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Evaluation of the use of species of Brazilian flora to inhibit corrosion of carbon steel in acidic medium: a review

Avaliação do uso de espécies da flora brasileira na inibição da corrosão do aço carbono em meio ácido: uma revisão

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RESUMO

Os inibidores são um dos métodos mais antigos de combate à corrosão, que ainda hoje é um problema desafiador e caro. Porém, os inibidores tradicionais são tóxicos e não biodegradáveis, o que vai contra os princípios da Química Verde, assim denominada por valorizar o uso de produtos biodegradáveis e ambientalmente amigáveis. Os inibidores podem ser obtidos de plantas e seus resíduos que, quando submetidos a algum tipo de processamento, geram extratos ou óleos com importante potencial inibitório. Na maioria dos casos, os inibidores verdes são compostos orgânicos classificados, quanto ao mecanismo, como mistos, possuindo grupos funcionais polares que atuam como centros de adsorção na superfície metálica. Este estudo teve como objetivo realizar uma revisão sistemática da literatura sobre inibidores de espécies da flora brasileira e utilizados em ambiente ácido para prevenir a corrosão em aço carbono. O Brasil apresenta uma rica biodiversidade de flora, com mais de 46.200 espécies catalogadas em 2020, o que proporciona um amplo campo de pesquisa nesta área. A revisão incluiu 28 espécies de plantas diferentes. Os resultados encontrados foram promissores, com destaque para o uso de partes de vegetais que normalmente são descartadas e mostram o potencial de desenvolvimento e aprimoramento de inibidores verdes para futura aplicação em escala industrial.

Palavras-chave: inibidor verde; flora brasileira; aço carbono; meio ácido.

ABSTRACT

Inhibitors are one of the oldest methods of mitigating corrosion, which is still a current challenging and costly problem. However, traditional inhibitors are toxic and non-biodegradable, which goes against the principles of Green Chemistry, so named for valuing the use of biodegradable and environmentally friendly products. They can be obtained from plants and their residues that, when subjected to some type of processing, generate extracts or oils with important inhibitory potential. In most cases, green inhibitors are organic compounds classified relating to the mechanism as mixed, having polar functional groups that act as adsorption centers on the metallic surface. This study aimed to carry out a systematic review of the literature on inhibitors from species of Brazilian flora and used in an acid environment to prevent corrosion in carbon steel in a specific medium. Brazil presents a rich biodiversity of flora, with more than 46,200 species cataloged in 2020, which provides a wide field of research in this area. The review included 28 different plant species. The results found were promising, with emphasis on the use of parts of vegetables that are normally discarded and show the potential for development and improvement of green inhibitors for future application on an industrial scale.

Keywords: green inhibitors; Brazilian flora; mild steel; acidic medium.

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1. INTRODUCTION

Corrosion is the spontaneous process of degradation of a material, usually metallic, by physical, chemical, or electrochemical action, causing the loss of its design properties, being, therefore, an undesirable occurrence. In an industrial environment, this process of deterioration has several implications such as: changing damaged equipment, scheduled maintenance routine, product contamination, and drop in process efficiency [1]. In a report published by the NACE International it was estimated that the global cost of corrosion corresponded to 3.4% of the world Gross Domestic Product (GDP), in 2013 [2]. Analyzing the panorama in Brazil, a study by the International Zinc Association (IZA), in partnership with the University of São Paulo (USP), pointed out that corrosion consumed 4% of the country's GDP in 2015 [3].

This is a continuous challenge to minimize corrosion and its negative impact on the economic and environmental spheres. Currently, there is a wide range of corrosion control methods, which are based on modification of the process and of the corrosive environment, on anodic or cathodic protection, and on protective coatings. Only the individual analysis of each case of corrosion, defining aspects such as mechanism, media, and morphology, can lead to the conclusion of which are the best methods to combat it, as there are no generalized solutions [1, 4]. Inhibitors, which are the focus of this article, are one of the oldest methods of controlling corrosion, based on the modification of the medium. Corrosion inhibitors are substances that, when added in small quantities to a potentially corrosive environment for the metal, will decrease the corrosion rate [5].

Several studies of corrosion inhibitors have adopted the acidic environment, with this choice having a practical basis. The use of an acid solution promotes the pickling of the metal surface. In several industrial processes, metal corrosion occurs due to contact with acid solutions [6]. Pickling consists of immersing the metal part in an acidic solution to remove incrustations of oxides or salts from its surface, enabling the execution of finishes, such as painting. Hydrochloric acid is one of the most used in pickling baths, in which inhibitors are added to minimize corrosion of the metal, which can be attacked after dissolving the encrusted layer [7]. Another relevant example is found in the oil industry, in the oil and gas extraction stage, specifically in the operation called matrix acidification. In this operation, an acid solution, consisting of hydrochloric acid or a mixture of it with hydrofluoric acid, is used to dissolve minerals from the interposed rock formation and increase the system's permeability. Corrosion inhibitors are added to the solution to maintain the integrity of the metallic materials present in the well structure [8]. A daily example of the use of acidic solutions is in the cleaning of metallic surfaces, in which formulations that combine the acid with wetting substances and detergents and promote the removal of contaminants. For this application, sulfuric and hydrochloric acids are the most used, as they can be recovered [7]. Hydrochloric acid generates localized corrosion on passivating metals and sulfuric acid produces generalized corrosion on metal surfaces. Once again, the use of corrosion inhibitors in the cleaning solution is necessary for the preservation of the metal [5, 9]. The acidic medium is used in several studies even though it is not the medium of service of the material, as it is an aggressive medium, accelerating the corrosion evaluation tests.

Considering the mechanism, interface inhibitors, which are those in which a film is formed on the metal surface, in the liquid phase, are classified as: anodic, cathodic, or mixed. Anodic inhibitors slow down the oxidation reaction of the metal, through the production of a passive layer, with chromates, tungstates, and molybdates being some of the substances that exhibit this behavior. Cathodic inhibitors act by inhibiting the reduction reaction, which may hinder the recombination of hydrogen on the metal surface, promote passivity through the precipitation of insoluble compounds, or even react with the dissolved oxygen in the medium. Selenides, bismuth, antimony, sulfates, sulfites, and hydrazine are some substances used as cathodic inhibitors. However, most inhibitors are organic compounds that, in turn, cannot be characterized as anodic or cathodic, being designated as a mixed inhibitor. In this case, the process responsible for decreasing the corrosion rate is the adsorption of organic compounds on anodic and/or cathodic areas of the metal surface. This protection process can take place in three different ways: physisorption, which occurs by electrostatic or Van der Waals forces, chemisorption, which involves charge transfer or sharing, or film formation, due to surface reactions. In acidic media, mixed inhibitors are often used, in which the hydrophilic functional group of the organic compound binds to the metal, while the hydrophobic group limits water access to it. Urea, amines, aldehydes, and nitrogenous heterocyclic compounds are some examples of mixed inhibitors [1, 10–12].

Traditional corrosion inhibitors are non-biodegradable and highly toxic compounds, which goes against the environmental sustainability principles of Green Chemistry, so named for valuing the use of biodegradable and environmentally friendly products. Green Chemistry is the design of chemical products and processes that reduce or eliminate the use or generation of hazardous substances. In this context, there is a growing demand for so-called green inhibitors, which generate little or no negative impact on the environment [13]. Green inhibitors can be inorganic [14], such as rare earth metals [15], or organic [14], such as ionic liquids [16], biopolymers [17], surfactants [18], amino acids [19], drugs [20], plant extracts [13], and oils [21]. It is known that about 80% of organic green inhibitors are classified as mixed, a condition in which their compounds have polar functional groups that act as adsorption centers on the metallic surface. The presence of heteroatoms of N, O, S, P, Se and As, in addition to the possible occurrence of unsaturation, characterize these functional groups, being some examples: –OH, $–NH_2$, $–NO_2$, –C≡C–, $–CONH_2$, –COOH, –N=N–N–, –S– and –P–. It is also common for organic green inhibitors to have heterocyclic aliphatic or aromatic rings, which can also act as nuclei of interaction with the metal [14]. Substances such as phenolic compounds, catechins, flavonoids, alkaloids, terpenoids, and tannins, which have a favorable chemical structure, can be obtained from plant extracts, and inhibit corrosion of various metals in an acidic medium [14]. The nature of the inhibition process can be better understood by adjusting pairs of surface coverage data and inhibitor concentration to classic models of adsorption isotherms, such as Langmuir, Temkin, Frumkin, Freudlich, Virial Parson, and Bockris-Swinkels [22]. To evaluate a given compound as a corrosion inhibitor, or even compare it with another, it is necessary to quantify the inhibition efficiency (η). In studies involving green inhibitors, this efficiency is usually calculated by measures of mass loss by total immersion (MLTI), Potentiodynamic Polarization (PP), and Electrochemical Impedance Spectroscopy (EIS), according to Equations 1–3, respectively [14].

$$\eta_{PMIt} = \frac{w_0 - w_1}{w_0} \times 100\% \tag{1}$$

$$\eta_{PP} = \frac{j_{corr\ (0)} - j_{corr\ (1)}}{j_{corr\ (0)}} \times 100\% \tag{2}$$

$$\eta_{EIE} = \frac{R_{tc(1)} - R_{tc(0)}}{R_{tc(0)}} \times 100\% \tag{3}$$

where w = corrosion rate, j_{corr} = corrosion current density, R_{tc} = resistance to charge transfer, sub-index 0 → absence of inhibitor and sub-index 1 → presence of inhibitor.

According to data from the National Flora Conservation Center (CNCFlora), there are more than 46,200 species that make up the Brazilian flora in 2020, including native, cultivated, and naturalized species [23]. All of this biodiversity provides a huge research field for green corrosion inhibitors to be explored in Brazil.

2. THE USE OF SPECIES OF BRAZILIAN FLORA IN THE CORROSION INHIBITION OF CARBON STEEL IN ACID MEDIUM

To carry out a systematic review of the literature on the evaluation of the use of species of Brazilian flora to inhibit corrosion of carbon steel in an acidic environment, some search restrictions were established to delimit the theme. One of them is that the process of obtaining the inhibitor was of a physical nature, not involving chemical reactions of any kind. Therefore, studies that investigate plant extracts, vegetable oils, and products derived from other types of physical processing of species, such as crushing or pressing, are included. However, the works that carry out the saponification of vegetable oil, for example, were not considered. The other constraint is that there was a calculation of the inhibition efficiency to allow comparative analyzes.

Literature in the past 15 years studied corrosion inhibitors from 28 different plant species from Brazilian flora. A summary of the data from these articles is shown in Table 1.

The selected papers have similar methodological construction. In general, there is a stage of obtaining the inhibitor from the plant species, as shown in Figure 1, and then, corrosion tests are carried out, as shown in Figure 2. Finally, the results are analyzed from a qualitative point of view, through comparative observations, and quantitative, through the calculation of the inhibition efficiency. Some studies have carried out techniques for the characterization of the inhibitor obtained to verify the presence of substances with the potential to reduce corrosion. Other works sought to understand the inhibition mechanism, testing the correspondence of its experimental data with the classic models of adsorption isotherms. But a very frequent approach was the evaluation of the influence of different process factors on the inhibition efficiency, such as the method of inhibitor production, its concentration in the medium, time and temperature in the MLTI tests, among others. Next, the main fundaments and results portrayed in the articles of this review are discussed.

Babassu coconut oil (*Orbignya oleifera*) contains fatty acids such as oleic, lauric, and myristic acids. Due to the presence of the –COOH group at one end of the carbon chain, these substances would be able to

Table 1: Literature about the use of species of the Brazilian flora as steel corrosion inhibitors in acidic media.

REFERENCE	SPECIE	MEDIUM	STEEL TYPE	OBTAINING PROCESS	CORROSION TEST
[24]	Babassu coconut (Attalea speciosa)	HCl 1 mol/L	AISI 1020	Cold pressing	PP; EIS
[25]	White tea (Camellia sinensis (L.) Kuntze)	HCl 1 mol/L	AISI 1020	Infusion	MLTI; PP; EIS; SMA
[26]	Fresh leaves and dried buttercup flowers (*Unxia kubitzkii* H. Rob.)	HCl 1 mol/L	P110	Maceration; Reflux extraction	MLTI
[27]	Black pepper (Piper nigrum)	HCl 1 mol/L	AISI 1020	Soxhlet extraction	MLTI
[28]	Grape marc	HCl 1 mol/L	AISI 1020	Hydroalcoholic extraction	PP; EIS; SMA
[29]	Hummingbird Hibiscus Leaves (*Malvaviscos arboreus*)	H_2SO_4 1 mol/L	AISI 1020	Maceration	MLTI
[30]	Silver banana peel (*Musa* AAB subgrupo Prata)	HCl 1 mol/L	AISI 1020	Maceration	MLTI; PP; SMA
[31]	Green tea (Camellia sinensis (L.) Kuntze)	HCl 1 mol/L	AISI 1020	Infusion; Ultrasound extraction; Soxhlet extraction	PP; EIS
[32]	Yerba mate leaves (*Ilex paraguariensis*); Gorse leaves (Baccharis trimera)	HCl 1 mol/L	AISI 1020	Infusion; Ultrasound extraction	PP; EIS
[33]	Cranberry leaf tea and flour	HCl 1 mol/L	AISI 1020	Decoction	PP; EIS
[34]	White and green tea (Camellia sinensis (L.) Kuntze)	HCl 1 mol/L	AISI 1020	Decoction	MLTI; PP; EIS
[35]	Murici leaves (Byrsonima sericea)	HCl 1 mol/L	AISI 1040	Maceration	MLTI; PP; SMA
[36]	White tea (Camellia sinensis (L.) Kuntze)	HCl 1 mol/L	P110	Soxhlet extraction	MLTI; PP; EIS
[37]	Orange skin; Mango peel; Passion fruit peel; Cashew shell; Grape marc	HCl 1 mol/L	SAE 1020	Hydroalcoholic extraction; Infusion; Soxhlet extraction	MLTI; SMA
[38]	Coffee leaves (Coffea sp.)	HCl 1 mol/L	SAE 1020	Infusion	MLTI
[39]	Palm Oil Pie (*Elaeis guineenses* Jacq.)	HCl 0,5 mol/L	ASTM 1020	Milling	MLTI; PP; EIS; SMA
[40]	Papaya seed (Carica papaya)	HCl 1 mol/L	1020	Infusion	MLTI; PP; EIS; SMA
[41]	Olive leaves (Olea europaea L.)	HCl 1 mol/L	AISI 1020	Water bath extraction	MLTI; PP; LPR; EIS; SMA
[42]	Precious bark stem (*Aniba canelilla* (HBK) Mez)	H_2SO_4 1 mol/L	UNS G10200	Soxhlet extraction	MLTI; PP; EIS
[43]	Bilberry leaves (*Plectranthus barbatus* Andrews)	HCl 1 mol/L; H_2SO_4 0,5 mol/L	P110	Maceration; Decoction	MLTI

REFERENCE	SPECIE	MEDIUM	STEEL TYPE	OBTAINING PROCESS	CORROSION TEST
[44]	Coffee leaves (Coffea sp.)	HCl 1 mol/L	AISI 1020	Water bath extraction	EIS
[45]	Bark of the ipe-purple stalk (*Tabebuia impetiginosa*)	HCl 1 mol/L	P110	Decoction	MLTI
[46]	Green Yerba Mate (*Ilex paraguariensis*)	HCl 1 mol/L	SAE 1020	Infusion	MLTI; PP; EIS; AMS
[47]	Garlic peel	HCl 1 mol/L	SAE 1020	Infusion	MLTI; PP; EIS; SMA
[48]	Orange skin; Mango peel; Passion fruit peel; Cashew	HCl 1 mol/L	SAE 1020	Infusion	MLTI; PP; EIS; SMA
[49]	Commercial ground coffee (*Coffea arabic* e *Coffea canephora*)	HCl 1 mol/L	SAE 1020	Decoction; Infusion	MLTI; PP; EIS; SMA
[50]	Grape marc	HCl 1 mol/L	SAE 1008	Hydroalcoholic extraction	MLTI; PP; EIS; SMA
[51]	Orange skin; Mango peel	HCl 1 mol/L	SAE 1020	Soxhlet extraction	MLTI; PP; EIS
[52]	White tea (Camellia sinensis (L.) Kuntze)	HCl 1 mol/L	P110	Soxhlet extraction	MLTI
[53]	Coffee grains (Coffea canephora)	HCl 1 mol/L	SAE 1020	Infusion	MLTI; PP; EIS; AMS
[54]	Coffee leaves (*Coffea sp.*)	HCl 1 mol/L	AISI 1020	Extraction	MLTI; PP; EIS; SMA
[55]	Papaya seed papaya (*Carica papaya*)	HCl 1 mol/L	AISI 1020	Infusion; Maceration	MLTI; PP; EIS; SMA
[56]	Castor peel (Ricinus communis)	HCl 0.5 mol/L	AISI 1020	Maceration	MLTI; PP; EIS; SMA
[57]	Avocado seed (Persea Americana)	HCl 0.5 mol/L	SAE 1008	Maceration	MLTI; PP; EIS; SMA
[58]	Cocoa Almond Shell (Theobroma cacao L.)	HCl 0.5 mol/L	SAE 1008	Maceration	MLTI; PP; EIS
[59]	Guarana peel (Paullinia Cupana)	HCl 1 mol/L; H_2SO_4 0.5 mol/L	Medium carbon steel	Milling	MLTI; ENT; SMA

MLTI = Mass loss by total immersion; PP = Potentiodynamic Polarization; LRP = Linear resistance polarization; EIS = Electrochemical impedance spectroscopy; ENT = Electrochemical Noise Technique; SMA = Surface morphological analysis

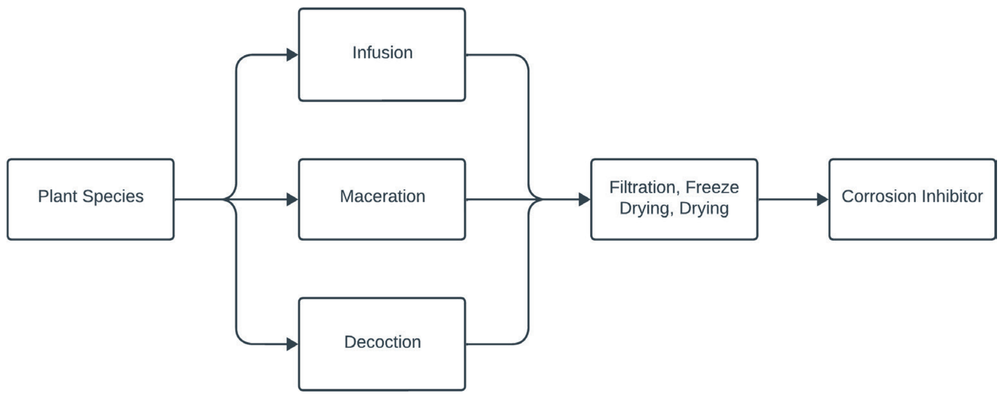

Figure 1: Methodology of obtaining a corrosion inhibitor.

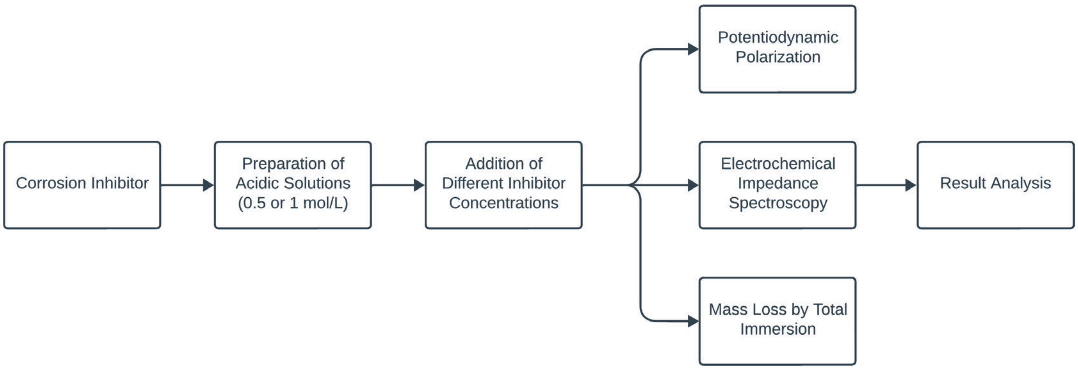

Figure 2: Testing procedure of corrosion inhibitors.

adsorb on the metal surface, replacing water molecules, which allows this inhibitor to be classified as mixed. The parameter varied by PERES *et al.* [24] was the concentration of the inhibitor in the corrosion medium, with the values of 3, 4 and 5 g/L being adopted. A reduction in the inhibition efficiency was observed with the increase of this concentration, including the intensification of the corrosion of the metal in the medium with 5 g/L of inhibitor in comparison with the medium in the absence of this substance. Among the experimental conditions tested, the highest inhibition efficiency achieved was 56.45%, obtained by using the EIS analysis, for an inhibitor concentration of 3 g/L [24].

Camellia sinensis (L.) Kuntze is a plant from which it is possible to obtain four types of teas, according to the drying process of the vegetable parts, namely: white tea, green tea, black tea, and oolong tea. It has alkaloids such as caffeine, and polyphenols such as catechins, flavonoids, anthocyanins, and phenolic acid. The inhibitory activities of the extracts of white and green tea were researched, and it was demonstrated that the inhibition efficiency increases with the growth of the concentration of these products in the corrosive environment. TEIXEIRA *et al.* [25], when studying the aqueous infused extract of white tea, found that the increase in temperature in the MLTI assay caused a decrease in the inhibition efficiency, which suggested a mechanism of physical adsorption. The highest inhibition efficiency achieved was 89%, in PP and EIS assays, for an inhibitor concentration of 1500 ppm. D'ELIA *et al.* [31] compared the following green tea extraction methods: aqueous infusion, acid infusion, acidic ultrasound and in a Soxhlet device. The best method was the extraction in a Soxhlet apparatus, which provided an inhibition efficiency of 93.7%, measured by using the EIS analysis, for an inhibitor concentration of 500 ppm [31]. The data adjusted well to the Langmuir isotherm [31]. TEIXEIRA *et al.* [34] observed higher values of inhibition efficiency for shorter extraction times by decoction of white and green teas, with the polarization curves indicating that the inhibitors would be mixed. The highest inhibition efficiencies achieved were 92.37% for white tea extract at a concentration of 40 mL of inhibitor per 100 mL of 1 mol/L HCl, measured by using a PP assay, and 94.76%, for green tea extract in a concentration of 40 mL of inhibitor per 80 mL of 1 mol / L HCl, measured in an MLTI assay (24 h; room temperature, both for an extraction time of 30 min. OLIVEIRA and CARDOSO [36, 52] achieved a maximum inhibition efficiency of 95% for white tea extract at a concentration of 300 ppm, obtained by using the EIS analysis; the inhibition mechanism being explained by the adsorption of metabolites on the metal surface.

The buttercup (*Unxia kubitzkii* H. Rob.) had two of its parts evaluated by FERNANDES *et al.* [26]: fresh leaves and dried flowers. The extraction of the leaves was by maceration and that of the flowers was under reflux, using distilled water or ethanol as solvent. Numerous secondary metabolites were identified in the investigated products, with emphasis on the tannins in the aqueous extracts of both parts, and the phenolic compounds in the ethanolic extract of the leaves, whose chemical structures favor the adsorption on the metal surface. The study varied the concentration of inhibitor in the medium in the values of 1, 5, and 15% v/v, having observed an increase in the inhibition efficiency with the growth of this concentration. The inhibition efficiency of the ethanolic extract of the flowers is much lower than that of the ethanolic extract of the leaves. Considering the set of conditions tested, the highest inhibition efficiency achieved was 93.67%, obtained in an MLTI test (2 h; 25 ºC), for an inhibitor concentration of 15% v/v [26].

Black pepper (*Piper nigrum*) is a well-known spice whose extract contains a combination of alkaloids that play an important anti-corrosive action, with piperine being the main constituent. ASSIS *et al.* [27] varied the concentration of crude black pepper extract in the range of 1000-6000 mg/L and of isolated piperine in the range of 60-360 mg/L, to compare their inhibitory effects by using MLTI assays (24 h; T_{amb}). Regarding the crude extract, the inhibition efficiency was not affected by the inhibitor concentration in the tested range, having stabilized at 97% from 2000 mg/L. In the case of piperine, there was an oscillation in the inhibition efficiency, with an excellent result of 72%, for the concentration of 180 mg/L. The authors attributed the better performance of the crude extract, compared to pure piperine, to the presence of other substances that contribute to the prevention of corrosion in its composition. Regarding the mechanism of inhibition of piperine, the data showed good correspondence with the Langmuir isotherm and the standard free energy value of adsorption (ΔGads) suggested a process of chemisorption [27].

The grape, when processed for the production of wine and juice, generates a residue composed of skin, seeds, and stem, which is called grape marc. The research carried out the hydroalcoholic extraction of this vegetable residue, subjecting the final product to concentration in a nanofiltration system, to obtain two types of grape marc extracts: crude and concentrate. The higher inhibition efficiencies were associated with higher inhibitor concentrations. ROCHA *et al.* [28] achieved a maximum inhibition efficiency of 92%, by using the EIS test, for 3% v/v of concentrated grape marc extract. ROCHA and GOMES [37] achieved 97% inhibition efficiency, in an MLTI test (24 h; 25 ºC), for 2% v/v of concentrated grape marc extract. This was also the best result of ROCHA *et al.* [50], who also found that the inhibition efficiency increased with the increase of time, from 4-24 h, and decreased with the temperature growth, in the range of 25-60 ºC, in tests of MLTI. This study highlighted flavonoids as substances responsible for the adsorption process, which adjusted well to a Langmuir isotherm.

The hummingbird hibiscus (*Malvaviscus arboreus*) is a shrub widely used as an ornamental plant. VALBON *et al.* [29] identified the presence of porphyrin class metabolites in the leaf extract of this species, which would explain the chelating behavior concerning iron ions. The extract was tested in concentrations of 35, 50, 125 and 500 ppm, in MLTI assays, with an increase in inhibition efficiency with the increase of the inhibitor content. The duration of these tests was also varied, in the values of 3, 24 and 48 h, for a temperature of 308 K, showing a reduction in the inhibition efficiency with increasing time, an effect that is attenuated in higher concentrations. Another factor evaluated was the temperature of the MLTI test, which was examined through its variation in the values of 308, 318, 328, and 338 K, for an inhibitor concentration of 500 ppm and a time of 3 h, occuring a drop in inhibition efficiency for temperatures above 328 K. The highest inhibition efficiency achieved in the study was 97.5%, obtained in MLTI assays (3, 24, and 48 h; 308 K), for an inhibitor concentration of 500 ppm. Regarding the mechanism, the Langmuir isotherm was the one with the best suitability.

The banana is one of the most produced fruits in Brazil, and its peel is a part that is not generally used. EURIDES *et al.* [30] crushed the silver banana peel (*Musa* AAB subgroup Silver) and extracted its components by maceration in water, testing the product obtained in the range of 0.4-1.0 g/L, in an acidic medium. In MLTI assays (48 h; Also), a saturation of inhibition efficiency around 90% was noted for inhibitor concentrations greater than 0.6 g/L. The characterization of the extract indicated the presence of polyphenolic compounds, such as galocatechin, and catechin. The result of the polarization curves led the authors to classify this inhibitor as cathodic.

Yerba mate (*Ilex paraguariensis*) is a plant typically consumed as a tea. One of the studies, by SOUZA *et al.* [46], Measured a significant concentration of total phenolic compounds in the aqueous infused extract of this plant, whose adsorption capacity on the metal surface can promote the formation of a protective layer against corrosion. In MLTI assays, the concentration of the inhibitor in the medium was varied in the range of 100-1000 mg/L, with an increase in inhibition efficiency being noticed with the growth of this parameter. In all investigated concentrations and at a temperature of 25 ºC, it was verified that the inhibition efficiency for a 24 h immersion time was greater than for a period of only 4 h. The temperature variation in these tests did not result in any effect on the results. The highest inhibition efficiency achieved in this study was 93%, obtained in MLTI assays (24 h; 25 ºC) when adopting an inhibitor concentration of 1000 mg/L. D'ELIA *et al.* [32] worked with a higher concentration range, from 1-10 g/L, in which the same effect on the inhibition efficiency was observed, however in a milder way, since the system seemed to approach saturation. A comparison was made between two extraction methods: infusion and ultrasound. The inhibition efficiency, which in this paper was always calculated by using the EIS tests, was better in extracts obtained by ultrasound. Among the conditions tested, the highest inhibition efficiency achieved was 99.5%, given an inhibitor concentration of 10 g/L and an extraction by ultrasound. Both studies concluded that the yerba mate extract acts as a mixed inhibitor, following the Langmuir isotherm [32, 46].

The gorse (*Baccharis trimera*) is an herb widely used for medicinal purposes. D'ELIA *et al.* [32] performed the extraction of the leaves of this plant by alternative methods of infusion or ultrasound, obtaining inhibitors tested by using EIS tests, with concentrations that varied in the range of 1–10 g/L. Unlike what happened with yerba mate, the extraction method did not affect the final result. The inhibition efficiency increases with increasing inhibitor concentration, except for the 2.5 g/L condition. The highest efficiency of inhibition measured was 98.5%, referring to the inhibitor concentration of 10 g/L and extraction by infusion. The data were well represented by a Langmuir isotherm.

The blackberry is the fruit of the mulberry tree, which presents itself in nature in a great diversity of species. SILVA *et al.* [33] made the extraction by a decoction of two products that use mulberry as a raw material: leaf tea and flour. In PP tests and using an inhibitor concentration of 40% v/v, inhibition efficiencies of 91.24% were obtained for tea extract from mulberry leaves and 70.18% for an extract of blackberry flour. This fact would be explained by the higher concentration of phenolic compounds in the leaves compared to the fruits of the mulberry. The study classified the inhibitors evaluated as mixed.

Murici (*Byrsonima sericea*) is a tree native to the North and Northeast regions of Brazil. ANDRADE NETO *et al.* [35] obtained the ethanolic extract of murici leaves by maceration and tested at concentrations of 260 and 1000 ppm in an acid medium. The highest inhibition efficiency achieved was 60%, in a PP assay, for an inhibitor concentration of 1000 ppm. The phytochemical tests carried out indicated that a series of organic compounds present in the crude extract degrade with the addition of HCL 1 mol/L, leaving only the tripertenoids and saponins, among those analyzed. The authors pointed out that the inhibition mechanism is probably mixed.

The orange, mango, passion fruit and cashew fruits have peels rich in antioxidant substances such as polyphenols, carotenoids, and vitamins C and E. In the research, the compounds were extracted from the peels by an aqueous infusion or using a sequence of solvents with different polarities in a device Soxhlet type, and this choice did not significantly change the results. ROCHA and GOMES [37] obtained maximum inhibition efficiencies, at a concentration of 400 ppm, in MLTI tests (24 h; 25 ºC), the efficiencies are of 95% for orange, 97% for mango, 96% for passion fruit, and 93% for cashew. The other studies in this review that dedicated to researching these species obtained the same result concerning maximum efficiencies and, when investigating other aspects, reached the following common conclusions: the adsorption data followed the Langmuir isotherm and the inhibition efficiency increased with the increase of concentration and time, in the range of 1–24 h, in MLTI assays. ROCHA *et al.* [48] observed that the inhibition efficiency decreases with increasing temperature, in the range of 25–60 ºC, indicating a physical adsorption process. ROCHA *et al.* [51] studied orange and mango peels and pointed out an adsorption mechanism involving hydrogen bonds and substitution of water molecules on the metal surface.

The coffee tree (*Coffea sp.*) is a shrub whose seeds are known as coffee beans. Brazil is the world's largest coffee producer. The articles covered in this review presented studies on the inhibitory action of aqueous extracts of the coffee leaf. DIAS *et al.* [38] achieved an inhibition efficiency of 89.43%, in an MLTI test (24 h; 25 ºC), for an infused extract concentration of 5% v/v, demonstrating in tensile tests the attenuation of the loss of mechanical properties with the use of the inhibitor. CHAGAS *et al.* [44] found that the increase in both temperature and extraction time in a water bath contributes positively to the inhibition efficiency, with the maximum measured value being 97.05%, by using the EIS analysis, for a concentration of 16% v/v, under the following extraction conditions: 40 min and 80 ºC. SCHOLZ *et al.* [54] observed that the increase in the extract concentration, in the range of 5–15% v/v, generated a decrease in the inhibition efficiency and that, for the lowest concentration in this range, the increase in the extraction temperature in a water bath from 80 ºC also caused a reduction of this parameter, the highest value obtained was 92.24%, in an MLTI test (24 h; T_{amb}), for an inhibitor concentration of 5% v/v and an extraction temperature of 80 ºC.

The oil palm (*Elaeis guineensis* Jacq.) is a palm tree whose fruit is known as a palm oil, and palm kernel is a co-product of the production of palm oil. SANTOS *et al.* [39] crushed the palm kernel pie in a knife mill, obtaining a powder, in which functional groups were identified, such as O–H, N–H, C = O, C–N, C–O, and aromatic rings, which favored adsorption on the metal surface. The authors varied the concentration of inhibitor in the medium in the range of 0.44–1.11 g/L, verifying an increase in the inhibition efficiency with the growth of this factor, the highest value being 90%, using the EIS analysis, for an inhibitor concentration of 1.11 g/L. This work has the differential of using the powder directly, without any type of extraction and uses the technique of SVET (scanning vibrating electrode technique) that identifies anodic and cathodic areas on the metal surface.

Papaya (*Carica papaya*) has seeds rich in phenolic compounds, such as p-cumaric, caffeic and ferulic acids. TORRES *et al.* [40, 55] obtained a maximum inhibition efficiency of 94%, in an MLTI test (24 and 48 h: 25 ºC), for a papaya seed extract concentration of 400 mg/L. The authors made the following observations: adsorption is well represented by the Langmuir isotherm, the inhibition efficiency is higher at higher inhibitor concentrations, in the range of 10–1000 mg/L. In MLTI assays, the inhibition efficiency of papaya seed extract is increasing for times of up to 24 h, being stable in the 24–48 h interval, in addition to varying negatively with the temperature increase of 35–65 ºC [40, 55]. TORRES *et al.* [55] still showed that better results are possible when adopting aqueous infusion instead of acid maceration as an extraction method.

The olive tree (*Olea europaea L.*) is the tree whose fruits are olives, used for the production of olive oil. SÁ *et al.* [41] made aqueous extracts in a water bath of the olive leaves, at temperatures of 60, 80 and 100 ºC, verifying that the inhibition efficiency increases as the bath is heated. The highest value achieved for this efficiency was 92.80%, in an MLTI test (2 h; T_{amb}), corresponding to an inhibitor extracted at 100 ºC and applied in the concentration of 40 mL of extract per 100 mL of 1 mol HCl/L. The authors attributed the inhibitory activity to the presence of phenolic compounds in olive leaves such as oleuropein and hydroxytyrosol, which can be adsorbed on the metal surface.

The precious bark (*Aniba canelilla* (HBK) Mez) is a source tree of an essential oil, which in Brazil is found in the states of Amazonas and Pará. BARROS *et al.* [42] investigated the ethanolic extract of the bark of this species, obtained in a Soxhlet apparatus, in concentrations that varied in the range of 50–300 mg/L, observing an increase in the inhibition efficiency with the increase of this factor. For the concentration of 100 mg/L and the temperature of 25 ºC, they verified in the MLTI tests that the inhibition efficiency increased in the time interval of 2–24 h but decreased after 24–72 h of immersion. In addition, for the same concentration and a duration of the MLTI test of 2 h, the inhibition efficiency increased for temperatures up to 35 ºC, after which the inhibition efficiency decreased. The highest inhibition efficiency in this study was 86.9%, measured in a PP test and corresponding to a concentration of 300 mg/L. The authors confirmed that the data fitted well to the Langmuir isotherm and identified the alkaloids reticulin and N-methylclocaurine in the extract.

Boldo (*Plectranthus barbatus* Andrews) is a shrub with medicinal applications widely grown in Brazil. OLIVEIRA *et al.* [43] performed ethanolic maceration and aqueous decoction of bilberry leaves, adding the extracts obtained in varying concentrations, in the range of 1–30% v/v, in aqueous solutions of HCl 1 mol/L and H_2SO_4 0.5 mol/L. In MLTI tests, the authors found an increase in the inhibition efficiency with the increase of the extract concentration. For the same corrosive medium, the results were better when the extraction was by using the aqueous decoction and, finally, they concluded that, for the highest concentration of inhibitor, the inhibition efficiency of the aqueous extract surpassed that of the ethanolic in the presence of HCl, whereas, in the medium with H_2SO_4, the situation was reversed. The highest inhibition efficiency achieved was 96.8%, measured in an MLTI test (2 h; 25 ºC), for an inhibitor concentration of 30% v/v, extracted by ethanolic maceration, in 0.5 mol H_2SO_4/L.

The ipe-purple (*Tabebuia impetiginosa*) is a species found in all regions of Brazil and widely used in urban afforestation. SANTOS and CARDOSO [45] operated the decoction extraction of the bark of the ipe-purple and subsequently evaluated the product at concentrations ranging from 0.1-2.5 g/L in the corrosive medium, in MLTI tests (2 h; 25 ºC). There was an increase in the inhibition efficiency with the increase in inhibitor concentration, with the highest achieved value of efficiency being 89.26%. The data showed good correspondence with the Frumkin isotherm and the calculated ΔGad's value suggested a spontaneous process of physisorption. The inner bark of the ipe-purple is rich in compounds such as flavonoids and saponins, which contribute to the inhibitory action.

Garlic has a skin rich in organosulfur compounds capable of being chemisorbed on the metal surface, protecting it from corrosion. PEREIRA *et al.* [47] tested the aqueous infused extract of garlic peel by using MLTI assays and observed that the inhibition efficiency increases with the growth of the extract concentration, in the range of 10–1000 mg/L, with increasing temperature in the test, in the range of 25–55 ºC, and with the immersion time up to 48 h, instant from which the efficiency starts to decrease. The highest value of inhibition efficiency achieved was 98%, by using the EIS analysis, for an inhibitor concentration of 1000 mg/L. The process followed the Langmuir isotherm.

A commercial ground coffee (*Coffea arabic* and *Coffea canephora*), studied by TORRES *et al.* [49], was extracted conventionally in an electric coffee maker and dried in an oven before being subjected to the processes of decoction or aqueous infusion. The authors found that the increase in concentration in the range of 100–400 mg/L provided an increase in inhibition efficiency and that, for the highest concentration in this range, in MLTI assays, an increase in this factor was observed with the heating of the medium from 25 ºC up to 55 ºC, and up to 24 h, after which the inhibition efficiency dropped. It was also noticed that the results when using infusion as an extraction method were better than when using decoction, which was justified by the slightly higher content of phenolic compounds in general and by the presence of individual chlorogenic acids in the infused extract. The highest inhibition efficiency achieved in this study was 97%, in an MLTI assay (24 h; 25 ºC), for an inhibitor concentration of 400 mg/L, using infusion. SOUZA *et al.* [53]carried out the extraction by infusion of roasted and ground coffee beans (*Coffea canephora*), destining part of the product to an ultrafiltration process to separate the high molecular weight fraction (FAPM), rich in melanoidins, the substances that are probably responsible for inhibitory action of roasted coffee extract. In MLTI assays for an inhibitor concentration of 400 mg/L, the authors confirmed the positive effect of temperature, varying in the range of 35–65 ºC, on the inhibition efficiency. However, they diverged when showing its increasing value until 48 h. Higher inhibition efficiencies were achieved with FAPM compared to the infused extract, but as the concentration increased there was no longer a significant difference. The highest inhibition efficiency measured in this study was 96%, in an MLTI assay (48 h; T_{amb}), for an infused extract concentration of 400 mg/L. Both studies confirmed that the inhibition mechanism occurs through chemisorption and the data adjusted to the Langmuir isotherm [49, 53].

Castor bean (*Ricinus communis*) is a fruit from which an oil is extracted, which has application in the cosmetics and food industries, its skin being one of the residues of this process. SANTOS *et al.* [56] carried out the washing, drying, and grinding of the castor bean peel, obtaining a powder whose characterization indicated the presence of N and O atoms in different functional groups that adsorb on the metal surface. The inhibitor was tested in a concentration range of 0.44–1.77 g/L, in MLTI (120 min; T_{amb}) and EIE assays, with an increase in inhibition efficiency with the increase of this factor, reaching a maximum value of 83% by both techniques. MLTI tests have also shown a reduction in corrosion protection over time up to 24 h. The Langmuir isotherm represents well the adsorption of the inhibitor produced from the powder of the castor bean skin.

Avocado (*Persea Americana*) is a fruit whose main purpose is to feed the population, the seed being a residue. JESUS *et al.* [57] dried and crushed the avocado seed, producing a powder inhibitor that, when characterized, indicated the probable presence of aromatic rings, and N and O atoms in functional groups. The inhibitor concentration varied in the range of 0.44–1.77 g/L, being verified, in MLTI (120 min; T_{amb}) and EIE tests, that this factor positively influences the inhibition efficiency. The highest efficiency achieved was 98%, b using the EIE analysis, for an inhibitor concentration of 1.77 g/L. Protection stability over time was monitored by using EIS assays, observing a peak of inhibition after 24 h of immersion, after which a probable barrier deterioration of protection occurred. There is possibly a physical adsorption process, and the data adjusted well to the Langmuir isotherm.

Cocoa (*Theobroma cacao L.*) is a fruit that is a raw material for the production of chocolate, with the shell of its almond being a residue generated in the roasting stage of processing. CARVALHO and CAPELOSSI [50] obtained the inhibitor, in the form of a powder, by drying and crushing the cocoa bean shell. The characterization of the inhibitor identified the presence of aromatic rings and the groups O–H, C = O, C = C, C = N and C–O, which are responsible for adsorption on the metal surface. The concentration of inhibitor in the corrosive medium varied in the range of 0.44–1.77 g/L, observing that the growth of this parameter increased the corrosion efficiency. The highest efficiency achieved was 97%, in a MLTI assay (2 h; T_{amb}), for an inhibitor concentration of 1.77 g/L [58].

Guarana (Paullinia Cupana) is a vine native to the Amazon Rainforest region whose fruit is used to make powders, syrups, and soft drinks. FARIA NETO *et al.* [59] dried and crushed the guarana bark, to obtain a powder that was extracted directly in the corrosive media chosen for testing, which were solutions of HCl 1.0 mol/L and H_2SO_4 0.5 mol/L. In the MLTI assays (4 h; 25 ºC), it was observed that the inhibition efficiency increased with the growth of the inhibitor concentration in the range of 1–20 g/L in both media, becoming approximately constant for higher concentrations. In the electrochemical noise test, in which the inhibitor concentration was fixed at 20 g/L, a current proportional to that of corrosion was obtained through the load curves over time, generated from the integration of the noise data of electrochemical current. Then, this current was used to calculate the inhibition efficiency, which by this method exhibited the maximum value found in all work, being 66.47% in H_2SO_4 and 89.03% in HCl. FTIR analysis of corrosive media with inhibitor showed that some compounds of the inhibitor are oxidized when in contact with H_2SO_4, which explains their inferior performance in this medium. The data showed a good fit to the Langmuir isotherm and the value of ΔG^0ads indicated that the inhibitor is physically adsorbed to the metal surface [59].

3. CONCLUSIONS

The remarkable variety of plant species present in Brazil, whose extraction of components presents good results in inhibiting the corrosion of carbon steel in acidic medium, points to the great possibility of expanding the research of green inhibitors in the country. However, it is important to note that many of the raw materials used in the production of these inhibitors are already used for human consumption, such as coffee beans and powder, which are widely traded and exported, and boldo, whose leaves have been used in the manufacture of herbal medicines for the treatment of gastric disorders. Therefore, in order to avoid competition for raw materials with other areas and increasing the sustainability of production, it is interesting to highlight studies that use residues from other industries in the production of inhibitors, such as fruit peels, palm kernel pie and seeds, which are not used for human consumption.

In the survey carried out it was possible to observe that the studies, although most of the time they present the compounds most responsible for the inhibition of corrosion, rarely show the mechanisms, steps and chemical reactions that lead to the inhibitory effect. A complete understanding of these steps is of paramount importance to understand how the intermediate products of the reactions affect the inhibition, or even if there is the formation of harmful residues to the environment due to this process, which goes against the goal of producing green inhibitors. That said, it is believed that there should be a deeper study of the action mechanisms of inhibitory compounds by the authors, so that it goes beyond just the proposition that they act through adsorption on metal as the main mechanism. Substances derived from plants are usually classified as mixed inhibitors, which protect the metal by adsorbing on its surface, replacing water molecules. Mixed inhibitors work by reducing anodic and cathodic currents. In literature the deeper characterization of green inhibitors and the identification of active ingredients of natural compounds are topics to be better elucidated.

Finally, since the green inhibitors included in this review are in an early stage of development, the forms of extraction, the best plant species and the inhibition efficiency are being studied on a laboratory scale, and experiments on a pilot or industrial scale will be necessary. As research progresses, it is important to develop these tests, in addition to assessing the costs related to the production of green inhibitors, as well as their durability in practical environments and possible reuse.

4. ACKNOWLEDGMENTS

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