Arabian Journal of Chemical and Environmental Research

Vol. 04 Issue 1 (2017) 46–62

An overview on plant extracts as potential green corrosion inhibitors for metals and alloys

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Received 15 Sept 2017, Revised 22 Dec 2017, Accepted 27 Dec 2017

Abstract

Metal corrosion is a phenomenon that has existed for a long time. Considering the economic losses caused by this phenomenon; different protection techniques are used, such as corrosion inhibitors which can be harmful to the environment. Reduced environmental risk, lower cost, wide spread availability and high corrosion inhibition effectiveness make the plant extracts as suitable candidates to replace the expensive and toxic traditional synthetic corrosion inhibitors. Literature survey reveals that different extracts issued from leaf, root, stem, bark, pulp, fruit, etc. have been effectively proposed as sustainable inhibitors for the corrosion of different metallic materials. This review article aims to demonstrate the use of new corrosion inhibitors based on extract or an oil natural plant, which is biodegradable and eco-friendly substances, called "green inhibitors" and describes the collection of published work that has been carried out on the topic: plant extract as corrosion inhibitors for metals and alloys in aggressive aqueous solutions. The inhibitory action of these natural mixtures is essentially due to the presence of polyphenols, catechol and gallic tannins, flavonoids, saponins, terpenoids …

Keywords: corrosion, green inhibitor, metals, biodegradable

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ISSN : 2458-6544

1. Introduction

Metals are materials used in all fields, domestic and even industrial fields. They are exposed to degradation under the influence of a surrounding environment such as the use of acid solutions. These solutions are widely used in various fields such as pickling or acid cleaning, stimulation of oil wells, and the removal of localized deposits (scale, rust, bacterial deposits) [1].

The economic losses caused by corrosion are significant, for example according to the World Corrosion Organization WTO: « It is estimated that each year a quarter of steel production is destroyed by corrosion, which corresponds approximately to 150 million tons / year or even 5 tons / second». This damage has led to the use of corrosion inhibitors as a means of protection; some of them are toxic to the environment, such as chromate. This has allowed researchers to study new inhibitory substances extracted from plants called "green inhibitors".

2. Corrosion inhibitor

National Association of Corrosion Engineers (NACE) describes an inhibitor as follow: «An inhibitor is a chemical substance which, when added in small concentrations to an environment, effectively checks, decreases, or prevents the reaction of the metal with the environment » [2].

3. Inhibitor proprieties

In general, an inhibitor should: [3]

- Lower the corrosion rate of a metal, without affecting its physico-chemical characteristics.
- Have to be effective at low concentrations.
- Have to be compatible with non-toxicity standards.
- Have to be inexpensive

4. Inhibitor class

There are several possibilities for classifying inhibitors [4] :

- Either from the nature of the products (organic or mineral inhibitors).
- Either from their electrochemical mechanism of action (cathodic inhibitors, anodic or mixed).
- Either from their interface mechanisms and principles of action (adsorption to the surface metal and / or formation of a protective film).
- Either from the application domain.

The corrosion inhibitors mostly used are from synthetic origin or originate from organic compounds with hetero-atoms, such as nitrogen, sulfur, phosphorus or oxygen in their aromatic system or in their carbon chain; but most of these substances are toxic to humans and the environment. Over time and through centuries, medicinal plants have proven their use as a preventive or curative of diseases thanks to their very varied chemical composition [5].

5. Green inhibitor

The toxicity drawbacks caused by synthetic inhibitors have led to search for natural substances which may be effective as metal corrosion inhibitors.

The utilization of herbal compounds had a great interest; they are biodegradable, ecological and available. Green inhibitors are the different compounds obtained by plants, such as flavonoids and tannins [5]. The organic green corrosion inhibitor active groups are phytochemical constituents which have functional groups with N, O, S, P, or heteroatoms via which they are attached on the metal surface [6]. Several researchers have studied the inhibitory effectiveness of natural extracts obtained from different parts of plants (stems, leaves and seeds), in different corrosive mediums.

6. Use of green inhibitors in different corrosive mediums

The richness of the Algerian flora has enabled researchers to study the inhibitory efficiency of certain plant compounds. In 2012, Selles *& al;* tested the aqueous extract of *Anacyclus pyrethrum* L. (Asteraceae) (leaves and stems) on mild steel corrosion, in 0.5M (H_2SO_4) medium. The result showed 87% of inhibition efficiency [7]. Moreover, polarization curves show that the action of various components of extract acted as anodic type inhibitors [8].

Another compound of plants extract was studied by Benali& al; it was the tannin extract of *Chamaerops Humilis*, tested on mild steel in 0.5M H_2SO_4 solution, the inhibition efficiency was important, and reached 86.51% with 100mg/L of green inhibitor [9]. The valorisation of the methanol extracts of Chamaerops humilis L. of Benslimane, Morocco showed that near a significant antioxidant activity leading to 81% of inhibition, also exhibited the protection of steel corrosion [10]. The addition of *Chamaerops Humilis* can effectively decrease the reinforcement steel corrosion, enhance the stability of the film formed in alkaline solution and lead to the formation of a more compact protective layer on the surface [11]. These various actions are essentially due to its phytochemical screening rich in Polyphenols, Catechol and Gallic Tannins, Flavonoids, Saponins, Terpenoids, Anthracenosides and Cardiac glycosides [9-13].

Mentha Pulegium widely growing in Mediterranean was also studied at various applications [14-18]. Then, *Mentha Pulegium* extract was studied as green corrosion inhibitor for steel in 1M HCl medium, by Khadraoui *& al;* the result showed that this plant extract had significant effect as inhibitor with 88% of inhibitor efficiency [19]. The pulegone is extracted starting from oil of Pennyroyal Mint (Mentha pulegium) and its pulegone oxide prepared by oxidation of pulegone, both were tested as inhibitors steel corrosion in molar hydrochloric acid using weight loss measurements, potentiodynamic polarization, and impedance spectroscopy (EIS) methods. The inhibition efficiency was found to increase with the inhibitor content to attain 81 and 75% at 5g/L for pulegone and pulegone oxide, respectively [20, 21]. The corrosion inhibition characteristics of essential oil of *Mentha pulegium* leaves have been studied as a green inhibitor of corrosion of aluminum in 2 M phosphoric acid using potentiodynamic polarization and electrochemical impedance spectroscopy [22].

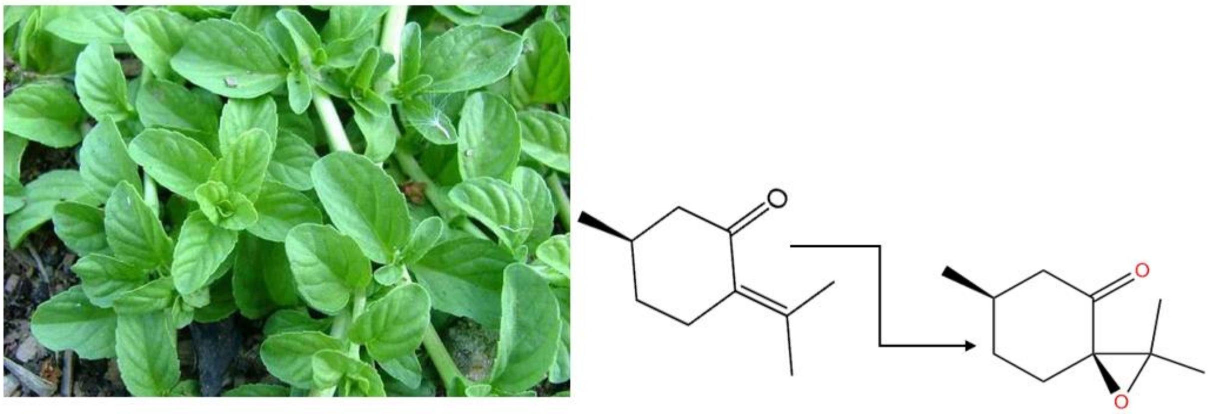


Figure 1 : *Mentha Pulegium* to pulegone and of pulegone oxide

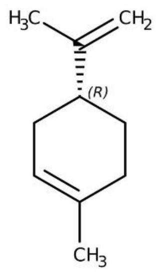
In 2016, Khadraoui & al. studied the inhibitor effect of *Thymus Algeriensis* extract on 2024 Aluminium alloy. The result reached 78.7% of inhibitor efficiency in 1M HCl medium [23]. *Thymus Algeriensis* oil was studied to protect steel corrosion in acid solution besides its chemical composition composed byborneol (28%), camphene (20.9%) and camphre (15.7%) as the major compounds [24]. TA extract and oil were studied as corrosion inhibitors, antioxidant or medical uses as anti-tumor activity [25-28]. Aqueous Extract of *Zygophyllum album L* Leaves was studied by Derfouf *et al.* in 2017. This green inhibitor was tested on carbon steel corrosion in 1M HCl solution; the inhibition efficiency attained 89% [29]. Extract of *Zygophyllum-album* was also investigated as corrosion inhibitor of mild steel in 1 M H_2SO_4 using conventional weight loss, electrochemical polarizations, electrochemical impedance spectroscopy and scanning electron microscopic techniques [30]. *Zygophyllum* (Z. album) is one of the common species growing in North Africa from Morocco to Egypt. Different specific amount of the chemical constituents of terpene, flavonoids, phenolic, tannin, saponine, alkaloids and reducing sugars were identified. The higher content of flavonoids, phenolic and antioxidants was also recorded. These findings were sufficient to justify the application of *Z.album* as Anti hyperglycemic, and antioxidant activities [31-35].

Eugenol (Eug) and its derivative acetyleugenol (AcEug) were extracted from the nail of clove to be tested on the corrosion of steel in molar hydrochloric acid has been studied using weight loss measurements, electrochemical polarisation and EIS methods. The inhibition efficiency was found to increase with acetyl eugenol content to attain 91% at 0.1737 g/L. Eugenol compounds act as mixed type inhibitors [36-38].

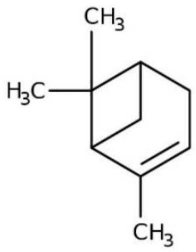
Corrosion inhibition effect of black pepper (BP) extract and its piperine isolated from BP on corrosion of C38 steel in 1 M HCl solution was investigated by weight loss method. Piperine was isolated by ethanol in yield 6 from ground BP. Results obtained from weight loss measurements indicate that the natural compounds tested exhibit higher efficiency exceeding 95% at 2g/L. It was studied by Dahmani *& al* [39]. Lahhit *& al;* used Essential oil from fennel (Foeniculum vulgare) as green inhibitor for carbon steel in 1M HCl medium. The global rate of corrosion estimated by weight loss measurements confirms the above results. The inhibition efficiency attains a maximum of 76 % at 3 mL/L, but decreases with the rise of temperature [40].

Photo 1 : dried fennel (Foeniculum vulgare)

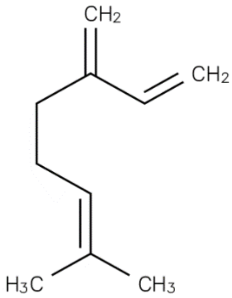
The analysis of FM oil, obtained by hydrodistillation, using Gas Chromatography (GC) and Gas Chromatography/Mass Spectrometry (GC/MS) is a good tool to explain how natural oil can act on the metal surface. The analysis allowed the identification of 21 components which accounted for 96.6% of the total weight. Effectively, analysis showed that the major components were limonene, pinene, myrcene, fenchone... The adsorption of these molecules could take place via interaction with the vacant d-orbitals of iron atoms via a competitive or concurrently action by a synergistic called intermolecular effect [40-43].



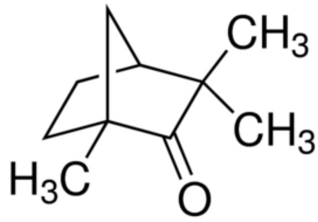
limonene



pinene



myrcene



fenchone

These molecules having double bonds and Oxygen atom, can be the active centers of adsorption on the metal surface to form a barrier against aggressive ions as H^+ or Cl^- as well as dissolved oxygen.

Ben Hmamou & al; studied carbon steel (C38) corrosion in 1M HCl, they used a plant extract of Chamomile as inhibitor; the inhibition efficiency reached 88 % with 7g/L of extract [44].

The major compounds of Chamomile were determined by Abdel-Gaber et al. in 2006 (Figure 2) [45]. To explain the role of each compound on the inhibitory effect, theoretical calculations suggested that Quercetin showed the most inhibition efficiency as compared to the other compounds, because they have the low E_{LUMO}. Therefore, it is expected that inhibitors can form a strong interaction with Fe to act as cathodic inhibitors. All inhibitors show spontaneous energy and suggest that Chamomile extract have inhibitive action which is mainly from Quercetin [46].

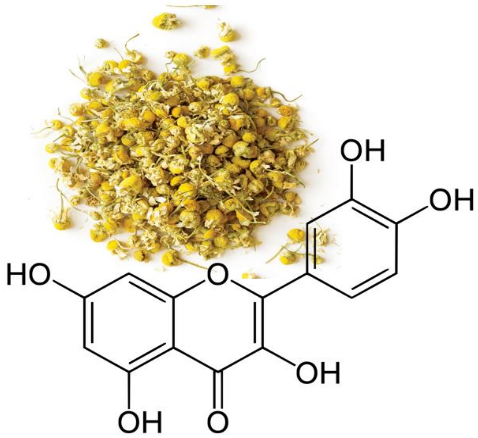


Photo 2: dried Chamomile and Quercetin as major compound

Quercetin is known for its antioxidant activity in radical scavenging and anti-allergic properties characterized by stimulation of immune system, antiviral activity, inhibition of histamine release, decrease in pro-inflammatory cytokines, leukotrienes creation, and suppresses interleukin IL-4 production [45]. Quercetin, a common flavonol, has been shown to be an effective antioxidant in several in vitro systems, including oxygen radical absorbance capacity, ferric reducing antioxidant power [50-54].

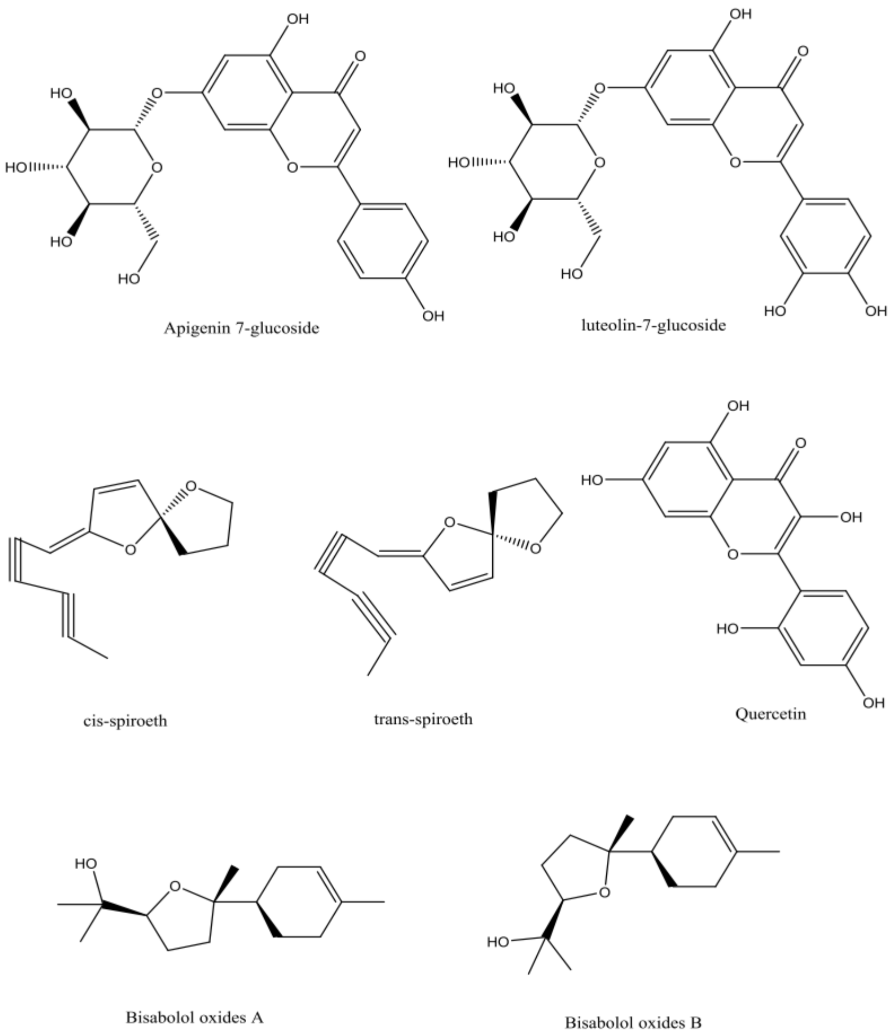


Figure 2: Structures of main chemical constituents of Chamomile [45-49]

Also, limonene extract was studied by Chaieb *et al.* [55]. It was extracted from citrus and orange fruit, the effectiveness of inhibition increased with the increase in concentration of limonene and this exceeded 72% at 0.22 g/L. The inhibition efficiency is temperature independent in the temperature range of 298-328 K.

In 2014, Khadraoui *& al;* tested the *Ruta Chalepensis L.* Oil on steel in acid medium, the result showed 77% of corrosion inhibition efficiency [56]. Also, a study on Oil of *Mentha Rotundifolia* showed that it had an inhibitory action for carbon steel in acid medium; it shows 89% of efficiency reached for 2mL/L of the green inhibitor [57]. Moreover, Khadraoui *& al.* in 2016, studied the inhibitory action of *Mentha Pulgium* Oil. It was tested on carbon steel in 1M HCl. The result showed 81% of efficiency achieved with 3 mL/L of the green inhibitor oil at 298 K. It was concluded that this green inhibitor is effective and eco-friendly corrosion inhibitor [58].

In 2016, The corrosion and inhibition behaviours of mild steel in hydrochloric, sulphuric, and nitric acid in the presence of *Cyamopsis tetragonoloba* (guar gum) have been studied using the weight loss, gas chromatography mass spectrometry (GC-MS), and thermometric techniques.

The maximum efficiency (90.56%) has been observed at highest inhibitor concentration 1000 ppm at 72 h at temperature 304 K in 1N HCl, 87.4% for 24 h on 1000 ppm in 1N H_2SO_4, and 73.67% for 24 h on 250 ppm in 1N HNO_3 medium. The worth of inhibition falling with high temperature represents physical adsorption mechanism. This is indicating that this green corrosion inhibitor can be efficient for stainless steel [59].

On the other hand, there are several leaf extracts that have been reported as green and efficient corrosion inhibitors for mild steel in hydrochloric as well as in sulphuric acid solutions. Some of the common and important leaf extracts investigated as inhibitors for mild steel in boththe acidic solutions are Ginkgo (*Chinesename Yingxing*) [60], *Occimum viridis* (OV) [61], *Dodonaea viscose* [62], *Trifoliate fenugreek* [63], *Kigelia pinnata* [64], *Citrus aurantiifolia* [65=52], *Jatropha Curcas* [66], *Hibiscus sabdariffa* [67], *Gnetum africanum*, *Gongronemalati folium*, *Chromolena odaratum* [68], *Phyllanthus amarus* [69], lupine (*Lupinousalbus*) [70]. The following table recaps some other examples for the use of green corrosion inhibitors in the last decade:

Reference	Metal	Inhibitor	Medium	Inhibition efficiency (%)	Year
[71]	C38 steel	*Rosmarinus officinalis* oil	0.5 M H_2SO_4	61%	2010
[72]	Cold Rolled Steel	*Jasminum nudiflorum* Lindl	1M HCl	96,3%	2010
[73]	Zinc	*Aloe vera* (Leaf extract)	2M HCl	67%	2010
[74]	Acier C38	*Oxandraas beckii*	1M HCl	91%	2011
[75]	Aluminium	*Gossipium Hirsutum* L.	1M HCl	92%	2011
[76]	Iron	Clove Oil	0.5M HCl	98 %	2012
[77]	C38 Steel	*Verbena* Extract	1 M HCl	85.3%	2012
[78]	Carbon steel	BAMBOO	0.1M H_3PO_4	95.7%	2014
[79]	Brass	Eugenol Clove Essential Oil	3% NaCl	82,1%	2016
[80]	Stainless Steel	*Thymus Vulgari*	1M HCl	62.15%	2017
[81]	Aluminium	*Jasminum nudiflorum* Lindl.	1M HCl	93.6 %	2012

7. Phenomenon of synergism

Obviously, each plant extract contains several active phytochemicals/constituents that can adsorb at the surface of metal and electrolyte. However, addition of some inorganic salts such as KCl, KBr and KI in the corrosive medium can add the protection ability of plant extract towards metallic dissolution. Oguzie investigated the inhibition property of *Occimum viridis* (OV) extract on mild steel in 2 M HCl and 1M at 30 and 60°C temperatures [61]. From the results derived in the study several interesting things were observed. In hydrochloric acidic medium, addition of KCl decreases the protection power of the plant extract. Whereas, addition of KB rand KI particularly at lower temperature (30°C) protection efficiency of the extract increased significantly. In sulphuric acid medium, addition of KCl, KBr and KI enhances the protection ability of the extract. In both electrolytic media, the effectiveness of halides obeyed the order: KI > KBr > KCl. He observed similar observation for the inhibition effect of *Hibiscus sabdariffa* on mild steel corrosion in 2M HCl and 1M H_2SO_4 [82].

Khadraoui and co-workers described the synergistic effect of KI on the inhibition efficiency of *M. pulegium* in 1M HCl electrolytic solution. Using hydrogen weight loss method, these authors observed that addition of KI at all studied concentration of the extract causes noteworthy increase in the protection ability at both studied temperatures (308-338 K). The *M. pulegium* extract at 33 %v/v concentration showed inhibition efficiency of 84.34%. Addition of KI at 3mM concentration in the same solution causes increase in the inhibition efficiency up to 90.59% [83].

Conclusion

From the various studies made by the researchers, it can be concluded that herbal inhibitors have given satisfactory results, and they are effective in inhibiting the corrosion of metals. Natural Plants used as extracts or oils can be used safely as corrosion inhibitors for metals and alloys as well as in cement concrete to limit damage of aggressivity against metals. Rich in infinite compounds at different concentrations play an essential role in adsorption at the metallic surface. Thus, they are from a renewable, ecological, biodegradable, and inexpensive source, so they represent a means of preserving the environment. Over time and through centuries, medicinal plants have proven their use as a preventive or curative of diseases thanks to their very varied chemical composition.

Conflict of Interest

The authors declare that the research was conducted in the absence of any commercial or financial relationships that could be construed as a potential conflict of interest.

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