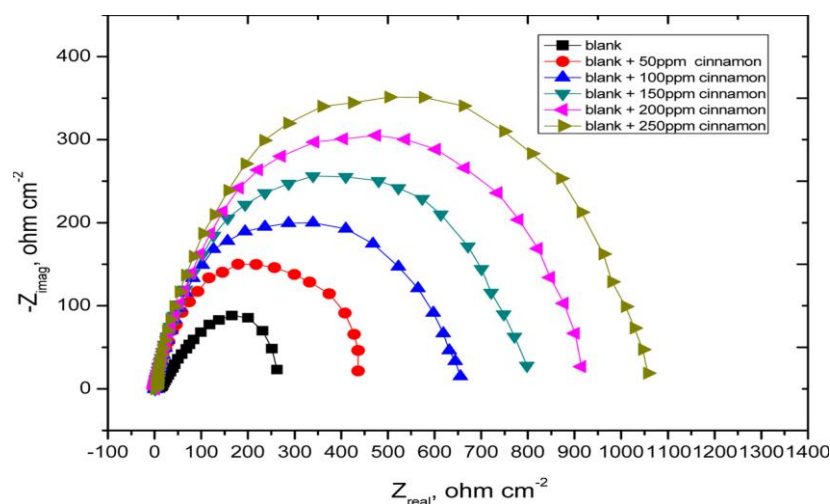
## Cinnamon

Dehghani et al., [1] examined the corrosion inhibitory effect of *cinnamon* extract powder on the carbon steel surface in 1 molar solution of HCl. The investigations of electrochemical impedance spectroscopy (EIS) and potentiodynamic polarization revealed that the inhibitor molecules are able to control the corrosion with cathodic and anodic reactions, and 88% efficiency was obtained after 2.5 h of metal immersion in 1 molar HCl solution containing 800 ppm green inhibitor. Alternatively, the results of scanning electron microscope (SEM) and atomic force microscope (AFM) analysis indicated that the steel has been significantly controlled by increasing an inhibitor concentration to the corrosive solution. Furthermore, it was observed that the molecules of *cinnamon* extract powder have physically and chemically adsorbed by following Langmuir isotherm on the metal surface.

Anupama and Joseph [30] have assessed the corrosion behavior of mild steel with and without *cinnamon* extract powder in different acidic media such as 0.5 molar  $\text{H}_2\text{SO}_4$  and 1 molar HCl by using polarization technique. The results of polarization demonstrated that *cinnamon* extract powder showed the excellent efficiency in both corrosive environments. That is to say, the inhibitory amounts were 99.73% and 96.55% in HCl and  $\text{H}_2\text{SO}_4$ , respectively. Moreover, both anodic and cathodic branches were shifted into lower current density in the presence of *cinnamon* extract powder, showing its mixed inhibition rate (anodic and cathodic). On the other hand, SEM clearly showed that adsorbed film of *cinnamon* extract has formed on the surface, leading increased resistance against corrosion.

Fouda et al., [31] investigated the corrosion inhibitory influence of *cinnamon* extract on the mild steel surface in 3.5% NaCl solution polluted with sulfate. In Figure 4, the results of Nyquist plot showed that the adsorbed molecules of this extract on the mild steel inhibited from dissolution and surface degradation and also resulted in the decreased amount of layer double capacitance ( $C_{dl}$ ) and increased charge transfer resistance ( $R_{ct}$ ). On the other hand, increasing the *cinnamon* inhibitor concentration by 250 ppm resulted in the high inhibition efficiency, reaching 81.3% level.

By using the polarization test, Bouraoui et al., [32] assessed the corrosion behavior of 304 stainless steel in the presence of *cinnamon* oil at 0.5-5% concentration in 0.1 molar HCl solution. The results of polarization showed that, increasing the concentration of the *cinnamon* oil by 5% suppressed the corrosion current density and shifted the corrosion potential to more positive amounts. Increasing corrosion current density and decreasing potential resulted in formation of inhibitor film on steel surface, blocking the active anodic and cathodic sites, and restricting the attack of corrosive species on the surface. Moreover, increasing the mentioned inhibitor concentration caused high inhibition efficiency, reaching 96.20% level.



**Figure 4.** Nyquist Plot of the samples of steel immersed in 3.5% NaCl solution polluted with sulfate containing different concentrations of *cinnamon* extract [31]

### Ginger

In another study, *Fouda et al.*, [33] investigated the *ginger* extract on mild steel corrosion in 3.5% NaCl solution polluted with sulfate. The results demonstrated that increasing concentration of *ginger* extract by 250 ppm caused low corrosion rate as 19.11 mm/year while the amount of surface coverage and inhibition efficiency increased and became 0.839 and 83.9%. The temkin adsorption isotherm clearly indicated that inhibitor molecules of *ginger* on the metal surface were physically adsorbed by electrostatic interaction.

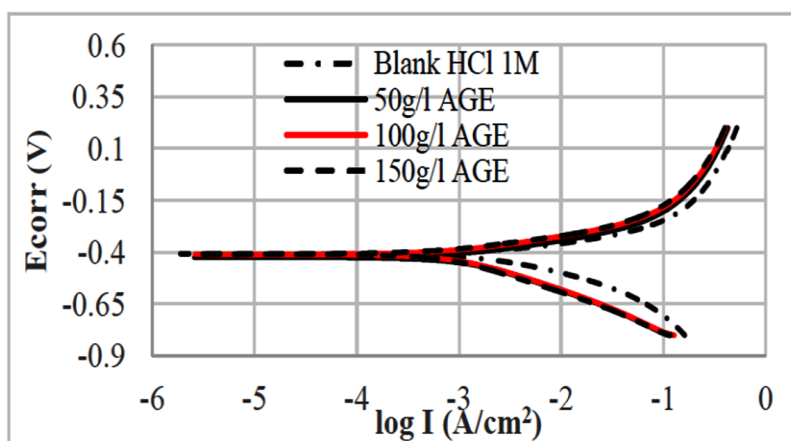
*Fidrusli et al.*, [34] carried out a research on the *ginger* extract on mild steel in 1 molar HCl solution and used the polarization method. According to Figure 5, the polarization results show that with the increase of *ginger* concentration to 150 g/L, the corrosion current density was decreased and inhibition efficiency was increased and were 0.054 mA/cm<sup>2</sup> and 86%, respectively.

*Al Fakih et al.*, [35] examined the effect of *ginger* extract on the corrosion behavior of mild steel in 1 molar HCl solution. The polarization results and weight loss indicated that by increasing the concentration of *ginger* inhibitor to 10 g/L, the inhibition efficiency was remarkably increased and reached 90%. Furthermore, based on Figure 6, SEM images represent the steel samples immersing in the acidic corrosive environment was suffered from corrosion without inhibitor. However, the corrosive solution with inhibitor prevented from corrosion due to the adsorption of the *ginger* molecule on the metal surface and creation of a protection layer.

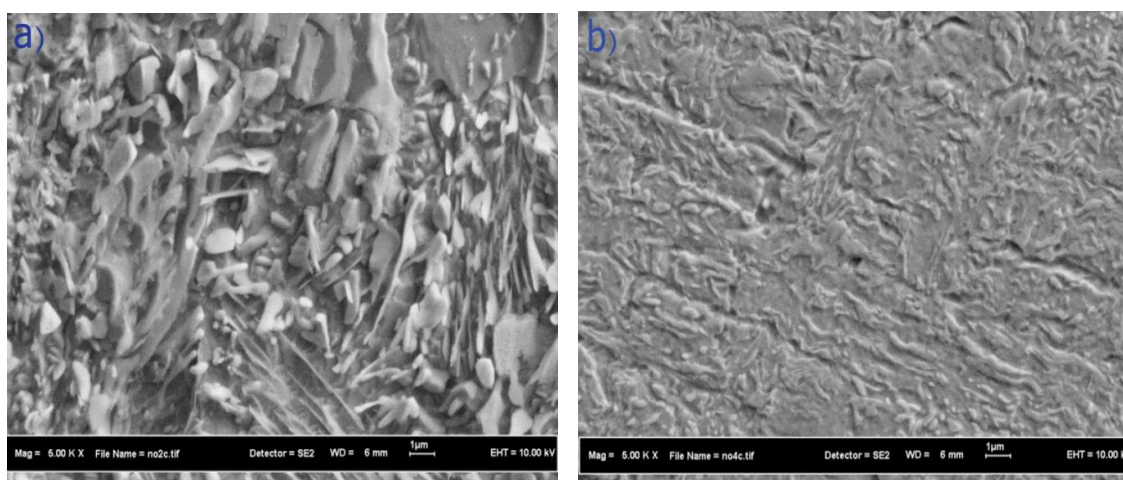
Gado and Motawea [36] conducted a research to examine the root extract of *ginger* on carbon steel corrosion in 1 molar HCl medium. The results of the EIS demonstrated that with the increase of mentioned inhibitor concentration up to 200 ppm, the level of charge transfer resistance ( $R_{ct}$ )



enhanced; however, admittance ( $Y_0$ ) and layer double capacitance ( $C_{dl}$ ) were decreased. The low level of these parameters  $Y_0$  and  $C_{dl}$  can be associated with the formation of inhibitor film or increased resistance against the corrosion.



**Figure 5.** The polarization curve associated with the samples of mild steel immersing in the different concentrations of *ginger* extract [34]



**Figure 6.** The SEM images of mild steel surface after 2 hours immersion in acidic solution: a) without inhibitor, b) with inhibitor [35]

### Other Green Inhibitors

In the distinct research, Eltre [37, 38] has examined and studied the anti-corrosion function of Indian *ginger* extract on aluminum substrate in the acidic environment and inhibitory properties of organic honey on a copper substrate in 0.5 molar NaCl medium. The results of weight loss and polarization tests indicated that Indian *ginger* extract shows effective inhibition and provides protection against pitting corrosion in the acidic solution. The findings obtained from mentioned

tests on organic honey showed that the inhibition action of this organic inhibitor gradually decreased in the neutral environment [37, 38]. Jokar et al., [39] investigated the inhibition effect of the extract taken from berry's leaf as a green inhibitor on steel corrosion in 1 molar acidic environment of HCl. The results of EIS represents 93% efficiency of this compound. Adding potassium iodide to this compound caused a 3% increase in inhibition efficiency, associating with the creation of a synergistic effect.

Sanaei et al., [40] synthesized a hybrid inhibitor based on the zinc acetate/chicory and investigated its anti-corrosion action on mild steel in 3.5% NaCl medium by using electrochemical tests and surface analysis. The results of the polarization analysis showed that the available oxygenated compounds in the organic inhibitor made a strong bond with zinc cations of mineral part, and both anodic and cathodic reactions were polarized with this pigment. Also, the corrosion resistance has been increased in a salt solution saturated with this pigment. Furthermore, surface analysis is the creation of a compressed film containing adsorption of organic molecules inhibitors through the creation of a bond with zinc/iron cations and approves the zinc hydroxide sediments on the cathodic areas.

Research such as that conducted by Nasibi et al., [41] demonstrated the inhibitory role of *nettle* extract on mild steel in 1 molar HCl solution. The results of this research indicated that the inhibitor being studied acts as a mixed inhibitor and decreases the corrosion rate of anodic and cathodic reactions by adsorption and forming a thin film on the metal surface, which has barrier properties. Regarding the high volume of studies conducted on the mechanism of green inhibitor, the most important actions of them have been shown in Table 1.

**Table 1.** The mechanism of action of green corrosion inhibitors in the different corrosive environments

Green Corrosion inhibitor	Type of inhibitor	Type of corrosive media	Type of metal	Maximum inhibition efficiency (%)	Adsorption mechanism	Ref.
Glycyrrhiza glabra leaves extracts	mixed	1 M HCl	Mild Steel	88	Physical and chemical adsorption	42
Persian liquorice extract	mixed	3.5 % NaCl	Mild Steel	98.8	Film formation on the active sites	43
Chinese gooseberry fruit shell extract	mixed	1 M HCl	Mild Steel	92	Physical adsorption	44

Lawsonia enermis	mixed	3.5 % NaCl	Aluminum	93	Physical adsorption	<a href="#">45</a>
Lawsonia inermis	mixed	0.5 M acetic acid	Mild Steel	86.27	Physical adsorption	<a href="#">46</a>
Lawsonia inermis	mixed	0.5 M HCl	Mild Steel	88.26	Physical adsorption	<a href="#">47</a>
Turmeric extract	mixed	3 % NaCl	Carbon Steel	78	Physical and chemical adsorption	<a href="#">48</a>
Turmeric extract	mixed	3.5 % NaCl	Mild Steel	81.6	Physical and chemical adsorption	<a href="#">49</a>
Turmeric extract	mixed	1 M HCl	Mild Steel	63	Physical and chemical adsorption	<a href="#">50</a>
Turmeric extract + Zh	mixed	3.5 % NaCl	Carbon Steel	93	Adsorption due to the formation of protective layer of corrosion inhibitor	<a href="#">51</a>
Juglans regia fruit shell extract	mixed	3.5% NaCl	Mild Steel	94	Adsorbed electrostatically and covalently on the metal surface	<a href="#">52</a>
Citrus aurantium leaves extracts	mixed	1 M H <sub>2</sub> SO <sub>4</sub>	Mild Steel	88.8	Physical adsorption	<a href="#">53</a>
Citrus aurantium leaves extracts	mixed	0.5 M H <sub>2</sub> SO <sub>4</sub>	Mild Steel	96.46	Physical adsorption	<a href="#">54</a>
Black pepper extract	mixed	1 M HCl	Mild Steel	98	chemical adsorption	<a href="#">55</a>
Black pepper extract	mixed	1 M HCl	Aluminum	97.66	chemical adsorption	<a href="#">56</a>
Black pepper extract	mixed	1 M HCl	Carbon Steel	95.8	chemical adsorption	<a href="#">57</a>
Primula Vulgaris flower aqueous extract	mixed	1 M HCl	Carbon Steel	95.5	Physical and chemical adsorption	<a href="#">58</a>
Nettle leaves extract+Zn <sup>2+</sup>	mixed	Chloride Solution	Carbon Steel	96	Adsorbed to surface of steel through	<a href="#">59</a>

					electronic donor-acceptor mechanism	
Thymus Vulgaris plant extract	mixed	1 M HCl	Stainless steel 304	62.75	Physical and chemical adsorption	60

## Conclusions

The extracts of plants can control the corrosion of different areas of metals by inhibitory mechanisms (anodic, cathodic, and mixed) in corrosive environments. These substances are mainly adsorbed by Langmuir adsorption isotherm on the metal surface, preventing the access of corrosive species to the surface by isolating water molecules from the metal surface. With respect to the high efficiency of some plants extract for controlling corrosion of some electrolyte/electrodes systems, these compounds can be used as a replacement of most harmful industrial organic inhibitor compounds for the environment.

## Disclosure Statement

No potential conflict of interest was reported by the authors.

## Orcid

Mohammad Banimahd Keivani  0000-0003-2481-778X

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